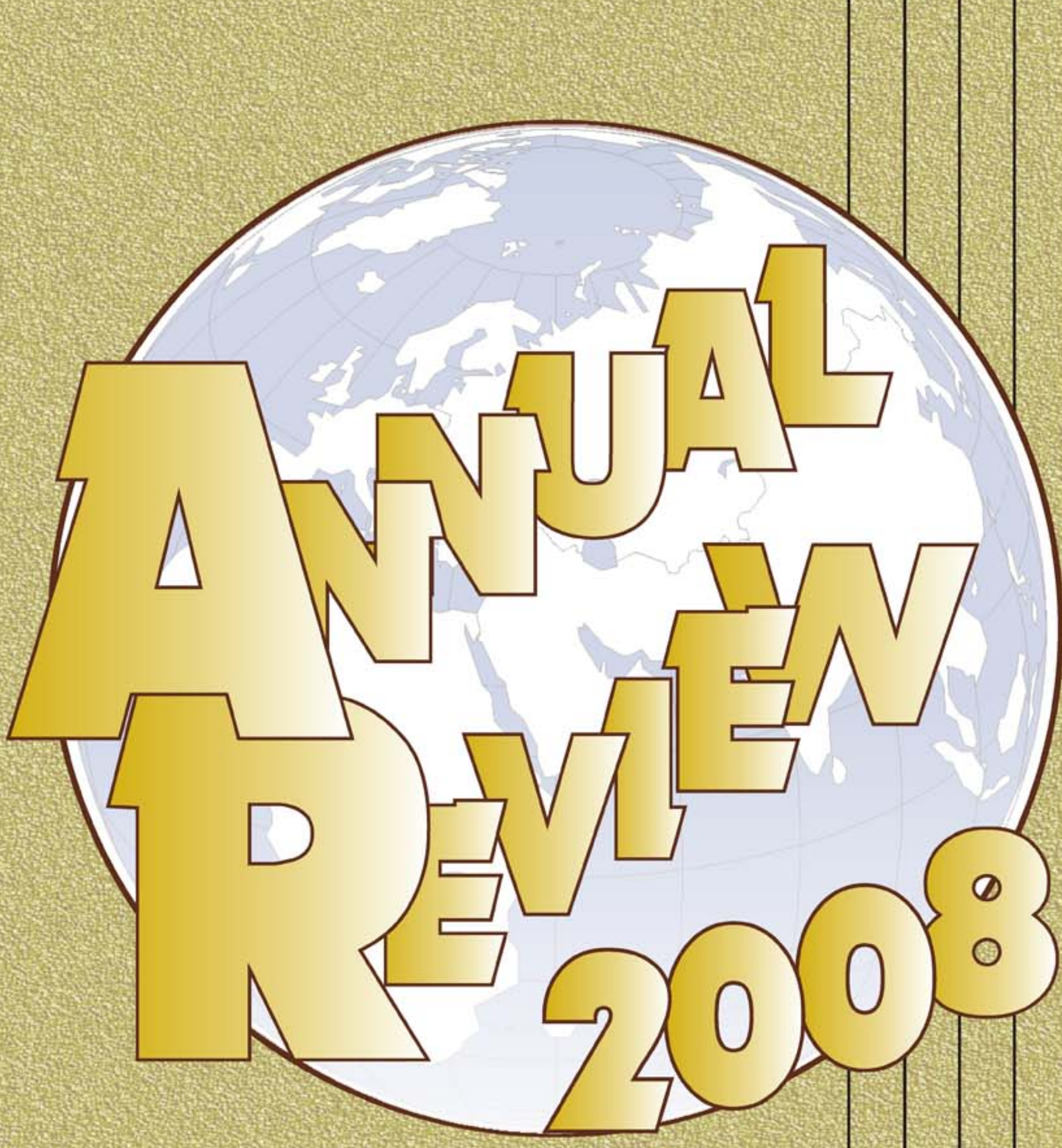
BORESKOV INSTITUTE OF CATALYSIS

SIBERIAN BRANCH OF THE RUSSIAN ACADEMY OF SCIENCES

50



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of Activities in Basic Research Areas
2007

BOREKOV INSTITUTE OF CATALYSIS

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Dear Colleagues,

This issue of the Annual Review of the Boreskov Institute of Catalysis, BIC, is appearing at the very important time for the Institute: the Institute celebrates at the year 2008 its 50-th anniversary. However, the issue covers the BIC's activity in the main fields of its academic and R&D activities during the year 2007 and reflects already more than the ten-year experience of the Institute in publishing such reviews.

The Boreskov Institute of Catalysis, or BIC, is well known to experts in both academic and industrial catalysis not only in Russia and the fSU countries, but also in many western and oriental countries. The BIC has been founded at the year 1958 in order to improve the transfer of scientific knowledge of the Academy of Science for the former USSR to the chemical industry. Since 1960's it has remained the largest Russian chemical research institution in the Asian part of Russia and had many deep and stable traditions in providing basic and applied research in nearly all fields of catalysis. The interests of BIC are spreading from the sophisticated areas of purely academic research to the real applied problems, including development of commercial catalysts and catalytic technologies.

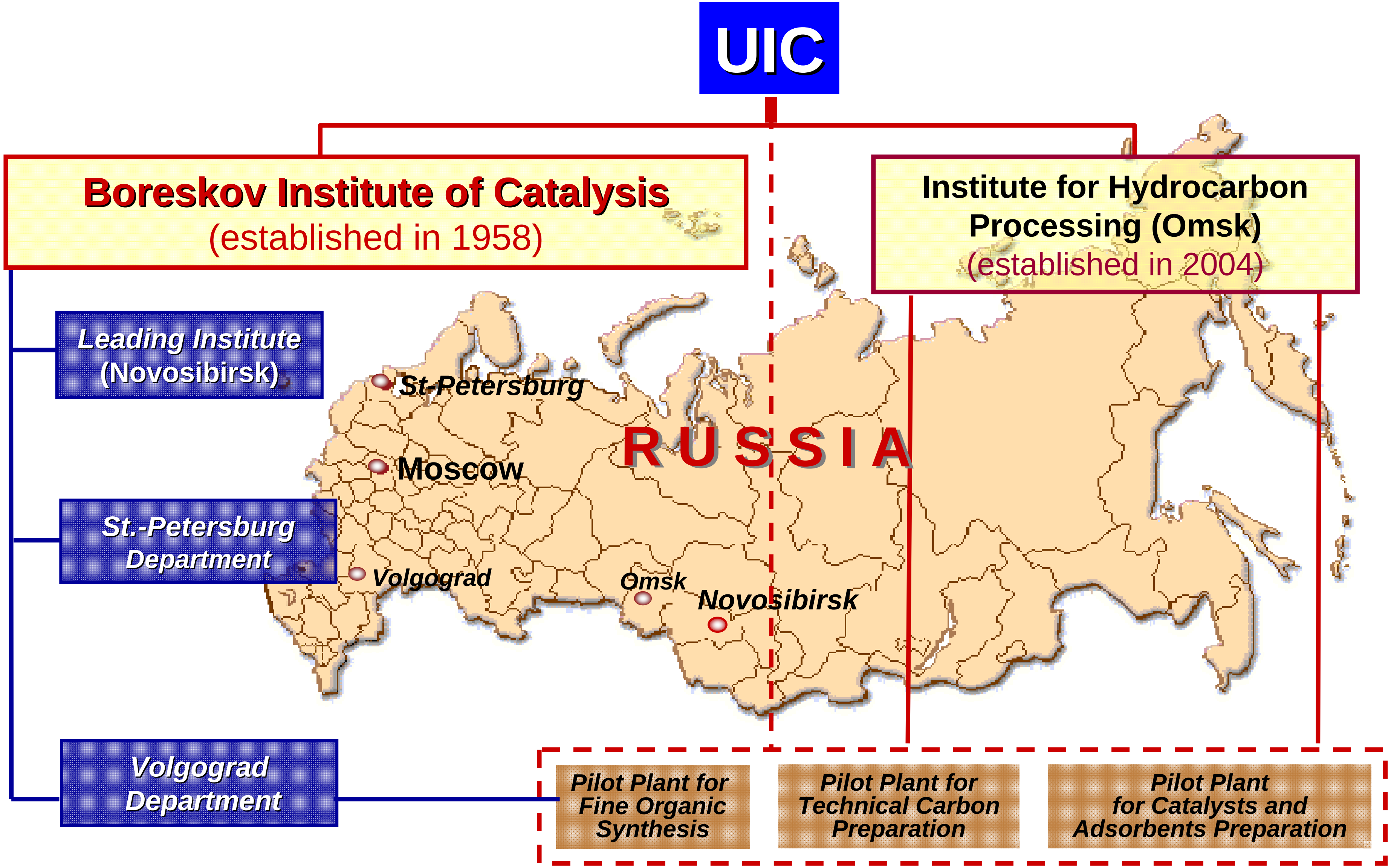
The current features of BIC reflect Russia entering into the market economy. BIC belongs to the Siberian branch of the Russian Academy of Sciences. However, according to new realities of Russia, a lot deal of activity of BIC is devoted not only to basic research, but also to numerous industrially oriented topics. The intensity and quality of the research and engineering activity at the Institute became at the new period of the Russian history even higher as before. Fortunately, the Institute gained in research in catalysis fundamentals, thus continuing the old traditions originated from the beginning of the Institute due to a very powerful starting impulse of its creator and first director Academician Georgii K. Boreskov, as well as the master ideas of the second director Academician Kirill I. Zamaraev. Hope, the readers of this Review will feel that themselves, when comparing the materials of this and previous issues of the Annual Review.

The year 2007, the same as all previous years, was very important for the life of BIC in many aspects. The main practical results of the year 2007 were successful industrial approach of the first genuine Russian catalyst for the deep desulfurization of diesel fuel, as well as successful industrial tests of a palladium based catalyst for the large scale hydrogenation of plant oils. The largest success for BIC is that the catalyst for the diesel fuel desulfurization has been developed in unusual short terms: starting from the nearly unknown for the BIC field of the catalysis, it took only half a year in order to transfer the technology to the industrial catalyst manufacture. Now, this catalyst is under exploiting at a big Russian oil refinery and demonstrates the parameters similar to those of the best known catalysts of the same family. The strategic collaboration with the Russian industries is nowadays, undoubtedly, the very important feature of the scientific activity of the Boreskov Institute, as well as of many other academic institutes of RAS.

Since 1998 the Institute has been united with few other R&D chemical institutions of the Siberian Branch of the Russian Academy of Sciences in the frame of a new R&D association which was called the United Institute of Catalysis (UIC). According to the UIC bylaw, the director of the Boreskov Institute of Catalysis has to be simultaneously the general director of the UIC. In 2004 there were some rearrangements in the UIC structure, since the Omsk Department of the BIC has separated from BIC as a new legal entity, the Institute of Hydrocarbons Processing (IHP), and the Volgograd Scientific center with a large flexible pilot facilities for fine organic synthesis has joined BIC as its Volgograd Department. Thus, now UIC consists of two large legal entities: BIC and IHP. The facilities of the UIC include three flexible and powerful semi-industrial plants capable of manufacturing the first industrial batches of various catalysts and specialty chemicals. Thus, BIC together with UIC is now one of the most powerful Russian specialized R&D organizations in the industry-oriented chemistry with unique facilities for scaling up numerous innovations for that industry. Indeed, the current management of the Institute realizes the existing problems of the large R&D structure operation and tries to take care of the maintenance of high R&D activity and productivity of BIC and UIC in both fundamental and applied areas as the goal of the highest priority.

Hope, the current issue of the Annual Review of the Boreskov Institute of Catalysis will succeed in exhibiting many sides of the Institute potentialities in basic research on catalysis, catalysts and catalytic technologies. As to the activity of the Institute in the applied areas, which is now much more active than ever in the past, we publish special leaflets. Also, we are inviting the readers to visit the BIC’s website **www.catalysis.ru**.

Valentin N. Parmon



UIC

Boreskov Institute of Catalysis
(established in 1958)

***Leading Institute* (Novosibirsk)**

St.-Petersburg Department

Volgograd Department

Institute for Hydrocarbon Processing (Omsk)
(established in 2004)

Pilot Plant for Fine Organic Synthesis

Pilot Plant for Technical Carbon Preparation

Pilot Plant for Catalysts and Adsorbents Preparation

BORESKOV INSTITUTE OF CATALYSIS OF THE SIBERIAN BRANCH OF THE RUSSIAN ACADEMY OF SCIENCES

The Boreskov Institute of Catalysis of the Siberian Branch of the Russian Academy of Sciences (BIC) still remains the largest specialized institute working in the field of catalysis not only at the territory of Asian part of Russia, but in the world too. It is the only institution in the country fully specialized on the problems of catalysis and encompasses all its aspects from fundamental and theoretical to industrial mastering of new catalysts.

The Institute was founded in 1958. Initiator of the Institute and its permanent director up to 1984 was Academician Georgii K. Boreskov (1907-1984), an eminent scientist with international recognition, an outstanding physical chemist and chemical technologist. In 1992 the Institute of Catalysis was named after Academician G.K. Boreskov.

In 1976 Professor Kirill I. Zamaraev has been invited by Academician G.K. Boreskov to join the Institute of Catalysis as a Deputy Director. Kirill Ilyich has come to Novosibirsk at early 1977 together with a large team of young chemical physicists, thus largely widening the studies of elementary catalytic reactions on molecular-atomic level through application of modern physical and kinetic instrumental methods of investigation.

In 1984 Professor Zamaraev became the Director of the Institute of Catalysis, and since that time he was the real leader of all research and development activity of this huge (up to 1000 people in staff) Institute. K.I. Zamaraev, the former President of the International Union of Pure and Applied Chemistry, Foreign Fellow of the Indian National Science Academy and of the Korean Academy of Science and Technology, holder of the Silver Medal of the Royal Chemical Society of Great Britain as a centenary lecture, has passed away in the prime of his brilliant scientific career on 26 June 1996.

In 1995 Professor Valentin N. Parmon, prominent specialist in the field of catalysis, chemical kinetics and photocatalysis, headed the third generation of the directorate of the Institute.

Since 1998 the Institute has been united with some other R&D chemical institutions of the Siberian Branch of the Russian Academy of Sciences in the frame of a new R&D association: the United Institute of Catalysis headed by the director of the Boreskov Institute of Catalysis. Now the United Institute of Catalysis (UIC) includes the Boreskov Institute of Catalysis with its St. Petersburg Department, Volgograd Department and the Institute of Hydrocarbons Processing in Omsk.

STRUCTURE OF THE INSTITUTE

The Boreskov Institute of Catalysis (BIC) unites more than 1000 employees, among its 400 researchers being 1 academician, 2 Correspondidng Members RAS, more than 60 Drs.Sci. and 200 PhD's. The Institute structure comprises 8 research departments, the Department of Applied Catalysis and the Information Center. Service, production and management departments maintain creative activities of the all the Institute personnel.

The Institute is a unique teamwork of experts in various fields of science and engineering who are capable of solving any catalytic problems from fundamentals to design of industrial catalysts and processes.

The Center for Catalyst Characterization and Testing has a bank of catalyst samples, supports and adsorbents, a set of State Standard Samples to calibrate and certify instruments for measurements of specific surface areas of various dispersed and porous materials.

The main directions of its activity are:

- Complex investigation and characterization of the catalysts by a set of modern physicochemical methods.
- Joint scientific projects with foreign firms and organizations on studying certain catalytic processes and catalysts for the purpose of their improvement.
- Development of modern standard experimental methods and equipment for catalyst studies and tests. Design of catalysts and materials with certificated properties.

The successful research activities are based on the overall access to the latest and reliable scientific data. There is the **Information and Library Center** in the Institute, and the library is one of the best scientific libraries in Siberia.

Department of Applied Catalysis with a complex bench of adoption and testing of processes and catalysts provides commercialization of new developments in shortest time.

The **Scientific Organization Department** deals with scientific events such as conferences, seminars, schools, meetings, presentations of various companies and equipment producers, discussions. There is a museum where the information on catalytic science is collected.

Advertising and Exhibits Department of the Institute is active in competence, perfect service in the field of organization of exhibitions of the new inventions and successive achievements of the Institute.

STRUCTURE OF THE INSTITUTE

Directorate:
I.A. Kamolkin, S.E. Glaznev,
O.N. Martyanov, A.S. Noskov,
B.S. Bal'zhinimaev, V.N. Parmon,
V.I. Bukhtiyarov, V.N. Novikov

DIRECTOR

Directorate

Scientific Council

Research Departments

DEPARTMENT OF PHYSICOCHEMICAL METHODS FOR CATALYST INVESTIGATION
Head: Prof. Valerii I. Bukhtiyarov

DEPARTMENT OF NONTRADITIONAL CATALYTIC PROCESSES AND TECHNOLOGIES
Head: Acad. Valentin N. Parmon

DEPARTMENT OF HETEROGENEOUS CATALYSIS
Head: Prof. Aleksandr A. Khassin

DEPARTMENT OF EXPLORATORY AND APPLIED INVESTIGATIONS
Center for Catalyst Characterization and Testing
Head: Prof. Bair S. Bal'zhinimaev

DEPARTMENT OF CATALYTIC PROCESS ENGINEERING
Head: Prof. Alexander S. Noskov

DEPARTMENT OF CATALYTIC PROCESSES OF FINE ORGANIC AND BIOORGANIC SYNTHESIS
Head: Prof. Zinaida P. Pai

St. PETERSBURG DEPARTMENT OF THE BORESKOV INSTITUTE OF CATALYSIS
Head: Prof. Sergey S. Ivanchev

VOLGOGRAD DEPARTMENT OF THE BORESKOV INSTITUTE OF CATALYSIS
Scientific Director Dr. Aleksandr M. Beskopylnyi

Scientific-Supportive Units

Manufacturing Facilities

Administration and Services

DEPARTMENT OF PHYSICOCHEMICAL METHODS FOR CATALYSTS INVESTIGATION

Head of the Department Prof. Valerii I. Bukhtiyarov

Surface Science Laboratory
Head: Prof. Valerii I. Bukhtiyarov

Laboratory of Analytical Chemistry
Head: Prof. Vladimir N. Sidelnikov

Laboratory of Investigation of the Mechanisms of Catalytic Reactions
Head: Prof. Eugenii P. Talsi

Laboratory of Catalyst Texture Studies
Head: Dr. Maxim S. Melgunov

Laboratory of Quantum Chemistry
Head: Dr. Igor L. Zilberberg

Laboratory of Spectral Methods
Head: Prof. Dimitrii I. Kochubey

Laboratory of Structural Methods
Head: Prof. Sergey V. Tsybulya

Group of Low Temperature Catalysis by Metals
Head: Prof. Vladimir V. Gorodetskii

Group of Solid-State NMR Spectroscopy
Head: Prof. Olga B. Lapina

Group of Surface Compounds Synthesis

Head: Dr. Vladimir L. Kuznetsov

Group of NMR Spectroscopy for Catalytic Hydrocarbon Conversion

Head: Prof. Alexander G. Stepanov

DEPARTMENT OF NONTRADITIONAL CATALYTIC PROCESSES

Head of the Department Academician Valentin N. Parmon

Laboratory of Catalytic Methods of Solar Energy Conversion
Head: Academician Valentin N. Parmon

Laboratory of Adsorption
Head: Dr. Oleg N. Martyanov

Laboratory of Hydride Compounds Studying
Head: Prof. Valentina I. Simagina

Laboratory of Energy Accumulating Processes and Materials
Head: Prof. Yuriy I. Aristov

Laboratory of Catalytic Processes for Desulfurization
Head: Dr. Olga N. Kovalenko

Group of Hydrogenation Processes
Head: Dr. Galina A. Bukhtiyarova

Group of Adsorption-Catalytic Processes for Fuel Cells
Head: Dr. Alexey G. Okunev

Group of Aerosol Catalysis
Head: Dr. Valerii N. Snytnikov

Group of Biocatalysis
Head: Prof. Galina A. Kovalenko

Group of Catalysts on Carbon Supports
Head: Dr. Irina L. Simakova

Group of Small Angle Scattering
Head: Prof. Fedor V. Tuzikov

Group of Metal Complex Catalysis
Head: Prof. Nina I. Kuznetsova

Group of Sulfide Catalysts
Head: Prof. Anatolii N. Startsev

Group of Photocatalysis on Semiconductors
Head: Dr. Alexandr V. Vorontsov

Group of Energy-Chemical Processes and Technologies
Head: Prof. Vladimir I. Anikeev

DEPARTMENT OF HETEROGENEOUS CATALYSIS

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Laboratory of Catalytic Conversion of Carbon Oxides
Head: Prof. Aleksandr A. Khassin

Laboratory of Catalytic Processes in Fuel Cells
Head: Prof. Vladimir A. Sobyanin

Laboratory of Heterogeneous Selective Oxidation
Head: Dr. Vladimir I. Sobolev

Laboratory of Catalysts for Deep Oxidation
Head: Prof. Vladislav A. Sadykov

Laboratory of Ecological Catalysis
Head: Prof. Zinfer R. Ismagilov

Laboratory of Dehydrogenation
Head: Prof. Victor V. Molchanov

Laboratory of Oxidative Catalysis over Zeolites
Head: Prof. Gennadii I. Panov

Laboratory of Catalysts Preparation
Head: Prof. Aleksandra S. Ivanova

Group of Noble Metal Based Heterogeneous Catalysts
Head: Prof. Anatolii V. Romanenko

Group of Reactions of Oxidation on Metals
Head: Prof. Aleksandr V. Khasin

Group of Synthesis of Nanodispersed Materials
Head: Prof. Oleg P. Krivoruchko

DEPARTMENT OF CATALYTIC PROCESS ENGINEERING

Head of the Department Prof. Aleksandr S. Noskov

Laboratory of Unsteady-State Catalytic Methods for Gas Purification
Head: Prof. Aleksandr S. Noskov

Laboratory of Multiphase Processes Modeling
Head: Prof. Valerii A. Kirillov

Laboratory of Catalytic Hydrocarbon Conversion
Head: Prof. Gennadii V. Echevsky

Laboratory of Catalytic Polymerization
Head: Prof. Vladimir A. Zakharov

Sector of Complex Engineering Projects
Head: Dr. Victor A. Chumachenko

Group of Dynamics of Catalytic Processes
Head: Prof. Andrey N. Zagoruiko

Group of Catalytic Technologies for Carbon Materials Synthesis
Head: Dr. Vadim A. Yakovlev

Group of Kinetics of Catalytic Processes
Head: Dr. Sergei I. Reshetnikov

Group of Mathematical Methods for Catalytic and Adsorption Processes Modeling
Head: Dr. Natalia A. Chumakova

Group of Development and Optimization of Catalytic Processes with Catalyst Deactivation
Head: Dr. Vitalii N. Kashkin

Group of Processes in Fixed Catalyst Layer
Head: Dr. Ilya A. Zolotarskii

DEPARTMENT OF EXPLORATORY AND APPLIED INVESTIGATIONS

Head of the Department Prof. Bair S. Bal'zhinimaev

Center for Catalysts Characterization and Testing
Head: Prof. Bair S. Bal'zhinimaev

Laboratory of Zeolites and Acid Base Catalysis
Head: Prof. Eugenii A. Paukshtis

Laboratory of Catalysts Activity Testing
Head: Dr. Nikolai N. Bobrov

Group of Catalytic Conversion of Sulfur-Containing Compounds
Head: Prof. Anna V. Mashkina

Group of Catalysts and Supports for High Temperature Processes
Head: Prof. Lyubov A. Isupova

DEPARTMENT OF CATALYTIC PROCESSES OF FINE ORGANIC AND BIOORGANIC SYNTHESIS

Head of the Department Prof. Zinaida P. Pai

Group of Liquid-Phase Catalytic Oxidation of Organic Compounds
Head: Prof. Zinaida P. Pai

Group of Catalysts and Processes Based on Heteropoly Acids
Head: Prof. Elena G. Zhizhina

Group of Heterogeneous Catalysts for Selective Liquid-Phase Oxidation
Head: Prof. Oxana A. Kholdeeva

ST. PETERSBURG DEPARTMENT OF THE BORESKOV INSTITUTE OF CATALYSIS

Director and Scientific Manager
Prof. Sergey S. Ivanchev

Vice-Director Dr. Valery P. Tulmankov

Laboratory of Micellar Catalysis, Emulsion Polymerization and Processes for Obtaining Polymeric Composites
Head: Prof. Valery N. Pavlyuchenko

Laboratory of Novel Catalytic Systems for Olefin Polymerization and Copolymerization
Head: Prof. Sergey S. Ivanchev

VOLGOGRAD DEPARTMENT OF THE BORESKOV INSTITUTE OF CATALYSIS

Executive Director Dr. Andrey P. Kovalenko

Scientific Director Dr. Aleksandr M. Beskopylnyi

SCHOOLS OF THE BORESKOV INSTITUTE OF CATALYSIS

The Boreskov Institute of Catalysis has numerous and deep traditions and continues intensive fundamental and applied investigations. It possesses an actively and fruitfully working school of specialists in catalysis.

Many works by the school of **Academician Georgii K. Boreskov** in the field of deep and partial oxidation processes are well known in our country and far from its frontiers.

The principal importance has the concept of chemical nature of phenomenon the decisive role in which belongs to the intermediate chemical interaction between the reagents and catalysts. The fundamentals of the theory of heterogeneous oxidative catalysis are:

— The key importance of the energy of reagent bonding to the catalyst for the rate and rote of oxidation reactions;

— The influence of cation nearest environment on catalytic properties;

— A viewpoint on reaction medium and catalyst as an indivisible system;

— An idea of stepwise and concerted mechanisms of redox reactions.

Under the guidance of **Prof. Mikhail G. Slin'ko** in 60–70th years a school raised famous not only in Russia, but abroad as well.

M.G. Slin’ko developed a method of mathematical modeling of catalytic reactors and processes, based on hierarchical principle. This method was developed for multi-phase processes and reactors with fixed and fluidized beds.

Design of real industrial catalysts with optimum strength, pore structure; providing their reproducibility on a large scale are the necessary elements of all aspects in the field of applied catalysis. **Prof. Vera A. Dzis’ko** contributed significantly to the development of methods of preparation and studying the regularities of oxide catalysts formation.

The revealed correlation of preparation conditions and physico-chemical properties of oxide systems allowed to formulate the criteria to control phase composition, surface area, stability, and acid-base properties, that appeared to be the basis for creation of a number of industrial catalysts and carriers.

At the end of the 60s big progress was in the field of homogeneous catalysis and catalysis by transition metal complexes. Of essential importance in this field not only for our country were the works by **Prof. Yurii I. Yermakov**.

Yu.I. Yermakov formulated the conception of the surface metal-organic compounds as active sites precursors for the supported catalysts. It served as a basis for the new approach in catalysis – application of principles used in homogeneous catalysis to study fundamental problems of heterogeneous catalysis. Methods for purposeful synthesis of supported catalysts of desired composition and dispersity have been developed.

In the 1977-1978 a big team of chemical physicists of the younger generation headed by **Academician Kirill I. Zamaraev** came from Moscow. The flow of these specialists significantly enlivened the atomic-molecular studies of catalytic reaction mechanisms and catalytic properties of heterogeneous catalysis by physicochemical methods.

The school by K.I. Zamaraev contributed much to:

— *Development of theory of electron tunneling in condensed media;*

— *Coordination chemistry of metal-complexes solutions;*

— *Step-wise description of homogeneous catalytic reactions;*

— *The works in photochemistry, photocatalysis and solar-to-chemical energy conversion.*

The main principle of these works is the complex application of modern physico-chemical technique to mechanistic studies of heterogeneous and homogeneous reactions, studies of the structure and the active sites at the molecular level, search of new nontraditional directions in catalysis.

Prof. Roman A. Buyanov developed the home school of fundamentals for catalyst preparation and technology**.**

The main research areas covered are:

- *Design and application of new methods to increase solid phases activity at catalysts preparation and to control their activity and selectivity;*
- *Study of catalysts deactivation and carbon-mineral compositions formation;*
- *Development of the theory of non-branched radical chain reactions in the absence of oxygen (pyrolysis);*
- *Design of industrial catalysts.*

The works of school by **Academician Valentin N. Parmon** relate to:

— *Design and study of novel and nontraditional catalytic processes, first of all, photocatalytic and thermochemical methods for the direct conversion of solar energy and ionizing radiation energy;*

— *Development of an original approaches to description of catalytic processes by methods of nonequilibrium thermodynamics;*

— *Study of a possible impact of photocatalysis over the troposphere aerosols on the global chemistry of the Earth atmosphere.*

The school of **Prof. Georgii M. Zhidomirov** develops the quantum-chemical direction in theoretical spectroscopy and molecular theory of catalysis:

The methodological basis for molecular (cluster) modeling of catalytic systems is formulated, the cluster approximation is widely used to calculation of electronic structure and reactivity of active sites on zeolites and oxide catalysts.

Boreskov Institute of Catalysis

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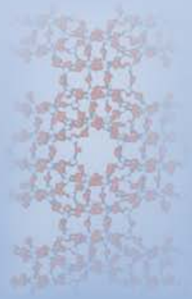
Countries > 20
Chemical & Petrochemical
Companies > 70
Foreign contracts:
R&D - 75%,
Licensing - 23%
Others - 2%

BIC - International Cooperation



2007 IC-GO-1 catalyst for deep hydrotreatment of diesel fractions

2006 Supported catalysts for olefines polymerization. Ultrahigh molecular weight polyethylene

2005
2004 Cracking catalysts LUXE and reforming catalysts of PR and RU series. In cooperation with the Institute for Hydrocarbons Processing (Omsk)

2002 Selective water sorbents

2000 New technology of loading the reactors with catalysts

1998 One-step technology for production of nicotinic acid
Degussa, Germany, 2000

1997 Non-platinum catalysts for production of nitric acid

1996 AlphOx™ technology (benzene + [N_2O] → phenol)
Developed in cooperation with Solutia Inc., USA, 1996

1995 Catalysts for FCC (oil cracking)

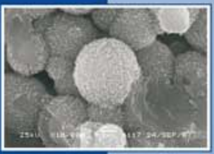
1993 SIBUNIT® - carbonaceous material
Sibunit for phosgene synthesis, Du Pont, USA, 1994

1988 Catalysts for propylene polymerization
DSM, the Netherlands, 1995

1982 REVERSE-Process
Enviro - Chem Systems, Inc. (A Monsanto - Company), USA, 1992

1978 Vanadium catalysts for sulfuric acid production

1958




R&D ACTIVITY OF THE INSTITUTE

Basic research

- ♦ *Determination of general regularities of catalysis*
- ♦ *Development of the scientific basis for molecular design the catalysts and materials*
- ♦ *Development of the theoretical basis to design the catalytic processes*
- ♦ *In-situ methods to study catalyst and mechanism of important catalytic reactions*
- ♦ *Methods and algorithms to predict the catalytic action*
- ♦ *Methods and facilities for the catalysts testing*
- ♦ *Nanotechnological approach to design the catalysts with predictable properties*

Applied catalysis

- *Design of highly efficient catalysts for production of key chemicals; oil & gas processing*
- *Design of the catalytic composites and technologies for applied purposes*
- *Catalysts and catalytic technologies for polymers production with specified properties*
- *Design of sorbents, catalysts and technologies for detoxication of hazardous man-caused wastes*
- *Selective oxidation of light C_1-C_4 paraffins by molecular oxygen*
- *Gas-phase propylene epoxidation by molecular oxygen*
- *Reactors and new processes based on membrane catalysts. Microchannel catalytic reactors*
- *Catalysts and processes related to hydrogen energy technologies*
- *Catalysis concerned to the environmental and energy problems*
- *New directions and opportunities of catalysis in utilization of renewable natural resources*

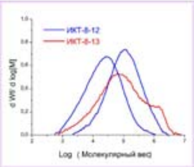
Some catalytic technologies owned by BIC

- **Reverse process** for catalytic gas purification
- **Zeoforming processes** for synthesis of gasoline
- **Alphox technology** for the one step phenol form benzene production
- **Adsoroption-catalytic purification** of industrial wastewaters
- **Nitric acid** production with the use of honeycomb catalysts
- **One-step** technology for the **nicotinic acid** production
- New technology of **loading the reactors** with catalysts
- **BIMT technology** for production of gasoline and diesel fuel without preliminary rectifying the hydrocarbon feedstock
- Reburning of Klaus **tail gases**
- The **BICYCLAR** technology for production of aromatic hydrocarbons with methane involvement
- Direct catalytic **oxidation of H_2S** to elemental sulfur

- Treating of mixed **radioactive organic wastes**
- Liquid-phase oxidative **treating of industrial wastewater**
- Technology for **formaldehyde** production
- Technology for **one step formic acid** production
- Technology for **reducing atmospheres** production
- Technique for preparation of **emulsifiers and water-bitumen emulsions**

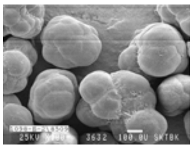
Some industrial catalysts by BIC

Vanadium pentoxide catalysts for sulfuric acid production



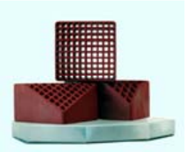
Supported catalysts ICT-8-12 and ICT-8-13 for polyethylene production

Catalyst ICT-8-20 for production of ultrahigh molecular weight polyethylene



Supported Ti-Mg catalyst IC-8-21 for polypropylene production

Microspherical zeolite-containing cracking catalysts

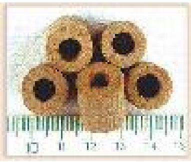


Non-platinum catalyst IC-42-1 for nitrogen acid production

Gasoline reforming catalysts of PR series

High silica zeolite catalysts

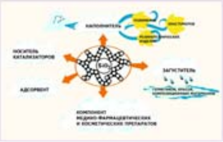
Hydrogenation catalysts of for manufacture of medical products and edible fats



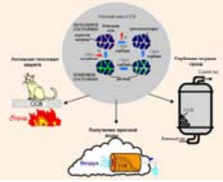
Thermostable catalyst ICT-12-40 for gas purification

Catalyst for hydropurification of terephthalic acid

Some catalyst supports by BIC



Superfine silica gels IC-01-2



Composite selective water sorbents SWS

Mesoporous carbonaceous material SIBUNIT

INSTRUMENTATION FACILITIES

The Institute engages the leading position in the world in the improvement and design of new physicochemical methods for catalyst study. The Boreskov Institute of Catalysis has specialized groups united into Department of Physicochemical Methods for Catalyst Investigation oriented at analytical and scientific studies. At the Department there are many specialists of high qualification specializing in the investigation of catalysts and nanomaterials.

Modern and, in some cases, unique physicochemical methods provide a basis for fundamental studies at the atomic-molecular level.

The types of analytical and research studies performed at the Institute and methods used for them are listed below.

Analytical (composition of catalysts and catalytic reaction products)

Bulk composition

Atomic emission spectrometry

Atomic absorption spectroscopy

X-Ray fluorescence analysis of macro- and microprobes

Phase analysis

X-Ray diffraction, including *in situ* diffraction

Differential dissolution

Thermally programmed reduction, oxidation, desorption
X-Ray diffusion scattering
EXAFS spectroscopy (for amorphous materials)

Morphology
Transmission electron microscopy
High resolution transmission electron microscopy (HRTEM)
Scanning electron microscopy
Scanning tunneling microscopy
X-Ray small-angle scattering

Surface
X-Ray photoelectron spectroscopy (XPS)
Secondary ion mass-spectrometry (SIMS)
Determination of the surface acidity using IR spectroscopy of probe molecules

Molecular composition of individual compounds and their mixtures
Gas, gas-liquid and liquid chromatography on packed, capillary and polycapillary columns
Superrapid chromatography
Mass-spectrometry

Adsorptive (specific surface area, pore structure, adsorption heat)
Porosimetry
Calorimetry

Kinetic
Gradientless and integral differential reactors
Fast relaxation technique
Stop flow technique
Radiochemical and isotopic methods
Mass-spectrometric detection of free radicals.

Spectral methods
NMR spectroscopy on different nuclei, including high-temperature and *in situ*
ESR spectroscopy, including *in situ*
FMR spectroscopy, including *in situ*
VUV electron spectroscopy
UV-VIS electron spectroscopy
Vibrational spectroscopies (IR, including *in situ,* and RAMAN)
HREELS
LEED
X-Ray and NMR tomography

INTERNATIONAL COOPERATION

The scientific cooperation of the Boreskov Institute of Catalysis with the catalytic communities from various countries is effected in accordance with various forms of cooperation: conducting joint seminars on catalysis, exchanging the information and appropriate materials, exchanging research fellows, visiting scientific centers, and participating in congresses and symposia on theoretical and applied catalysis.

According to research programs, projects and grants, the fundamentals of catalysis are studied jointly with researchers from various universities, institutions, research laboratories and companies. BIC collaborates fruitfully on a commercial basis with the leading companies from more than 20 countries, sells licenses, know-how and performs research projects according to client requests.

Academician Valentin N. Parmon is the Russian representative in the European Federation of Catalytic Societies (EFCATS), Member of the International Association of the Catalysis Societies (IACS).

Visits of the specialists from the Boreskov Institute of Catalysis to foreign institutions in 2007

Austria	2	Germany	26	South Korea	3
Belarus	3	Greece	11	Saudi Arabia	6
Belgium	2	Ireland	1	Spain	5
Bulgaria	1	Israel	5	Switzerland	2
Brazil	2	Italy	15	Tunisia	1
China	9	Japan	6	Turkey	2
Cyprus	1	Kazakhstan	1	Ukraine	5
Czechia	1	Netherlands	13	United Kingdom	7
Finland	22	Poland	2	USA	13
France	16	Portugal	2		

Visits of foreign specialists to the Boreskov Institute of Catalysis in 2007

Bulgaria	1	Italy	5	South Korea	8
Finland	1	Japan	8	United Kingdom	1
France	4	Kazakhstan	4	USA	29
Germany	1	Netherlands	10		

SCIENTIFIC COOPERATION
WITHIN THE FRAMEWORK OF CONTRACTS AND AGREEMENTS
WITH FOREIGN ORGANIZATION

ITALY

The cooperation in the frame of the agreement between **Russian Academy of Sciences** (RAS) and **National Council on the Scientific Research** of Italy:

- *Istituto di Trasformazione e Accumulazione d'Energia* (CNR Institute for Transformation and Storage of Energy), Messina – *BIC*, Novosibirsk on the Programme ***"Catalysis for Solving the Energy Problem"***, *Project* ***"Adsorption and Catalysis for Advanced Power Technologies"***. Coordinators: **Prof. Yu. Aristov** (*BIC*) and **Prof. G. Cacciola** (*Istituto di Trasformazione e Accumulazione d'Energia*).

- *Istituto di Scienze e Tecnologie Molecolari* (Institute of Molecular Science and Technologies), Milano – *BIC*, Novosibirsk on the Project ***"Methods for Homo- and Heteronuclear Bonds Formation and Opening in Stoichiometric Reactions and Catalytic Reactions Using Transition Metal Complexes"***. Coordinators: **Prof. N. Ustynyuk** (*Institute of General and Inorganic Chemistry, Moscow*) and **Prof. R. Psaro** (*Istituto di Scienze e Tecnologie Molecolar*). The Project theme is ***"Process Development for Catalytic Stereoselective Methanol Synthesis"*** (**Dr. I. Simakova**, *BIC*).

FRANCE

According to the agreement between **RAS** and **CNRS** BIC collaborates with:

- *Institute de Recherches sur la Catalyse* (Research Institute on Catalysis), Villeurbanne in the frame of the Russian-French European associated Laboratory on Catalysis headed by Acad. V. Parmon and Dr. M. Lacroix. The Laboratory was established by an agreement signed December 6, 2004 in Moscow by RAS and CNRS. Four areas of research were identified:

- ***Catalysis and Energy: Production of Syngas and Hydrogen***
- ***Catalysis and Environment: Treatment of Waste Waters***
- ***Catalysis and Fine Organic Chemistry*****:** ***Selective Oxidation with Hydrogen Peroxide***
- ***Advanced Spectroscopic Methods: Mobility of Molecules in Porous Media****.*

- *Universite Louis Pasteur*, Strasbourg on the Project ***"Novel Spatially Ordered Nanomaterials for Energy Conversion"***. Coordinators **Dr. Francois Garin** (*Universite Louis Pasteur*) and **Prof. E. Savinova** (*BIC*).

INDIA

In the frame of Indo-Russian Integrated Long Term Programme of cooperation in science and technology (ILTP) BIC collaborates with:

- *Indian Institute of Chemical Technology*, Hyderabad, on the Project ***"Study and Development of Heterogeneous Photocatalytic Removal of Hazardous Compounds***

from Air and Water". Coordinators: **Dr. A. Vorontsov** (*BIC*) and **Dr. M. Sabramaniam** (*Indian Institute of Chemical Technology*).

- *National Chemical Laboratory,* Pune on the Project ***"Design of Bifunctional Supported Non-Iron Catalysts for Low Temperature Ammonia Synthesis"***. Coordinators: **Dr. B. Moroz** (*BIC*) and **Dr. A.V. Ramaswamy** (*National Chemical Laboratory*).

Coordinators on the Program "Catalysis" are Acad. V. Parmon and Dr. S. Sivaram.

POLAND

In the frame of **RAS-PAS** agreement BIC cooperates with:

- *Instytut Inżynierii Chemicznej PAN* (Institute of Chemical Engineering), Gliwice on the Project ***"Synthesis and Utilization of Hyperporous Solids"***. Coordinators: **Prof. Yu. Aristov** (*BIC*), **Acad. V. Parmon** (*BIC*) and **Prof. A. Burghardt** (*Instytut Inżynierii Chemicznej*).
- *Instytut Katalizy i Fizykochemii Powierzchni PAN* (Institute of Catalysis and Surface Chemistry PAS), Cracow on the Project ***"V-Nb Catalysts: Relationship between Structure and Catalytic Activity".*** Coordinators: **Prof. O. Lapina** (*BIC*) and **Prof. B. Grzybowska-Świerkosz** (*Instytut Katalizy i Fizykochemii Powierzchni*).

GERMANY

The cooperation in the frame of the agreement between **RAS** and **German Scientific Research Society** (GSRS) with

- *Universitat Konstanz, Konstanz* on the Project ***"NMR and UV-Vis Spectroscopic Studies on the Interaction between Metallocene Complexes and Modified MAO"***. *Coordinators* **Prof. E. Talsi** (*BIC*) and **Prof. Dr. H.-H. Brintzinger** (*Universitat Konstanz*)
- *Fritz-Haber-Institut der Max-Planck-Gesellschaft*, Berlin on the Project ***"Development of in situ Methods for Study of Solid Surfaces"***. Coordinators: **Prof. V. Bukhtiyarov** (*BIC*) and **Prof. R. Schlögl** (*Fritz-Haber-Institut der MPG*).

CZECH REPUBLIC

The cooperation with *J. Heyrovského Ústav Fyzikální Chemie AV ČR* (J. Heyrovsky Institute of Physical Chemistry ASCR) on the Projects

- ***"Study of Cation Distribution in Zeolites".*** Coordinators**: Prof. Z. Sobalik** (*J. Heyrovského Ústav Fyzikální Chemi*) and **Prof. O. Lapina** (*BIC*).
- ***"Electron Spectroscopy to Study Catalyst Surface".*** Coordinators: **Prof. P. Carsky**, **Prof. Z. Bastl** (*J. Heyrovského Ústav Fyzikální Chemi*), **Prof. A. Boronin** (*BIC*).

CHINA

The cooperation in the frame of **Associated Research Laboratory** which was established by an agreement signed December 4, 2004 by the Boreskov Institute of Catalysis and Heilundzyan University, Harbin. Chief Executive officers of Laboratory are:

Prof. V. Bukhtiyarov (*BIC*) and **Fu Hong-Gang** (*Heilundzyan University*) on the Project ***"Synthesis and Modification of ZSM-12 Zeolites. Zeolite ZSM-12 in Reaction of Naphthalene Alkylation with Methanol"***. Coordinators: **Prof. G.J. Sheng** (*Heilundzyan University*), **Prof. G. Echevsky** (*BIC*).

SPAIN

The cooperation with *Instituto de Catalisis y Petroleoquimica* (Institute of Catalysis and Petroleochemistry), Madrid on the Project ***"Fundamental and Technical Aspects of in situ Spectroscopic Studies of Oxide Catalysts"***. Coordinators: ***Prof. O. Lapina*** (*BIC*) and **M.A. Bañares** (*Instituto de Catalisis y Petroleoquimica*).

COOPERATION IN THE FRAME OF PROJECTS FINANCED BY INTERNATIONAL FOUNDATIONS

INTAS SUPPORTED PROJECTS

I. Novel Catalytic Process for Industrial Waste Water Treatment

Project Coordinator:

Dr. P. Gallezot, *Institut de Recherches sur la Catalyse*, Villeurbanne, France

Participants:

Acad. V. Parmon, *The Boreskov Institute of Catalysis*, Novosibirsk, Russia
Prof. B. Laskin, *Research Scientific Center for Applied Chemistry*, St. Petersburg, Russia.

II. Competitive Hydrogen from Agro-Forestry Residues

Project Coordinator:

Prof. G. Grassi, *European Biomass Industry Association (EUBIA)*, Brussels, Belgium

Participants:

Prof. V. Kirillov, *The Boreskov Institute of Catalysis*, Novosibirsk, Russia.

III. Sustainable Route to the Generation of Synfuels via Syngas Derived from Biomass

Project Coordinator:

Prof. J. Ross, *University of Limerick*, Limerick, Ireland

Participants:

Dr. K. Seshan, *Universiteit Twente*, Enschede, The Netherlands
Dr. O. Hazewinkel, *Techno Invent Ingenieursbureau voor Milieutechniek b.v.*, Zoetermeer, The Netherlands
Prof. V. Sadykov, *The Boreskov Institute of Catalysis*, Novosibirsk, Russia
Prof. A. Rozovskii, *Topchiev Institute of Petrochemical Synthesis*, Moscow, Russia.

INTAS - SB RAS Supported Project

Electromagnetic Response Properties of Carbon Onions and Carbon Onion-Based Composites

Project Coordinator:

Dr. Ph. Lambin, *Facultes Universitaires Notre-Dame de la Paix*, Namur, Belgium

Participants:

Belarus State University, Minsk, Belarus; *University of Joensuu*, Finland; *Institute for Technical Physics and Materials Science*, Budapest, Hungary; *The Boreskov Institute of Catalysis*, Novosibirsk, Russia (**Dr. V. Kuznetsov)**, *Nikolaev Institute of Inorganic Chemistry*, Novosibirsk, Russia.

EUROPEAN COMMUNITY SIXTH FRAMEWORK PROGRAM

I. International Partnership for a Hydrogen Economy for Generation of New Ionomer Membranes

Coordinator: **Dr. R. Mallant**, *Energy Research Centre of The Netherlands*, Petten, The Netherlands

Partners:

Daimler Chrysler; *FuMA-Tech GmbH*; *CNRS Montpellier*; *Dohgyue Chenzhou New Materials Company*; *Shanghai Jiao Tong University*, Shanghai, China; *The Boreskov Institute of Catalysis*, Novosibirsk, Russia (**Prof. V. Bukhtiyarov**).

II. Co-Processing of Upgraded Bio-Liquids in Standard Refinery Units

Coordinator: **Dr. Y. Solantausta**, *VTT Processes*, Espoo, Finland

Partners:

Rijksuniversiteit Groningen, The Netherlands; The Boreskov Institute of Catalysis, Novosibirsk, Russia (**Prof. V. Kirillov**); Uhde Hochdrucktechnik GmbH, Germany; BTG Biomass Technology Group BV, The Netherlands; University of Twente, The Netherlands; STFI-PACKFORSK AG, Sweden; Institute of Wood Chemistry, Hamburg, Germany; Slovenian Institute of Chemistry, Slovenia; Arkema SA, France; Helsinki University of Technology, Finland; ALMA Consulting Group SAS, France; Centre National de la Recherche Scientifique, France; Chimar Hellas SA, Greece; Albermarle Catalysts Company BV, The Netherlands; Metabolic Explorer, France; Shell Global Solutions International, The Netherlands.

III. Non-Noble Catalysts for Proton Exchange Membrane Fuel Cell Anodes

Coordinator:

Dr. G. Tsotridis, *Institute for Energy, Joint Research Centre*, Petten, The Netherlands

Partners:

Technical University of Denmark, Lyngby, Denmark; *The Boreskov Institute of Catalysis*, Novosibirsk, Russia (**Acad. V. Parmon)**; *Southampton University*, United Kingdom; *Technical University of Munich*, Germany; *Bavarian Center for Applied Energy Research*; *Umicore, AG & Co KG*, Germany.

IV. *Novel Materials for Silicate-Based Fuel Cells*

Coordinator: **Dr. Ch. Arguirusis**, *Technische Universität Clausthal*, Clausthal, Germany

Partners:

University of Aveiro, Aveiro, Portugal; *Foundation of Research and Technology Hellas*, Greece; *Katholieke University of Leuven*, Belgium; *Max-Plank Institute of Colloids and Interfaces*, Munchen, Germany; *The Boreskov Institute of Catalysis*, Novosibirsk, Russia (**Prof. V. Sadykov**); *Ceramics and Refractories Technological Development Company*, Greece; *Ceramiques Techniques et Industrielles*, France.

NATO PROGRAMME: SCIENCE FOR PEACE

I. Solid Oxide Fuel Cells for Energy Security

NATO Country Project Director:

Prof. N. Orlovskaya, *Drexel University*, Philadelphia, United States

Partner Country Project Director:

Prof. O. Vasiliev, *Frantcevych Institute for Problems of Material Science*, Kiev, Ukraine

Project Co-Directors:

Prof. V. Sadykov, *The Boreskov Institute of Catalysis*, Novosibirsk, Russia
Prof. J. Irvine, *University of St. Andrews*, St. Andrews, United Kingdom
Prof. N. Sammes, *University of Connecticut*, Storrs, United States
Prof. R. Hasanov, *Azerbaijan State Oil Academy*, Baku, Azerbaijan
Dr. A. Schokin, *State Committee for Energy Saving of Ukraine*, Kiev, Ukraine
Prof. John Kilner, *Imperial College*, London, United Kingdom.

II. Mixed Conducting Membranes for Partial Oxidation of Natural Gas to Synthesis Gas

NATO Country Project Director:

Prof. J. Frade, *Departamento de Engenharia Cerâmica e do Vidro, Universidade de Aveiro*, Aveiro, Portugal

Partner Country Project Director:

Dr. V. Kharton, *Institute of Physicochemical Problems, Belarus State University*, Minsk, Belarus

Project Co-Directors:

Dr. J. Irvine, *School of Chemistry*, *University of St. Andreas*, Scotland, United Kingdom
Dr. T. Norby, SMN, *Universitetet i Oslo*, Oslo, Norway
Dr. J. Jurado, *Instituto de Cerámica y Vidrio*, CSIC, Madrid, Spain
Prof. V. Sobyanin, *The Boreskov Institute of Catalysis*, Novosibirsk, Russia
Prof. V. Kozhevnikov, *Institute of Solid State Chemistry*, Ekaterinburg, Russia
Dr. L. Boginsky, *Institute for Personal Development and Staff Retraining in New Areas of Techniques, Technologies and Economics of the Belarus Ministry of Education*, Minsk, Belarus.

NATO PROGRAMME: SCIENCE FOR PEACE AND SECURITY

Capture and Decontamination of Chemical & Biological Agents by Novel Catalysts and Millisecond Jet Reactors

Project Coordinator from a NATO Country:

Prof. P. Smirniotis, *University of Cincinnaty*, Cincinnaty, United States

Project Coordinator from a Partner Country:

Dr. A. Vorontsov, *The Boreskov Institute of Catalysis*, Novosibirsk, Russia.

NATO COLLABORATIVE LINKAGE GRANTS

Oxide Catalysts for Air and Water Quality Control Ammonia Removal

Project Coordinator from a NATO Country:

Dr. M. Banares, *Instituto de Catalisis y Petroleoquimica*, Madrid, Spain
Dr. M. Ziolek, *Adam Mickieiwcz University*, Poznan, Poland

Project Coordinator from a Partner Country:

Prof. O. Lapina, *The Boreskov Institute of Catalysis*, Novosibirsk, Russia.

INTERNATIONAL SCIENCE AND TECHNOLOGY CENTER (ISTC)

I. Development of Catalysts and Reactors for Syn-Gas Production from Diesel Fuel and for Selective NO_x Reduction with Syn-Gas in Diesel Exhausts

Project Manager from BIC **Prof. V. Kirillov**.

II. Development of High-Performance Oxygen-Containing Membranes and Compact Syn-Gas Generators on Their Base

Project Manager from BIC **Prof. V. Sadykov**.

III. Synthesis and Investigation of the Metal Oxide Catalysts for Photocatalytic Degradation of Harmful Gases Resulted from Terrorist Acts and Man-Caused Catastrophes

Project Manager from BIC **Dr. A. Vorontsov**.

IV. Catalytic Production of SO_3 for Conditioning of Electrostatic Precipitators Using in Russia and the Newly Independent States (NIS)

Project Manager from BIC **Prof. A. Zagoruiko**.

V. Development of an Integrated Separator for Direct Reforming of Hydrocarbons in High-Temperature Fuel Cells

Project Manager from BIC **Prof. Z. Ismagilov**.

CONFERENCE AND EXHIBITION ACTIVITIES

Participation in exhibitions, discussion of commercial prospects of the displayed innovations illustrates the research and social activities of an institution and promotes an increase in its rating, expanding of the potential market for new technologies and materials. Besides, the level and consumer properties of the scientific achievements may be evaluated in more realistic manner.

In 2007, the most important BIC's achievements in the area of applied Catalysis were presented at the Exhibitions:

- ***Exhibition "Oil. Gas. Chemistry. Ecology"***, February 14-16, Naberezhnye Chelny, Russia
- ***International Exhibition "Sibnedra. Mining. Sibneftegas. Metalls of Siberia. Foundry, Metal Working, Welding"***, March 27-29, Novosibirsk, Russia
- ***II Siberian Energy Congress "Fuel-Energy Complex. Modern Technique and Equipment. Siberia-2007"***, April 11-12, Novosibirsk, Russia
- ***"High Technologies of XXI Century"***, April 23-26, "Expocentr", Moscow, Russia
- ***VII International Exhibition of Military and Equipment, Technologies and Armament, "VTTV-Omsk-2007"***, June 5-9, Omsk, Russia
- ***III Anniversary Ural-Siberian Scientific-Industrial Fair***, June 9-21, Yekaterinburg, Russia
- ***14th International Exhibition of Chemical Industry and Science – KHIMIA'2007***, September 3-7, "Expocentr", Moscow, Russia
- ***Exhibition of Achievements of the Institutes of SB RAS in the frame of Russian-Kazakh Forum***, October 4-5, Novosibirsk, Russia
- ***International Industrial Exhibition "SIBPOLYTECH: Science of Siberia"***, October 23-26, Novosibirsk, Russia
- ***Exhibition-Presentation of Scientific and Technical Developments of the Institutes of SB RAS***, October 25-27, Urumchi, China
- ***Exhibition devoted to 70th Anniversary of SB RAS***, October 31-November 2, Novosibirsk, Russia.

Solution of many specific problems requires joint efforts in order to promote its fastest accomplishing. International congresses, symposia, conferences, and seminars are the most productive to familiarize scientists with innovations outside their own research and to disseminate the information about the results achieved to a wider audience. One of the directions of the scientific-organizational work carried out in the Institute is aimed on conducting conferences and seminars with the participation not only of Russian scientists and researchers from NIS, but foreign participants as well.

CHEMISTRY AND CATALYSIS

All-Russian Conference of the Laureates of the Zamaraev International Charitable Scientific Foundation - to the 10th Anniversary of Founding

May 16-19, Novosibirsk, Russia

Organizers:

- Zamaraev International Charitable Scientific Foundation
- The Boreskov Institute of Catalysis, Novosibirsk, Russia.

120 participants, among them 34 laureates of the Foundation, took part in the Conference. The scientific program included 5 plenary lectures, 45 oral presentations and 35 posters.

Plenary lectures:

A.G. Stepanov (The Boreskov Institute of Catalysis, Novosibirsk, Russia) – *“In situ NMR Spectroscopy in Heterogeneous Catalysis”*

E.M. Moroz (The Boreskov Institute of Catalysis, Novosibirsk, Russia) – *“Determination of Local Structure of Nanosystems by Radial Distribution of Electron Density (RDED) Method”*

L.M. Levchenko, V.N. Mitkin* (Nikolaev Institute of Inorganic Chemistry, Novosibirsk, Russia; *JSC “Novosibirsk Chemical Concentrates Plant”, Novosibirsk, Russia) – *“New Generation of Nanocomposite Carbon-Carbon and Carbon-Fluorocarbon Matrices and Functional Materials on Their Base for Sorption and Catalytic Technologies”*

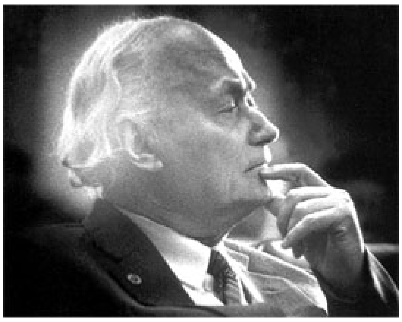
V.I. Bukhtiyarov (The Boreskov Institute of Catalysis, Novosibirsk, Russia) – *“Size Effects in Catalysis by Supported Metal Nanoparticles”*

A.G. Okunev (The Boreskov Institute of Catalysis, Novosibirsk, Russia) – *“Low-Temperature Fuel Cells and Power Installation on Its Base”.*

Memorial seminar in honor of Academician K. Zamaraev was organized in the frame of the Conference.

Contest **UMNIK** (“The Participant of Youth Scientific Innovative Contest-2007”) has been held during the Conference. “The Foundation for Assistance to Small Innovative Enterprises”, “FASIE”, every year carries out this contest and selects the best proposals of scientific conferences, contests, seminars and other events, hold in Russian Federation. The aim of the program UMNIK is to reveal the youth aiming toward self-actualization through innovation activity, and encouragement their scientific-technical and innovative activity by financial support of innovative projects.

The winner of the UMNIC contest became **Dr. Sergey V. Semikolenov** for the work *“Synthesis of Liquid Polyketones by Carboxidation of Rubbers with Nitrous Oxide”* by S.V. Semikolenov, K.A. Dubkov, D.P. Ivanov, L.G. Echevskaya, M.A. Matsko, D.E. Babushkin, G.I. Panov.



III INTERNATIONAL CONFERENCE “CATALYSIS: FUNDAMENTALS AND APPLICATION” dedicated to the 100th anniversary of Academician Georgii K. Boreskov

July 4-8, Novosibirsk, Russia

Conference organizers:

- The Siberian Branch of Russian Academy of Sciences
- Scientific Council on Catalysis RAS
- Boreskov Institute of Catalysis,
  Novosibirsk, Russia
- Ministry of Education and Science of the Russian Federation, Moscow, Russia
- Russian Foundation for Basic Research, Moscow, Russia
- Russian Mendeleev Chemical Society, Novosibirsk Department, Russia.

379 participants, among them 90 from 17 foreign countries and 22 from NIS took part in the event.

Leading specialists in the field of catalysis were invited as plenary and key lecturers. The Program of the Conference consisted of 8 plenary and 16 keynote invited lectures, 83 oral presentations and 278 posters.

The oral presentations were scheduled in 3 parallel sessions:

- Mechanisms of Heterogeneous and Homogeneous Catalysis at Molecular Level
- Design of Heterogeneous and Homogeneous Catalysts
- Environmental and Industrial Catalysis.

World recognized scientists have accepted the invitation of the Organizing Committee to give the plenary lectures:

A.T. Bell (Department of Chemical Engineering, University of California, Berkeley, CA, USA) – *"The Science of Catalysis"*

R.K. Grasselli (Center for Catalytic Science and Technology, University of Delaware, Newark, USA; Department of Chemistry, Technische Universität München, Garching, Germany) – *"Design of Selective Heterogeneous Oxidation Catalysts Based on the "Seven Pillars of Oxidation Catalysis"*

M. Misono (National Institute of Technology and Evaluation (NITE), Tokyo, Japan) – *"Green Catalytic Technology: Recent Developments of Heteropolyacid and Perovskite Catalysts"*

G.I. Panov (The Boreskov Institute of Catalysis, Novosibirsk, Russia) – *"Oxidation Catalysis: Active Oxygen in Selective Oxidation"*

A. Holmen (Department of Chemical Engineering, Norwegian University of Science and Technology, Trondheim, Norway) **–** *"CO Hydrogenation on Cobalt Catalysts"*

Yu. Matros, G. Bunimovich (Matros Technologies, Inc., Chesterfield, USA) – *"Forced Unsteady-State Catalytic Processes. Positive and Negative Experience"*

M.J. Ledoux (Laboratoire des Matériaux, Surfaces et Procédé pour la Catalyse, CNRS-Université Louis Pasteur-ECPM-ELCASS, Strasbourg, France) – *"Assisted Heterogeneous Catalysis, a Look to the Future"*

F. Schüth (Max-Planck-Institut für Kohlenforschung, Mülheim, Germany) – *"Colloidal Metal Particles for the Synthesis of Catalytic Materials"*.

The Conference Program also included the School-Session of the Young scientists *"Functional Nanomaterials in Catalysis and Energy-Saving"* - 20 oral presentations were given.

THE RUSSIAN-GERMAN SEMINAR ON CATALYSIS "BRIDGING THE GAP BETWEEN MODEL AND REAL CATALYSIS"

July 9-12, Novosibirsk - Altai Mountains, Russia

The Seminar followed the III International Conference *"Catalysis: Fundamentals and Application"* dedicated to the 100th Anniversary of Academician G.K. Boreskov.

The Organizers of the Seminar:

- The Boreskov Institute of Catalysis, Novosibirsk, Russia
- Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany.

47 scientists participated in the Seminar including 15 participants from Germany and 32 from Russia.

The session topics covered:

- Study of the Mechanisms of Heterogeneous Catalytic Reactions at Atomic-Molecular Level
- Application of the Surface Sensitive Methods to Study the Formation, Activation and Degradation of the Active Sites in Heterogeneous Catalysts
- Computational Chemistry to Study the Catalytic Reactions.

Scientific program included 7 invited plenary lectures and 20 oral presentations. The main attention was paid for application of various physical methods for catalyst and reaction mechanism studying.

Plenary lectures:

I.L. Zilberberg (The Boreskov Institute of Catalysis, Novosibirsk, Russia) – *"Oxygen Electronic States on the Surface of Catalysts: A Quantum-Chemical View"*

Sh. Shaikhutdinov (Fritz-Haber-Institut der Max-Planck-Gesellschaft, Abteilung Chemische Physik, Berlin, Germany) – *"Towards an Understanding of Structure-Reactivity Relationships in Catalysis: Model Studies"*

V.B. Kazansky, I.R. Subbotina, F.C. Jentoft*, R. Schlogl* (Zelinsky Institute of Organic Chemistry, Moscow, Russia; *Fritz Haber Institute of the Max Planck Society, Department of Inorganic Chemistry, Berlin, Germany) - *"Intensities of IR Stretching Bands as a New Spectral Criterion of Chemical Activation of Adsorbed Molecules via Their Polarization by the Active Sites in the Acid Heterogeneous Catalysis"*

M. Havecker, D. Teschner, E. Vass, **A. Knop-Gericke,** S. Zafeiratos, H. Gabasch, P. Schnorch, R. Schlogl (Fritz-Haber-Institut der Max-Planck-Gesellschaft, Department of Inorganic Chemistry, Berlin, Germany) – "*The Role of Subsurface Species in Heterogeneous Catalytic Reactions"*

J.A. Lercher (Technische Universität München, Department of Chemistry, Garching, Germany) – "*Activating Light Alkanes – New Opportunities through Molecular Understanding"*

E.R. Savinova (The Boreskov Institute of Catalysis, Novosibirsk, Russia; Universite Louis Pasteur, Strasbourg, France) – "*Application of in situ Methods for the Investigation of Electrocatalytic Reactions"*

A. Stakheev, W. Grunert*, O. Tkachenko, N. Telegina (Zelinsky Institute of Organic Chemistry, Moscow, Russia; *Ruhr-University Bochum, Bochum, Germany) – "*Revealing Structure-Activity Relationship in Supported Metal Catalysts by Analysis of XPS Line Shape"*.

RUSSIAN CONFERENCE (WITH THE PARTICIPATION OF THE WESTERN PARTICIPANTS) “CATALYTIC TECHNOLOGIES FOR ENVIRONMENTAL PROTECTION IN INDUSTRY AND TRANSPORTATION”

December 11-14, St. Petersburg, Russia

The Conference was organized by

- The Boreskov Institute of Catalysis, Novosibirsk, Russia
- FSUE RSC “Applied Chemistry”, St. Petersburg, Russia
- Center for Ecological Safety RAS, St. Petersburg, Russia
- Federal Agency of Science and Innovation of the Russian Federation
- Scientific Council on Catalysis RAS
- Scientific Council on Scientific Bases of Chemical Technology RAS
- The Federal Service on Ecological, Technological and Nuclear Control of RF
- Novosibirsk Regional Committee of the Environment and Natural Resources Preservation
- St. Petersburg Scientific Center RAS.

Three-day scientific program of the Conference comprised 6 plenary invited lectures, 5 key, 42 oral and 45 poster presentations on the following topics:

- Catalytic Exhaust Gas Purification of Industrial Enterprises from:
  - Carbon oxides and organic compounds
  - Nitrogen oxides
  - Sulfur oxides and Sulfur Compounds
- Catalytic Exhausts Purification form Specially Dangerous and Specific Pollutants
- Catalytic Methods for the Detoxification of Automobile Transport Waste Emissions

- Catalytic and Sorption Methods for Industrial Waste Purification
- Utilization of Solid Wastes from Industrial, Extractive and Agricultural Enterprises
- Innovative Ecologically Safe Catalytic Technologies
- Analytic Control of Man-Made Pollution of Industrial Enterprises and Automobile Transport.

Plenary lectures:

Z.R. Ismagilov (The Boreskov Institute of Catalysis, Novosibirsk, Russia) – “*Catalytic Fuel Combustion. From Fundamental Studies to Industrial Production*”

M. Rodkin, A. Blatov*, S. Aleshin* (BASF Catalyst LLC, USA; *ZAO “BASF”) –“*State-of-the-Art in Developments of Catalytic Purification of Exhaust Gas from Mobile Sources*”

G.B. Manelis (Institute of Chemical Physics, Chernogolovka, Russia) – “*Chemical Problems of Thermal Power Engineering*”

A.G. Tumanovskii, **S.N. Anichkov** (The All-Russian Thermal Engineering Institute, Moscow, Russia) – “*Ecological Problems in Thermal Power Engineering*”

S.B. Putin, V.D. Samarin (Corporation “Roskhimzaschita”, Tambov, Russia) – “*Modern Technologies of Adsorptive and Catalytic Air Purification as the Base for Production of Personal and Collective Protective Equipment from Damaging Action of Chemical and Biological Nature*”

A.A. Solovyanov (Committees for Nature Management and Ecology (Chamber of Commerce and Industry; Russian Union of Industrialists and Entrepreneurs, Moscow) – “*Legislative and Regulatory Basis for Environmental Activity in Russian Federation: Problems and Perspectives*”.

II INTERNATIONAL CONFERENCE BIOSPHERE ORIGIN AND EVOLUTION

October 28 - November 2, Loutraki, Greece

The Conference was organized by:

- Trofimuk United Institute of Geology, Geophysics & Mineralogy, Novosibirsk, Russia
- The Boreskov Institute of Catalysis, Novosibirsk, Russia
- Institute of Cytology and Genetics, Novosibirsk, Russia
- Paleontological Institute, Moscow, Russia.

Four-day scientific program of BOE-2007 comprised 10 plenary invited lectures, 10 key-note presentations, 46 oral presentations and 70 posters on the following topics:

- Problems of Abiogenic Synthesis and Evolution of the Matter under Conditions of Pregeological Stages of the Earth Evolution. Astrochemistry, Astrocatalysis
- Astrobiology. RNA World
- Biogeological Problems of the Evolution of the Archean–Proterozoic and Phanerozoic Biosphere
- Biomineral Systems
- Genetic and Ecosystem Problems of the Evolution
- Mechanisms of Anthropogenesis and Inhabitation of Humans.

The Posters Session was a significant part of the program, followed by the Round Table discussions.

120 scientists from 6 countries (Russia, USA, Germany, Ukraine, Belarus, Kazakhstan) participated in the Conference.

EDUCATIONAL ACTIVITIES

With that deep belief that people are a decisive factor and that the best way to educate is to teach others, many researchers of the Institute participate in the training of specialists in chemistry and, particularly, in catalysis, ensuring closest and time-tested links of the Institute with the main sources of researchers – Novosibirsk State University (NSU), Novosibirsk State Technical University (NSTU), Tomsk State University (TSU), Tomsk Polytechnical University (TPU). Students studying at the following chairs realize their graduate works within the Boreskov Institute of Catalysis:

- Catalysis and Adsorption, Department of Natural Sciences, NSU
- Physical Chemistry, Department of Natural Sciences, NSU
- General Chemistry, Department of Natural Sciences, NSU
- Chemical and Biological Physics, Department of Physics, NSU
- Physical Methods to Study Solids, Department of Physics, NSU
- Environmental Engineering, Aircraft Department, NSTU
- Technological Processes and Apparatuses, Department of Mechanics and Technology, NSTU
- Physical and Colloid Chemistry, Chemical Department, TSU
- Fuel Chemical Technology and Chemical Cybernetics, TPU.

About 70 scientists combine teaching and their job at the Research Institute. They present lectures, give seminars and practical classes; participate in the organization and renovation of the educational process.

From the first years, the strategy of the training of students was built up by leading scientists of BIC and harmonically combines two aspects: on the one hand, curricula provide the students with deep theoretical and practical knowledge in catalysis, i.e. theory of the prevision of the catalytic action of substances, kinetics of catalytic reactions, scientific bases of the preparation of catalysts, bases of the technology of catalytic processes, application of modern physical and mathematical methods for investigation of catalysts and catalysis; on the other hand, the educational process arrangement allows to wide significantly the sphere of the postgraduate activity of the students. They can work as technologists, production managers, researchers at various research organizations, teach at universities.

All these features allow students to prepare their diploma at the highest scientific level and become widely informed specialists in chemistry and catalysis. The graduates of the chair of adsorption and catalysis are the principal source of recruitment of the personnel of the Institute.

BIC – NSU

Novosibirsk State University was established as an essential part of the Siberian Branch of the Academy of Sciences; it embodies the idea of the close integration of education and science. The chief mission of the University is to train professionals for scientific institutions of the country.

From its earliest age, the University pursues *three fundamental principles.* The *first* one is that the teachers are experts engaged in the real science. The intellectual basis of the University is more than 5000 scientists of 30 research institutes of the Siberian Branch. The *second* principle is a thorough mathematical background provided to students of all the departments. And the *third* principle means that the students master theoretical disciplines during their first three years and do their practical research in academic institutes of the Siberian Branch during the last years.

The Boreskov Institute of Catalysis is a key element in the training of specialists in chemistry at NSU. Graduation works of the students are conducted at the laboratories of the Institute and are supervised by experienced researchers. Their results are usually published in scientific journals and form the basis of their future Ph.D. theses.

The *Chair of Catalysis and Adsorption* is of special importance for the Institute. It is only 5 years younger than Novosibirsk State University itself. The organization of the chair was initiated in 1964 by Academician G.K. Boreskov – the founder and first director of the Boreskov Institute of Catalysis SB RAS. The chair is located at the Institute, which provides it with everything required for efficient training of students specializing in catalysis and technology of catalytic processes.

Courses of lectures:

- Catalysis
- Adsorption and Porous Structure
- Scientific Bases for Catalysts Preparation
- Kinetics of Heterogeneous Catalytic Reactions
- Engineering Chemistry of Catalytic Processes
- Catalysis, Environment and Sustainable Development of Civilization
- Molecular Design of Catalysts
- Modern Technique of Catalytic Experiments
- Analytical Methods
- Quantum-Chemical Methods in Catalysis
- Magnetic Resonance Spectroscopy
- Optical Spectroscopy
- X-Ray Technique
- Thermodynamics of Working Catalyst
- Physical Methods in Catalysis
- Computer Application for Catalytic Studies

The *Chair of Physical Chemistry* prepares highly skilled specialists in the field of chemical kinetics and thermodynamics, application of physical methods for catalysis.

Courses of lectures:

- Physical Chemistry
- Chemical Thermodynamics
- Chemical Kinetics

- Nonequilibrium Thermodynamics
- Matter Structure
- Terminal System: Computer Modeling of Processes and Physical-Chemical Phenomena
- NMR and ESR Spectroscopy for Catalysis

Educational Scientific Centers (*ESC*) was established according to the Government concept for the education modernization . Educational, scientific and innovational complexes give the possibility to increase quality of professional education by providing the unity of educational, scientific and technical innovational activity at every stage of training specialists.

Educational Scientific Center “Adsorption and Catalytic Technologies” was made on the basis of Chair of Catalysis and Adsorption and BIC’s laboratories. It includes educational, scientific and innovational constituents.

The ***ESC on Photochemistry*** *was made* on the basis of Chair of Physical Chemistry, laboratories of the Institute of Chemical Kinetics and Combustion and BIC. It gives the opportunities to train specialists in the field of physical chemistry, photochemistry and photocatalysis.

BIC – NSTU

Novosibirsk State Technical University is one of the largest research and educational centers in Russia. The University trains and retrains qualified specialists for research and industrial complex of Siberia and the Far East. Joint training of specialists at NSTU and institutes of the Siberian Branch of RAS in the field of technology of catalytic processes is of special importance for practical application of fundamental knowledge in the field of catalysis. The basic training is conducted by the *Chair of Environmental Engineering* founded at the Boreskov Institute of Catalysis.

Courses of lectures:

- Technology Bases for Environmental Protection
- Processes and Apparatuses for Environmental Protection
- Catalytic Methods for Environment Protection
- Methods and Devices for Environmental Control, Ecological Monitoring
- Mathematical Modeling of Chemical Processes and Reactors
- Organic Chemistry
- Industrial Ecology
- Technologies for Utilization of Industrial Wastes
- Oil-Gas Branch
- Analytic Chemistry and Physical-Chemical Methods of Analysis

Chemical engineering is a science-intensive industry. The engineers will be able to successfully work in the priority investment areas. The close co-operation of the *Chair of*

Technological Processes and Apparatuses with research institutes of the Siberian Branch of Russian Academy of Science is the key to train high-skilled specialists.

Dissertation Council Activities

Besides teaching of the students in many Russian universities, researchers of the Institute participate in the training of young scientists of the highest qualification capable to provide original basic and applied research in the field of catalysis. Their training has priority importance for the Institute and is directly related to its development and well being. The Institute solves these problems through the postgraduate school of SB RAS and postgraduate school of NSU. Annually 20-30 postgraduates are trained at the graduate school working at the Institute. Supervised by the researchers of the Institute, postgraduate students conduct their Ph.D. studies in chemical kinetics and catalysis, physical chemistry, chemical engineering in the BIC laboratories.

Dissertation Council D 003.012.01 was confirmed by Supreme Certifying Commission (SCC) of Russia on December 8, 2000. Dissertation Council is allowed to uphold a thesis to receive an academic degree Doctor of Science on specialties: 02.00.15 "Catalysis" within chemical and technical sciences and 02.00.04 "Physical Chemistry” within chemical sciences. Head of the Dissertation Council is the director of the Institute – Academician V. Parmon.

Dissertation Council К 003.012.01 was confirmed by SCC on December 8, 2000. Dissertation Council is allowed to uphold a thesis to receive an academic degree on specialties 02.00.15 "Catalysis" within chemical sciences and 05.17.08 "Processes and Apparatuses in Chemical Technology” within technical sciences. Head of the Dissertation Council deputy director of the Institute Prof. Vladimir A. Sobyanin.

1 Doctoral thesis and 9 Ph.D. theses have been defended in 2007.

Doctoral theses:

Galina A. Kovalenko – *“Catalysis by Enzymes and Non-Growing Bacterial Cells, Immobilized on Inorganic Supports”*

Ph.D. theses:

Natalia V. Shtertser - *“Synthesis and Dehydrogenation of Methanol over Copper-Containing Catalysts”*

Olga V. Zalomaeva - *“Oxidation of Functionalized Phenols and Naphthols to the Corresponding Quinones over Titan- and Iron-Containing Catalysts Using* H_2O_2*”*

Aleksandr N. Simonov - *“The Catalytic Synthesis of Monosaccharides from Formaldehyde in Aqueous Solutions”*

Sukhe D. Badmaev – *“Catalytic Dimethyl Ether Conversion to Hydrogen-Rich Gas”*

Ekaterina V. Matus – *“Synthesis and Investigation of Mo/ZSM-5 Catalysts for Methane Dehydroisomerization”*

Eugenii V. Kovalev – *“Statistical Lattice Model for the Physicochemical Processes Proceeding over the Supported Catalysts Taking into Account Surface Morphology and Shape of the Particles”*

Anna N. Usoltseva – *“The Physico-Chemical Basis for Catalytic Carbon Nanotubes Synthesis”*

Andrey I. Studnichenko – *“Study of Adsorbed Oxygen on the Surface of Polycrystalline and Nanodispersed Gold”*

Ilya N. Mazov – *“Development of Physico-Chemical Basis of Catalytic Synthesis of Filamentous SiC Crystals and Study of Their Properties”*.

There is a great demand for specialists in catalysis trained at the Boreskov Institute of Catalysis. Many of them apply their knowledge in these fields at various research centers, universities and manufacturing companies.

Joint Educational Scientific Laboratories

3 Joint Laboratories were created with the purpose of integration of high education and fundamental science to increase the efficiency of joint efforts aimed to the training of high skilled specialists.

Joint **Laboratory for Catalytic Processing of Light Hydrocarbons** of the *Boreskov Institute of Catalysis* (Novosibirsk) and *Institute of Petroleum Chemistry* (Tomsk), created in 2000. Scientific Coordinators are **Prof. Aleksandr S. Noskov** (The Boreskov Institute of Catalysis) and **Dr. Aleksandr V. Vosmerikov** (Institute of Petroleum Chemistry).

Main areas of scientific research of the Laboratory are:

- Development of the Scientific Basis for the Technology for Light Hydrocarbons Catalytic Processing
- Engineering Elaboration of Catalytic Processes and Devices for Light Hydrocarbons Processing
- Study of Combined Catalytic Processes for Oil and Natural Gas Processing.

Laboratory acts in close contact with Institution of Higher Education of Tomsk.

Joint **Laboratory of Catalytic Processes and Apparatuses** of the *Boreskov Institute of Catalysis* and *Novosibirsk State Technical University,* created in 2002. Scientific Coordinators are **Prof. Aleksandr S. Noskov** (The Boreskov Institute of Catalysis) and **Prof. Gennadii G. Kuvshinov** (Novosibirsk State Technical University).

Main areas of scientific research of the Laboratory are:

- Mass and Thermo Transport Processes in Apparatuses for Chemical Engineering
- Design and Development of New Processes and Apparatuses for Energy- and Resource- Saving Chemical Technologies
- Development of the Scientific Basis for the Technology of Filament Carbon Synthesis
- Simulation of Apparatuses with Moving Bed.

Joint **Laboratory of Sorption and Catalytic Processes** of the *Boreskov Institute of Catalysis* (Novosibirsk) and *Butlerov Institute of Chemistry, Kazan State University* (Kazan) created in 2003. Scientific Coordinators are **Prof. Aleksandr S. Noskov** (The Boreskov Institute of Catalysis) and **Prof. Aleksandr A. Lamberov** (Kazan State University).

Main areas of scientific research of the Laboratory are:

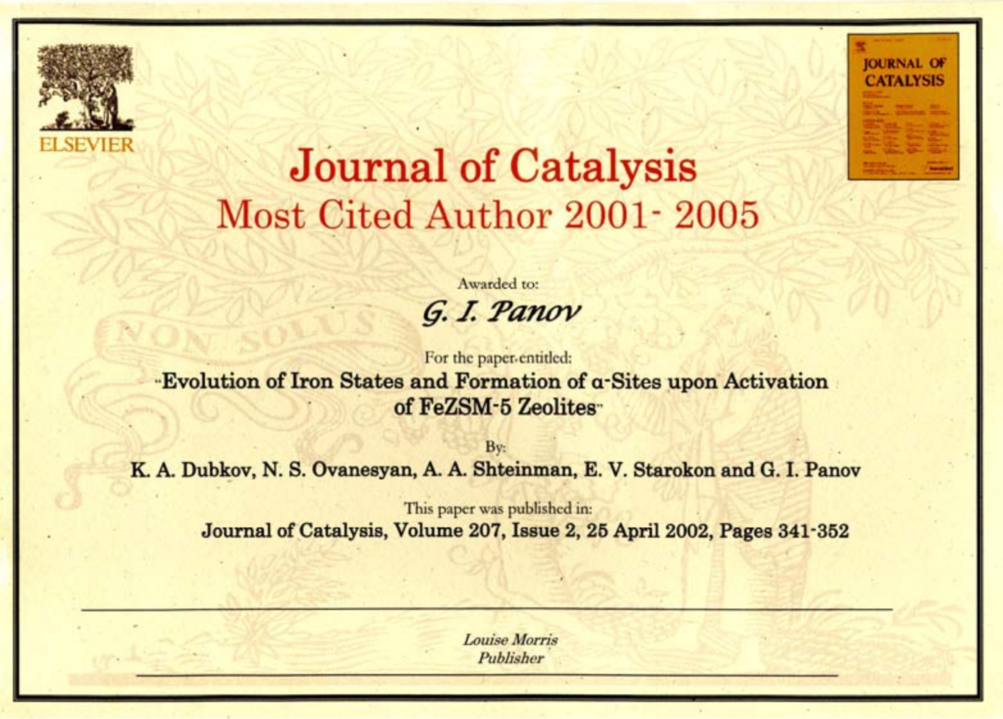
- Design of the Catalysts and Sorbents for Petrochemistry and Oil Refinery in Russia
- Scientific Basis of the Mechanism of Support Texture Formation within Industrial Synthesis and Working-Off
- Comparative Testing of Supports, Sorbents and Catalysts
- Development of New Highly Efficient Energy- and Resource-Saving Technologies for Petrochemistry and Oil Processing.

SCIENTIFIC SOCIAL LIFE

June, St. Petersburg

First vice-premier of RF Government Sergey Ivanov, Minister of Education and Science Andrey Fursenko, Head of the Federal Agency on Science and Innovation Sergey Mazurenko, Head of the federal target program in the field of nanotechnologies Corresponding Member of the RAS Mikhail Kovalchuk are familiarized with the characteristics of “warm skating-rink” produced in Special Economic Zone in Tomsk from ultra-high molecular weight polyethylene (UHMWPE), designed by the Boreskov Institute of Catalysis.

Prof. Gennadii I. Panov was awarded by *Journal of Catalysis* as the Most Cited Author 2001-2005 for the paper entitled *“Evolution of Iron States and Formation of α-Sites upon Activation of FeZSM-5 Zeolites”* by K.A. Dubkov, N.S. Ovanesyan, A.A. Shteinman, E.V. Startokon and G.I. Panov. This paper was published in Journal of Catalysis, volume 207, issue 2, 25 April 2002, Pages 341-352.



ELSEVIER

JOURNAL OF CATALYSIS

Journal of Catalysis
Most Cited Author 2001- 2005

Awarded to:

G. I. Panov

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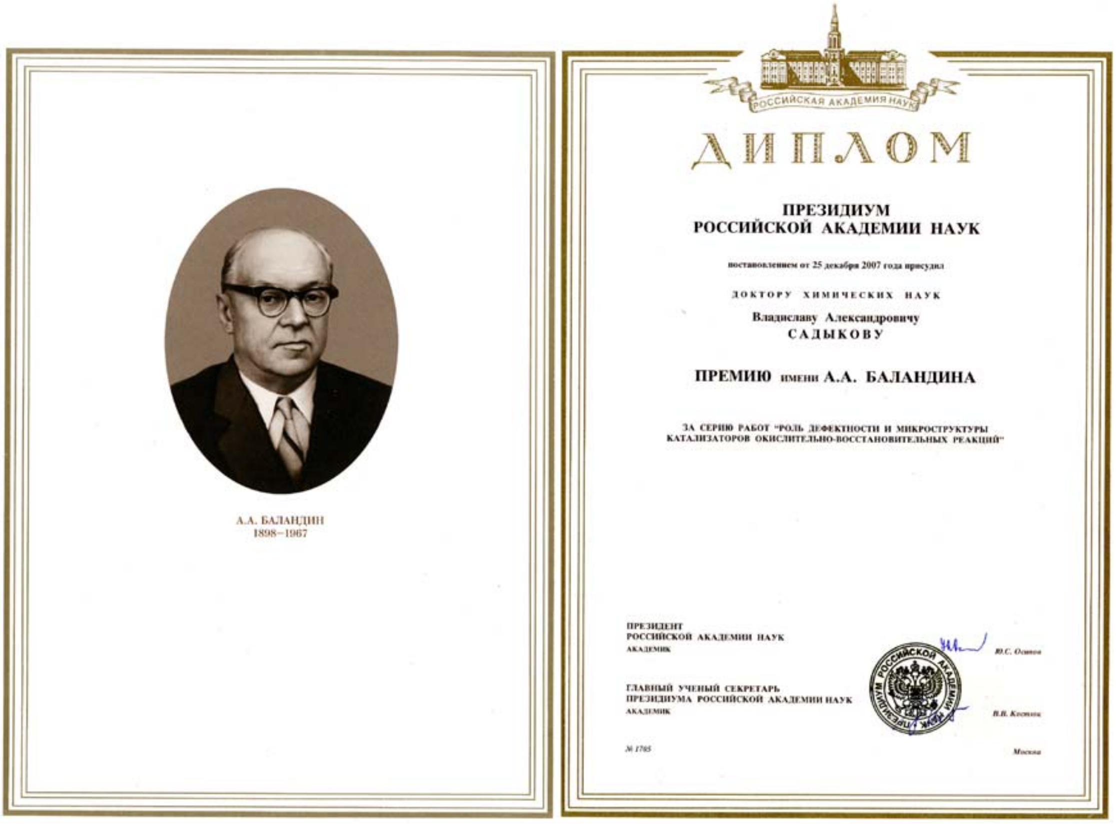
This paper was published in:

Journal of Catalysis, Volume 207, Issue 2, 25 April 2002, Pages 341-352

Louise Morris
Publisher

The **A.A. Balandin**. **Prize 2007** for the outstanding works in the field of catalysis was awarded to **Prof. Vladislav A. Sadykov** for the series of works *“Effect of Deficiency and Microstructure of the Catalysts for Redox Reactions”*.



А.А. БАЛАНДИН
1898–1967

РОССИЙСКАЯ АКАДЕМИЯ НАУК

ДИПЛОМ

ПРЕЗИДИУМ
РОССИЙСКОЙ АКАДЕМИИ НАУК

постановлением от 25 декабря 2007 года присудил

ДОКТОРУ ХИМИЧЕСКИХ НАУК
Владиславу Александровичу
САДЫКОВУ

ПРЕМИЮ имени А.А. БАЛАНДИНА

ЗА СЕРИЮ РАБОТ "РОЛЬ ДЕФЕКТНОСТИ И МИКРОСТРУКТУРЫ КАТАЛИЗАТОРОВ ОКИСЛИТЕЛЬНО-ВОССТАНОВИТЕЛЬНЫХ РЕАКЦИЙ"

ПРЕЗИДЕНТ
РОССИЙСКОЙ АКАДЕМИИ НАУК
АКАДЕМИК
Ю.С. Осипов

ГЛАВНЫЙ УЧЕНЫЙ СЕКРЕТАРЬ
ПРЕЗИДИУМА РОССИЙСКОЙ АКАДЕМИИ НАУК
АКАДЕМИК
В.В. Костюк

№ 1705
Москва

For the merits in fortifying the defense of the Country, outstanding contribution to the development of science and in connection with 50th Anniversary of the Siberian Branch of the Russian Academy of Sciences

- The Boreskov Institute of Catalysis was awarded with the **Order of Alexander Nevsky** (1st class).

- Director of the Boreskov Institute of Catalysis **Academician Valentin N. Parmon** was decorated with the **Order of Lomonosov**.

Anna V. Nartova became the silver winner of the “Samsung Electro-Mechanics 3rd '1nside Edge' International Thesis Competition for the work “*STM/XPS Study of Model Catalysts - Pt Nanoparticles on Alumina*”.

The International Conference *“Molecular and Nanoscale Systems for Energy Conversion”* (October 1-3, 2007, Moscow, Russia) awarded Certificate to **Ekaterina A. Kozlova** for the paper *“Photocatalytic Hydrogen Emission from Water Solution of Organophosphorous Compounds”* by E.A. Kozlova, A.V. Vorontsov, V.N. Parmon.

Dr. Vladimir I. Zaikovskii has received Diploma for the best scientific publication in the journals issued by the International Academic Publishing Company (MAIK) «Nauka/Interperiodica» (2006).

Dr. Ekaterina V. Parkhomchuk is among the winners of the RF Presidential Grant for Young Russian Scientists Competition for the work *“Advanced Oxidation Technology for Treatment of the Water Contraining Organic Pollutants”*.

Integrated Long Term Programme of Cooperation (ILTP) in Science & Technology between India and Russian Federation is one of the most exhaustive bilateral collaborative R&D programmes, covering all aspects of science and technology. ILTP provides for scientist to scientist and institute to institute interaction in frontier areas of science and technology. Main instrument of cooperation is joint R&D projects of mutual interest which are identified by scientists and are supported by the two sides based on their merit. Other modes of cooperation include holding of thematic workshops, organizing exploratory visits of senior scientists, fellowships for young Russian scientists and participation in major scientific events on either side. **Valentin N. Parmon** is Area Coordinator from the Russian side in the field of catalysis.

On the *14th session of Indo-Russian Joint Council for ILPT* **Dr. S. Sivaram,** Director of National Chemical Laboratory, Pune and **V. Parmon** made a joint presentation on the activities in this area. The Joint Council recommended that new proposals for collaboration in basic science area related to environmental catalysis, catalysis of fine chemical processes and catalysis for energy technologies be considered for future collaboration. To facilitate closer understanding between Russia and India, The Joint Council endorses the idea of organizing joint Workshops regularly.

On **August, 4, 1997 Zamaraev International Charitable Scientific Foundation** has been organized in the memory and honor of Professor Kirill I. Zamaraev.

The main Founders of the Foundation from the Russian side are:

- The Boreskov Institute of Catalysis, Novosibirsk
- Institute of Semiconductors Physics, Novosibirsk
- Russian D.I. Mendeleev Chemical Society, Moscow
- Scientific-Industrial Company «Altay», Byisk
- «Ecogeochem Ltd.», Moscow
- Association «Russian House for International Scientific and Technological Cooperation»
- Other scientific, commercial and financial institutions.

The Foundation is supported by IUPAC and some Russian sponsors.

Zamaraev Foundation is grateful to all sponsors for their contribution and invites Russian and foreign organizations (as well as wealthy persons) to become the honorary participants of the campaign.

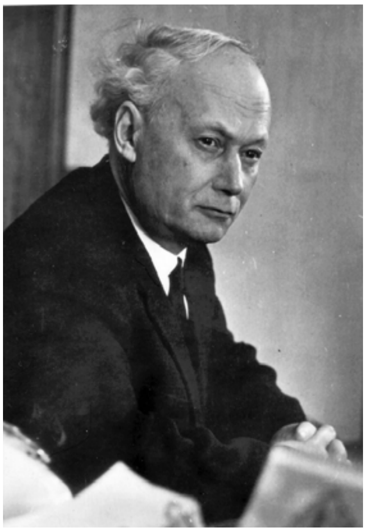
The main aim of the Foundation is to support gifted Russian scientific youth working in the field of chemical catalysis, physical methods of its research and chemical physics.

Major part of Foundation means is spent on Zamaraev post-graduate scholarships, prizes and grants. Foundation plans to support the international cooperation of scientific youth in the field of chemical catalysis and chemical physics, i.e., their participation in international scientific conferences and seminars. In 2007 **S. Kuntsevich and E. Kozlova** have got the post-graduate scholarship**; A. Nadeev, O. Lyakin, A. Shutilov, L. Kibis** - incentive post-graduate scholarships.

- 16 Ph.D. students and scientific employees of the Institute form the personnel of the **Council of Scientific Youth.** The main principle of the Council is submission of the interests of the youth in the Institute. It includes organization of competition of the youth projects, support of the young lecturers of higher educational establishments – employees of the Institute, organization of training for qualifying examinations for the Ph.D. degree, improvement of living conditions of the young scientists, and other directions of activity connected with the improving of a position of the young employees in the Institute.

Memorial Rooms of Academician Georgii K. Boreskov and Academician Kirill I. Zamaraev

The **Memorial Room of Academician Georgii K. Boreskov**, the founder and first director of the Institute of Catalysis, was opened in June 1987, on the occasion of his 80th anniversary. Georgii Konstantinovich always emphasized that the foundation of the Institute was the most important cause of his life. In 1991, the Institute of Catalysis was named after Georgii K. Boreskov.



There is a small exhibition based on the documents, awards, books, Boreskov's belongings in one of the rooms of Laboratory of Oxidation, which was headed by Georgii Konstantinovich.

A series of photos dated 1930's through 1960's tell about an extremely intense and fruitful activity of G.K. Boreskov in the fields of research and science management. He headed the Laboratory of Catalysis at the Chemicoradiological Institute (Odessa) in 1932 through 1937 (the laboratory registry dated 1931 with records by Georgii Konstantinovich concerning the preparation and characterization of vanadium catalysts was kept); the Laboratory of Catalysis at the Research Institute for Fertilizers and Insecto-Fungicides (Moscow) in 1937 through 1946, the Laboratory of Technological Catalysis at the Karpov Institute of Physical Chemistry (Moscow), and in 1958 became the Director of the Institute of Catalysis (Novosibirsk).

For his contribution to the Russian science, distinguished scientific, educational, managing and social activities, his great role in establishing the Siberian Branch of the USSR Academy of Sciences, Georgii Boreskov was awarded the Badge of Honor, three Orders of Lenin, Order of the Red Banner of Labor, Golden Medal of Hammer and Sickle, he was twice USSR State Prize winner, Ukrainian SSR State Prize winner, he was awarded the title of a Hero of Socialist Labor. Among the others, these decorations are exhibited in the Museum.

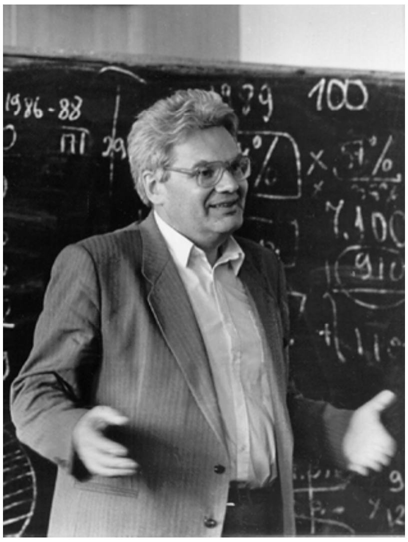
Boreskov's activity in the area of catalysis was recognized worldwide. He was a foreign member of the National Academy of Sciences of German Democratic Republic, honorary member of the New York Academy of Sciences, Doctor HONORIS CAUSA of the Wroclaw Polytechnic Institute (Poland) and of the Poitiers University (France). Government of Bulgarian Republic awarded the Order of Kirill and Mephodium to G.K. Boreskov. Visitors can see an unusual exposition displaying the full attributes of ceremonies of awarding the HONORIS CAUSA titles to Georgii Boreskov in Wroclaw in 1976 and in Poitiers in 1981, these are black mantles and caps, an ermine scarf and leather tubules for storing the Diplomas.

Of particular interest among the exhibit is the personal book collection of Georgii Konstantinovich including more than 1000 scientific volumes and 15 sets of Russian and foreign scientific journals. There are especially memorable copies with dedicatory inscriptions by the authors and the rarities published in the end of XIX and beginning of XX century in the collection. The books capable of telling about Boreskov's educatory activity are displayed in one of the showcases, these are course descriptions and lecture summaries, books used for preparing the lectures, and the like.

There is his writing table, case, bureau and an old push-button typewriter.

Albums, arranged by the colleagues and friends in different periods of time and presented by foreign colleagues, tell about numerous scientific events, where Boreskov presented lectures, about his keenness to winter fishing and to summer “mushroom hunting”, as well as about many other interesting events.

We see the same Georgii Konstantinovich in the portraits and photos, as he was – an intellectual and charming individual.



The **Memorial Room of Academician Kirill I. Zamaraev**, the Director of the Institute of Catalysis from 1984 through 1995, was opened on the 20th of May 1999, the day of 60th anniversary of K. Zamaraev. Establishment of the Memorial Room was promoted by the International Charitable Zamaraev Foundation and directly participated by Kirill Ilyich’s widow, Lyudmila Aleksandrovna Zamaraeva. This small museum exposition relates to the life and activities of Academician Kirill Ilyich Zamaraev, who was a hereditary scientist.

Diplomas, awards, photo archives and belongings of Kirill Ilyich are displayed in showcases. A unique scientific book collection gathered by him is also available here.

Kirill demonstrated his brilliant potentialities in sciences as early as his school years. He finished school as the Golden Medal winner; the medal is also stored in the Museum. He continued his education in the Mendeleev Institute of Chemical Technology in Moscow, but after the third year moved to the Moscow Physical and Technological Institute (MPhTI) in order to get a strong background in chemical physics. In the Museum, there are laboratory registries of post-graduate student Kirill Zamaraev and EPR spectra recorded by him during his post-graduate years.

The Moscow period (1956-1976) of his life and work is illustrated by the photo expositions: here is Kirill Zamaraev as a junior, then senior researcher at the Institute of Chemical Physics of the USSR Academy of Sciences; he heads a laboratory and, at the same time, he is an assistant professor in MPhTI.

Early in 1977 Kirill Ilyich was invited by Academician G.K.Boreskov; with his family and a team of young MPhTI graduates, he moved to Novosibirsk. Owing to the activity of Kirill Zamaraev and his colleagues, investigations of mechanisms of catalytic reactions at the atomic and molecular levels were considerably extended and intensified in the Institute of Catalysis.

He combined his vast research and management activity with teaching in the Novosibirsk State University.

The decade from mid-1980’s to mid-1990’s was the time of international recognition of his scientific achievements. The exhibition demonstrates his state awards, honorary medals of the USSR Academy of Sciences, as well as the certificate of full member of the USSR Academy of Sciences. He became a foreign member of Indian and Korean Academies of Sciences, awarded the Silver Medal by the Royal Chemical Society as an “Outstanding Lecturer of the Century”, and was elected a member of Academy of Europe. In 1994 K.I. Zamaraev became a President of IUPAC, International Union of Pure and Applied Chemistry, and in 1995 was honored with the German prestigious Karpinski Award for Russian Scientists. There are diplomas and medals, as well as a number of photos of that time in the Museum.

Everyone, who knew personally Kirill Ilyich, remember his exceptional ability not to loose courage at any, even very severe, circumstances. He had a really iron character, and it was not without vain that the sport he went in for was mountaineering.

A smiling, talented, brilliant and courageous man appears before visitors to the Memorial Room.

RESEARCH ACTIVITY

Quantum-Chemical Investigations

A COMPUTATIONAL STUDY OF H_2 DISSOCIATION ON SILVER SURFACES: THE EFFECT OF OXYGEN IN THE ADDED ROW STRUCTURE OF Ag(110)

A.B. Mohammad*, K.H. Lim*, I.V. Yudanov, K.M. Neyman*, N. Rösch* (**Technische Universität München, Garching, Germany*)

Phys. Chem. Chem. Phys.
9(10) (2007) pp. 1247-1254.

The activation of H_2 on clean planar (111), (110) and stepped (221) as well as oxygen pre-covered silver surfaces using a density functional slab model approach was studied computationally. In line with previous data clean silver was determined to be inert towards H_2 dissociation, both thermodynamically and kinetically. The reaction is endothermic by approximately 40 kJ mol^{-1} and exhibits high activation energies of approximately 125 kJ mol^{-1}. However, oxygen on the surface, modeled by the reconstructed surface p(2x1)O/Ag(110) that exhibits -O-Ag-O- added rows, renders H_2 dissociation clearly exothermic and kinetically feasible. The reaction was calculated to proceed in two steps: first the H-H bond is broken at an Ag-O pair with an activation barrier E_a approximately 70 kJ mol^{-1}, then the H atom bound at an Ag center migrates to a neighboring O center with E_a approximately 12 kJ mol^{-1}.

MECHANISM OF OLEFIN EPOXIDATION WITH TRANSITION METAL PEROXO COMPLEXES: DFT STUDY

I.V. Yudanov

J. Struct. Chem.,
supply_48 (2007) pp. S111-S124.

Using density functional calculations over the last decade led to considerable progress in understanding the mechanism of olefin epoxidation with Ti, V, Mo, W, and Re peroxo complexes. According to calculations, the reaction occurs by direct electrophilic transfer of one of the atoms of the peroxo group to the olefin. The alternative stepwise mechanism, which has been discussed for a long time and suggested the formation of a metallocyclic intermediate, is characterized by higher activation barriers than direct transfer. The electrophilic character of the direct transfer of oxygen was interpreted at the level of molecular orbital analysis as interaction between the HOMO of the olefin π(C-C) and the LUMO of the peroxo group σ*(O-O). The factors determining the activity of various metal complexes in epoxidation were examined in relation to the ligand environment and the structure of the peroxo group.

ELECTRONIC STRUCTURE AND BONDING OF $\{Fe(PhNO_2)\}^6$ COMPLEXES: A DENSITY FUNCTIONAL THEORY STUDY

O. Isayev*, L. Gorb*, I.L. Zilberberg, J. Leszczynski* (**Jackson State University, Jackson, USA*)

J. Phys. Chem. A,
111(18) (2007) pp. 3571-3576.

Reduction of nitro-aromatic compounds (NACs) proceeds through intermediates with a partial electron transfer into the nitro group from a reducing agent. To estimate the extent of such a transfer and, therefore, the activity of various model ferrous-containing reductants toward NAC degradation, the unrestricted density functional theory (DFT) in the basis of paired Lowdin-Amos-Hall orbitals has been applied to complexes of nitrobenzene (NB) and model Fe(II) hydroxides including cationic $[FeOH]^+$, then neutral $Fe(OH)_2$, and finally anionic $[Fe(OH)_3]^-$. Electron transfer is considered to be a process of unpairing electrons (without the change of total spin projection Sz) that reveals itself in a substantial spin contamination of the unrestricted solution. The unrestricted orbitals are transformed into localized paired orbitals to determine the orbital channels for a particular electron-transfer state and the weights of idealized charge-transfer and covalent electron structures. This approach allows insight into the electronic structure and bonding of the $\{Fe(PhNO_2)\}^6$ unit (according to Enemark and Feltham notation) to be gained using model nitrobenzene complexes. The electronic structure of this unit can be expressed in terms of π-type covalent bonding $[Fe^{+2}(d^6, S = 2) - PhNO_2(S = 0)]$ or charge-transfer configuration $[Fe^{+3}(d^5, S = 5/2) - \{PhNO_2\}^- ((\pi^*)^1, S = 1/2)]$.

DENSITY FUNCTIONAL THEORY STUDIES OF NITROUS OXIDE ADSORPTION AND DECOMPOSITION ON Ga-ZSM-5

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Int. J. Quantum Chem.,
107(13) (2007) pp. 2417-2425.

In this study, density functional theory (DFT/B3LYP) was used to assess a possible reaction pathway for the catalytic dissociation of N_2O. The active centers were taken to be mononuclear $[Ga]^+$ and

$[Ga=O]^+$, and the surrounding portion of the zeolite was represented by a 3T cluster, namely ($AlSi_2O_4H_8$). The first step of N_2O decomposition involves the formation of $[GaO]^+$ and the release of N_2. The metal-oxo species produced in this step then react with N_2O again, to release N_2 and form GaO_2. The calculated activation energies for N_2O dissociation in Ga-ZSM-5 and GaO-ZSM-5 at B3LYP/6-31+G* level are 22.2 and 24.9 kcal/mol, respectively. The calculated energy of the molecular oxygen elimination from 3T-(GaO_2) cluster is ΔH (298 K) = +46.5 kcal/mol and ΔG (298 K) = 35.9 kcal/mol.

SWIFT HOPPING GALLIUM $[AlO_4]^-$ TETRAHEDRA IN Ga/ZSM - A DFT STUDY

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Int. J. Quantum Chem.,
107(13) (2007) pp. 2434-2441.

Density functional theory calculations were carried out to investigate gallium species (Ga^+, $[GaH_2]^+$, and $[GaO]^+$) stabilization in Ga-exchanged HZSM-5, using cluster modeling approach. Three isomeric gallium positions over $[AlO_4]^-$ zeolite fragment at T12 position were found. These isomers are turning into each over with low activation energy barrier and gallium fragment revolves around $[AlO_4]^-$ tetrahedron by hopping between cationic positions. Activation energies of gallium fragment hopping were computed and compared for different gallium containing cations. Those barriers were found to be times less than the activation energies of catalytic processes on gallium-exchanged zeolite.

MODELS OF ACTIVE SITES IN SUPPORTED Cu METAL CATALYSTS IN 1,2-DICHLOROETHANE DECHLORINATION. DFT ANALYSIS

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J. Struct. Chem.,
supply_48 (2007) pp. S160-S170.

It is suggested that a set of discrete Cu nanoclusters satisfying the conditions of structural and electron stability should be used as models of active sites on supported metal catalysts. The close-packed Cu_{20} tetrahedral nanocluster that satisfies these two conditions was taken as a base model of active sites on supported copper catalysts. Theoretical analysis of two possible mechanisms of C-Cl bond dissociation of 1,2-dichloroethane on copper catalysts was performed by the density functional theory method. The first mechanism involves sequential splitting of C-Cl bonds in the molecule in three stages with further stabilization of chloroalkyl intermediates (stepwise mechanism). All these stages are activated. The limiting stage is the one that corresponds to dissociation of the first C-Cl bond with an activation energy of $E^\#$ =34.3 kcal/mol. The second mechanism corresponds to the simultaneous elimination of two chlorine atoms from 1,2-dichloroethane with liberation of ethylene in the gas phase; this is a one-stage process with an activation energy of $E^\#$=26.1 kcal/mol (direct mechanism). A comparison of the two reaction routes shows that the direct mechanism is most probable on copper catalysts.

DFT ANALYSIS OF THE MECHANISM OF 1,2-DICHLOROETHANE DECHLORINATION ON SUPPORTED Cu-Pt BIMETALLIC CATALYSTS

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J. Struct. Chem.,
supply_48 (2007) pp. S171-S183.

The reaction routes of 1,2-dichloroethane dechlorination to ethylene on discrete nanoclusters that served as models of the active sites of supported Cu-Pt catalysts were calculated. Two reaction pathways were predicted. The first route corresponds to sequential elimination of the chlorine atoms from 1,2-dichloroethane; this is a three-stage reaction that occurs via two stable intermediates (stepwise mechanism). The limiting stage is the stage that corresponds to the dissociation of the first C-Cl bond. The second channel corresponds to a simultaneous one-stage elimination of two chlorine atoms (direct mechanism). Both reaction routes are thermodynamically possible, but the stepwise process is more probable, in contrast to the process on monometallic Cu catalysts. For the stepwise process, the vibrational spectra of stable intermediates were calculated for identification of the latter. A set of spectral data characteristic for the stepwise mechanism were determined. The three-step molecular mechanism suggested for 1,2-dichloroethane dechlorination to ethylene is compared with several kinetic schemes known from the literature. Possible modifications of the reaction route that forms ethane and monochloroethane are analyzed.

DENSITY FUNCTIONAL THEORY MOLECULAR CLUSTER STUDY OF COPPER INTERACTION WITH NITRIC OXIDE DIMER IN Cu-ZSM-5 CATALYSTS

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J. Phys. Chem. C,
111(7) (2007) pp. 3080-3089.

The various quantum chemical models of catalytic active site in Cu-ZSM-5 zeolites are analyzed. The density functional theory (DFT) is used to calculate the electronic structure of molecular cluster $(HO)_3$Al-O-Cu-O-Cu modeling the catalytic active site in Cu-ZSM-5 zeolites and study the interaction and decomposition of NO. It is assumed that the rate-determining stage of the low-temperature selective catalytic reduction of NO is the formation of the π-radical $(N_2O_2)^-$ on electron donor sites of Cu-ZSM-5 catalysts. This is in good agreement with the high electron affinity of the molecular dimer ONNO (E_a = -1.5 eV) and is confirmed by the experimental data on the formation of surface anion π-radical $(N_2O_2)^-$ on electron donor sites of supported organo-zirconium surface complex. The DFT calculated electronic structure and excitation energy spectra for the model system $(HO)_3$Al-O-Cu-O-Cu show that it is a satisfactory model for description of experimental UV-vis spectra of Cu-ZSM-5, containing (-O-Cu-O-Cu-) chain structures in the zeolite channels. The calculated reaction energy profile of ONNO adsorption and decomposition on the model catalytic active site shows the possibility of the low-temperature decomposition of dimer $(NO)_2$ with low activation energy and the important role of copper oxide chains (-O-Cu-O-Cu-) in the channels of Cu-ZSM-5 zeolite during selective reduction of NO.

DFT QUANTUM-CHEMICAL CALCULATIONS OF NITROGEN OXIDE CHEMISORPTION AND REACTIVITY ON THE Cu(100) SURFACE

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J. Struct. Chem.,
supply_48 (2007) pp. S147-S159.

A DFT quantum-chemical study of NO adsorption and reactivity on the Cu_{20} and Cu_{16} metal clusters showed that only the molecular form of NO is stabilized on the copper surface. The heat of monomolecular adsorption was calculated to be $\Delta H_m = -49.9$ kJ/mol, while dissociative adsorption of NO is energetically unfavorable, $\Delta H_d = +15.7$ kJ/mol, and dissociation demands a very high activation energy, $E_a = +125.4$ kJ/mol. Because of the absence of NO dissociation on the copper surface, the formation mechanism of the reduction products, N_2 and N_2O, is debatable since the surface reaction ultimately leads to N-O bond cleavage. As the reaction occurs with a very low activation energy, E_a = 7.3 kJ/mol, interpretation of the NO direct reduction mechanism is both an important and intriguing problem because the binding energy in the NO molecule is high (630 kJ/mol) and the experimental studies revealed only *physically adsorbed forms* on the copper surface. It was found that the formation mechanism of the N_2 and N_2O reduction products involves formation (on the copper surface) of the $(O_{ad}N\text{-}NO_{ad})$ dimer intermediate that is *chemisorbed* via the oxygen atoms and characterized by a stable N-N bond ($r_{N\text{-}N}$ ~1.3 Å). The N-N binding between the adsorbed NO molecules occurs through electron-accepting interaction between the oxygen atoms in NO and the metal atoms on the “defective” copper surface. The electronic structure of the $(O_{ad}N\text{-}NO_{ad})$ surface dimer is characterized by excess electron density $(ON\text{-}NO)^{\delta-}$ and high reactivity in N-O_{ad} bond dissociation. The calculated activation energy of the destruction of the chemisorbed intermediate $(O_{ad}N\text{-}NO_{ad})$ is very low (E_a = 5–10 kJ/mol), which shows that it is kinetically unstable against the instantaneous release of the N_2 and N_2O reduction products into the gas phase and cannot be identified by modern experimental methods of metal surface studies. At the same time, on the MgO surface and in the individual $(Ph_3P)_2Pt(O_2N_2)$ complex, a stable $(O_{ad}N\text{-}NO_{ad})$ dimer was revealed experimentally.

QUANTUM CHEMICAL EXAMINATION OF INTERACTION OF CYTOSTATIC FLUOROURACIL WITH DEOXYRIBONUCLEIC ACIDS

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Int. J. Quantum Chem.,
107(13) (2007) pp. 2384-2388.

Within the framework of semiempirical method of quantum chemical PM3, the possibility of formation of paired stack structures under interaction of fluorouracil with pyrimidine and purine nitrogenous bases of nucleotides has been examined. Possible mechanism of transformation of

2-deoxyuridine-5-monophosphate into metabolite-5-fluorin-2-deoxyuridine-5-monophosphate has been given. The calculations that were made allow to suppose that biotransformation of 5-FU in 5-fluorin-2-deoxyuridine-5-monophosphate, most likely, is carried out not in free nucleotides, but in the structure of DNA in two nucleotide triplets UUC and UGU, including the case when directly two nucleotides of deoxyuridine monophosphate, are transformed into 5-fluorin-2-deoxyuridine-5-monophosphate.

Cytostatic ability of 5-FU is increased by its capacity to be selectively embedded into nucleotide triplets creating new chemical compounds that violate matrix RNA formation and accordingly violate protein synthesis.

CHEMICAL BONDING AND ELECTRONIC STRUCTURE OF $LaMnO_3$ AND $La_{0.75}MnO_3$ ORTHORHOMBIC CRYSTALS

V.M. Tapilin

J. Struct. Chem.,
48(2) (2007) pp. 212-218.

The electronic structure of the $LaMnO_3$ orthorhombic crystal of a stoichiometric composition and of $La_{0.75}MnO_3$ crystals with a La vacancy in the unit cell is calculated in the LSDA+U approximation of density functional theory. The calculations showed that $LaMnO_3$ is an insulator with a forbidden gap of 0.5 eV and with antiferromagnetic ordering of magnetic moments. The magnetic moment on the manganese ions is 3.78 BM. The La atom has ionic bonds in the lattice, while the bond between oxygen and manganese is covalent. After lanthanum has been removed, geometry optimization of the unit cell leads to $La_{0.75}MnO_3$ stable structures. In one of the structures, which is lower in energy, the states of manganese may be attributed to Mn^{4+} ions. In both structures with removed lanthanum, the oxygen ions have reduced effective charge, so that one can speak about O^- ions appearing along with O^{2-} in the structure. The oxygen, as well as lanthanum and manganese, ions are nonequivalent in these structures; their nonequivalence is primarily reflected by the local densities of states. This leads to charge and magnetic nonequivalence of ions. In $La_{0.75}MnO_3$ crystals, the degree of bond covalence between manganese and oxygen decreases.

THEORETICAL AND EXPERIMENTAL DAPS STUDY OF CLEAN, OXYGEN, AND HYDROGEN COVERED Pt(100) SINGLE CRYSTAL SURFACE

A.R. Cholach, V.M. Tapilin

Phys. Chem.: An Indian J.,
2(1) (2007) 6 pages.

Local density of states is calculated for the three-fold Pt(100) slab surface covered with various coverage of hydrogen and oxygen. The formation of new surface states region below the platinum conduction band is responsible for covalent contribution to the bond formation between adsorbed species and surface Pt atoms. Resonant states around the substrate Fermi level give an ionic constituent to the adsorbate-surface chemical bond due to considerable DOS difference between adsorbed and surface platinum atoms. Calculated and experimental extended disappearance potential spectra related to different H and O coverage are in a good agreement in spite of baffling complexity of spectral structures. Remarkable similarity between theoretical and experimental results evidences for reliability of obtained data on the LDOS structure of adsorbed system considered.

Monte-Carlo Simulations to Study Physicochemical Processes

HYSTERESIS IN OSCILLATORY BEHAVIOUR IN CO OXIDATION REACTION OVER Pd(110) REVEALED BY ASYNCHRONOUS CELLUAR AUTOMATA SIMULATION

V.I. Elokhin, A.V. Matveev, V.V. Gorodetskii, E.I. Latkin

Lect. Notes Comp. Sci.,
“Parallel Comp. Technol.”,
4671 (2007) pp. 401-409.

The dynamic behaviour of the CO oxidation reaction over Pd(110) has been studied by means of probabilistic asynchronous cellular automata (Dynamic Monte-Carlo). The influence of the internal parameters on the shapes of surface concentration waves obtained in simulations under the limited surface diffusion intensity conditions has been studied. The hysteresis in oscillatory behaviour has been found under step-by-step variation of oxygen partial pressure. Two different oscillatory regimes could exist at one and the same parameters of the reaction. The parameters of oscillations (amplitude, period and the shape of spatio-temporal patterns on the surface) depend on the kinetic prehistory of the system. The possibility for the appearance of the cellular and turbulent patterns, spiral, target and stripe oxygen waves on the surface in the cases under study has been shown.

SIMULATION OF METHANE OXIDATION ON Pt

V.P. Zhdanov, P.-A. Carlsson*, B. Kasemo* (**Chalmers University of Technology, Göteborg, Sweden*)

J. Chem. Phys., 126(23) (2007) pp. 234705 (6 pages).

The authors present a generic model of CH_4 oxidation on Pt with the emphasis on the role of surface-oxide formation. The latter process is treated in terms of the theory of first-order phase transitions. The corresponding Monte Carlo simulations indicate that the surface-oxide formation may result in stepwise features in the reaction kinetics. Specifically, with increasing CH_4 pressure and/or decreasing O_2 pressure, the model predicts a sharp transition from a low-reactive state with the surface completely covered by oxide to a high-reactive state with the surface covered by chemisorbed oxygen. In the former case, the reaction is first order in CH_4 and zero order in O_2. In the latter case, both reaction orders are positive. All these findings help in interpreting available experiments.

LIPID EXCHANGE DURING CONTACT BETWEEN OPPOSITELY CHARGED LIPID BILAYERS

V.P. Zhdanov, B. Kasemo* (**Chalmers University of Technology, Göteborg, Sweden*)

J. Phys. Chem. B, 111(32) (2007) pp. 9428-9430.

The mechanistic details of processes occurring in and during contact between lipid bilayers are still poorly understood due to their complexity on the mesoscopic scale. Here, lipid exchange during contact of oppositely charged lipid bilayers is analyzed. Specifically, a generic mechanism is explored, where this process occurs via diffusion of individual lipids between the layers. The estimates indicate that this scenario is feasible on the time scale of conventional experiments and also on the time scale of biochemical processes in cells.

ENHANCEMENT OF PROTEIN ADSORPTION INDUCED BY SURFACE ROUGHNESS

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Langmuir, 22(26) (2006) pp. 10885-10888.

Using quartz crystal microbalance with dissipation and ellipsometry, it is shown that during adsorption of fibrinogen on evaporated tantalum films the saturation uptake increases with increasing root-mean-square roughness (from 2.0 to 32.9 nm) beyond the accompanying increase in surface area. This increase is attributed to a change in the geometrical arrangement of the fibrinogen molecules on the surface. For comparison, the adsorption of a nearly globular protein, bovine serum albumin, was studied as well. In this case, the adsorption was less influenced by the roughness. Simple Monte Carlo simulations taking into account surface roughness and the anisotropic shape of fibrinogen reproduce the experimentally observed trend.

KINETIC MODEL OF HIV INFECTION

V.P. Zhdanov

J. Exp. Theor. Phys., 105(4) (2007) pp. 856-860.

The recent experiments, clarifying the details of exhaustion of the CD8 T cells specific to various strains of human immunodeficiency virus (HIV), are indicative of slow irreversible (on the one-year time scale) deterioration of the immune system. The conventional models of HIV kinetics do not take this effect into account. Removing this shortcoming, it is shown the likely influence of such changes on the HIV escape from control of the immune system.

CONTROLLED RADIAL DISTRIBUTION OF NANOSCALE VESICLES DURING BINDING TO AN OSCILLATING QCM SURFACE

M. Edvardson*, V.P. Zhdanov, F. Hook** (**Chalmers University of Technology, Göteborg, Sweden; **Lund University, Lund, Sweden*)

Small, 3(4) (2007) pp. 585-589.

The article contains experimental and theoretical data illustrating radial distribution of nanoscale vesicles during binding to an oscillating QCM surface.

EFFECT OF CELL-CELL COMMUNICATION ON THE KINETICS OF PROLIFERATION AND DIFFERENTIATION OF STEM CELLS

V.P. Zhdanov

Chem. Phys. Lett., 437(4-6) (2007) pp. 253-256.

A generic mean-field kinetic model describing the interplay between proliferation, differentiation, death and communication in an ensemble of stem and differentiated cells is presented. The model takes into account the influence of signals between cells on the rate and outcome of division of stem cells. The cellular growth kinetics calculated exhibit three phases, including the initial lag phase, transient exponential growth, and steady state. Due to the effect of cell-cell communication on the division outcome, the growth may be slightly non-monotonous. Under certain conditions, the model predicts stable oscillations as well.

STEM CELL PROLIFERATION AND DIFFERENTIATION AND STOCHASTIC BISTABILITY IN GENE EXPRESSION

V.P. Zhdanov

J. Exp. Theor. Phys., 104(1) (2007) pp. 162-169.

The process of proliferation and differentiation of stem cells is inherently stochastic in the sense that the outcome of cell division is characterized by probabilities that depend on the intracellular properties, extracellular medium, and cell-cell communication. Despite four decades of intensive studies, the understanding of the physics behind this stochasticity is still limited, both in details and conceptually. Here, it is suggest a simple scheme showing that the stochastic behavior of a single stem cell may be related to (i) the existence of a short stage of decision whether it will proliferate or differentiate and (ii) control of this stage by stochastic bistability in gene expression or, more specifically, by transcriptional “bursts”. Monte Carlo simulations indicate that proposed scheme may operate if the number of mRNA (or protein) molecules generated during the high-reactive periods of gene expression is below or about 50. The stochastic-burst window in the space of kinetic parameters is found to increase with decreasing the mRNA and/or regulatory-protein numbers and increasing the number of regulatory sites. For mRNA production with three regulatory sites, for example, the mRNA degradation rate constant may change in the range ± 10%.

EFFECT OF mRNA DIFFUSION ON STOCHASTIC BURSTS IN GENE TRANSCRIPTION

V.P. Zhdanov

JETP Lett., 85(6) (2007) pp. 302-305.

The positive feedback between messenger ribonucleic acid (mRNA) and regulatory-protein production may result in bistability and stochastic "bursts"' in gene transcription. The temporal kinetics of such bursts has already been simulated in detail. It is (i) shown that the applicability of the temporal models may be limited due to relatively slow mRNA and/or protein diffusion and (ii) presented the first 3D Monte-Carlo simulations indicating that, with explicit mRNA diffusion, the bursts may be much more irregular; the periods between bursts may be much shorter, and, depending on the circumstances, the burst window may be reduced or extended.

STOCHASTIC BISTABLE KINETICS OF GENE TRANSCRIPTION DURING THE CELL CYCLE

V.P. Zhdanov

JETP Lett., 84(11) (2007) pp. 632-634.

The positive feedback between messenger ribonucleic acid (mRNA) and regulatory-protein production may result in bistability of gene transcription. If in addition the mRNA and/or protein numbers in a cell are low, one can observe stochastic transcriptional “bursts” provided that the intracellular conditions are steady. Monte Carlo simulations showing what may happen with the bursts under transient conditions during the cell cycle are reported.

OSCILLATORY KINETICS OF GENE TRANSCRIPTION DURING THE CELLULAR GROWTH

V.P. Zhdanov

Techn. Phys. Lett.,
86(7) (2007) pp. 563-566.

The negative feedback between mRNA and regulatory-protein production may result in oscillations in the kinetics of gene transcription. If the mRNA and/or protein number are low, the oscillations may be irregular. Mean-field calculations and Monte-Carlo simulations showing evolution of such oscillations during the cellular growth and division are presented. The oscillations are found to be fairly stable with respect to the cellular growth.

EFFICIENCY OF THE PLASMON-MEDIATED ENHANCEMENT OF PHOTOEXCITATION OF ADSORBATE ON *nm*-SIZED METAL PARTICLES

V.P. Zhdanov, B. Kasemo* (**Chalmers University of Technology, Göteborg, Sweden*)

Chem. Phys. Lett., 433(1-3) (2007) pp. 132-135.

Photoexcitation of molecules adsorbed on a *nm*-sized metal particle can be appreciably enhanced due to plasmon oscillations. The price for this enhancement is characterized by the ratio of the rates of light absorption by plasmons and an adsorbed molecule. The equations are presented making it possible to estimate this ratio for monochromatic light and also in the case when the photon frequency is distributed over an interval which is wider than the plasmon damping width.

SIMULATION OF PROCESSES RELATED TO H_2-O_2 PEM FUEL CELLS

V.P. Zhdanov

J. Electroanal. Chem., 607(1-2) (2007) pp. 17-24.

The understanding of elementary processes occurring in low-temperature H_2–O_2 polymer-electrolyte-membrane (PEM) fuel cells is still limited due to their complexity on atomic and mesoscopic scales. Here, Monte Carlo analysis of two related problems is presented. (i) Due to phase separation of the hydrophobic polymer chains and water, polymer electrolytes such as wetted NAFION are heterogeneous on the scale of about 10 nm and accordingly can be viewed as a porous solid filled by water. One of the physically reasonable models of such pores can be constructed using 3D Ostwald ripening of a lattice gas with attractive interactions. Employing this model, simulations of random-walk diffusion of guest particles in pores were performed. The tracer diffusion coefficient is demonstrated to rapidly decrease with decreasing porosity and near the percolation threshold to be described by the power law. The threshold porosity and the corresponding dynamic exponent are found to be 0.23 ± 0.01 and 0.82 ± 0.01. (ii) The experiments show that the kinetics of O_2 reduction on Pt (with NAFION as electrolyte) is first order in O_2 in a wide range of reaction conditions. In addition, there are indications that during this reaction the atomic-oxygen coverage may be appreciable. These two findings can be adjusted taking into account the effect of repulsive lateral interactions between oxygen adatoms on the reaction kinetics. The surface mobility of these atoms may be low. It is shown that the influence of the latter factor on the reaction kinetics is minor.

OSTWALD RIPENING OF CLOSE-PACKED AND HONEYCOMB ISLANDS DURING COADSORPTION

V.P. Zhdanov

Phys. Rev. B, 76(3) (2007) 033406.

To mimic the self-assembly of hexagonally shaped molecules such as cyanuric acid and melamine, Monte Carlo simulations of the growth of close-packed and honeycomb islands during adsorption and coadsorption of A and B particles on a triangular lattice are presented. Each particle occupies a single site and is characterized by its orientation on the lattice. The orientation is changed by rotation by 60 degrees, so that the rotation by 120 degrees does not change the Hamiltonian. The model predicts slow or nearly negligible island growth at $t \leq 10^3$ Monte Carlo steps (MCSs) and then faster growth. On the late stage ($10^5 \leq t \leq 10^6$ MCSs), the growth exponent is found to be 0.28-0.30 in the case of adsorption of particles of one type and 0.24-0.29 in the case of coadsorption. These values are somewhat or appreciably lower than the Lifshitz-Slyozov exponent $x=1/3$.

KINETICS OF THE N_2O-CO REACTION ON Rh(110)

V.P. Zhdanov, O. Nakagoe*, T. Matsushima* (**Hokkaido University, Sapporo, Japan*)

Surf. Sci., 601(10) (2007) L49-L54.

Recent studies of the steady-state kinetics of the N_2O-CO reaction on Rh(1 1 0) indicate that at CO excess, the reaction rate increases with increasing temperature. At N_2O excess, the reaction rate is nearly independent of temperature at $T < 520$ K and rapidly decreases with increasing temperature at $T > 520$ K. Present analysis of the relevant data indicates that the latter feature seems to be related to surface-oxide formation. Following this line, it is proposed a mean-field kinetic model making it possible to describe and clarify the experiment.

SIMULATION OF THERMAL DESORPTION SPECTRA OF N_2 OBSERVED DURING N_2O DECOMPOSITION ON Rh(100)

V.P. Zhdanov, T. Matsishima* (**Hokkaido University, Sapporo, Japan*)

Surf. Sci.,
601(11) (2007) pp. 2373-2377.

The fulfilled measurements indicate that under temperature-programmed conditions the NO decomposition occurs on Rh(100) between 60 and 140 K and results in the appearance of two N_2 desorption peaks related to N_2 molecules leaving the surface during and after N_2O dissociation events, respectively. Both peaks are observed even at low initial coverages. This and other features of N_2 desorption are explained by using Monte Carlo simulations taking into account attractive N_2O-O lateral interactions stabilizing N_2O adsorption. The results presented are compared with those obtained earlier for Rh(110).

INTERPLAY OF FIRST-ORDER KINETIC AND THERMODYNAMIC PHASE TRANSITIONS IN HETEROGENEOUS CATALYTIC REACTIONS

V.P. Zhdanov, T. Matsishima* (**Hokkaido University, Sapporo, Japan*)

Phys. Rev. Lett., 98(3) (2007) 036101.

Using the rate constants obtained on the basis of independent transient measurements and density functional theory calculations, Monte Carlo (MC) simulations of the bistable kinetics of the N_2O-CO reaction on Pd(110) at 450 K were performed. In the absence of lateral interactions, the MC technique predicts a wide hysteresis loop in perfect agreement with the mean-field analysis. With attractive substrate-mediated lateral interactions resulting in the formation of (1 x 2) O islands and reducing the reaction rate inside islands, the hysteresis is found to be dramatically (about 5 times) narrower. This finding explains why the first-order kinetic phase transition experimentally observed in this reaction is not accompanied by hysteresis.

CRITERION FOR EXTINCTION OF CO OXIDATION IN A CATALYTIC MONOLITH

V.P. Zhdanov

Catal. Lett., 116(1-2) (2007) pp. 23-26.

The extinction of CO oxidation occurring on the Pt catalyst in a monolithic reactor depends on the interplay of the reaction kinetics and mass and heat transfer. In particular, it may be related to a first-order kinetic phase transition from the high-reactive regime to the low-reactive regime. For this scenario, the criterion for extinction are obtained.

SIMULATION OF THE EFFECT OF SURFACE-OXIDE FORMATION ON BISTABILITY IN CO OXIDATION ON Pt-GROUP METALS

V.P. Zhdanov

J. Chem. Phys., 126 (2007) 074706 (9 pages).

The kinetics of CO oxidation on Pt-group metals are known to often exhibit bistability. During the low-reactive regime observed at relatively high CO pressure, the surface is primarily covered by CO and the reaction rate is controlled by O_2 dissociation. During the high-reactive regime at relatively low CO pressure, in contrast, the surface is mainly covered by oxygen and the reaction rate is proportional to CO pressure. In the latter case, the adsorbed oxygen may be in the chemisorbed state and/or may form surface oxide. The experiments indicate that the formation of surface oxide often occurs via the island growth and accordingly should be described in terms of the theory of first-order phase transitions. Here, the author proposes a generic lattice-gas model satisfying this requirement and allowing one to execute the corresponding Monte Carlo simulations. Systematically varying the model parameters determining the oxide stability, he classifies the likely scenarios of the bistable reaction kinetics complicated by oxide formation.

KINETIC EXPLOSION AND BISTABILITY IN ADSORPTION AND REACTION OF ACETIC ACID ON Pd(110)

M. Bowker*, C. Morgan*, V.P. Zhdanov (**School of Chemistry, Cardiff University, Cardiff, UK*)

Phys. Chem. Chem. Phys.,
9(42) (2007) pp. 5700-5703.

The adsorption and reaction of acetic acid with Pd(110) have been studied using thermal molecular beam reaction measurements and temperature-programmed desorption. Acetic acid adsorption results in the formation of acetate species which decompose to produce coincident CO_2 and H_2 desorption from the surface. C is deposited on the surface from the dehydrogenation of the methyl group. In combination, these steps are found to exhibit unusual kinetics including (i) a “surface explosion” during heating and (ii) bistability in the reaction profile for heating and cooling curves. This is the first report of such behaviour for a complex system during *in situ* reaction.

THREE-DIMENSIONAL MONTE CARLO SIMULATIONS OF INTRACELLULAR DIFFUSION AND REACTION OF SIGNALING PROTEINS

V.P. Zhdanov

J. Chem. Phys., 127 (2007) 035101 (4 pages).

It is shown that the Monte Carlo technique makes it possible to perform three-dimensional simulations of intracellular protein-mediated signal transduction with realistic ratio of the rates of protein diffusion and association with genes. Specifically, it is illustrated that in the simplest case when the protein degradation and phosphorylation/dephosphorylationthe are negligible the distribution of the first passage time for this process is close to exponential provided that the number of target genes is between 1 and 100.

BORESKOV-HORIUTI-ENOMOTO RULES FOR REVERSIBLE HETEROGENEOUS CATALYTIC REACTIONS

V.P. Zhdanov

Surf. Rev. Lett., 14(3) (2007) pp. 419-424.

In the middle of the previous century, G.K. Boreskov and independently J. Horiuti and S. Enomoto showed that for reversible reactions, running via a one-route mechanism with a rate-limiting step, there exist general relationships between the reaction rates in the forward and backward directions and also between the corresponding apparent activation energies and reaction heat. Their treatments are formally applicable to gas- and liquid-phase reactions and also to heterogeneous catalytic reactions (HCR) occurring in an ideal overlayer adsorbed on a uniform surface. In reality, HCR often run on heterogeneous surfaces and the HCR kinetics are complicated by adsorbate-adsorbate lateral interactions. It is explicitly demonstrated that in such situations the Boreskov-Horiuti-Enomoto rules are applicable as well.

Studying of Active Sites, Mechanism and Reaction Kinetics

MODERN TRENDS IN THE DEVELOPMENT OF SURFACE SCIENCE AS APPLIED TO CATALYSIS. THE ELUCIDATION OF THE STRUCTURE–ACTIVITY RELATIONSHIPS IN HETEROGENEOUS CATALYSTS

V.I. Bukhtiyarov

Russ. Chem. Rev.,
76(6) (2007) pp. 553-581.

A fundamental approach to the development of heterogeneous catalysts consisting of the study of the structure–activity relationships using surface science methods and techniques is proposed. According to this approach, a study should start with an investigation into well-ordered single crystals under ultrahigh vacuum conditions and then move along the way of both increasing the gas phase pressure (to address the ′pressure gap' problem) and complicating the samples under study from single crystals to supported clusters (to address the ′material gap' problem). The case studies cited in the review prove the efficacy of the proposed approach to investigations into the reactivity of active component particles for the subsequent molecular design of catalysts with improved performance (activity, selectivity and stability) and demonstrate that it conforms with the modern trends in the development of surface science as applied to heterogeneous catalysis.

TRANSIENT RESPONSE STUDY OF THE FORMALDEHYDE OXIDATION TO FORMIC ACID ON V-Ti-O CATALYST: FTIR AND PULSE STUDY

G.Ya. Popova, T.V. Andrushkevich,
Yu.A. Chesalov, V.N. Parmon

J. Mol. Catal.,
268(1-2) (2007) pp. 251-256.

The mechanism of the formaldehyde oxidation to formic acid on a V-Ti-O catalyst has been studied by pulse and spectrokinetic (*in situ* FTIR) methods. Bidentate symmetrical formates and asymmetric formates are observed by formaldehyde adsorption both in the presence and in the absence of dioxygen in the temperature range between 100 and 200°C. Formic acid may form by two parallel pathways: by an associative mechanism through the oxidative elimination of bidentate formates and by a stepwise redox Mars – van Krevelen mechanism through the decomposition of asymmetric formates; the associative mechanism seems to be preferable.

INVESTIGATION OF THE REACTION MECHANISM OF METHYLPYRAZINE AMMOXIDATION ON VANADIA-TITANIA CATALYST BY FTIR *IN SITU*

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E.A. Paukshtis, N.A. Paukshtis, A.A. Budneva,
V.N. Parmon

J. Mol. Catal. A.,
269(1-2) (2007) pp. 240-245.

The mechanism of methylpyrazine ammoxidation on a vanadia-titania catalyst has been studied by *in situ* FTIR spectroscopy. The structure of surface intermediates has been identified and the sequence of their transformation in the temperature range of 150-230°C has been ascertained. The interaction of methylpyrazine with catalyst surface includes a consecutive transformation of coordinatively bound methylpyrazine into oxygenated surface compounds, viz., an aldehyde-like complex and an asymmetrical carboxylate. The main reaction product, amidopyrazine, is formed through the interaction of the surface oxyintermediates with adsorbed ammonia species.

HIGH-TEMPERATURE CARBOXIDATION OF CYCLOPENTENE WITH NITROUS OXIDE

E.V. Starokon, K.S. Shubnikov, K.A. Dubkov, A.S. Kharitonov, G.I. Panov

Kinet. Catal., 48(3) (2007) pp. 376-380.

It is demonstrated by the example of cyclopentene that the noncatalytic oxidation of alkenes with nitrous oxide to carbonyl compounds (carboxidation), which is known to occur in the liquid phase at 150–250°C, can also take place in the gas phase at higher temperatures (300–475°C). Like liquid-phase carboxidation, the gas-phase reaction likely proceeds via a dipolar 1,3-addition mechanism. However, 4-pentenal is formed along with cyclopentanone in the gas phase. The 4-pentenal selectivity increases from 2.5 to 23% as the reaction temperature is raised. High-temperature cyclopentene carboxidation can be carried out in a paraffin melt (bp ~ 400°C). Filling the reactor with paraffin accelerates the reaction and reduces its activation energy.

LIQUID-PHASE NONCATALYTIC OXIDATION OF MONOTERPENOIDS WITH NITROUS OXIDE

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Russ. Chem. Bull.,
56(6) (2007) pp. 1239-1243.

A series of monoterpenoids differing in the number of double bonds and the pattern of their substitution were tested in the liquid-phase noncatalytic oxidation with nitrous oxide (N_2O). The structure of olefins has a significant effect on the oxidation route. In the case of terpenoids containing 1,1-disubstituted double bond, *nor*-carbonyl compounds are formed with high selectivity.

INVESTIGATION OF UNSTEADY-STATE REDOX MECHANISMS OVER CERIA BASED CATALYSTS FOR PARTIAL OXIDATION OF METHANE

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Stud. Surf. Sci. Catal.,
167 (2007) pp. 287-292.

The dynamics of transient experiments carried out on nanocrystalline CeO_2-ZrO_2 and Pt/CeO_2-ZrO_2 materials in POM appear to be associated with i) at least two types of oxygen forms differing by their reactivity and ii) a fast surface/bulk oxygen diffusion, which might be determined by a high density of domain boundaries and Frenkel-type defects. The profile of these transients is clearly related to the state of the catalyst (preoxidized or pre-reduced). The fast oxygen transfer from the support to Pt sites favors preferential routes for POM reactions over Pt clusters, although the mixed oxide alone displays a reasonably high activity. Domain boundaries might be involved also in storage of carbonates and hydroxyls which affects observed transients as well. All these mechanistic conclusions provide new trends for designing catalysts specifically adapted to applications involving essentially transient regimes.

CERIA-BASED FLUORITE-LIKE OXIDE SOLID SOLUTIONS PROMOTED BY PRECIOUS METALS AS CATALYSTS OF METHANE TRANSFORMATION INTO SYNGAS

V.A. Sadykov, T.G. Kuznetsova, G.M. Alikina, Yu.V. Frolova, A.I. Lukashevich, V.S. Muzykantov, V.A. Rogov, L.Ch. Batuev, V.V. Kriventsov, D.I. Kochubey, E.M. Moroz, D.A. Zyuzin, E.A. Paukshtis, E.B. Burgina, S.N. Trukhan, V.P. Ivanov, L.G. Pinaeva, Yu.A. Ivanova, V.G. Kostrovskii*, S. Neophytides, E. Kemnitz***, K. Scheurel***, C. Mirodatos****** (**Institute of Solid-State Chemistry and Mechanochemistry, Novosibirsk, Russia; **Institute of Chemical Engineering and High-Temperature Processes, Patras, Greece; ***Humboldt University, Berlin, Germany; ****Institut de Recherches sur la Catalyse CNRS, Villeurbanne, France*)

In "*New Topics in Catalysis Research*",
Ed. D. McReynolds, 2006,
Nova Science Publishers Inc., pp. 97-196.

For catalysts comprised of Pt supported onto dispersed complex fluorite-like oxides (ceria doped by Pr, Gd, Sm, or CeO_2-ZrO_2 doped by La, Gd or Pr), the effects of the oxygen mobility in supports and Pt dispersion on the performance in methane selective oxidation into syngas at short contact times were elucidated using combination of kinetic and spectroscopic methods. While in general any simple universal relation between the oxygen mobility, Pt dispersion and the rate of methane transformation into syngas was not found, for some series, a good correlation was observed agreeing with the bifunctional scheme of the methane selective oxidation into syngas.

METHANE OXIDATION OVER MIXED-CONDUCTING $SrFe(Al)O_{3-\delta}$–$SrAl_2O_4$ COMPOSITE

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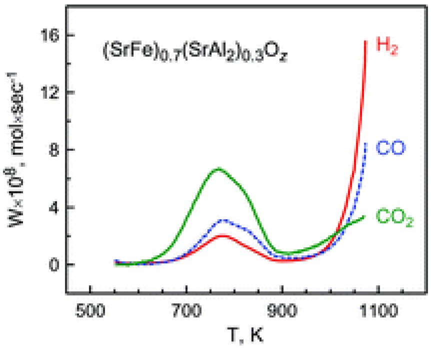
Phys. Chem. Chem. Phys., 9(21) (2007) pp. 2744-2752.

The steady-state CH_4 conversion by oxygen permeating through mixed-conducting $(SrFe)_{0.7}(SrAl_2)_{0.3}O_z$ composite membranes, comprising strontium-deficient $SrFe(Al)O_{3-\delta}$ perovskite and monoclinic $SrAl_2O_4$-based phases, occurs *via* different mechanisms in comparison to the dry methane interaction with the lattice oxygen. The catalytic behavior of powdered $(SrFe)_{0.7}(SrAl_2)_{0.3}O_z$, studied by temperature-programmed reduction in dry CH_4 at 523–1073 K, is governed by the level of oxygen nonstoichiometry in the crystal lattice of the perovskite component and is qualitatively similar to that of other perovskite-related ferrites, such as $Sr_{0.7}La_{0.3}Fe_{0.8}Al_{0.2}O_{3-\delta}$. While extensive oxygen release from the ferrite lattice at 700–900 K leads to predominant total oxidation of methane, significant selectivity to synthesis gas formation, with H_2/CO ratios close to 2, is observed above 1000 K, when a critical value of oxygen deficiency is achieved. The steady-state oxidation over dense membranes at 1123–1223 K results, however, in prevailing total combustion, particularly due to excessive oxygen chemical potential at the membrane surface. In combination with surface-limited oxygen permeability, mass transport limitations in a porous layer at the membrane permeate side prevent reduction and enable stable operation of $(SrFe)_{0.7}(SrAl_2)_{0.3}O_z$ membranes under air/methane gradient. Taking into account the catalytic activity of $SrFeO_{3-\delta}$-based phases for the partial oxidation of methane to synthesis gas and the important role of mass transport-related effects, one promising approach for membrane development is the fabrication of thick layer of porous ferrite-based catalyst at the surface of dense $(SrFe)_{0.7}(SrAl_2)_{0.3}O_z$ composite.



METHANE OXIDATION BY LATTICE OXYGEN OF $CeNbO_{4+\delta}$

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Catal. Commun., 8(3) (2007) pp. 335-339.

The reactivity of methane with lattice oxygen of cerium niobate, $CeNbO_{4+\delta}$, was studied by temperature-programmed reduction (TPR) in dry CH_4 flow at 523–1073 K. Phase transformations and reduction of cerium niobate at 900–1023 K lead to a massive release of hyperstoichiometric oxygen, in amounts determined by the intermediate-temperature phase composition dependent on thermal history. In this temperature range, CH_4–TPR shows prevailing formation of carbon monoxide and steam, suggesting that the synthesis gas generation occurs in parallel with extensive oxidation of H_2 on the cerium niobate surface. At 1073 K when $\delta \to 0$, the reaction of methane with $CeNbO_{4+\delta}$ selectively yields synthesis gas with H_2/CO ratio close to two.

METHANE CARBONYLATION WITH CO ON SULFATED ZIRCONIA: EVIDENCE FROM SOLID-STATE NMR FOR THE SELECTIVE FORMATION OF ACETIC ACID

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J. Phys. Chem. C, 111(28) (2007) pp. 10624-10629.

Using ^{13}C and ^{1}H solid-state NMR it has been shown, that methane can be carbonylated with carbon monoxide to give acetic acid on solid acid catalyst, sulfated zirconia. The carbonylation occurs at 473-573 K with high selectivity and essential conversion. The reaction proceeds both in the absence and in the presence of molecular oxygen. In the presence of oxygen, the catalyst can be used for the carbonylation of further portion of methane without reactivation in air. The mechanism of the reaction is discussed. The reaction observed opens up new possibilities of using sulfated zirconia-based solid catalysts for the synthesis of acetic acid from methane and carbon monoxide.

LIQUID-PHASE OXIDATION OF α-PINENE WITH OXYGEN CATALYZED BY CARBON-SUPPORTED PLATINUM METALS

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Kinet. Catal., 48(1) (2007) pp. 38-44.

The liquid-phase oxidation of α-pinene with oxygen at 70–90°C is studied in the presence of Pd, Pt, Ru, Rh, and Ir supported on carbon. The conversion of α-pinene and the selectivity of formation of the main reaction products, namely, verbenol (**1**), verbenone (**2**), and α-pinene oxide (**3**), depends on the nature of the metal, on its oxidation state and extent of dispersion, and on the admixtures introduced into the system. In the presence of the Pt catalysts and promoting admixtures of tetrahexylammonium chloride (Hex_4NCl), the selectivity of formation of the most valuable oxidation products **(1 + 2)** reaches 50% at an α-pinene conversion of 20–30%. The fraction of resinlike oxidation products decreases in the presence of the catalysts. The results obtained are discussed in the framework of the radical mechanism of α-pinene oxidation.

CATALYTIC HYDRODECHLORINATION ON PALLADIUM-CONTAINING CATALYSTS

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Kinet. Catal.,
48(1) (2007) pp. 60-66.

The catalytic liquid-phase hydrodechlorination of chlorobenzene on supported palladium-containing catalysts has been investigated. The following processes capable of deactivating the catalyst occur during the liquid-phase hydrodechlorination: the coarsening of supported metal particles, the washoff of the active component with the reaction medium, and potassium chloride deposition on the catalyst surface. The effects of the active component composition and of the preparation method on the hydrodechlorination activity and deactivation stability of the catalysts have been studied. The catalysts have been characterized by several physical methods.

EXPERIMENTAL STUDY OF THE GAS-PHASE HYDROFLUORINATION OF PERCHLOROETHYLENE OVER A CHROMIUM-BASED CATALYST

A.A. Zirka, S.I. Reshetnikov

React. Kinet. Catal. Lett.,
90(1) (2007) pp. 213-214.

Gas-phase hydrofluorination of perchloroethylene to pentafluoroethane in the presence of a chromium-magnesium catalyst at 0.4 MPa and 330-390°C has been studied. A reaction scheme taking into account the formation of by-products is suggested.

Ti- AND Zr-MONOSUBSTITUTED POLYOXOMETALATES AS MOLECULAR MODELS FOR STUDYING MECHANISMS OF OXIDATION CATALYSIS

O.A. Kholdeeva, R.I. Maksimovskaya

J. Mol. Catal. A: Chem.,
262(1-2) (2007) pp. 7-24.

Recent developments in the application of titanium- and zirconium-monosubstituted Keggin-type polyoxometalates (Ti- and Zr-POMs) as molecular models for studying mechanisms of H_2O_2-based selective oxidation catalysis are reviewed. The similarity in the catalytic behavior of Ti- and Zr-POMs and heterogeneous Ti(IV) and Zr(IV) single-site selective oxidation catalysts is demonstrated. The progress on the synthesis of Ti- and Zr-POMs, studying their interaction with H_2O and H_2O_2, and characterization of the peroxo species formed are surveyed, with special emphasis being placed on the role of protons in the activation of the peroxo species.

THE NATURE OF AUTOCATALYSIS IN THE BUTLEROV REACTION

A.N. Simonov, O.P. Pestunova, L.G. Matvienko, V.N. Parmon

Kinet. Catal., 48(2) (2007) pp. 245-254.

The effect of the nature of an initiator on the kinetics of formaldehyde consumption and on product composition in the Butlerov reaction was studied in a stirred flow reactor and a batch reactor. It was found that, under flow conditions, the kinetics and the product composition of this reaction are independent of the nature of the initiator. The reaction schemes proposed previously for an autocatalytic process mechanism based on the formation of glycolaldehyde from two formaldehyde molecules are incorrect. A correlation between the initiating activities of various monosaccharides and the rates of their conversion into an enediol form was found with the use of a batch reactor. Solid enediol complexes with Ca^{2+} ions were isolated for glucose, fructose, ribose, and sorbose; the initiating activity of these complexes was found to be much higher than the initiating activity of pure monosaccharides. A self-consistent mechanism was proposed for Butlerov reaction initiation. The formation of the enediol forms of monosaccharides followed by degradation to lower carbohydrates plays a key role in this mechanism. In turn, the initiating activity depends on the position of the carbonyl group in the monosaccharide molecule. The condensation reactions of glycolaldehyde, glyceraldehyde, and dihydroxyacetone with each other were studied. Based on data on the condensation products of lower carbohydrates, a scheme was proposed for the Butlerov reaction. According to this reaction scheme, C_2 and C_3 carbohydrates mainly undergo an aldol condensation reaction with formaldehyde, whereas the formation of higher monosaccharides occurs by the aldol condensation of lower C_2–C_3 carbohydrates with each other.

SYMMETRIC OXIDATION OF SULFIDES WITH H_2O_2 CATALYZED BY TITANIUM COMPLEX AMINOALCOHOL DERIVED SCHIFF BASES

K.P. Bryliakov, E.P. Talsi

J. Mol. Catal. A.,
264(1-2) (2007) pp. 280-287.

Sulfoxidation catalysts generated *in situ* from titanium(IV) isopropoxide and enantiopure Schiff bases promote the enantioselective oxidation of alkyl aryl sulfides to the corresponding sulfoxides at low catalyst loadings (<1 mol%), 30% aqueous hydrogen peroxide being the terminal oxidant. Upon screening of several ligands derived from β-aminoalcohols and salicylaldehydes, a catalyst affording sulfoxides with over 90% chemoselectivity and up to 60% ee was found, and the kinetics of the catalytic reaction was analyzed by ^{1}H NMR.

THE ACTIVE INTERMEDIATES OF NON-HEME-IRON-BASED SYSTEMS FOR CATALYTIC ALKENE EPOXIDATION WITH H_2O_2/CH_3COOH

E.A. Duban, K.P. Bryliakov, E.P. Talsi

Eur. J. Inorg. Chem.,
6 (2007) pp. 852-857.

Iron complexes with aminopyridine ligands $[Fe^{II}(BPMEN)(CH_3CN)_2](ClO_4)_2$ (1) (BPMEN = N,N'-dimethyl-N,N′-bis(2-pyridylmethyl)-1,2-diaminoethane) and $[Fe^{II}(TPA)(CH_3CN)_2](ClO_4)_2$ (2) (TPA = tris(2-pyridylmethyl)amine) are rare examples of iron-based catalysts for selective olefin epoxidation with H_2O_2/CH_3COOH combination. Using ^{1,2}H NMR and EPR spectroscopy, it was shown that the active species of the catalytic systems $1/H_2O_2/CH_3COOH$ and $2/H_2O_2/CH_3COOH$ are mononuclear iron(IV)-oxo complexes $[(BPMEN)Fe^{IV}{=}O(S)]^{2+}$ and $[(TPA)Fe^{IV}{=}O(S)]^{2+}$, respectively (S = solvent molecule). These intermediates are formed via decomposition of the corresponding acylperoxo complexes $[(L)Fe^{III}(O_3CCH_3)]^{2+}$ (L = BPMEN or TPA). The acylperoxo complex $[(BPMEN)Fe^{III}(O_3CCD_3)]^{2+}$ was identified in the catalytic system $1/H_2O_2/CD_3COOD$ by ^{2}H NMR spectroscopy. The improved epoxidation activity and selectivity of the catalytic systems $1/H_2O_2/CH_3COOH$ and $2/H_2O_2/CH_3COOH$, with respect to $1/H_2O_2$ and $2/H_2O_2$, is caused by the effective formation of intermediates $[(L)Fe^{IV}{=}O(S)]^{2+}$ via decomposition of the acylperoxo complexes $[(L)Fe^{III}(O_3CCH_3)]^{2+}$.

IRON CATALYZED OXIDATION OF THIOETHERS BY IODOSYLARENES: STEREOSELECTIVITY AND REACTION MECHANISM

K.P. Bryliakov, E.P. Talsi

Chem. Eur. J., 13(28) (2007) pp. 8045-8050.

Catalytic properties of a series of iron(III)-salen (salen=N,N'-bis(salicylidene)ethylenediamine dianion) and related complexes in asymmetric sulfoxidation reactions, with iodosylarenes as terminal oxidants, have been explored. These catalysts have been found to efficiently catalyze oxidation of alkyl aryl sulfides to sulfoxides with high chemoselectivity (up to 100 %) and moderate-to-high enantioselectivity (up to 84 % with isopropylthiobenzene and iodosylmesitylene), the TON (TON=turnover number) approaching 500. The influence of the ligand (electronic and steric effects of the substituents), oxidant, and substrate structures on the oxidation stereoselectivity has been investigated systematically. The structure of the reactive intermediates (complexes of the type $[Fe^{III}(ArIO)(salen)]$ and the reaction mechanism have been revealed by both mechanistic studies with different iodosylarenes and direct in situ ^{1}H NMR observation of the formation of the reactive species and its reaction with the substrate.

MECHANISM OF THE REACTION NO + H_2 ON THE Pt(100)-HEX SURFACE UNDER CONDITIONS OF THE SPATIALLY NONUNIFORM DISTRIBUTION OF REACTING SPECIES

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Kinet. Catal.,
48(6) (2007) pp. 853-863.

The interaction of hydrogen with $NO_{ads}/1{\times}1$ islands produced by NO adsorption on the reconstructed surface Pt(100)-hex was studied by high-resolution electron energy loss spectroscopy (HREELS) and the temperature-programmed reaction (TPR) method. The islands are areas of the unreconstructed surface Pt(100)-1 × 1 saturated with NO_{ads} molecules. The hexagonal phase around these islands adsorbs much more hydrogen near room temperature than does the clean Pt(100)-hex surface. It is assumed that hydrogen is adsorbed on the hexagonal surface areas that are adjacent to, and are modified by, the $NO_{ads}/1 \times 1$ islands. The reaction of adsorbed hydrogen atoms with NO_{ads} takes place upon heating and has the character of so-called surface explosion. The TPR peaks of the products of this reaction—nitrogen and water—occur at T_{des}~ 365–370 K, their full width at half-maximum being ~5–10 K. In the case of the $NO_{ads}/1 \times 1$ islands preactivated by heating in vacuo above the NO desorption onset temperature (375–425 K), after the admission of hydrogen at 300 K, the reaction proceeds in an autocatalytic regime and the product formation rate increases monotonically at its initial stage. In the case of activation at 375 K, during the initial, slow stage of the reaction (induction period), hydrogen reacts with nitric oxide molecules bound to structure defects (NO_{def}). After activation at 425 K, the induction period is characterized by the formation and consumption of imido species (NH_{ads}). It is assumed that NH_{ads} formation involves N_{ads} atoms that have resulted from NO_{ads} dissociation on defects upon thermal activation. The induction period is followed by a rapid stage of the reaction, during which hydrogen reacts with $NO_{1{\times}1}$ molecules adsorbed on 1×1 areas, irrespective of the activation temperature. After the completion of the reaction, the areas of the unreconstructed phase 1×1 are saturated with adsorbed hydrogen. The formation of H_{ads} is accompanied by the formation of a small amount of amino species (NH_{2ads}).

REACTION OF CO OXIDATION ON PLATINUM, RHODIUM, A PLATINUM-RHODIUM ALLOY, AND A HETEROPHASE BIMETALLIC PLATINUM/RHODIUM SURFACE

A.V. Kalinkin, A.V. Pashis, V.I. Bukhtiyarov

Kinet. Catal., 48(2) (2007) pp. 298-304.

The reaction of CO oxidation on thin metal films of platinum, rhodium, and their alloy and on a heterophase bimetallic Pt/Rh surface that consisted of platinum particles of size 10–20 nm on the surface of rhodium was studied in the region of low reactant pressures (lower than 2×10^{-5} mbar). At low temperatures (T < 200°C), the activity of samples increased in the order Rh > Pt/Rh > Pt–Rh alloy > Pt. Above 200°C, the rate of reaction on the heterophase Pt/Rh surface was almost twice as high as the sum of the rates of reaction on the individual metals; this fact is indicative of a synergistic effect. The nature of this effect is considered.

THE MOLECULAR MECHANISM OF LOW-TEMPERATURE DECOMPOSITION OF HYDROGEN SULFIDE UNDER CONJUGATED CHEMISORPTION-CATALYSIS CONDITIONS

I.I. Zakharov, A.N. Startsev, O.V. Voroshina,
A.V. Pashigreva, N.A. Chashkova, V.N. Parmon

Russ. J. Phys. Chem. A,
80(9) (2006) pp. 1403-1410.

The molecular mechanism of interaction of two hydrogen sulfide molecules with the $(Co^{III}\text{-}H_o)_2S_2(SH_2)_4$ model active center containing occluded hydrogen was studied by the density functional theory method with the B3P86 hybrid exchange-correlation functional. The reaction was found to occur in the following elementary steps: molecular adsorption of hydrogen sulfide ⇒ dissociative chemisorption ⇒ S-S bond formation in the surface intermediate $\{2Co^{III} - (\mu\text{-}S_2) + 2H(ads)\}$ with the release of the first hydrogen molecule into the gas phase $H_2(g)$ ⇒ the release of the second hydrogen molecule into the gas phase $H_2(g)$ ⇒ the formation of cyclooctasulfur in the reaction $4S_2(ads) \rightarrow S_8(ads)$. The first three steps occur spontaneously at room temperature, the thermodynamic driving force of the process being the stoichiometric reaction of S-S bond formation at the stage of conjugated chemisorption of two hydrogen sulfide molecules on two adjacent metal ions with the release of the first hydrogen molecule into the gas phase. The catalytic cycle is terminated by the recombination of molecular sulfur S_2 into cyclooctasulfur S_8 in the adsorption layer and the release of the second hydrogen molecule into the gas phase.

EFFECTS OF THE PROPERTIES OF SO_4/ZrO_2 SOLID CATALYSTS ON THE PRODUCTS OF TRANSFORMATION AND REACTION MECHANISM OF *R*-(+)-LIMONENE DIEPOXIDES

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J. Mol. Catal. A: Chem., 269(1-2) (2007) pp. 72-80.

Transformations of limonene diepoxides on solid SO_4/ZrO_2 catalysts in dried methylene chloride at room temperature were investigated by varying the amounts of supported sulfate ions and zirconia polymorphs. A method for estimating acid centers from DRIFTS spectra of adsorbed pyridine on the solid catalyst in a dried methylene chloride solution has been developed. It is shown that the mechanism of diepoxide transformations is changed by varying the type and amount of acid centers. At a 0.9–3% level of sulfation, Lewis centers are mainly responsible for the initial cleavage of the 8,9-epoxy group of limonene diepoxides (route B). At higher contents of sulfate ions (up to 30 wt.% SO_4), the key transformations proceed with cleavage of the 1,2-epoxy group (route A) and involve the Brønsted centers. The type of support affects the structural features of the sulfate ions, which govern the rearrangement of limonene diepoxides by route A or B. Transformation of limonene diepoxides on the Lewis centers formed on sulfated alumina follows mainly route A. The type of acid centers also affects the stereochemical composition of the product mixture.

VANADIUM SPECIES IN NEW CATALYSTS FOR THE SELECTIVE OXIDATION OF METHANE TO FORMALDEHYDE: ACTIVATION OF THE CATALYTIC SITES

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Catal. Today, 128(3-4) (2007) pp. 176-182.

New vanadium oxide supported on mesoporous silica catalysts for the oxidation of methane to formaldehyde were investigated by infrared and Raman spectroscopies to identify and characterize the molecular structure of the most active and selective catalytic sites. *In situ* and operando experiments have been conducted in order to understand the redox and hydroxylation/dehydroxylation processes of the vanadium species. $(SiO)_2VO(OH)$ species were identified in these catalysts in reaction conditions and shown to undergo a deprotonation at 580°C under vacuum, leading to a site giving a photoluminescence band at 550 nm attributed to reverse radiative decay from the excited triplet state: $V^{4+}–O^{-})^{*} \leftrightarrows (V^{5+}=O^{2-})$. An activation mechanism of vanadium monomeric species with electrophilic oxygen species is proposed.

STATE OF PALLADIUM IN PALLADIUM-ALUMINOSILICATE CATALYSTS AS STUDIED BY XPS AND THE CATALYTIC ACTIVITY OF THE CATALYSTS IN THE DEEP OXIDATION OF METHANE

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Kinet. Catal., 48(5) (2007) pp. 728-734.

Palladium catalysts based on Siralox and AS aluminosilicate supports for the deep oxidation of methane were studied. With the use of XRD analysis, it was found that they were heterophase systems consisting of an amorphous aluminosilicate and γ-Al_2O_3 stabilized against agglomeration. It was found that the catalytic activity of palladiumaluminosilicate catalysts in the deep oxidation of methane at 500°C depended on the support precalcination temperature. X-ray photoelectron spectroscopy (XPS) was used to study the states of the AS-30 aluminosilicate support calcined at 600, 800, or 1000°C and palladium supported on it. It was found that the action of an acid impregnation solution of palladium nitrate on the aluminosilicate calcined at 800°C resulted in a structural rearrangement of the aluminosilicate surface. This rearrangement resulted in the stabilization of both palladium oxide and palladium metal particles at surface defects and the incorporation of these particles into the aluminosilicate after catalyst calcination. As a result, an anomalous decrease in catalytic activity was observed in aluminosilicate samples calcined at 800°C. According to XPS data, palladium in the catalyst was stabilized in the following three phases: metal ($E_b(Pd3d_{5/2})$ = 334.8 eV), oxide ($E_b(Pd3d_{5/2})$ = 336.8 eV), and "interaction" ($E_b(Pd3d_{5/2})$ = 335.8 eV) phases. The ratio between these phases depended on support and catalyst calcination temperatures. The interaction phase, which consisted of PdO_x clusters stabilized in the aluminosilicate structure, was responsible for the retention of activity after calcination at high temperatures (800°C). Based on an analysis of XPS data, it was hypothesized that palladium in the interaction phase occurred in a charged state with the formal charge on the Pd atom close to 1+ (δ+ phase).

ON THE INVOLVEMENT OF RADICAL OXYGEN SPECIES O^- IN CATALYTIC OXIDATION OF BENZENE TO PHENOL BY NITROUS OXIDE

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J. Catal., 245(2) (2007) 466-469.

Catalytic oxidation of benzene to phenol by nitrous oxide over Fe-MFI zeolites was studied in relation to the active oxygen species taking part in the oxidation. A linear dependence of the reaction rate on the concentration of independently identified active sites generating O^- radicals (α-sites) has been obtained within a broad range of values. The dependence is interpreted as a convincing evidence of the O^- involvement in the catalytic (not only stoichiometric) oxidation of benzene to phenol. This conclusion is of particular importance in connection with a long discussion in the literature on a possible role of O^- radicals in selective oxidation catalysis over V and Mo oxides. Reliable evidence of the catalytic role of O^- obtained with zeolites may renew a general interest in the once suggested but not recognized radical oxygen idea in the oxidation over widely used metal oxide catalysts.

ACTIVE SURFACE FORMATION ON A PROMOTED COPPER CATALYST OF THE PARTIAL OXIDATION OF ETHANOL

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Russ. J. Phys. Chem. A,
80(5) (2006) pp. 706-713.

The catalytic properties, chemical composition, and morphology of the surface of electrolytic copper crystals promoted with phosphorus as catalysts of the partial oxidation of ethanol into acetaldehyde were studied. A mechanism of active copper surface formation under the action of the promoting admixture was suggested. Promotion was shown to cause the formation of copper polyphosphates on the surface. Particles of copper metal responsible for selective alcohol transformation are formed in a promoter layer under the action of the reaction medium.

BIFUNCTIONAL CATALYSTS FOR HYDROGEN PRODUCTION FROM DIMETHYL ETHER

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Stud. Sur. Sci. Catal.,
167 (2007) pp. 445-450.

The production of hydrogen directly from dimethyl ether (DME) was performed on bifunctional Cu-Ce/Al_2O_3 catalysts. The catalysts were characterized by XRD, HRTEM, EDX and IR spectroscopy of low-temperature CO adsorption. The high hydrogen productivity up to 600 mmol g^{-1} h^{-1} may be explained by assuming that (1) DME dehydration occurs on acid sites of γ-Al_2O_3 and (2) methanol steam reforming takes place on mixed oxide phase CuO-CeO_2, solid solution of copper ions in cerium dioxide with ratio Cu/Ce from 12/86 to 33/67 at.%.

ACTIVITY OF $MoSe_2/Al_2O_3$ CATALYSTS IN DECOMPOSITION OF THIOPHENE AND SELENOPHENE

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React. Kinet. Catal. Lett.,
90(1) (2007) pp. 167-177.

The rate of thiophene decomposition was shown to be independent of the type of chalcogens used in catalysts MoX_2/Al_2O_3, where X = S, Se. On the contrary, the rate of selenophene decomposition was shown to be higher on catalysts $MoSe_2$ than that on MoS_2. This observation suggests that the decomposition proceeds on anion vacancies. The decomposition of either thiophene over $MoSe_2$ or selenophene over MoS_2 results in the formation of partially substituted chalcogenides. At that, the molar ratios of the substituted chalcogen to Mo were shown to coincide in both cases. The fact that the rate of the thiophene decomposition does not depend on the degree of anion exchange indicates that the decomposition is not associated with hydrogenolysis.

STUDY OF METHANE DEHYDROAROMATIZATION ON IMPREGNATED Mo/ZSM-5 CATALYSTS AND CHARACTERIZATION OF NANOSTRUCTURED Mo PHASES AND CARBONACEOUS DEPOSITS

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Ind. Eng. Chem. Res., 46(12) (2007) pp. 4063-4074.

Methane dehydroaromatization was studied over the series of impregnated Mo/ZSM-5 catalysts with different molybdenum contents and Si/Al atomic ratios in the parent H-ZSM-5 zeolites. The maximum catalytic activity (CH_4 conversion ~ 14%) and benzene formation selectivity (~ 70%) were observed for the samples with 2-5% Mo. The activity and selectivity are improving when Si/Al is decreased from 45 to 17. After the pre-treatment in argon and reaction at 720°C, the catalysts have been characterized by textural methods, XRD, DTA, HRTEM and EDX. As determined by HRTEM, after the pre-treatment the MoO_x phase is highly dispersed on the external surface of zeolite. During the reaction, Mo carbide nanoparticles of 2-15 nm in size are forming on the external surface, and the Mo-containing clusters - in the zeolite channels. The carbonaceous deposits (CD) are forming as the graphite layers on the surface of Mo_2C nanoparticles larger than 2 nm, and as the friable layers with disordered structure - on the external surface of zeolite. According to EDX, XRD and DTA, the content of CD and extent of their condensation (C/H ratio) are increasing with time-on-stream. For the all studied Mo contents (1-10%) and time-on-stream values (0.5-6 h), the CD formed on the catalysts with Si/Al = 17 are characterized by one maximum of the exothermic burn-out effect in DTA, whereas on the catalysts with Si/Al = 30 and 45 – by two such maxima. A correlation between the catalyst activity, selectivity vs. nanostructure and location of the Mo phases and CD is discussed.

Co-Pt BIMETALLIC CATALYSTS FOR THE SELECTIVE OXIDATION OF CARBON MONOXIDE IN HYDROGEN-CONTAINING MIXTURES

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Kinet. Catal., 48(2) (2007) pp. 276-281.

The performance of a Co–Pt powder and of Co–Pt catalysts supported on ν-Al_2O_3 and on the graphite-like carbon material Sibunit in selective CO oxidation in hydrogen-containing mixtures is considered. Fine particles of metal–metal solid solutions and intermetallides were obtained by the decomposition of a Co- and Pt-containing double complex salt in a hydrogen atmosphere at ~400°C. As compared to their Pt and Co monometallic counterparts, the bimetallic catalysts are more active and allow the CO concentration in hydrogen-containing mixtures to be reduced from 1 to 10^{-3} vol %. This effect is likely due to the formation of bimetallic particles of a Co–Pt solid solution on the support surface.

COPPER-CERIUM OXIDE CATALYSTS FOR THE SELECTIVE OXIDATION OF CARBON MONOXIDE IN HYDROGEN-CONTAINING MIXTURES. I. CATALYTIC ACTIVITY

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Kinet. Catal., 48(3) (2007) pp. 439-447.

A series of copper–cerium oxide catalysts were prepared, and their properties toward the reaction of CO oxidation in hydrogen-containing gas mixtures were studied. It was found that the copper–cerium oxide catalysts are stable, active, and selective in this reaction. The conditions under which these catalysts decreased the concentration of CO from 1 to $<10^{-3}$ vol % in hydrogen containing water vapor and carbon dioxide were determined.

COPPER-CERIUM OXIDE CATALYSTS FOR THE SELECTIVE OXIDATION OF CARBON MONOXIDE IN HYDROGEN-CONTAINING MIXTURES. II. PHYSICOCHEMICAL CHARACTERIZATION OF THE CATALYSTS

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Kinet. Catal., 48(3) (2007) pp. 448-456.

The copper–cerium oxide catalysts were characterized using a set of physicochemical techniques including *in situ* FTIR spectroscopy, XPS, and XRD. It was found that copper segregated on the surface of cerium oxide and its states were labile and dependent on catalyst pretreatment conditions. Copper in a dispersed state was responsible for the reaction of CO oxidation in the presence of H_2 on the copper–cerium oxide catalysts. It is likely that this state of copper was composed of two-dimensional or three-dimensional surface clusters containing Cu^+ ions.

STEAM REFORMING OF DIMETHYL ETHER TO HYDROGEN-RICH GAS OVER BIFUNCTIONAL $CuO\text{-}CeO_2/\gamma\text{-}Al_2O_3$-CATALYSTS

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Chem. Sustain. Devel.,
15(1) (2007) pp. 117-121.

The steam reforming of dimethyl ether (DME) (SR) to a hydrogen-rich gas over a bifunctional $CuO\text{-}CeO_2/\gamma\text{-}Al_2O_3$-catalysts, containing surface sites responsible for proceeding of the reactions of DME hydration and steam methanol conversion was studied. The catalysts were shown to possess high activity and selectivity in DME steam reforming. The hydrogen production rate in DME SR was found to reach 0,47-0,61 mol $H_2/(g_{cat}\cdot h)$ at 350^oC, gas hourly space velocity of 10000 h^{-1} and molar ratio $H_2O/DME = 3$. The vol.% of CO in the hydrogen-rich gas is lower than equilibrium value and not exceeding 1 %.

STEAM REFORMING OF DIMETHYL ETHER TO HYDROGEN-RICH GAS

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React. Kinet. Catal. Lett.,
90(1) (2007) pp. 205-211.

The steam reforming of dimethyl ether (DME) (SR) to a hydrogen-rich gas over a mechanical mixture of WO_x/ZrO_2 (the DME hydration catalyst) and $CuZnAlO_x$ (the methanol SR catalyst) was studied. The mechanically mixed catalyst was shown to provide almost complete conversion of DME to the hydrogen-rich gas containing <0.5 vol.% of CO at 300°C, atmospheric pressure, gas hourly space velocity (GHSV) of 10000 h^{-1} and molar ratio $H_2O/DME = 3$. The hydrogen production rate in DME SR was found to reach 180–250 mmol $H_2/(g_{cat}\cdot h)$ at 250–300°C.

HYDRATION OF DIMETHYL ETHER TO METHANOL OVER SOLID ACIDS

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React. Kinet. Catal. Lett.,
90(1) (2007) pp. 197-204.

The hydration of dimethyl ether (DME) to methanol over various solid acids was studied. The acidity of the catalysts is determined by FTIR spectroscopy. The hydration is found to occur predominantly on the Brönsted acid sites. Among the catalysts studied, WO_x/ZrO_2 and H-ZSM-5 appeared to be most active and selective.

ELUCIDATION OF THE NATURE OF ACTIVE OXYGEN IN THE REACTION OF LOW-TEMPERATURE OXIDATION OF CO ON SINGLE CRYSTAL SURFACES PLATINUM AND PALLADIUM

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Russ. J. Phys. Chem. B,
1(2) (2007) pp. 130-137.

The role of the surface reconstruction, subsurface oxygen (O_{subs}) and concentration of CO_{ads} molecules was revealed in the low-temperature CO oxidation on Pt (100), Pt (410), Pd (111) and Pd (110) surfaces by HREELS, TPR and molecular beams methods. The possibility of the formation of so-called *"hot"* oxygen atoms, which arise at the surface at the instant of dissociation of O_{2ads} molecules and can react with CO_{ads} at low temperatures (~150 K) to form CO_2, was examined. It was revealed that, when present in high concentration, CO_{ads} initiates the phase transition of the Pt(100)-(hex) reconstructed surface into the (1×1) non-reconstructed one and blocks fourfold hollow sites of oxygen adsorption ($Pt_4\text{-}O_{ads}$), thereby initiating the formation of weakly bound oxygen ($Pt_2\text{-}O_{ads}$), highly-active in CO oxidation. For the Pt(410), Pd(111), and Pd(110) surfaces, the reactivity of O_{ads} with respect to CO was demonstrated to be dependent on the surface coverage of CO_{ads}. The $^{18}O_{ads}$ isotope label was used to determine the nature of active oxygen reacting with CO at ~150-200 K. It was examined why a CO_{ads} layer produces a strong effect on the reactivity of atomic oxygen. The experimental results were confirmed by theoretical calculations based on the minimization of the Gibbs energy of the adsorption layer. According to these calculations, the CO_{ads} layer causes a decrease in the apparent activation energy E_{act} of the reaction due to changes in the type of coordination and in the energy of binding of O_{ads} atoms to the surface.

EFFECT OF THE MODIFICATION OF ZrO_2-CONTAINING PILLARED CLAY WITH Pt AND Cu ATOMS ON THE PROPERTIES OF INORGANIC COMPLEX INTERMEDIATES IN THE SELECTIVE CATALYTIC REDUCTION OF NITROGEN OXIDES WITH PROPYLENE ACCORDING TO *IN SITU* IR-SPECTROSCOPIC DATA

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Kinet. Catal., 48(1) (2007) pp. 74-83.

It was found that only bridging and bidentate nitrate complexes were formed on the surface of Pt,Cu/ZrO_2-pillared interlayered clay (ZrO_2-PILC) upon the interaction with a flow of the (0.2% NO + 2.5% O_2)/N_2 mixture, whereas monodentate and nitrosyl complexes were not detected. The concentration of nitrate complexes on Pt,Cu/ZrO_2-PILC was higher and the strength of their bond to the surface was weaker than those on unmodified ZrO_2-PILC. Isopropoxide and acetate complexes and coordinatively bound acetone were formed on the surface in the interaction of Pt,Cu/ZrO_2-PILC with a flow of the (0.2% C_3H_6 + 2.5% O_2)/N_2 mixture. The supporting of Pt and Cu onto zirconium dioxide pillars resulted in considerable changes in the concentration and the temperature region of the existence of hydrocarbon surface compounds, as compared with ZrO_2-PILC. Under reaction conditions at relatively low temperatures, isopropoxide and nitrate intermediates on the surface of Pt,Cu/ZrO_2-PILC formed a complex structurally similar to adsorbed dinitropropane. At elevated temperatures, a surface nitromethane complex was formed in the interaction of the acetate complex with nitrate species. The spectrokinetic measurements demonstrated that the apparent rate constants of consumption of nitrate and nitroorganic complexes considerably increased on going from ZrO_2-PILC to Pt,Cu/ZrO_2-PILC. Moreover, the constants of consumption of nitroorganic and nitrate complexes were similar for both of the catalysts. This fact suggests that, on the test catalysts, nitroorganic complexes were reaction intermediates in the selective catalytic reduction of NO_x (NO_x SCR) with hydrocarbons. The found differences in the activation species and thermal stabilities of reactants can explain different activities of ZrO_2-PILC and Pt,Cu/ZrO_2-PILC in the SCR reaction of NO_x with propylene in an excess of oxygen.

SPECIFICS OF THE DESORPTION OF OXYGEN FROM A SILVER SURFACE PROMOTED BY PHOSPHATES

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Russ. J. Phys. Chem. A, 81(5) (2007) pp. 788-793.

The interaction of oxygen with the surface of a polycrystalline silver catalyst promoted with phosphorus-containing compounds was studied by the temperature-programmed desorption method. It was demonstrated that the introduction of a phosphorus-containing promoter gives rise to the formation of predominantly high-temperature forms of oxygen, which are more strongly bonded to the catalyst surface. The chemical composition of the active silver surface at various stages of its evolution was determined by IR spectroscopy. Under the action of temperature and a reducer, the polyphosphate promoter layer was observed to produce metal silver particles, which participate in the formation of active sites at the catalyst surface.

KINETICS OF OXYGEN EXCHANGE OVER CeO_2-ZrO_2 FLUORITE-BASED CATALYSTS

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J. Phys. Chem. A, 111(20) (2007) pp. 4498-4505.

The kinetics of $^{18}O/^{16}O$ isotopic exchange over CeO_2-ZrO_2-La_2O_3 and Pt/CeO_2-ZrO_2 catalysts have been investigated under the conditions of dynamic adsorption–desorption equilibrium at atmospheric pressure and temperature range 650-850°C. The rates of oxygen adsorption/desorption on Pt sites, support surface, oxygen transfer (spillover) from Pt to the support as well as the amount of oxygen accumulated in the oxide bulk and oxygen diffusion coefficient were estimated. The nanocrystalline structure of lanthana doped ceria-zirconia prepared via the Pechini route with a developed network of domain boundaries and specific defects guarantees a high oxygen mobility in the oxide bulk ($D = (1.5\div2.0)\cdot10^{-18}\ m^2s^{-1}$ at 650^0C) and allows accumulation of over-stoichiometric/excess oxygen. For Pt/CeO_2-ZrO_2

oxygen transfer from Pt to support (characteristic time $< 10^{-2}$ s) was shown to be responsible for the fast exchange between the gas phase oxygen and oxygen adsorbed on the mixed oxide surface. The rate of direct exchange between the gas phase and surface oxygen is increased as well, due to the increased concentration (up to 2 monolayers) of surface/near subsurface oxygen species accumulated on the oxygen vacancies (originated from the incorporation of highly dispersed Pt atoms). The characteristic time of diffusion of the oxygen localized in the subsurface layers is about 1 s. The overall quantity of over-stoichiometric oxygen and/or hydroxyl groups accumulated in the bulk can reach the equivalent of 10 monolayers, and characteristic time of oxygen diffusion within the bulk is about 20 s. All these kinetic data are required for the further step of modeling partial oxidation of hydrocarbons under steady- and unsteady-state conditions.

REGIOSELECTIVE H/D EXCHANGE OF PROPANE ON Zn/H-MFI ZEOLITE

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Catal. Lett., 114(1-2) (2007) pp. 85-90.

The hydrogen exchange for propane-d_8 adsorbed on zeolite Zn/H-MFI has been studied by ^{1}H MAS NMR spectroscopy *in situ* within the temperature range of 420–490 K. Kinetic measurements of the H/D exchange between the acidic hydroxyl groups of the zeolite and the adsorbed deuterated propane molecules show that only methyl groups of the alkane are involved in the exchange. Two mechanisms are proposed to rationalize the regioselectivity of the exchange: (i) propane dehydrogenation on Zn-sites followed by protonation of propene by acidic OH groups in accordance to the Markovnikov's rule and abstraction of deuteride ion from another propane molecule; (ii) the reversible heterolytic dissociative adsorption of propane to form Zn-propyl species and acidic OH groups.

DYNAMICS OF LINEAR *n*-C_6 - *n*-C_{22} ALKANES INSIDE 5A ZEOLITE STUDIED BY ^{2}H NMR

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J. Phys. Chem. C,
111(11) (2007) pp. 4393-4403.

The dynamic behavior of deuterated analogues of linear alkanes, *n*-C_6-*n*-C_{22}, adsorbed in zeolite 5A has been studied by deuterium solid-state NMR (^{2}H NMR). Temperature dependences of spin-lattice (T_1) and spin-spin (T_2) relaxation times of the deuterium located in the CD_3 groups of the adsorbed n-alkanes were rationalized on the basis of a model derived for the motion of n-alkanes located in the pores of the zeolite. The model implies that the adsorbed molecules consist of two ensembles: diffusing (or stretched) and temporarily blocked from diffusion (or coiled). The possible intramolecular motions for the alkane chains were taken into account based on both the finite size of the zeolite cage and the allowable hydrocarbon chain conformations. The coiled molecules are involved in two modes of motion: isotropic reorientation and intramolecular conformational isomerization, whereas the stretched molecules are additionally involved in a diffusion process. Dynamics parameters for different modes of motion and a proportion of the blocked and stretched molecules were derived from the analysis of relaxation data. The estimated proportion of the diffusing molecules correlates with the alkanes diffusivities earlier obtained by neutron spin echo measurements.

KINETIC MODEL AND MECHANISM OF THE SELECTIVE OXIDATION OF CO IN THE PRESENCE OF HYDROGEN ON PLATINUM CATALYSTS

P.V. Snytnikov, V.D. Belyaev, V.A. Sobyanin

Kinet. Catal., 48(1) (2007) pp. 93-102.

The reaction kinetics of the selective oxidation of carbon monoxide in the presence of hydrogen on a Pt/carbon support catalyst was studied. It was found that this catalyst exhibited high activity and decreased the concentration of CO in a hydrogen-containing gas from 0.6–1.0 vol % to less than 10 ppm at the inlet concentration ratio $O_2/CO = 1.0–1.5$. A kinetic model of the reaction was proposed to describe quantitatively the experimental results.

IN SITU NMR SPECTROSCOPY IN HETEROGENEOUS CATALYSIS: KINETIC STUDY OF HYDROCARBON CONVERSION MECHANISMS

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Kinet. Catal., 48(4) (2007) pp. 521-534.

The potential of high-resolution solid-state NMR spectroscopy for kinetic and mechanistic studies of hydrocarbon conversion on solid acid catalysts between 20 and 300°C is considered. The use of this technique is illustrated by the elucidation of the mechanisms of hydrogen exchange and ^{13}C label transfer in alkanes and olefins, *n*-butane isomerization on sulfated zirconia, and ethane aromatization on zinc-containing zeolite beta. The kinetic parameters determined in these studies provide a basis for quantum chemical calculations of possible hydrocarbon activation and conversion pathways and for evaluating the reliability and accuracy of these theoretical calculations.

HOMOGENEOUS HIGH-TEMPERATURE OXIDATION OF METHANE

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React. Kinet. Catal. Lett., 91(2) (2007) pp. 273-282.

The kinetics of homogeneous deep oxidation of methane in lean mixtures (up to 2 vol % CH_4 in air) in ceramic tubes and fixed beds of ceramic spheres was studied. Experiments with the homogeneous reaction have shown that the methane oxidation occurs *via* a consecutive scheme through CO formation. The reaction rate of CH_4 oxidation was found to depend upon the equivalent pass diameter with a significant reaction inhibition in packing of small tubes and spheres, reflecting the influence of mass transfer on the radical-chain termination at the ceramic surfaces. It was also found that CO oxidation practically does not depend upon the mass exchange conditions, but it is visibly inhibited by methane. Recommended kinetic equations and their parameters are presented.

KINETICS OF ALUMINUM POWDER OXIDATION BY WATER AT 100°C

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React. Kinet. Catal. Lett., 92(1) (2007) pp. 83-88.

The kinetics of aluminum powder oxidation by water at 100°C has been investigated using three types of aluminum powders. A “shrinking sphere” model was used for the description of experimental data at the initial reaction stage, whereas a diffusion “parabolic” equation was used at the second stage. Variation of the specific reactivity is discussed.

AROMATIZATION OF LIGHT NAPHTA FRACTION ON ZEOLITES. 3. MODEL OF COKE FORMATION

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CI&CEQ, 13(2) (2007) pp. 51-54.

Previously the authors reported on the reaction kinetic and catalyst deactivation models for the Zeoforming process. The significant coking takes place, and two types of coke are formed. In this work the equations of coke accumulation are derived and its relations with relative activities are presented and discussed.

NONOXIDATIVE METHANE CONVERSION INTO AROMATIC HYDROCARBONS ON TUNGSTEN-CONTAINING PENTASILS

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Kinet. Catal., 48(3) (2007) pp. 409-413.

The effects of the tungsten concentration and of the method of tungsten introduction into ZSM-5 pentasils with different SiO_2/Al_2O_3 molar ratios on the acidity and the activity of the resulting catalysts in non-oxidative methane conversion into aromatic hydrocarbons are considered. The catalysts obtained from the SiO_2/Al_2O_3 = 40 pentasil and a nanosized tungsten powder are the most active and the most stable. The maximum methane conversion and the highest yield of aromatic hydrocarbons are achieved on the zeolite containing 8.0 wt % tungsten nanopowder.

ZrFe INTERMETALLIDES FOR FISCHER-TROPSCH SYNTHESIS: PURE AND ENCAPSULATED INTO ALUMINA-CONTAINING MATRICES

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Stud. Surf. Sci. Catal., 163 (2007) pp. 153-175.

Performance of the bulk hydrogenated ZrFe intermetallides both pure and encapsulated into alumina-containing matrix (Al_2O_3/Al, Al_2O_3) was studied in catalysis of Fischer-Tropsch synthesis. Their structural, textural and surface properties were characterized by combination of such methods as XRD, SEM, TEM, nitrogen adsorption-desorption isotherms and XPS, and impact of these properties on catalytic activity and selectivity was analyzed. The highest activity per the surface Fe atom ($\sim 5.10^{-19}$ CH_x/at.Fe.h) was obtained for pure active component, while the highest activity per the unit of volume (~168 g C^{5+}/l.h) was revealed for a composite catalyst at 300°C, 3 MPa and space velocity ~7000 h^{-1}.

Application of Physicochemical Methods for Characterization of Catalysts, Supports, Synthesized Substances and Materials

THE USE OF RDF ANALYSIS IN THE STUDY OF POWDER SAMPLES: ZIRCONIA COMPLEX SUPPORTED BY CLAY AND PSEUDOBOEHMITE

E.M. Moroz, D.A. Zyuzin

Z. Kristallogr. Suppl., 26 (2007) pp. 273-278.

Two examples illustrating the use of radial distribution functions of electronic density to determine the structure of supported zirconia complex and particularities of the pseudoboehmite structure are reported.

RDF METHOD APPLICATION TO PHASE ANALYSIS OF X-RAY AMORPHOUS PRODUCTS OF GIBBSITE THERMAL ACTIVATION

K.I. Shefer, E.M. Moroz, L.I. Isupova, I.V. Kharina, Yu.Yu. Tanashev

Z. Kristallogr., suppl_26 (2007) pp. 279-282.

The phase compositions of finely dispersed initial and rehydrated products of gibbsite thermal activation prepared by different technological methods – thermochemical activation (TCA) and centrifugal thermal activation (CTA) were studied using radial distribution function (RDF) method. The initial phase composition was shown to depend on the preparation method and determine the composition of the rehydrated products. Nuclei of different hydroxide phases are formed in addition to χ-alumina – pseudoboehmite in the case of the TCA process and bayerite for the CTA process. This leads to different phase compositions of rehydrated products. For TCA, pseudoboehmite is the primary rehydration product, whereas bayerite is formed for CTA.

METHOD FOR CONSTRUCTING THE RADIAL DISTRIBUTION CURVES

E.M. Moroz, D.A. Zyuzin, K.I. Shefer

J. Struct. Chem., 48(2) (2007) pp. 262-266.

A method for constructing the model radial distribution curves of electron density is described. The utility of this technique for the refinement of structure and phase composition for highly disperse materials is demonstrated by reference to several examples.

RADIAL DISTRIBUTION MODEL CURVES OF ELECTRON DENSITY FOR ALUMINUM OXIDES AND HYDROXIDES

E.M. Moroz, D.A. Zyuzin, K.I. Shefer, L.A. Isupova

J. Struct. Chem., 48(4) (2007) pp. 704-707.

Radial distribution model curves of electron density were constructed for various aluminum oxides and hydroxides that can be used for the refinement of the phase composition and structural features of these highly disperse materials employed as catalyst supports.

SUPPORTED MODIFIED MANGANESE-PALLADIUM CATALYSTS OF MPK-1 TYPE FOR CO OXIDATION REACTION

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Catal. Ind., 3 (2007) pp. 3-9.

The process of formation of active Mn-oxide phase in dependence of its concentration and type of carriers: SHN (γ-, χ-Al_2O_3) and SHN-2M (κ-Al_2O_3, $CaO*2Al_2O_3$) using X-ray phase and thermal analyses was studied. Optimal catalyst: 4-5 Mn, 0,05 Pd(mass%)/SHN-2M corresponds by composition to pilot sample of catalyst MPK-1. Calcination in air at 900^0C, 10 h leads to decrease of the mechanical solidity of the sample MPK-1/SHN-2 from 7,5 to 3,2 MPa and decrease of its specific surface area from 98,0 to 10,0 m^2/g. However, for the sample MPK-1/SHN-2M calcination results in decrease of its specific surface area to 50,0 m^2/g, but the mechanical solidity of this catalyst does not change. This treatment leads to increase of temperature of reaching the 75 % degree of CO conversion at 130^0C and 50^0C for catalysts MPK-1/SHN-2 and MPK-1/SHN-2M, respectively. For both carriers formation of solid solution of Mn^{4+} was observed. Catalysts were examined in the systems of cleaning of exhaust gases of automotive engines and recommended for further practical use.

VACANCIES ORDERING IN $La^{3+}_{1-x}Ba^{2+}_xFeO_{3-\delta}$ PEROVSKITES

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Z. Kristallogr.,
suppl_26 (2007) pp. 381-386.

Using powder X-ray diffraction the $La^{3+}_{1-x}Ba^{2+}_xFeO_{3-\delta}$ perovskite related solid solutions were investigated. A morphotropic phase transition from orthorhombic to cubic was shown for the sample with x ~ 0.3. High temperature XRD patterns of $La_{0.2}Ba_{0.8}FeO_{3-\delta}$ obtained under air and vacuum revealed new diffraction peaks related to local ordering of the oxygen vacancies. Such ordering remained at room temperature only after cooling in vacuum.

HIGH-TEMPERATURE STUDIES OF $La_{1-x}Sr_xFeO_{3-\delta}$ SOLID SOLUTIONS USING SYNCHROTRON RADIATION

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J. Struct. Chem.,
48(6) (2007) pp. 1105-1109.

A series of $La_{1-x}Sr_xFeO_{3-\delta}$ samples with a perovskite structure were investigated by high-temperature X-ray analysis using synchrotron radiation. In this series, one can observe a morphotropic phase transition ($0.3\le x\le 0.4$) from the orthorhombic ($0\le x<0.3$) to cubic ($0.4<x\le 0.75$). modification. The samples from the morphotropic transition region (MTR) at room temperature have a highly disordered microblock structure related to their phase heterogeneity; according to high-temperature X-ray analysis data, this is the result of particle stratification of the high-temperature homogeneous solid solution formed at the temperature of the synthesis (1200°C) in the course of cooling.

CHARACTERIZATION OF NEW CATALYSTS BASED ON URANIUM OXIDES

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V.A. Ushakov

Kinet. Catal., 48(4) (2007) pp. 511-520.

Catalysts based on uranium oxides were systematically studied for the first time. Catalysts containing various amounts of uranium oxides (5 and 15%) supported on alumina and mixed Ni–U/Al_2O_3 catalysts were synthesized. The uranium oxide catalysts were characterized using the thermal desorption of argon, the low-temperature adsorption of nitrogen, X-ray diffraction analysis, and temperature-programmed reduction with hydrogen and CO. The effects of composition, preparation conditions, and thermal treatment on physico-chemical properties and catalytic activity in the reactions of methane and butane oxidation, the steam and carbon dioxide reforming of methane, and the partial oxidation of methane were studied. It was found that a catalyst containing 5% U on alumina calcined at 1000°C was most active in the reaction of high-temperature methane oxidation. For the Ni–U/Al_2O_3 catalysts containing various uranium amounts (from 0 to 30%), the introduction of uranium as a catalyst constituent considerably increased the catalytic activity in methane steam reforming and partial oxidation.

STRUCTURAL STUDY OF HIGHLY DISPERSED CERIUM DIOXIDES

V.P. Kolko, E.M. Moroz, V.V. Kriventsov, D.A. Zyuzin

Bull. Russ. Acad. Sci.: Phys.,
71(5) (2007) pp. 695-698.

Cerium dioxide samples of different dispersivity, obtained by different methods, have been investigated by X-ray diffraction (XRD) analysis, full-profile analysis of diffraction patterns, calculation of radial distribution function (RDF) of electron density, and extended X-ray absorption fine-structure (EXAFS) spectroscopy. According to the XRD data, the samples studied can be identified in the fluorite structure with conventional unit-cell parameters. However, the local structure of highly dispersed samples, determined by the RDF and EXAFS methods, changes: the coordination spheres for the Ce–O distances are represented by two distances.

STRUCTURAL DETERMINATION OF CERIA–ZIRCONIA NANOSYSTEM DOPED BY Gd

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Nucl. Instrum. Methods Phys. Res., Sect. A,
575(1-2) (2007) pp. 91-95.

The work is devoted to the structural study of doped Gd ceria–zirconia nanosystem by the EXAFS, WAXS (wide angle X-ray scattering) and XRD methods. A polymerized complex precursor method (Pechini route) was used for synthesis of ceria–zirconia doped by Gd. It was found by XRD analysis that samples of ceria–zirconia doped by Gd have a CeO_2-reference structure (a cubic fluorite-like structure). However, analysis of the EXAFS and WAXS revealed the existence of two solid solutions with different content of zirconium cations, this implies heterogeneity on local level of structure.

STRUCTURE OF THE MIXED OXIDES $Me_x(Ce_{0.5}Zr_{0.5})_{1-x}O_y$ (Me = Gd, Pd)

V.P. Kolko, D.A. Zyuzin, V.A. Sadykov, V.V. Kriventsov, E.M. Moroz

Glass Phys. Chem.,
33(4) (2007) pp. 335–339.

The structure of the mixed oxides $Me_x(Ce_{0.5}Zr_{0.5})_{1-x}O_y$ (x = 0, 0.1–0.30; $y < 2$) doped with trivalent Gd and Pr metals is investigated using the X-ray powder diffraction analysis, the full-profile analysis of the diffraction pattern, and EXAFS spectroscopy. The mixed oxides are solid solutions with a structure similar to the CeO_2 structure. An increase in the content of the introduced trivalent metal brings about an increase in the unit cell parameter and a disordering of the structure of the oxides under investigation.

STRUCTURE STABILIZING EFFECT OF TUNGSTEN IN MIXED MOLYBDENUM OXIDE WITH Mo_5O_{14}-TYPE STRUCTURE

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Catal. Today, 126(1) (2007) pp. 112-118.

Bulk structural properties of single phase crystalline $(Mo_{0.91}V_{0.09})_5O_{14}$ and $(Mo_{0.68}V_{0.23}W_{0.09})_5O_{14}$ materials were investigated using *in situ* X-ray diffraction and *in situ* X-ray absorption spectroscopy at three metal edges. Temperature programmed experiments in reducing (propene) and oxidizing (oxygen) atmosphere and isothermal redox experiments at 773 K revealed differences in the bulk properties of the two phases studied. A structure stabilizing effect of tungsten in $(MoVW)_5O_{14}$ under oxidizing conditions was found. Moreover, tungsten centers in a (MoVW) oxide material exert a structure-directing effect towards re-oxidation to a Mo_5O_{14}-type structure.

XAS STUDY ON MICROSTRUCTURE OF Au NANOPARTICLES DEPOSITED ON ALUMINA

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Nucl. Instrum. Methods Phys. Res., Sect. A,
575(1-2) (2007) pp. 105-108.

X-ray adsorption spectroscopy (XAS) was applied to the characterization of the Au/δ-Al_2O_3 catalysts with different mean Au particle sizes ($\langle d_l \rangle$ = 2-28 nm) prepared *via* adsorption of anionic Au(III) hydroxide complexes and calcined in air at 673 K. The parameters of local atomic surrounding for Au atoms in these catalysts were determined using XANES and EXAFS techniques at 12 and 296 K. In all samples, the only phase detected was metallic gold. A contraction of interatomic Au-Au distance (R_1) and a gradual decrease in the coordination number for the first coordination shell of Au atoms (N_1), as well as an increase in the Debye-Waller factor (σ) were observed with decreasing mean Au particle size.

XPS AND TEM STUDIES ON THE ROLE OF THE SUPPORT AND ALKALI PROMOTER IN Ru/MgO AND Ru-Cs^+/MgO CATALYSTS FOR AMMONIA SYNTHESIS

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J. Phys. Chem. C, 111(26) (2007) pp. 9426-9437.

The chemical state of ruthenium in the Ru/MgO and Ru–Cs^+/MgO catalysts prepared by the incipient wetness technique with the use of $Ru(OH)Cl_3$ and Cs_2CO_3 as the catalyst and promoter precursors, respectively, is characterized by X-ray photoelectron spectroscopy (XPS). The influence of the final state effects (the differential charging and variations of the relaxation energy) on the binding energy of Ru $3d_{5/2}$ core level measured for supported Ru nanoparticles is estimated by comparison of the Fermi levels and the modified Auger parameters determined for the Ru/MgO and Ru–Cs^+/MgO catalysts with the corresponding characteristics of polycrystalline Ru foil. High-resolution transmission electron microscopy (HRTEM) in combination with energy dispersive X-ray (EDX) microanalysis reveals that Ru particles in the Ru–Cs^+/MgO catalyst are covered with an amorphous layer containing cesium. XPS data show that the layer subjected to the prolonged treatment with H_2 at 450°C consists of cesium suboxide $Cs_{2+x}O$. The shift of the Ru $3d_{5/2}$ binding energy toward lower values is found for the Ru–Cs^+/MgO catalyst (279.7 eV) with respect to metallic Ru (280.2 eV) and the Ru/MgO catalyst (280.5 eV). It is assumed that the shift results from a decrease in the work function of ruthenium under the action of Cs^+ cations located on the Ru surface. The data obtained are used to explain the sharp difference in the activities of the Ru/MgO and Ru–Cs^+/MgO catalysts for ammonia synthesis at 250–400°C.

X-RAY PHOTOELECTRON SPECTROSCOPIC STUDY OF THE INTERACTION OF SUPPORTED METAL CATALYSTS WITH NO_X

M.Yu. Smirnov, A.V. Kalinkin, V.I. Bukhtiyarov

J. Struct. Chem.,
48(6) (2007) pp. 1053-1060.

The reactions of the platinum and rhodium model catalysts applied to aluminum oxide with NO_x (10 Torr NO + 10 Torr O_2) were studied by X-ray photoelectron spectroscopy. The reaction conducted at room temperature formed on the surface of the oxide support the $NO_{3,s}^-$ nitrate ions characterized by the N1*s* line at 407.4 eV and O1*s* line at 533.1 eV and the $NO_{2,s}^-$ nitrite ions characterized by the N1*s* line with a binding energy of 404.7 eV. At the same time, the Pt4*f* and Rh3*d* lines of the supported platinum particles are shifted toward higher binding energies by 0.5–1.0 eV and 0.7–1.2 eV, respectively. It is assumed that the binding energies increase due to changes in the chemical state of the platinum metal in which oxygen is dissolved. The reaction of NO_x with Pt/Al_2O_3 at 200°C forms platinum oxide defined by the $Pt4f_{7/2}$ line with a binding energy of 72.3 eV.

MIXED LAYERED Ni–Mn–Co HYDROXIDES: CRYSTAL STRUCTURE, ELECTRONIC STATE OF IONS, AND THERMAL DECOMPOSITION

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J. Power Sources,
174(2) (2007) pp. 735-740.

It has been shown that varying conditions of co-precipitation processes, two types of mixed Ni–Mn–Co hydroxides: either brucite, or hydrotalcite-like (LDHs) - with different Ni/Mn/Co ratio can be prepared. According to XPS study, these hydroxides are mixed-valence materials: Ni ions present in 2^+ oxidation state, whereas Mn and Co ions are in $3^+/4^+$ and $2^+/3^+$ state, respectively; their oxidation state increases with an increase of their content in hydroxides. For Ni-rich hydroxides, a strong effect of nickel segregation is observed indicating that chemistry of the surface and of the bulk is different. Mixed Ni–Mn–Co hydroxides decompose in two steps resulting in formation of NiO bunsenite and $(Ni,Co,Mn)_3O_4$ spinel.

OPTIMIZATION OF Ni^{2+}/Ni^{3+} RATIO IN LAYERED Li(Ni,Mn,Co)O_2 CATHODES FOR BETTER ELECTROCHEMISTRY

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J. Power Sources,
174(2) (2007) pp. 965-969.

A study of the $LiNi_{1-x-y}Co_xMn_yO_2$ ($x = y = 0.1$; 0.2; 0.33) cathode materials prepared from mixed hydroxides and LiOH using the method of mechanical activation has been performed. All as prepared materials were indexed in the *R*-3*m* space group. The cell volume and the degree of structural disordering decrease with reducing of Ni content. According to XPS study, the main electronic state of d-ions in the surface layer of particles corresponds to Ni^{2+}, Co^{3+} and Mn^{4+}. However, the $Ni2p_{3/2}$ and $Co2p_{3/2}$ binding energy increases versus Ni content, indicating the increased Me-bond covalency. The highest specific discharge capacity was observed for $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ - 170 mAh g^{-1}. The position of redox peaks on the differential capacity curves depends on the composition of the sample: it shifts towards lower voltage with increased Ni content.

$LiNi_{1-x-y}Co_xMn_yO_2$ ($x = y = 0.1, 0.2, 0.33$) CATHODE MATERIALS PREPARED USING MECHANICAL ACTIVATION: STRUCTURE, STATE OF IONS, AND ELECTROCHEMICAL PERFORMANCE

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Inorg. Mater.,
43(2) (2007) pp. 185-193.

$LiNi_{1-x-y}Co_xMn_yO_2$ ($x = y = 0.1, 0.2, 0.33$) cathode materials synthesized from mechanically activated mixtures of lithium hydroxide and nickel cobalt manganese hydroxide were investigated. The materials have a layered structure (sp. gr. *R*-3*m*). Their unit-cell volume and the degree of disordering in their structure decrease with decreasing nickel content. According to XPS data, the major states of the transition-metal ions in the surface layer of the materials are Ni^{2+}, Co^{3+}, and Mn^{4+}. With increasing nickel content, the Ni $2p_{3/2}$ and Co $2p_{3/2}$ binding energies increase, attesting to changes in M–O bond covalence. The highest specific electrochemical capacity, ~170 mA h/g, is offered by $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$. The position of redox peaks in the differential capacity curves of the three materials depends on composition: with increasing nickel content, the peaks shift to lower voltages.

XAFS AND NEUTRON DIFFRACTION STUDY OF $La_{1-x}Sr_xCo_{1-y}Nb_yO_3$

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Nucl. Instrum. Methods Phys. Res., Sect. A,
575(1-2) (2007) pp. 176-179.

The effect of hole doping on the crystal and electronic structure of $La_{1-x}Sr_xCo_{1-y}Nb_yO_3$ (y = 0.0-0.25) at the x = 0.0; 0.2; 0.5 has been studied by neutron diffraction and X-ray absorption spectroscopy at the Co K and $L_{2,3}$-edges. The preliminary analysis of the Co K and $L_{2,3}$-edges X-ray absorption near edge structure (XANES) in $La_{1-x}Sr_xCo_{1-y}Nb_yO_3$ series suggests that upon niobium doping, an increase of the intermediate Co^{3+} spin state and a decrease of low spin Co^{4+} states contributions occur. The possible explanation of the observed changes of crystal and electronic structure in these cobaltites is discussed.

XAFS AND NEUTRON DIFFRACTION STUDY OF $La_{1-x}Sr_xCo_{1-y}O_3$

V.V. Efimov*, E. Efimova*, D. Karpinsky, D.I. Kochubey, V.V. Kriventsov, A. Kuzmin***, S. Molodtsov****, V. Sikolenko*,*****, S. Tiutiunnikov*, I.O. Troyanchuk**, A.N. Shmakov, D. Vyalikh******** (**Joint Institute for Nuclear Research, Dubna, Moscow Region, Russia; **Institute of Solid State and Semiconductor Physics, Minsk, Belarus; ***Institute of Solid State Physics, Riga, Latvia; ****Institut für Kristallographie und Festkörperphysik, Dresden, Germany; *****Hahn-Meitner-Institut, Berlin Germany; ******Universita'di Trento, Trento, Italy*)

Phys. Status Solidi C, 4(3) (2007) pp. 805-808.

The effects of hole doping on the crystal and electronic structure of the $La_{1-x}Sr_xCoO_3$ (x = 0.0÷0.5) by neutron Rietveld analysis and its correlation with the X-ray-absorption spectroscopy data have been studied. The abrupt decrease of the Co-O distance and an increase of the Co-O-Co angle upon substitution of La^{3+} by Sr^{2+} in $La_{1-x}Sr_xCoO_3$ are attributed to a change in the band structure at the transition from semiconducting to metallic state. Upon strontium doping, a variation of Co $L_{2,3}$-edges in $La_{1-x}Sr_xCoO_3$ series suggests an increase of the mixed low Co^{4+} and high or intermediate Co^{3+} spin states. The possible explanation of the observed changes of the crystal and electronic structure in these cobaltites is discussed.

NEUTRON DIFFRACTION AND SYNCHROTRON RADIATION STUDIES OF $La_{1-x}Sr_xCoO_3$ MAGNETIC PROPERTIES

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J. Magn. Magn. Mater.,
310(2) (2007) pp. e181-e183.

Magnetic properties of the $La_{1-x}Sr_xCoO_3$ solid solutions have been studied by neutron diffraction and X-ray absorption near edge structure (XANES) measurements. Our data are in agreement with the following scenarios. The first scenario is about the magnetic phase separation into regions with different content of Co^{3+}/Co^{4+} ions in different spin states. The second one is about the gradual thermal excitation of the Co^{3+} ions from low- to intermediate- or high-spin state in wide temperature range.

PLUTONIUM AND NEPTUNIUM SPECIATION BOUND TO HYDROUS FERRIC OXIDE COLLOIDS

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Comptes Rend. Chem.,
10(10-11) (2007) pp. 1060-1066.

The speciation of plutonium and neptunium in colloid suspension collected from the contaminated aquifer of the "Mayak" site was studied by nano-SIMS, XPS and XANES. At tracer level concentration found in original groundwater sample Pu is sorbed onto hydrous ferric oxide colloids, while at higher concentration relevant to near-field conditions it forms intrinsic $PuO_2 \times nH_2O$ colloids. The formation of such colloids occurs through the reduction of Pu(V) to Pu(IV) upon its sorption.

SPECTROSCOPY OF HOMOGENEOUS DEUTERATED HYDROCARBON FILMS REDEPOSITED FROM T-10 TOPKAMAK PLASMA DISCHARGE

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*J. Surf. Invest. X-Ray,
Synchrotron and Neutron Techn.*,
1 (2007) pp. 17-22.

Homogeneous deuterated hydrocarbon films redeposited from a deuterium plasma discharge inside the T-10 tokamak vacuum chamber are studied using photoluminescence methods; EXAFS, EPR, and IR spectroscopies; and temperature measurements. The photoluminescence excitation spectra of sp^3-sp^2 nanostructures of tokamak films are compared with those of sp^2 nanostructures of C^{60} fullerite films. The effect of defect states on photoluminescence and its temperature quenching is discussed. It is concluded that the model of temperature luminescence quenching for homogeneous deuterated tokamak films is similar to that for amorphous a-C:H films.

XAFS STUDY OF HIGH-DISPERSE Pd-CONTAINING NANOSYSTEM SUPPORTED ON TiO_2 OXIDE MATRIX

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Nucl. Instrum. Methods Phys. Res. A,
575(1-2) (2007) pp. 180-184.

Recently, great efforts are devoted to develop new methods of preparation of high-disperse Pd-containing nanosystems (composed of metal and/or oxide substance) stabilized on oxide matrix. New approach of synthesis is usage of $PdCo(\mu\text{-}OOCMe)_4(NCMe)$ complex and $Ti(OBu)_4$ as precursors to anchor Pd on the surface of oxide matrix surface in a highly dispersed form. The present work is devoted to the structural study of this high-disperse Pd-containing nanosystem by the XAFS spectroscopy. The strong interaction between Pd and Co cations takes place in the studied system. Seemingly, Co cations incorporate into TiO_2 oxide matrix, forming mixed oxides. This allows to anchor Pd cations, with the formation of the palladium oxide structures, modified by interaction with Co and Ti cations. These compounds are non-stochiometric and have typical structural features of mixed oxides. All possible structural models are discussed in detail.

FEATURES OF LOCAL ATOMIC STRUCTURE OF NANOCRYSTALLINE DISORDERED Fe–M (M=Ge, Sn, Al, Si) ALLOYS: EXAFS-STUDY

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Nucl. Instrum. Methods Phys. Res. A,
575(1-2) (2007) pp. 189-192.

The EXAFS-study (Fe, Sn and Ge K-edges) of disordered by mechanical activation binary supersaturated nanocrystalline solid solutions Fe–Al, Fe–Ge, Fe–Sn and Fe–Si is presented. The EXAFS-spectra are processed by solving the inverse binary problem, using the EXAFS-spectrum of the Fe K-edge only or combining the EXAFS-spectra on two K-edges, Fe and Ge or Fe and Sn. The parameters of partial correlation functions indicate chemical short-range ordering, high local static distortions in the lattice, increasing with metalloid content. The macrostructure of other type is forming through an initial, “local” stage within the *bcc* lattice.

X-RAY EMISSION AND X-RAY PHOTOELECTRON SPECTROSCOPIC STUDIES OF FULLERENE FLUORIDE $C_{60}F_{24}$

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Phys. Solid State, 49(5) (2007) pp. 1195-1200.

The structure of the fullerence fluoride $C_{60}F_{24}$ of the T_h symmetry contains two types of chemically different carbon atoms, namely, atoms of isolated double bonds and atoms of CF groups. X-ray photoelectron and X-ray emission spectroscopic studies of $C_{60}F_{24}$ revealed a difference in the widths of the X-ray bands corresponding to these types of atoms. Nonempirical quantum-chemical calculations performed for $C_{59}NF_{24}{}^{+}$ ions with a hole in the C 1*s* core level of the fullerence fluoride showed that the difference in the bandwidths may be due to the fact that the vibrational states of the system are different when 1*s* electrons are removed from chemically nonequivalent atoms.

XPS STUDIES OF THE OXYGEN STATE DURING OXIDATION OF POLYCRYSTALLINE GOLD SURFACE

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Herald of the Moscow State University,
ser. 2, Chem.,
48(6) (2007) pp. 418-426.

X-ray photoelectron spectroscopy (XPS) was used to study oxide films on bulk gold obtained by high-frequency activation of O_2. It was established that this method is effective for the formation of oxide films thicker than 3–5 nm. Inspection of the Au4f spectra revealed that the gold atoms are in 3+ state during the surface oxidation. The complex structure of XPS spectra of oxygen comprise 4 individual features that characterize various non-equivalent states of oxygen in the obtained oxide films. Presumably, two main oxygen states (E_b(O1s) = 529.0 eV and 530.0 eV) relate to oxygen atoms in the composition of two- and three-dimensional gold oxide Au_2O_3. Oxygen species with high bonding energies (E_b(O1s) = 531.8 eV and 535.2 eV) are most likely to be characterized by the molecular oxygen state in the composition of peroxide and superoxide groups, respectively.

TEMPERATURE EFFECTS ON THE IR SPECTRA OF CRYSTALLINE AMINO ACIDS, DIPEPTIDES, AND POLYAMINO ACIDS.
I. GLYCINE

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J. Struct. Chem.,
48(2) (2007) pp. 332-339.

The IR spectra of the α, β, and γ polymorphic modifications of pure glycine (without diluents) have been studied in the temperature range 93–433 K; changes in the IR spectra caused by variations of temperature are correlated with previously obtained diffraction data about polymorphic transitions, anisotropic compression of structure, and changes in hydrogen bond parameters.

X-RAY CRYSTAL STRUCTURE OF [BPMEN(Cl)Fe^{III}OFe^{III}(Cl)BPMEN]$(ClO_4)_2$, [BPMEN = N,N'-DIMETHYL-N,N'-BIS (2-PYRIDYLMETHYL)ETHANE-1,2-DIAMINE] AND THE ASSIGNMENT OF ITS ^{1}H NMR PEAKS IN CD_3CN

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Mendeleev Commun.,
17(5) (2007) pp. 291-293.

The complex [BPMEN(Cl)Fe^{III}OFe^{III}(Cl)BPMEN] $(ClO_4)_2$ [BPMEN = N,N'-dimethyl-N,N'-bis (2-pyridylmethyl)ethane-1,2-diamine] was prepared and characterised by X-ray crystallography; the ^{1}H NMR peaks of this complex in CD_3CN were assigned based on the integration and proximity of particular protons to a paramagnetic centre.

EPR IDENTIFICATION OF Zr(III) COMPLEXES FORMED UPON INTERACTION OF $(2\text{-PhInd})_2ZrCl_2$ AND *rac*-$Me_2Si(1\text{-Ind})_2ZrCl_2$ WITH MAO AND MMAO

O.Y. Lyakin, K.P. Bryliakov, V.N. Panchenko, N.V. Semikolenova, V.A. Zakharov, E.P. Talsi

Macromol. Chem. Phys.,
208 (2007) pp. 1168-1175.

Reactions of **1** and **2** with MAO and MMAO were monitored by EPR. It was found that MMAO is a stronger reducing agent than MAO. **1** is more prone to reduction than **2**. The reduction of Zr^{IV} to Zr^{III} seems to be the essential pathway of some zirconocene catalysts' deactivation. Zr^{III} species with the following proposed structures can be identified in the **1**/MMAO system: $(2\text{-PhInd})_2Zr^{III}(^i\text{Bu})$, $(2\text{-PhInd})_2Zr^{III}(\mu\text{-Cl})_2Al^i\text{Bu}_2$, $(2\text{-PhInd})_2Zr^{III}(\mu\text{-Cl})(^i\text{Bu})Al^i\text{Bu}_2$, and $[(2\text{-PhInd})_2Zr^{III}]^+[\text{Me-MAO}]^-$. The degree of reduction of Zr^{IV} species determined by EPR in the catalytic system **2**/MMAO can be masked by the formation of diamagnetic Zr^{III}/Zr^{III} dimers. Addition of monomers to the **2**/MAO system promotes reduction of the zirconium species.

^{1}H AND ^{13}C NMR STUDIES OF CATIONIC INTERMEDIATES FORMED UPON ACTIVATION OF “OSCILLATING” CATALYSTS $(2\text{-PhInd})_2ZrCl_2$ WITH MAO, MMAO, AND $AlMe_3/[CPh_3]^+[B(C_6F_5)_4]$

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Organomet.,
26(6) (2007) pp. 1536-1540.

The conformationally dynamic unbridged metallocene $(2\text{-PhInd})_2ZrCl_2$ (**1-Cl**) was activated with methylaluminoxane (MAO), modified methylaluminoxane (MMAO), and $AlMe_3/[CPh_3]^+[B(C_6F_5)_4]^-$. The following ion pairs were characterized by ^{1}H and ^{13}C NMR: $[(2\text{-PhInd})_2Zr(\mu\text{-Me})_2AlMe_2]^+[\text{Me-MAO}]^-$ (**III**), $[(2\text{-PhInd})_2Zr(\mu\text{-Me})_2AlMe_2]^+[B(C_6F_5)_4]^-$ (**III**'), $[(2\text{-PhInd})_2Zr(\mu\text{-Me})_2AlMe^i\text{Bu}]^+[\text{Me-MAO}]^-$ ($\mathbf{III_{Me\mathit{i}Bu}}$), $[(2\text{-PhInd})_2Zr(\mu\text{-Me})_2Al^i\text{Bu}_2]^+[\text{Me-MAO}]^-$ ($\mathbf{III_{\mathit{i}Bu\mathit{i}Bu}}$), and $[(2\text{-PhInd})_2ZrMe^+\cdots\text{Me-MAO}^-]$ (**IV**). In the temperature range -50 to 20°C, the rotation of indenyl ligands of complexes **III**, **III**', and $\mathbf{III_{Me\mathit{i}Bu}}$ is faster than the evaluated rate of propylene insertion, whereas for $\mathbf{III_{\mathit{i}Bu\mathit{i}Bu}}$ the rate of indenyl ligand rotation is comparable to or slower than the rate of propylene insertion. The ion pair $\mathbf{III_{\mathit{i}Bu\mathit{i}Bu}}$ demonstrates the fundamental possibility of the existence of intermediates with the "locked" conformation of 2-PhInd ligands in **1-Cl**/MAO or **1-Cl**/MMAO systems. For the catalytic system **1-Cl**/MAO, both outer sphere ion pair **III** and inner sphere ion pair **IV** are present in the reaction solution at high $[Al]_{MAO}/[Zr]$ ratios. For the catalytic system **1-Cl**/MMAO, the concentration of inner sphere ion pairs of type **IV** is much smaller (below the NMR detection limit), and only outer sphere ion pairs $\mathbf{III_{Me\mathit{i}Bu}}$ and $\mathbf{III_{\mathit{i}Bu\mathit{i}Bu}}$ are observed.

^{129}Xe NMR STUDY OF THE LOCALIZATION OF $PdCl_2$ SUPPORTED ON CARBON NANOTUBES

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React. Kinet. Catal. Lett.,
90(2) (2007) pp. 355-364.

Localization of $PdCl_2$ clusters supported on multi-wall carbon nanotubes (MWCNT) has been investigated using ^{129}Xe NMR of adsorbed xenon. As-made MWCNTs with channels initially inaccessible for adsorption and ball-milled MWCNTs with the totally accessible internal surface were used as supports. The observed ^{129}Xe NMR spectra were determined by the dynamics of xenon exchange between the aggregate pores and nanotube channels. No considerable changes of the ^{129}Xe NMR spectrum with the concentration of supported $PdCl_2$ were observed for the as-made MWCNT, while an additional resonance appeared for the ball-milled nanotubes. The ^{129}Xe NMR experiments evidenced the supported species to be localized on the internal surface of the ball-milled MWCNT.

IS ^{129}Xe NMR A USEFUL TECHNIQUE FOR PROBING THE PORE STRUCTURE AND SURFACE PROPERTIES OF CARBONACEOUS SOLIDS?

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Micropor. Mesopor. Mater.,
105 (2007) pp. 118-123.

Generally the interpretation of ^{129}Xe NMR data obtained for porous carbons is complicated. There are several reasons: structural disorder, distribution of the crystallite size or presence of amorphous domains, heterogeneity of surface properties, strong paramagnetic sites, etc. Thus it would appear that structural disorder, bad reproducibility of properties and various impurities make ^{129}Xe NMR ineffective when applied to porous carbon materials. This problem is re-examined for a series of microporous carbons with similar surface chemical properties and mesoporous carbons with a wide range of pore diameters. In both cases there is no relationship between the value, δ_S, of the chemical shift extrapolated to zero concentration and the pore diameter. However, the virial coefficient, δ_{Xe-Xe}, arising from binary xenon collisions in the free volume, varies linearly with the micropore size. In the case of mesoporous carbons there is a monotonic decrease of this coefficient with the increase of the pore size, if one take into account binary xenon collisions at the pore surface.

MULTINUCLEAR MAGNETIC RESONANCE IMAGING AS A MULTIFUNCTIONAL TOOL FOR THE INVESTIGATION OF THE PROPERTIES OF MATERIALS, TRANSPORT PROCESSES AND CATALYTIC REACTIONS

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Russ. Chem. Rev.,
76(6) (2007) pp. 583-598.

New physicochemical applications of multinuclear magnetic resonance imaging that will determine the development of the method in the near future are considered. These include studies of solids, investigations of mass transfer processes in multiphase systems, studies on *in situ* reactors, heterogeneous hydrogenation with para-hydrogen and spectroscopy of long-lived singlet states. It is noted that in addition to an extension of the range of objects and processes of interest, the magnetic resonance imaging technique itself undergoes considerable changes.

MULTINUCLEAR MAGNETIC RESONANCE IMAGING IN CATALYTIC RESEARCH: RECENT ADVANCES AND FUTURE PROSPECTS

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Kinet. Catal., 48(4) (2007) pp. 464-468.

The most important applications of magnetic resonance imaging in catalysis and related areas are considered. In combination with other advanced instrumental methods, this technique can provide essential information about the properties of catalysts and reactors and about processes occurring there. The examples given in the report include the preparation and characterization of porous supports; loading of supports with an active component by impregnation; and investigation of the structure of a granular catalyst bed, various mass transfer processes, and the operation dynamics of a model reactor.

APPLICATION OF MULTINUCLEAR MRI AND SOLID STATE MRI IN HETEROGENEOUS CATALYSIS

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Catal. Today,
126(1-2) (2007) pp. 37-43.

This paper presents some of the existing applications of the NMR imaging (MRI) technique relevant to catalytic research. Through the examples presented, it is demonstrated that MRI is a powerful addition to other modern techniques employed to characterize properties of catalysts and catalytic reactors and their performance. It is shown that the existing MRI approaches can be applied to get insight into a broad range of characteristics of catalysts and reactors. The examples presented include preparation and characterization of porous catalyst supports, dynamic studies of supported catalysts preparation by solution impregnation, packing and structure of catalyst beds and reactors, and various types of mass transport in the catalyst particles as well as in the entire reactor. Last but not least, it is shown that rapid MRI strategies are useful to study dynamic processes in operating catalytic reactors, with the multiphase catalytic hydrogenation reaction at elevated temperatures used as a representative example. These *in situ* studies demonstrate a number of essential phenomena including ignition of individual catalyst particles and their mutual influence, while combination of the spectroscopic and imaging modalities of the NMR technique can be useful for the characterization of the chemical conversion under reactive conditions.

^{27}Al NMR/MRI STUDIES OF THE TRANSPORT OF GRANULAR Al_2O_3

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Diffusion Fundam.,
5 (2007) pp. 2.1-2.7

The NMR/MRI techniques are applicable to the studies of motion of granular solids, providing information on the velocities, effective diffusivities and correlation times of the moving particles. The studies of transport of granular solids reported to-date are based on the detection of the ^{1}H NMR signal of the liquid phase of liquid-containing solid materials. Yet, the solid phase of many granular solids contains magnetic nuclei, providing in principle an opportunity to study motion of such solids by directly detecting the NMR signal of the solid phase. In this paper, it is demonstrated that this can be performed with the use of conventional echo pulse sequences in combination with the conventional motion encoding schemes. The detection of the ^{27}Al NMR signal of the Al_2O_3 powder was used to obtain velocity maps of the powder packed in a spinning cylinder, and to measure the velocity distribution (average propagator) for the gravity driven transport of the same powder in a vertical pipe.

NUCLEAR MAGNETIC RESONANCE IMAGING OF AN OPERATING GAS–LIQUID–SOLID CATALYTIC FIXED BED REACTOR

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Chem. Eng. J., 130(2-3) (2007) pp. 101-109.

This work reports pioneering application of the nuclear magnetic resonance imaging (MRI) technique to the dynamic *in situ* studies of gas–liquid–solid reactions carried out in a catalytic trickle bed reactor at elevated temperature. The major advance of these studies is that MRI experiments are performed under reactive conditions. MRI have been applied to map the distribution of liquid phase inside a catalyst pellet as well as in a catalyst bed in an operating trickle-bed reactor. In particular, the studies have revealed the existence of the oscillating regimes of the heterogeneous catalytic hydrogenation reaction caused by the oscillations of the catalyst temperature and directly demonstrated the existence of the coupling of mass and heat transport and phase transitions with chemical reaction. The existence of the partially wetted pellets in a catalyst bed which are potentially responsible for the appearance of hot spots in the reactor has been also visualized. The combination of NMR spectroscopy with MRI has been used to visualize the spatial distribution of the reactant-to-product conversion within an operating reactor.

^{95}Mo AND ^{17}O NMR STUDIES OF AQUEOUS MOLYBDATE SOLUTIONS

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Inorg. Chem.,
46(9) (2007) pp. 3688-3695.

Aqueous molybdate solutions with molybdenum concentrations of [Mo] = 1, 0.4, and 0.2 M have been studied by NMR at pH 7-1 and in 0.3-6 M $HClO_4$. The ^{95}Mo NMR spectrum of isopolyanion (IPA) $Mo_7O_{24}^{6-}$ (**I**) at pH = 5 consists of a signal at 210 ppm and two overlapping peaks at 32 and ~15 ppm with the intensity ratio ~1:4:2, and that of β-$Mo_8O_{26}^{4-}$ (**II**) consists of two signals at ~100 and 10 ppm with the intensity ratio ~1:3. A broad ^{95}Mo NMR line at around 0 ppm was observed in the pH range of IPA $Mo_{36}O_{112}^{8-}$ (**III**), and a signal of cationic oxospecies including MoO_2^{2+} (**IV**) was observed from -62 to -69 ppm. Two protonation sites of IPA **I** have been identified from ^{17}O NMR spectra, which suggests binding of up to two protons. The distribution diagram, derived from the ^{95}Mo NMR spectra, is given for [Mo] = 0.4 M. The ^{95}Mo NMR signals shift to lower frequencies with increasing number and strength of the Mo-O terminal bonds.

SOLID STATE NMR CHARACTERIZATION OF INDIVIDUAL COMPOUNDS AND SOLID SOLUTIONS FORMED IN Sc_2O_3–V_2O_5–Nb_2O_5–Ta_2O_5 SYSTEM

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Magn. Reson. Chem.,
45(11) (2007) pp. 962-970.

In this study, ^{51}V, ^{45}Sc and ^{93}Nb MAS NMR combined with satellite transition spectroscopy analysis were used to characterize the complex solid mixtures: $VNb_{9(1-x)}Ta_{9x}O_{25}$, $ScNb_{(1-x)}Ta_xO_4$ and $ScNb_{2(1-x)}Ta_{2x}VO_9$ (x = 0, 0.3, 0.5, 0.7, 1.0). This led to describe the structures of Sc and V sites. The conclusions were based on accurate values for ^{51}V quadrupole coupling and chemical shift tensors obtained with ^{51}V MAS NMR/SATRAS for VNb_9O_{25}, VTa_9O_{25} and $ScVO_4$. The ^{45}Sc NMR parameters have been obtained for Sc_2O_3, $ScVO_4$, $ScNbO_4$ and $ScTaO_4$. On the basis of ^{45}Sc NMR and data available from literature, the ranges of the ^{45}Sc chemical shift have been established for ScO_6 and ScO_8. The gradual change of the ^{45}Sc and ^{51}V NMR parameters with x confirms the formation of solid solutions in the process of synthesis of $VNb_{9(1-x)}Ta_{9x}O_{25}$ and $ScNb_{(1-x)}Ta_xO_4$, in contrast to $ScNb_{2(1-x)}Ta_{2x}VO_9$. The cation sublattice of $ScNb_{(1-x)}Ta_xO_4$ is found to be in octahedral coordination. The V sites in $VNb_{9(1-x)}Ta_{9x}O_{25}$ are present in the form of slightly distorted tetrahedra. The ^{93}Nb NMR parameters have been obtained for VNb_9O_{25}.

INVESTIGATION OF THE FORMATION PROCESS OF NANOSIZED PARTICLES OF Ru(III)

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J. Struct. Chem.,
48(1) (2007) pp. 144-149.

^{17}O, ^{35}Cl, ^{133}Cs NMR spectroscopy, static magnetic susceptibility measurements, EXAFS, UV spectroscopy, pH measurements, and electron microscopy are applied to investigate alkaline hydrolysis of aqueous $K_2[RuCl_5H_2O]$. It is found that on addition of a base ligand substitution is accompanied by polycondensation processes which afford filament structures consisting of $Ru(OH)_4Cl_2$ octahedra bonded by single OH-bridges. When polynuclear compounds are sorbed on the surface of carbonic substrates, the filaments aggregate into nanosized particles which have a diameter of approximately 2.0 nm with a narrow size distribution.

THE CHEMICAL COMPOSITION AND STRUCTURE OF SUPPORTED SULFATED ZIRCONIA REGULATED SIZE NANOPARTICLES

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AIP Conference Proceedings,
X-Ray Absorption Fine Structure - XAFS13,
882 (2007) pp. 648-650.

A set of model skeletal isomerization catalysts — sulfated zirconia nanoparticles of controlled thickness anchored on different supports — was prepared using colloidal solutions of Zr salt on titania as support. The nanoparticles of zirconia (1–5 nm) are epitaxially connected to the support surface, with S/Zr ratio equals to 1.3–1.5. It was shown by EXAFS that nanoparticles of non-stoichiometric zirconium sulfate $Zr(SO_4)_{1+x}$, where x<0.5, are formed on the support surface. Its structure looks like half-period shifted counterdirected chains built-up by zirconium atoms linked by triangle pyramids of sulfate groups. Considering catalytic data of skeletal n-butane isomerisation at 150°C, one can suggest that these species behave as the active component of sulfated zirconia. They are formed in subsurface layers as zirconium hydroxide undergoes sulfation followed by thermal treatment.

SYNTHESIS OF NICKEL-DOPED ALKALI HALIDE MICROCRYSTALS AND THEIR SPECTRAL AND STRUCTURAL ANALYSIS

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J. Struct. Chem.,
48(6) (2007) pp. 1099-1104.

Nickel-doped alkali halide microcrystals were obtained by various methods, including an original synthetic procedure. Their spectral and structural characteristics were investigated. The crystal field strength was calculated from the reflection spectra recorded for the microcrystals for different impurity concentrations. In contrast to single crystals, higher impurity concentrations can be obtained for lower degrees of aggregation in microcrystals.

MODIFICATION OF METHYLALUMINOXANE-ACTIVATED *ANSA*-ZIRCONOCENE CATALYSTS WITH TRIISOBUTYLALUMINUM - TRANSFORMATIONS OF REACTIVE CATIONS STUDIED BY NMR SPECTROSCOPY

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Chem. Eur. J.,
13(18) (2007) pp. 5294-5299.

When triisobutylaluminum ($\mathrm{Al}i\mathrm{Bu}_3$) is added to solutions containing methylaluminoxane (MAO) and *rac*-$[\mathrm{Me_2Si(ind)_2ZrCl_2}]$ (ind: indenyl) in C_6D_6, NMR spectra show that methyl-bridged mixed-alkylaluminum dimers $\mathrm{Al(\mu\text{-}Me)_2Me}_{4\text{-}x}i\mathrm{Bu}_x$ predominate. These dimers react with MAO under partial transfer of isobutyl groups and induce a conversion of the initially prevailing cationic trimethylaluminum adduct *rac*-$[\mathrm{Me_2Si(ind)_2Zr(\mu\text{-}Me)_2AlMe_2}^+]$ to *rac*-$[\mathrm{Me_2Si(ind)_2Zr(\mu\text{-}Me)_2AlMe}i\mathrm{Bu}^+]$ and *rac*-$[\mathrm{Me_2Si(ind)_2Zr(\mu\text{-}Me)_2Al}i\mathrm{Bu_2}^+]$. These species are unstable and release isobutene under formation of zirconocene hydrides.

STATE OF RHODIUM(III) IN SULFURIC ACID SOLUTIONS

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Russ. J. Coord. Chem.,
33(2) (2007) pp. 136-139.

The formation of rhodium(III) sulfate complexes under moderately rigorous temperature conditions was studied by ^{103}Rh and ^{17}O NMR spectroscopy. The complexes $[\mathrm{Rh_2(\mu\text{-}SO_4)_2(H_2O)_8}]^{2+}$, $[\mathrm{Rh_2(*\mu\text{-}SO_4)(H_2O)_8}]^{4+}$, and $[\mathrm{Rh_3(\mu\text{-}SO_4)_3(\mu\text{-}OH)(H_2O)_{10}}]^{2+}$ were found to be the most stable species in aged solutions.

THE STUDY OF INDUCED AQUATION OF Rh(III) SULFATE COMPLEXE

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Russ. J. Coord. Chem.,
33(2) (2007) pp. 140-144.

The processes of aquation of Rh(III) complexes in the presence of $BaCl_2$ and $Ba(ClO_4)_2$ were studied by the ^{103}RH and ^{17}O NMR methods. In the first case, the final products were found to be the monomeric aqua chloride complexes, while in the second case, aqua hydroxo complexes were formed. The chloride ions present in the system significantly increase the process rate.

COMPLEXATION OF Rh(III) IN DILUTED SULFURIC ACID SOLUTIONS

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Russ. J. Coord. Chem.,
33(6) (2007) pp. 449-453.

Complex formation in a system $\mathrm{Rh(III)–H_2SO_4–H_2O}$ was studied by the ^{103}Rh and ^{17}O NMR spectroscopy at room temperature. The formation of two interrelated systems of mononuclear and polynuclear complexes was established in the above solutions. The predominant species in the first system is a labile ionic pair $\{\mathrm{Rh(H_2O)_6}^{3+}\mathrm{SO_4}^{2-}\}^+$, while in the second system, two inert binuclear complexes $[\mathrm{Rh_2(\mu\text{-}SO_4)_2(H_2O)_8}]^{2+}$ and $[\mathrm{Rh_2(\mu\text{-}SO_4)(\mu\text{-}OH)(H_2O)_8}]^{3+}$ prevail.

CHARACTERIZATION OF A Pd–Fe BIMETALLIC MODEL CATALYST

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Surf. Sci.,
601(10) (2007) pp. 2105–2116.

Small bimetallic Pd-Fe particles supported on a well ordered alumina film grown on NiAl (110) were studied focusing on the geometric, electronic, adsorption, as well as magnetic properties. The morphology, growth mode and surface composition were investigated by combining scanning tunneling microscopy (STM), temperature-programmed desorption (TPD) and infrared spectroscopy (IRAs) using CO as a probe molecule. Information on the electronic properties of the bimetallic systems was obtained by means of X-ray photoelectron spectroscopy (XPS). These measurements were amended by *in situ* ferromagnetic resonance spectroscopy to address the magnetic properties of the bimetallic particles. The subsequent deposition of the metals at 300 K varying the order of metal deposition resulted in two distinct bimetallic systems. Pd deposited on existing Fe particles forms a shell, however, FMR and XPS suggest that intermixing of Pd and Fe occurs to some extent. For the reverse order, a larger amount of Fe is required to coat Pd particles, due to the strong tendency of Pd to segregate to the surface of the particles.

THERMALLY ACTIVATED DISSIPATION IN A NOVEL FOAMED Bi-BASED OXIDE SUPERCONDUCTOR IN MAGNETIC FIELDS

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Supercond. Sci. Technol.,
20(6) (2007) pp. 491-494.

The transport properties of a novel foamed superconductor $Bi_{1.8}Pb_{0.3}Sr_{1.9}Ca_2Cu_3O_x$ have been studied. As the analysis of resistive transitions in a magnetic field shows, the dissipation follows a thermally activated flux creep model with the temperature-independent pinning potential in S-N-S-type Josephson junctions.

FMR FINE STRUCTURE - A TOOL TO INVESTIGATE THE SPATIAL MAGNETIC PHASE SEPARATION PHENOMENA IN MANGANITES

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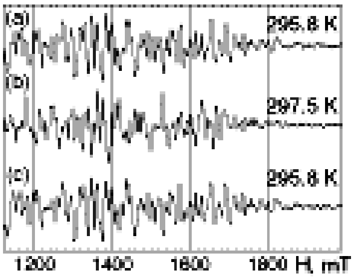
Phys. Status Solidi (RRL),
1(1) (2007) pp. R22-R24.

An original approach is proposed to study the magnetic phase separation phenomenon. It is based on the registration of the noise-like FMR Fine Structure (FMR FS) caused by the magnetic interparticle dipole-dipole interaction between spatially separated ferromagnetic regions. Data obtained for a $La_{0.7}Pb_{0.3}MnO_3$ single crystal point to the existence of spatially separated ferromagnetic regions. It is shown that FMR FS of the $La_{0.7}Pb_{0.3}MnO_3$ single crystal is temperature reversible and disappears at the maximum of magnetoresistance.



HIGHLY TEXTURED BISMUTH-CONTAINING HIGH-TEMPERATURE SUPERCONDUCTOR CERAMICS OBTAINED BY UNIAXIAL PRESSING IN LIQUID MEDIUM: FABRICATION AND PROPERTIES

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Techn. Phys. Lett.,
33(9) (2007) pp. 740-743.

A new method of fabricating textured ceramics with a composition of $Bi_{1.8}Pb_{0.3}Sr_{1.9}Ca_2Cu_3O_x$ is described, according to which the initial low-density ceramics is subjected to uniaxial pressing in a liquid medium followed by drying and annealing. Analysis of the X-ray diffraction and scanning electron microscopy data show evidence for a high degree of texture in high-temperature superconductor ceramics fabricated using the proposed method. The results of magnetic measurements indicate that the obtained material possesses anisotropy of the magnetic moment for the magnetic field **H** oriented parallel and perpendicular to the direction of uniaxial pressing. The textured ceramics also show high diamagnetic response for **H** parallel to the *c* axis of grains, which makes these ceramics promising materials for practical applications.

FERROMAGNETIC RESONANCE STUDY OF THIN FILM ANTIDOT ARRAYS: EXPERIMENT AND MICROMAGNETIC SIMULATIONS

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Phys. Rev. B,
75(17) (2007) 174429 (6 pages).

The dynamic magnetic properties of two-dimensional periodic Co antidot arrays were studied by X-band ferromagnetic resonance. The experimental results on geometrically scaled antidot arrays reveal a strong attenuation of the uniform ferromagnetic resonance mode in comparison to a continuous film, but an excitation of nonuniform in-plane spin-wave modes. Micromagnetic finite-element simulations show that the static magnetic structure in an antidot array depends on the direction of the external field with respect to the symmetry axes of the antidot lattice, even if the external field is strong enough to enforce a technically saturated magnetization state. The analysis gives evidence that characteristic inhomogeneities in the magnetization distribution around the antidots give rise to the changes of the resonance modes with the in-plane direction of the magnetization.

OXIDATION OF ALUMINA-SUPPORTED Co AND Co-Pd MODEL CATALYSTS FOR THE FISCHER-TROPSCH REACTION

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J. Phys. Chem.,
111(24) (2007) pp. 8566-8572.

Cobalt and bimetallic Co-Pd systems are well-known Fischer-Tropsch catalysts. Compared to Co, the bimetallic systems exhibit an increased activity toward CO hydrogenation and methane conversion, attributed to resistance against oxidation. To study the oxidation behavior, model catalysts have been generated by depositing either Co or first Co and subsequently Pd onto a thin epitaxial alumina film grown on NiAl(110). Pure Co particles and bimetallic particles with a Co core and a Pd shell have been studied before and after exposure to oxygen and after thermal treatments, using X-ray photoelectron spectroscopy (XPS), temperature-programmed CO desorption (TPD), ferromagnetic resonance (FMR), and infrared reflection absorption spectroscopy (IRAS) in ultrahigh vacuum. Large doses of O_2 (1000 langmuirs; 1 langmuir = 10^{-6} Torr·s) at 300 K lead to complete oxidation of Co particles. Upon annealing to temperatures above 530 K, XPS indicates that the cobalt oxide is mostly reduced by transfer of oxygen to the alumina support, resulting in its thickening. TPD, however, indicates the existence of persistent surface oxygen species, reducing the CO adsorption energy on the particles. Exposures to small doses of O_2 (30-50 langmuirs) were also studied by a careful comparison of XPS, TPD, and FMR data. In this case, XPS indicates Co in a metallic state, whereas TPD and FMR indicate oxidic behavior. It is concluded that small amounts of non-stoichiometric subsurface oxygen or subsurface and surface oxygen are present which are not detectable in the Co 2p XPS signal but have a pronounced effect on the surface chemistry and the magnetism, i.e., on certain bulk properties. In the case of bimetallic Co/Pd particles, an incomplete Pd shell on the Co particles even promotes oxygen uptake, while only a complete Pd shell inhibits oxidation.

INVESTIGATION OF THE HYDROGEN CAPACITY OF COMPOSITES BASED ON ZnOCu

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Crystallogr. Rep., 52(3) (2007) pp. 474-478.

The composites ZnOCuH(D) saturated with hydrogen (deuterium) to a content of ~1 wt % are investigated by the neutron scattering methods. Upon cooling of the samples (the ZnO matrix containing Cu crystals ~10 nm in size) from 300 to 4 K, hydrogen (deuterium) is condensed on the cluster surface and penetrates inside the clusters in which the atomic hydrogen content with respect to copper can be as high as 30% at 20 K. Simultaneously, hydrogen fills nanopores of the ZnO matrix. It is revealed that, at temperatures of 90–300 K, approximately one-third of the hydrogen amount participates in the fast diffusion (the diffusion constant is approximately equal to 8×10^{-5} cm^2/s) and the other two-thirds are immobilized. At 20 K, the fraction of mobile hydrogen decreases to ~10%. An analysis of the results obtained demonstrates that the energy barriers retaining hydrogen in defect regions are relatively low.

OPTICAL PROPERTIES AND MICROMORPHOLOGY OF THE ZrO_2/Si FILMS, PREPARED BY ION BEAM SPUTTERING DEPOSITION

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Fundament. Problems Modern Mater. Sci., 4(3) (2007) pp. 62-65.

ZrO_2 films have been deposited on Si(100) by ion beam sputtering deposition (IBSD) at temperature T = 70°C. Film structure properties and ZrO_2/Si interface were observed with reflection high energy electron diffraction (RHEED) and high resolution transmission electron microscopy (HRTEM). Optical properties of the film were studied by spectroscopic ellipsometry over the spectral range 250-900 nm.

MAGNETIC NANOPARTICLES BASED ON BORATE GLASSES DOPPED WITH Fe AND Mn

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Func. Mater., 13(2) (2006) pp. 293-299.

Magnetic properties, including magneto-optical Faraday Effect (FE) have been investigated in potassium-aluminum boron glasses doped with iron and manganese oxides. The formation of magnetic nanoparticles has been proved using high resolution electron microscopy. A wide variety of particle size, shape, structure and distribution in the matrix has been observed depending on the technological conditions. The narrow region of the conditions has been established providing formation of almost ideal manganese ferrite single crystals of about 40 nm size distributed rather homogeneously in the matrix. Therefore, the glasses can be considered as a nano-composite material. Thanks to the low concentration of paramagnetic elements, the glasses kept the optical transparence and demonstrated high FE in the infrared and a part of the visible spectral regions, thus making it possible to develop a new magneto-optical material for the 1,0-1,5 μm spectral region.

FROM MOLTEN GLASS TO CRYSTALLIZABLE MELT: THE ESSENCE OF STRUCTURAL EVOLUTION

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J. Crystal Growth, 294(1) (2006) pp. 22-28.

Within the frames of the model concept an attempt is made to reveal the principal differences in the structural self-assembling of melts responsible for their crystallization or vitrification. Results of *in situ* synchrotron diffraction and Raman spectroscopic study of melts, NMR spectroscopy of glasses, and computer modeling of predominant arrangement of cation and anion components of melts with different compositions were used as an experimental basis. The notions of active crystal-forming part of melt were introduced which is an assembly of cation–anion associative complexes and “free” structure-forming cations, and its passive part, which is fragments of polycondensed anion groupings of various scales. Crystallization is interpreted as cooperative interaction caused by the formation of coordination spheres of "free" cations inside precursor clusters resulting from the crystal-forming part of melt.

ANALYSIS OF THE PHASE COMPOSITION OF HUMAN KIDNEY STONES IN MODEL OBJECTS WITH THE USE OF SYNCHROTRON RADIATION DIFFRACTION

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Bull. Russ. Acad. Sci.: Phys.,
71(5) (2007) pp. 653–656.

The possibility of using synchrotron radiation for X-ray diffraction analysis of kidney stones directly in a human has been investigated. Experiments using a special phantom object imitating a human body have been performed. The experimental results obtained show that the significant thickness of the objects studied and the presence of liquids and biological polymers do not impose fundamental limitations on the possibility of in vivo synchrotron radiation diffraction analysis of human kidney stones.

MODEL EXPERIMENT OF *IN VIVO* SYNCHROTRON X-RAY DIFFRACTION OF HUMAN KIDNEY STONES

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Nucl. Instrum. Methods Phys. Res., sec. A,
575(1-2) (2007) pp. 221-224.

The diffraction of synchrotron radiation (SR) was used to explore the phase composition of kidney stones placed into a specific object phantom, which imitated the human body. As an imitation of the patient breath, the kidney stone was moved vertically and rotated to an angle of 15° during the recording of the X-ray pattern. It was shown that rotation and displacement did not distort the X-ray pattern.

APPLICATION OF SYNCHROTRON RADIATION FOR THE STUDY OF NONSTOICHIOMETRIC OXYGEN-PERMEABLE PEROVSKITES BY MEANS OF X-RAY DIFFRACTION

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Nucl. Instrum. Methods Phys. Res., sec. A,
575(1-2) (2007) pp. 144-148.

In the present work, applicability and advantages of the use of synchrotron radiation to the X-ray diffraction (XRD) study of the phase composition and microstructure of nonstoichiometric oxygen-permeable oxides having the composition $Sr_{1-x}A_xCo_{0.8-y}Al_yFe_{0.2}O_z$, (where A=La, Ca) and $SrFe_{1-x}M_xO_z$, (where M=W, Mo) have been demonstrated.

ADSORPTION OF MOLECULAR HYDROGEN ON ULTRAFINE MICROPOROUS/MESOPOROUS MATERIALS

V.Yu. Gavrilov

Kinet. Catal.,
48(6) (2007) pp. 799-802.

The comparative processing of H_2 adsorption isotherms obtained at 77 K is demonstrated to be applicable to the investigation of the microtexture of the ultrafine oxide materials MCM-41 and ZSM-5 and their mechanical mixtures. The H_2 sorption method allows the micropore volume to be determined correctly for mixed ultrafine microporous/mesoporous materials.

SPECIFIC FEATURES OF THE COMPARATIVE ADSORPTION ANALYSIS OF THE TEXTURE OF ULTRAFINE MICROPOROUS/MESOPOROUS MATERIALS

V.Yu. Gavrilov, E.E. Sokolov

Kinet. Catal.,
48(6) (2007) pp. 794-798.

N_2 adsorption on the uniformly mesoporous material MCM-41, zeolite ZSM-5, and their mechanical mixtures at 77 K is reported. The use of the differential comparative method in the analysis of adsorption isotherms for this class of materials is complicated by the weak sorbate–MCM-41 surface adsorption interaction and, for adsorbent mixtures, by the compensation effect caused by the introduction of adsorption sites with an increased adsorption potential. Specific features of sorption on the two-component system are analyzed in the framework of the conventional BET model.

IDENTIFICATION OF GOLD OXIDE CLUSTER STRUCTURES IN Au/Al_2O_3 CATALYSTS FOR LOW-TEMPERATURE CO OXIDATION

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Doklady Phys. Chem.,
413(2) (2007) pp. 75-80.

The chemical state of gold in Au/Al_2O_3 catalysts prepared by different methods (incipient wetness technique or absorption of gold(III) hydroxide complexes) was studied by diffuse reflectance (DR) UV-vis spectroscopy. Modeling of the plasmon absorption by small metallic gold particles as well as quantum-chemical calculations of the electronic spectra of hypothetical dimeric $[AuX(\mu_2–O)(H_2O)]_2$ (X = Cl или OH) complexes by the density functional theory (DFT) method were performed for the assignment of the absorption bands in the DR UV-vis spectra of gold catalysts. Comparison between the experimental and calculated data shows that the Au/Al_2O_3 catalysts calcined in air at 400°C contain both metallic gold nanoparticles and gold oxide clusters chemically anchored onto the support surface.

DISTORTION OF THE TETRAHEDRAL COORDINATION OF Fe(III) IONS STABILIZED IN ZSM-5 ZEOLITE FRAMEWORK

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(**Technical University Eindhoven, The Netherlands*)

J. Struct. Chem.,
48(5) (2007) pp. 855-861.

A simple qualitative method to analyze *d-d*-electronic transitions in cations of the transition elements in oxide matrices is proposed. In the particular case, all the excited states of interest differ only in the electronic configuration of *d*-orbitals, and the energies of transitions can be computed via the configuration interaction (CI) method restricted by the active space of five cation *d*-orbitals. An ordinary cluster model that takes into account the first coordination sphere of transition metal ion consisting of the framework of oxygen ions is sufficient for this purpose. The systematic overestimation error of transition energies can be corrected through the empirical factor calculated to fit experimental UV-vis spectra. The physical meaning of the scaling factor proposed is the dynamic part of electron correlation that remains unaccounted for in the chosen active CI space. The observed *d-d*-transitions of Fe^{3+} ions in MFI zeolites are analyzed in detail. It is suggested that the specifics of the observed electronic spectra are caused by the distortion of the tetrahedron of oxygen atoms around Fe^{3+}. The latter can be easily taken into account when selecting an appropriate Fe^{3+} cluster model in the framework. It is shown that the occurrence of the weak low-frequency band below 21,000 cm^{-1} indicates the distortion of the tetrahedral environment around Fe^{3+}.

STUDIES OF OXIDATIVE CAPACITY OF CERIUM-CONTAINING OXIDES OF VARIOUS COMPOSITIONS FOR CATALYSTS OF THE AUTOMOTIVE EXHAUST GASES CLEANING

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Catal. Ind., 6 (2007) pp. 39-45.

Introducing of cerium-containing oxide components into the catalysts for exhaust gases purification leads to improving the catalyst action in the conditions of variable composition of redox gas mixture. Promotion action of cerium containing oxides is conditioned by their ability to accumulate and give back oxygen (during transition of Ce^{+3} and Ce^{+4}) in dependence of partial pressure of oxygen in gas phase. The qualitative estimation of efficiency of the action of cerium containing component can be based on dynamic oxygen capacity. The dynamic oxygen capacity of cerium containing catalysts of various compositions: $CeO_{2-\delta}$; $Ce_{0,8}Zr_{0,2}O_{2-\delta}$; $Ce_{0,65}Zr_{0,2}Pr_{0,15}O_{2-\delta}$; $Ce_{0,8}Nd_{0,2}O_{2-\delta}$ and $Ce_{0,8}Gd_{0,2}O_{2-\delta}$ was characterized with the purpose of election the most optimal component for the catalysts for exhaust gases purification. Specific surface, porosity, phase composition and structure of the synthesized oxides were characterized using XPS, XPA, PEEM. Dynamic oxygen capacity (OSC) of oxides was evaluated in temperature interval 250-500°C in reaction of CO + O_2 oxidation based on data on degree of CO oxidation in the presence of mixed sample (Pt/Al_2O_2 + oxide) and of catalyst Pt/Al_2O_2 supported on cordierite block of honeycomb structure. It was stated that oxides $Ce_{0,8}Zr_{0,2}O_{2-\delta}$ and $Ce_{0,65}Zr_{0,2}Pr_{0,5}O_{2-\delta}$ revealed the most dynamic oxygen capacity, low changes after hydrothermal ageing at 1050°C. These allowe these oxides to be recommend for the use for preparation of catalysts for cleaning of exhaust gases.

SMALL-ANGLE X-RAY CHARACTERIZATION OF THE NUCLEOPROTEIN COMPLEXES RESULTING FROM DNA-INDUCED OLIGOMERIZATION OF HIV-1 INTEGRASE

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Nucleic Acids Res., 35(3) (2007) pp. 975-987.

HIV-1 integrase (IN) catalyses integration of a DNA copy of the viral genome into the host genome. Specific interactions between retroviral IN and long terminal repeats (LTR) are required for this insertion. To characterize quantitatively the influence of the determinants of DNA substrate specificity on the oligomerization status of IN the small angle X-ray scattering (SAXS) technique was used. Under certain conditions in the absence of ODNs IN existed only as monomers. IN preincubation with specific ODNs led mainly to formation of dimers, the relative amount of which correlated well with the increase in the enzyme activity in the 30-processing reaction. Under these conditions, tetramers were scarce. Non-specific ODNs stimulated formation of catalytically inactive dimers and tetramers. Complexes of monomeric, dimeric and tetrameric forms of IN with specific and non-specific ODNs had varying radii of gyration (*R*g), suggesting that the specific sequence-dependent formation of IN tetramers can probably occur by dimerization of two dimers of different structure. From data it can be concluded that the DNA-induced oligomerization of HIV-1 IN is probably of importance to provide substrate specificity and to increase the enzyme activity.

ELECTRONIC STRUCTURE OF HEAVY-ELEMENT OXIDES AND FLUORIDES: XANES L_{III} SPECTROSCOPY AND DFT CALCULATIONS

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Nucl. Instrum. Methods Phys. Res. A,
575(1-2) (2007) pp. 159-161.

XANES L_{III} spectra of series Tl, Pb and Bi compounds are studied. The data of DFT–ZORA calculations are in satisfactory agreement with the results of XANES spectroscopy of heavy-element fluorides and oxides. The fine structure of L_{III} absorption near edge for Tl^{3+}, Pb^{4+} and Bi^{5+} ions is associated with transitions to the atomic $6s_{1/2}$, $6p_{1/2}$ (relativistic allowed) and 6d–7s terms. The “superfluous” lines observed in XANES spectra of Tl^{+}, Pb^{2+} and Bi^{3+} oxides and fluorides are associated with the mix of $6s_{1/2}$ and $6p_{3/2}$ states of heavy $6s^2$ ions.

THE PREPARATION OF GOLD NANOPARTICLES IN TRITON N-42 REVERSE MICELLES AFTER PRELIMINARY CONCENTRATION FROM ACID SULFATE–CHLORIDE SOLUTIONS

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Russ. J. Phys. Chem.,
80(12) (2006) pp. 1980-1985.

The distribution of Au(III) between an acid (3 mol/l HCl) aqueous phase and a micellar Triton N-42 solution in *n*-decane was studied as a function of the concentration of Na_2SO_4 (0–3.55 mol/l). The high distribution coefficients (200–500) allow 50-fold absolute concentration to be performed at a 90% extraction. Au^0 nanoparticles were obtained by the injection solubilization of a reducing agent (hydrazine) into micellar extracts. At low solubilization capacities (≤1 vol %) and high reducing agent concentrations (≥0.2 mol/l) in Triton N-42 micelles, stable systems of Au^0 nanoparticles suitable for the spectrophotometric determination of gold were obtained. At high solubilization capacities and low hydrazine concentrations, reduction was accompanied by coagulation and sedimentation processes. These processes were studied by spectrophotometry and the static and dynamic light scattering methods.

PHASE ANALYSIS OF PRODUCTS OF MECHANOCHEMICAL TREATMENT BY REFERENCE-FREE STOICHIOGRAPHY METHOD BASED ON THE DIFFERENTIAL DISSOLUTION

V.V. Malakhov, L.S. Dovlitova, A.A. Vlasov, N.N. Boldyreva

Chem. Sustain. Devel.,
15(2-1) (2007) pp. 117-125.

Main principles of reference-free stoichiography method based on the Differential Dissolution (DD) are reported. This method allows determine stoichiometry and quantitative content of crystalline and amorphous substances of constant and variable composition without using reference samples of corresponding individual phases. High selectivity and sensibility of DD method, as well as special equipment provides simplicity and rapidity of DD-analysis of various objects. Products of mechanochemical treatment as usual are characterized by unknown phase composition, defect crystal or amorphous structure. Application of X-ray, IR spectroscopy and other methods for phase analysis often appears to be inefficient in these cases. At the same time DD method allows to solve most of problems appeared upon investigation of phase composition, structure and properties of such substances. At present this method is applied for investigation of about 20 systems, the results are summarized in the paper.

EFFECTS OF EVAPORATION AND MELTING ON NONSTOICHIOMETRY AND INHOMOGENEITY OF $LiInSe_2$ CRYSTALS

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J. Therm. Anal. Calorim.,
90(2) (2007) pp. 601-605.

Evaporation and compositional changes of the liquids above the melting point of $LiInSe_2$ crystals have been characterized quantitatively by using special techniques of a rapid thermal analysis and differential dissolution. The occurrence of a liquid immiscible region in the Li_2Se-rich side of the $Li_2Se–In_2Se_3$ diagram and incongruent evaporation with the preferential evaporation of In_2Se_3 rising markedly above boiling point were determined from the peaks on the thermal curves and from precise control over the composition of the vapour and residual solid as a function of temperature. It was shown that both the processes could be the sources of nonstoichiometry and inhomogeneity of the $LiInSe_2$ crystals.

ACIDIC PROPERTIES OF FIBERGLASS MATERIALS

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E.A. Paukshtis

React. Kinet. Catal. Lett.,
92(2) (2007) pp. 303-309.

Acidic properties of fiberglass materials were investigated using the adsorption of NH_3 and the rate of isopropanol dehydration. It is shown that the specific catalytic activity of such materials and amount of the Brönsted acid sites per their surface unit (100 Å^2) exceed considerably those in zeolite HZSM-5.

DIRECT OBSERVATION AND MODELING OF ORDERED HYDROGEN ADSORPTION AND CATALYZED *ortho–para* CONVERSION ON ETS-10 TITANOSILICATE MATERIAL

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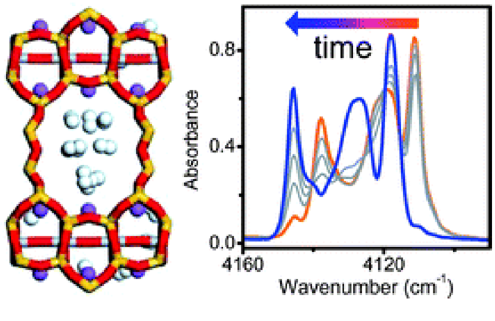
Phys. Chem. Chem. Phys.,
9(21) (2007) pp. 2753-2760.

Hydrogen physisorption on porous high surface materials is investigated for the purpose of hydrogen storage and hydrogen separation, because of its simplicity and intrinsic reversibility. For these purposes, the understanding of the binding of dihydrogen to materials, of the structure of the adsorbed phase and of the *ortho–para* conversion during thermal and pressure cycles are crucial for the development of new hydrogen adsorbents. The direct observation by IR spectroscopic methods of structured hydrogen adsorption on a porous titanosilicate (ETS-10) is reported, with resolution of the kinetics of the *ortho–para* transition, and an interpretation of the structure of the adsorbed phase based on classical atomistic simulations. Distinct infrared signals of *o*- and *p*-H_2 in different adsorbed states are measured, and the conversion of *o*- to *p*-H_2 is monitored over a timescale of hours, indicating the presence of a catalyzed reaction. Hydrogen adsorption occurs in three different regimes characterized by well separated IR manifestations: at low pressures ordered 1 : 1 adducts with Na and K ions exposed in the channels of the material are formed, which gradually convert into ordered 2 : 1 adducts. Further addition of H_2 occurs only through the formation of a disordered condensed phase. The binding enthalpy of the $Na^+–H_2$ 1 : 1 adduct is of $–8.7 \pm 0.1$ kJ mol^{-1}, as measured spectroscopically. Modeling of the weak interaction of H_2 with the materials requires an accurate force field with a precise description of both dispersion and electrostatics. A novel three body force field for molecular hydrogen is presented, based on the fitting of an accurate PES for the $H_2–H_2$ interaction to the experimental dipole polarizability and quadrupole moment. Molecular mechanics simulations of hydrogen adsorption at different coverages confirm the three regimes of adsorption and the structure of the adsorbed phase.



ADSORPTION PROPERTIES OF OXIDIZED GALLIUM-MODIFIED ZEOLITE ZSM-5 FROM DIFFUSE REFLECTANCE IR AND QUANTUM-CHEMICAL DATA.

I. INTERACTION WITH HYDROGEN AND ETHANE

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Kinet. Catal., 48(5) (2007) pp. 735-741.

Diffuse reflectance IR spectroscopy was used to study adsorption and subsequent high-temperature hydrogen and ethane transformations on gallium ions in gallium-modified ZSM-5 zeolite. The results were correlated with the corresponding quantum-chemical calculations. From the experimental and calculated data, it follows that trivalent gallium oxo ions are reduced to the univalent state in a hydrogen or ethane atmosphere even at moderate temperatures. Therefore, gallium oxo ions can function as active sites only at the early stages of light-paraffin aromatization. At the later stages, the dehydrogenation of light paraffins involves univalent gallium ions through the formation of intermediate gallium and alkylgallium hydrides.

BACTERIAL GENESIS OF CALCIUM PHOSPHATES IN HUMAN ORGANISM AND NATURE

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Lithology Mineral Resour.,
42(1) (2007) pp. 56-67.

Comparison of natural marine (nodular and granular) carbonate-apatites and pathogenic mineral structures formed in human cardiac valves by methods of scanning electron and high-resolution transmission microscopy revealed common morphological features corresponding to the bacterial origin of calcium phosphates. These features may be considered typomorphic ones regardless of their formation environments (in vivo or in nature).

POTENTIALS OF GAS CHROMATOGRAPHY IN THE DETERMINATION OF REACTION PRODUCTS IN THE CATALYTIC OXIDATION OF AMMONIA TO NITROGEN(II) OXIDE

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J. Anal. Chem., 62(12) (2007) pp. 1170-1175.

Peculiarities of the determination of nitrogen oxides and ammonia in the reaction products of ammonia oxidation are studied by gas chromatography. Particular attention was paid to the sampling problem. It is shown that in the course of the transportation of samples containing nitrogen(II) oxide and oxygen, the oxidation of nitrogen(II) oxide to nitrogen(IV) oxide takes place in the vapor phase. The conditions were found for suppressing or even eliminating the gas-phase reaction of NO oxidation to NO_2. A procedure for the gas-chromatographic analysis of the reaction products in the catalytic oxidation of ammonia is proposed using several samples and packed columns, including the determination of NO with respect to NO_2 formed in the gas-phase oxidation of NO.

Fundamental and Practical Approaches to Catalyst Preparation

MECHANOCHEMICAL SYNTHESIS OF SUPPORTED COPPER CATALYSTS FOR LACTIC ACID HYDROGENATION

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Chem. Sustain. Devel.,
15(2-1) (2007) pp. 169-173.

In the present work it was studied the opportunity of synthesis of nanodispersed copper catalysts by means of mechanochemical activation. The X-ray diffraction and IR-spectroscopy studies show, that interaction between aluminum and copper (II) oxide results in formation of dispersed metallic copper particles on the surface of alumina during very short activation time. The catalytic activity of obtained catalysts in lactic acid hydrogenation reaction is not high, but the selectivity of propylene glycol formation is significant. The mechanochemical activation of mixture of metallic copper and copper (II) oxide results in formation of copper (I) oxide that doesn't show catalytic activity even after reductive activation.

$LiCoO_2$-SUPPORTED CATALYSTS FOR HYDROGEN GENERATION FROM SODIUM BOROHYDRIDE SOLUTION

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Chem. Sustain. Devel.,
15(2) (2007) pp. 181-187.

Effect of $LiCoO_2$ support prepared by traditional ceramic and mechanochemical route on the activity of Rh and Pt catalysts in the reaction of $NaBH_4$ hydrolysis was studied and compared with traditional supports, such as γ-Al_2O_3, TiO_2 (anatase) and carbon (sibunit). Catalysts supported on $LiCoO_2$ prepared using mechanical activation were found to have the highest catalytic activity. However, the endurance tests of 1 % Rh-$LiCoO_2$ and 1 % Pt-$LiCoO_2$ catalysts showed their gradual degradation. To understand the reason of this phenomenon, the interaction of $LiCoO_2$ with $NaBH_4$ solution was studied by XRD, FTIR, DRS and TEM.

EFFECT OF THE NATURE OF THE ACTIVE COMPONENT AND SUPPORT ON THE ACTIVITY OF CATALYSTS FOR THE HYDROLYSIS OF SODIUM BOROHYDRIDE

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Kinet. Catal., 48(1) (2007) pp. 168-175.

The effect of the nature of an active component and a support on the rate of hydrolysis of aqueous sodium borohydride solutions was studied. It was found that the activity of supported catalysts, which were reduced in a reaction medium of sodium borohydride, decreased in the order Rh > Pt ≈ Ru ≥ Pd regardless of the nature of the support (γ-Al_2O_3, a Sibunit carbon material, or TiO_2). The catalysts based on TiO_2 exhibited the highest activity. As found by UV–vis diffuse reflectance spectroscopy, the composition and structure of the supported precursor of an active component depend on the nature of the support. It is likely that rhodium clusters with different reaction properties were formed on various supports under the action of a reaction medium.

REGULARITIES OF FORMATION OF CUBIC ZIRCONIA STABILIZED WITH CALCIUM, YTTRIUM, AND INDIUM OXIDES UNDER MECHANOCHEMICAL TREATMENT

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Bull. Russ. Acad. Sci.: Phys.,
71(5) (2007) pp. 614-617.

An effect of the composition of reacting components during mechanochemical activation on the ZrO_2 structure stabilized with additives of In_2O_3, CaO, and Y_2O_3 oxides has been revealed. It is established that mechanochemical activation of oxide mixtures leads to the formation of dispersed solid solutions based on cubic zirconia. It is shown that interaction depth of the components during mechanochemical activation increases in the series of oxides $In_2O_3 < CaO < Y_2O_3$.

FORMATION OF THE STRUCTURE OF CERIUM OXIDE-MODIFIED TITANIUM DIOXIDE

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Kinet. Catal., 48(5) (2007) pp. 742-748.

The formation of the structure of titanium dioxide containing 3–15 wt % CeO_2 in a wide temperature range (300–850°C) has been investigated by X-ray powder diffraction, electron microscopy, and adsorption methods. Modification of titanium dioxide with cerium oxide causes the formation of nanostructured Ce–Ti–O compounds consisting of incoherently intergrown fine anatase crystallites. The crystallites are separated by interblock boundaries in which cerium ions are stabilized. The nanostructure formed in the Ce–TiO_2 oxide system stabilizes the anatase phase, prevents the sintering of anatase particles at high temperatures, and allows modified anatase to retain a larger specific surface area and a higher porosity upon heat treatment than pure titanium dioxide does.

THE STRUCTURAL GENESIS OF A COMPLEX $(MoVW)_5O_{14}$ OXIDE DURING THERMAL TREATMENTS AND ITS REDOX BEHAVIOR AT ELEVATED TEMPERATURES

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Mater. Chem. Phys., 103(2-3) (2007) pp. 295-304.

The structural genesis of a $Mo_{0.68}V_{0.23}W_{0.09}$ oxide with Mo_5O_{14}-like structure has been examined. A precursor prepared by spray-drying of mixed aqueous metal salt solutions was calcined in air and subsequently treated in helium at different temperatures. X-ray diffraction, HRTEM, ^{51}V magic angle spinning (MAS) NMR, ESR, UV/vis DR spectroscopy and oxygen and hydrogen adsorption measurements have been applied to monitor the preparation procedure. It was found that a structure closely related to that of Mo_5O_{14} already appears at nano-scale level after calcinations of the spray-dried precursor in air at 350^0C. At this stage, the material comprises of crystalline particles less than 3 nm in size stabilized by an amorphous matrix. Further heating causes nanostructural rearrangements that lead to the formation of the final $Mo_{0.68}V_{0.23}W_{0.09}$ oxide with phase-pure polycrystalline structure. Molybdenum and tungsten ions are hexavalent and coordinated in an octahedral environment. Furthermore, vanadium is present as V^{4+} and V^{5+} ions occupying octahedral and highly distorted trigonal pyramidal sites, respectively. According to the results of H_2 and O_2 adsorption the crystalline ternary oxide does not possess accessible micropores. Oxygen pulses at 450^0C and reductive treatment with pure hydrogen at 300^0C did not cause noticeable changes of the bulk structure thus indicating a remarkable structural stability of the complex MoVW oxide under redox conditions at elevated temperature.

SYNTHESIS OF ALUMINUM OXIDES FROM THE PRODUCTS OF THE RAPID THERMAL DECOMPOSITION OF HYDRARGILLITE IN A CENTRIFUGAL FLASH REACTOR.
I. PHYSICOCHEMICAL PROPERTIES OF THE PRODUCTS OBTAINED BY THE CENTRIFUGAL THERMAL ACTIVATION OF HYDRARGILLITE

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Kinet. Catal., 48(1) (2007) pp. 153-161.

A variety of physicochemical methods were used to characterize the product of the rapid thermal decomposition of hydrargillite in a centrifugal flash reactor under the following conditions: the average particle size of the reactant, 80–120 μm; the temperature of the solid heating surface (plate or cylinder), 300-700°C; hot-zone residence time, ~1 s; transfer of the product to the cooled zone of the reactor. The composition of the product and the extent of decomposition of hydrargillite were determined as a function of the processing temperature. The centrifugal thermal activation (CTA) of hydrargillite affords an X-ray-amorphous, highly reactive product with a developed surface and a disordered and inhomogeneous porous structure. This structure is capable of forming different modifications of aluminum hydroxide and oxide. The properties of the CTA product are compared with the properties of the earlier reported hydrargillite rapid decomposition products obtained using a gaseous heat-transfer agent (thermochemical activation product) or a fluidized bed of a granular heat transfer agent (thermal dispersion product).

SYNTHESIS OF ALUMINUM OXIDES FROM THE PRODUCTS OF THE RAPID THERMAL DECOMPOSITION OF HYDRARGILLITE IN A CENTRIFUGAL FLASH REACTOR.
II. STRUCTURAL AND TEXTURAL PROPERTIES OF ALUMINUM HYDROXIDE AND OXIDE OBTAINED FROM THE PRODUCT OF THE CENTRIFUGAL THERMAL ACTIVATION OF HYDRARGILLITE (CTA PRODUCT)

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Kinet. Catal., 48(2) (2007) pp. 316-326.

The basic properties of aluminum hydroxide compounds, namely, the flash product and the centrifugal thermal activation (CTA) product both before and after heat treatment at 400–1100°C are considered. The hydration conditions for obtaining 100% pseudoboehmite from the CTA product are determined. The structure, morphology, and texture of pseudoboehmite aluminum hydroxide synthesized from the CTA product and of the aluminum oxides obtained from this hydroxide in the temperature range 600–1100°C are described.

SYNTHESIS OF ALUMINUM OXIDES FROM THE PRODUCTS OF THE RAPID THERMAL DECOMPOSITION OF HYDRARGILLITE IN A CENTRIFUGAL FLASH REACTOR.

III. PROPERTIES OF ALUMINUM HYDROXIDES AND OXIDES OBTAINED VIA THE MILD REHYDRATION OF THE PRODUCTS OF THE CENTRIFUGAL THERMAL ACTIVATION OF HYDRARGILLITE

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Kinet. Catal., 48(2) (2007) pp. 327-335.

The interaction between the amorphous product of the centrifugal thermal activation of hydrargillite (CTA HG) and aqueous electrolytes (pH 5–11) under mild conditions (15–35°C, atmospheric pressure) has been investigated by a variety of physicochemical methods. This interaction causes material morphologic and phase changes in CTA HG, and the product composition is governed by the pH of the electrolyte and by the hydration temperature and time. The product that forms in a basic medium or water in < 24 h contains up to 50% pseudoboehmite. Raising the pH or temperature or extending the hydration time results in the formation of bayerite as the major phase (~80%). An X-ray amorphous hydroxide forms in acid media. The heat treatment of this hydroxide at 550°C yields aluminum oxides differing from alumina prepared via hydroxide reprecipitation. Products with new, unusual properties can thus be obtained.

Ce-SILICA MESOPOROUS SBA-15-TYPE MATERIALS FOR OXIDATIVE CATALYSIS: SYNTHESIS, CHARACTERIZATION, AND CATALYTIC APPLICATION

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Appl. Catal., A, 317(1) (2007) pp. 1-10.

Cerium-containing mesoporous materials have been synthesized by hydrothermal method and characterized by IR, DR-UV–vis and DRIFT spectroscopy, XRD and N_2 adsorption methods. It was established that the d_{100} and unit-cell (a_0) parameter increase with the increase of cerium content up to 2% in SBA-15 and then tend to remain the same. According to DR-UV–vis spectroscopic data, an agglomeration of cerium atoms was observed in the form of fine CeO_2 crystallites. Important factors affecting the catalytic activity of Ce-SBA-15, namely the effect of cerium content, the state of cerium ions, the state of silanol groups on the surface of Ce-SBA-15, and stability of the catalyst have been studied in the cyclohexanol and cyclohexene oxidation with hydrogen peroxide.

EFFECT OF OXALIC ACID CONTENT AND MEDIUM OF THERMAL TREATMENT ON PHYSICOCHEMICAL AND CATALYTIC PROPERTIES OF MoVTeNb OXIDE CATALYSTS IN PROPANE AMMOXIDATION

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Appl. Catal., A, 328(2) (2007) pp. 195-200.

Catalytic characteristics of $MoV_{0.3}Te_{0.23}Nb_{0.12}O_x$ catalysts in propane ammoxidation depend strongly on the quantity of oxalic acid present during slurry preparation and on the redox conditions during thermal treatment of the catalyst. Two stage thermal treatment (320°C in air, 600°C in He) and oxalate anion to niobium ratio in the slurry $C_2O_4^{2-}/Nb = 3–5$ have been shown to result in the catalysts with the best catalytic properties. Optimal catalysts have been found to contain M1 and M2 crystalline phases in the ratio of 3:1 or more. When the molar ratio $C_2O_4^{2-}/Nb$ exceeds 3.5, tellurium is lost during calcination in He and other phases beside M1 and M2 are formed.

REINFORCED NICKEL AND NICKEL-PLATINUM CATALYSTS FOR PERFORMING THE THERMALLY COUPLED REACTIONS OF METHANE STEAM REFORMING AND HYDROGEN OXIDATION

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Kinet. Catal., 48(1) (2007) pp. 116-124.

The formation of composite nickel and nickel-platinum catalysts reinforced with steel gauze was studied. The catalysts were prepared by sintering powdered nickel metal and a supported nickel catalyst (GIAP-3 or NIAP-18) with a chromium oxide additive in the case of nickel-containing composite catalysts or by sintering powdered nickel, aluminum, and a supported platinum catalyst in the case of catalysts containing nickel-platinum. With the use of electron microscopy, mercury porosimetry, and X-ray electron probe microanalysis, it was found that a metal matrix, in the pores of which supported catalyst particles were distributed, was formed in the composite catalysts. The reinforced nickel catalysts prepared were active in the reaction of methane steam reforming, and the catalysts containing nickel-platinum were active in the reaction of hydrogen oxidation. An increase in the activity of reinforced nickel catalysts in the course of the reaction was found. It is believed that the increase of the activity was due to the reduction of nickel oxide from an inactive difficultto-reduce oxide film containing nickel and chromium oxides under the action of the reaction atmosphere.

DEPENDENCE OF THE PROPERTIES OF Ce-Zr-Y-La-M-O SYSTEMS ON SYNTHETIC CONDITIONS AND ON THE NATURE OF THE TRANSITION METAL M (Mn, Fe, Co)

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Kinet. Catal., 48(1) (2007) pp. 143-152.

The effects of synthetic conditions, component ratios, and the nature of the transition metal on the physicochemical and catalytic properties of Ce–Zr–Y–La–M–O (M = Mn, Fe, Co) systems are studied. The Ce–Zr–Y–La–M–O samples precipitated at ~23°C and calcined at 600°C are single-phase and are solid solutions with a fluorite structure, which persists upon calcination at 1150°C. The Ce–Zr–Y–La–Fe(Co)–O samples precipitated at 70°C and calcined at 1150°C consist of two solid solutions, one cubic, and the other tetragonal. The specific surface area (S_{sp}) of the samples precipitated at ~23°C and calcined at 600°C increases in the order Ce–Zr–Y–La–O < Ce–Zr–Y–La–Mn–O < Ce–Zr–Y–La–Co–O ≈ Ce–Zr–Y–La–Fe-O. The specific surface area of the samples precipitated at 70°C is independent of M and is ~110 m^2/g. Calcination at 1150°C reduces S_{sp} approximately by two orders of magnitude. The TPR of the unpromoted systems in H_2 proceeds in two steps at 600–650 and 750–840°C. The introduction of M decreases the reduction temperatures and gives rise to a lower temperature peak between 150 and 300°C. The most effective promoter is cobalt. The fluorite-type catalysts containing no noble metal are active in NO reduction ($X_{NO} \approx 100\%$) at $T_{react} = 400–450°C$. The cobalt-containing catalysts are the most active in the oxidation of CO ($X_{max} = 28\%$) and hydrocarbons ($X_{max} = 4.3\%$).

EFFECT OF THE CALCINATION TEMPERATURE ON THE PROPERTIES OF Fe_2O_3/SiO_2 CATALYSTS FOR OXIDATION OF HYDROGEN SULFIDE

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React. Kinet. Catal. Lett., 92(1) (2007) pp. 89-97.

The effect of the calcination temperature on the properties of supported iron oxide catalysts for hydrogen sulfide oxidation prepared by impregnation of silica with iron(III) nitrate has been studied. An increase in the calcination temperature was found to diminish the catalytic activity of the Fe_2O_3/SiO_2 catalysts in hydrogen sulfide oxidation. This behavior can be explained by the agglomeration of iron oxide particles and by a decrease in the surface concentration of active sites. It has been shown that an increase in the calcination temperature makes the catalyst more stable towards the sulfidation of the active component (Fe_2O_3) to the iron disulfide phase.

SELF-PROPAGATING SYNTHESIS OF $Pd-CeO_2/Al_2O_3$ AUTOMOTIVE MONOLITH CATALYSTS

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Kinet. Catal., 48(1) (2007) pp. 162-167.

$Pd–CeO_2$ catalysts on a monolith support with a honeycomb structure have been for the first time prepared by surface self-propagating thermal synthesis (SSTS). Decomposition routes for the cerium precursors are deduced from TG–DTA data and from the mass spectra of decomposition intermediates. The Pd-CeO_2/Al_2O_3 monoliths prepared by SSTS are more active in CO oxidation, total hydrocarbon oxidation, and nitrogen oxide reduction than the catalysts obtained by conventional impregnation. This is explained by the fact that the SSTS products have a larger specific surface area and their active component has a smaller particle size.

REGULARITIES IN FORMATION OF HONEYCOMB STRUCTURAL BLOCK TEXTURE ON BASIS OF DIFFERENT OXIDE MATERIALS

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Catal. Ind., 1 (2007) pp. 35-46.

Taken as an example of honeycomb structure carriers on the basis of aluminum oxide, the effects of the nature of oxide component, blinder, modifying agents and thermal treatment temperature (as high as 1300°C) on phase composition, specific surface, porous texture, and mechanical strength of ceramic materials are studied. The main regularities of texture formation were found using mercury porosimetry, XRD, SEM, and others. It has been shown that the textural changes of monolith samples are shown to be mostly caused by removal of capillary water (20-100°C), disappearance of small pores and consolidation of oxide particles (100-700°C), and phase transformation of oxide component or blinder (900-1300°C). Knowledge of the mechanism and the features of aluminum, titanium, zirconium and alumosilicate based honeycomb material texture formation allow giving the practical recommendations to select binding components and modifying additives as well as the carrier drying and thermal treating conditions thereby providing a necessary complex of operational properties: porous structure, specific surface, and mechanical strength. Using the dependences obtained, it is possible to control the textural properties of the honeycomb carriers on the basis of aluminum, titanium, zirconium oxides in a wide range of values, thus expanding the field of their application.

DEVELOPMENT OF MONOLITHIC CATALYSTS WITH LOW NOBLE METAL CONTENT FOR DIESEL VEHICLE EMISSION CONTROL

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Top. Catal.,
42-43(1-4) (2007) pp. 465-469.

The chemical composition of the catalyst containing Mn–Al–O and Pt has been developed on the basis of the synergetic effect of Pt and manganese oxides observed in the HC and CO oxidation reactions. This effect allows decreasing the Pt loading to 0.70 g/L in the catalytic systems of diesel engine exhaust gases and provides high activity in low temperature oxidation of light and heavy hydrocarbons, and high thermal stability. It has been found that the catalytic activity of Pt–Mn–Al monolithic catalysts in butane oxidation and DIESEL tests depends on the Pt precursor and Pt loading. At similar Pt loadings (1.06 g/L), the catalytic activity increases in the order H_2PtCl_6~$H_2[Pt(OH)_6]$ < $Pt(NO_2)_2(NH_3)_2$. When one Pt precursor is used, the catalytic performance improves with Pt loading increase from 0 to 1.06 g/L, and being nearly constant at higher Pt loading (2.65 g/L).

CATALYTIC PROPERTIES OF BIMETAL CATALYSTS BASED ON DOUBLE TRANSITION METAL COMPLEXES FOR THE FISCHER-TROPSCH SYNTHESIS

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Chem. Sustain. Devel., 15(6) (2007) pp. 683-693.

Potentialities of preparation of bimetal Fe–Ni, Fe–Co and Co–Cu catalysts by thermolysis of double complex salts (salts comprising a complex cation of one metal and a complex anion of another metal) and their catalytic properties for FT synthesis were studied. Supporting the double complex salts $[Ni(NH_3)_6]_3[Fe(CN)_6]_2$, $[Co(NH_3)_6][Fe(CN)_6]$ and $[Co(NH_3)_6]_2C_2O_4[Cu(C_2O_4)_2]_2$ on aluminium hydroxide followed by thermolysis of the resulting composition was shown to allow 15–40 nm metal particles to be obtained as anchored on alumina surface. In the case of double Ni–Fe and Co–Fe complexes, the resulting bimetal catalysts are 15–16 nm in size with fcc and bcc structures, respectively. Destruction of the complexes follows several sequential exothermic stages. Bimetal bcc Fe-Co particles produced by thermolysis of $[Co(NH_3)_6][Fe(CN)_6]$ + $Al(OH)_3$ composition demonstrate catalytic properties which are quite different from literature data on Co-Fe catalysts: They are less active to the secondary processes of olefin hydrogenation but very selective to olefins. An increase in the process pressure up to 10–20 atm results in a considerable changes in properties of the bimetal Co-Fe catalysts including a decrease in the selectivity to olefins and in the rate of the secondary process of steam CO conversion. Such a dependence is accounted for by changes in the structure of the catalytically active component under the action of the reaction medium.

INFLUENCE OF THE CONDITIONS OF MANUFACTURE OF NANOMERIC ZIRCONIUM DIOXIDE, STABILIZED WITH YTTRIUM OXIDE, ON ITS CATALYTIC PROPERTIES IN THE OXIDATION OF CO

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Theor. Exp. Chem., 43(2) (2007) pp. 102-107.

The morphological and catalytic properties of samples of nanomeric zirconium dioxide, stabilized with yttrium oxide, manufactured via the effect of a UHF field during the process of drying precipitated zirconium hydroxide and calcination at temperatures from 300 to 1000°C, were studied. It was shown that the highest activity in the oxidation of CO occurred with 40 nm particles of zirconium dioxide prepared at 1000°C.

PROPERTIES OF FERROUS-OXIDE CATALYST FOR AMMONIA OXIDATION DEPENDING ON RAW MATERIALS USED

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Catal. Ind., 2 (2007) pp. 46-53.

Effect of source raw material (which is prepared using sulphate or chloride technologies from commercial hematite) and Al_2O_3 – binder, prepared by re-precipitation or hydration of thermoactivated hydrargillite, on physical chemical properties (activity in ammonia oxidation reaction, durability and porous structure) of ferrous oxide catalyst for ammonia oxidation, IC-42-1, used in two-step process of ammonia oxidation in UKL-7 devices has been studied. The use of binder prepared by the hydration of thermoactivated hydrargillite has been shown to obtain the catalyst with a higher NO (II) yield with other conditions being the same. The use of «sulphate» α-Fe_2O_3 and calcination at 950°C provides better operation parameters of IC-42-1 catalyst. The results obtained are taken into account during preparation of industrial batches (up to 3 m^3/year) of IC-42-1 catalyst, which is delivered for use in UKL-7 devices.

FORMATION OF SUPPORTED SIZE-CONTROLLED NANOPARTICLES OF SULFATED ZIRCONIA

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React. Kinet. Catal. Lett.,
91(1) (2007) pp. 177-185.

Methods of the preparation of catalysts for alkane skeletal isomerization based on uniform nanoparticles of sulfated zirconia anchored to different supports were investigated. These catalysts were characterized by using the ICP, HRTEM and BET techniques. The activities of the catalysts in the reaction of n-butane isomerization were measured and compared with those of bulk catalysts.

STUDY OF PALLADIUM CATALYST DEACTIVATION IN SYNTHESIS OF 4,10-DIFORMYL-2,6,8,12-TETRAACETYL-2,4,6,8,10,12-HEXAAZAISOWURTZITANE

A.P. Koskin, I.L. Simakova, V.N. Parmon

React. Kinet. Catal. Lett.,
92(2) (2007) pp. 293-302.

Polycyclic nitramine hexanitrohexaazaisowurtzitane (HNIW, CL-20) is synthesized via hydrodebenzylation of a precursor over palladium-based catalyst. In the present work, it was studied the influence of the synthesis method of the palladium-based catalyst on the catalyst behavior during the hydrodebenzylation reaction of 2,4,6,8,10,12-hexabenzyl-2,4,6,8,10,12-hexaazatetracyclo[5.5.0.$0^{3,11}$.$0^{5,9}$] dodecane.

SPECIFIC FEATURES OF THE INTERACTION OF ALUMINIUM POWDER WITH WATER IN THE PRESENCE OF SURFACTANTS

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Herald of Belarusian Ac. Sci.,
2 (2007) pp. 27-33.

Kinetic and high-resolution solid state NMR (^{27}Al, ^{1}H) studies of the interaction of aluminium powder (ASD-1) with water demonstrated that the influence of surfactants on the alumina conversion depends on the specific reactivity of alumina and on the surfactant nature. Two reaction regions were identified: kinetic (oxidation rate is higher in solutions than in water) and diffusion (aluminium conversion is higher in water than in solutions). Surfactants were discovered to favor amorphization of the products of hydrothermal oxidation of aluminium, a decrease in the surface area and pore expanding in alumina. The method of statistical sorption of tetrachloromethane vapor revealed the transformation of hysteresis loop in the adsorption-desorption isotherms under the action of various surfactants that indicates reshaping of alumina mesopores.

ACTIVATED ALUMINUM: FEATURES OF PRODUCTION AND APPLICATION IN THE SYNTHESIS OF CATALYSTS FOR PERTOCHEMISTRY AND OIL PROCESSING

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Russ. J. General Chem.,
77(12) (2007) pp. 2320-2327.

The best known procedures for aluminum activation are discussed and systematized. The analysis of published data and of results of the proper physicochemical research reveals that the phenomena of surface distribution, intergrain penetration and the internal diffusion in the crystal grains underlie the nature of aluminum activation with liquid metal alloys based on gallium. The experimental investigation using X-ray fluorescent spectroscopy of diffusion processes in the systems Al-Ga-In were performed for the first time. The procedures for formation of catalytically active complexes of $AlCl_3$ type by treating the activated aluminum *in situ* in reaction media with chlorinated hydrocarbons were considered.

DESIGN OF SCHIFF BASE-LIKE POSTMETALLOCENE CATALYTIC SYSTEMS FOR POLYMERIZATION OF OLEFINS

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Proceed. Tula State University, ser. Chem.,
6 (2006) pp. 235-241.

The effect of rehydration of nanodisperse alumina aerogel (amorphous alumina) in water vapor medium results in the formation of pseudoboehmite. The following dehydration of pseudoboehmite results in the formation of disperse γ-Al_2O_3 which is stable to water vapor at temperatures below 100°C.

PREPARATION OF PURE MONODISPERSE CALCIUM CARBONATE PARTICLES

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A.G. Okunev

React. Kinet. Catal. Lett.,
90(1) (2007) pp. 151-157.

The non-template synthesis of monodisperse spherical particles of calcium carbonate is reported. Particles of a 3.5–4.5 μm size were produced by precipitation of calcium carbonate from alcohol solution at subzero temperatures.

SUPERCRITICAL CO_2 ASSISTED SYNTHESIS OF HIGHLY SELECTIVE NAFION–ZEOLITE NANOCOMPOSITE MEMBRANES FOR DIRECT METHANOL FUEL CELLS

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*J. Membr. Sc*i.,
297(1-2) (2007) pp. 1-4.

The synthesis of nafion–zeolite (Fe-silicalite-1) nanocomposite membranes is reported using either a supercritical CO_2 pre-treatment followed by impregnation with zeolite nanoparticles or *in situ* synthesis of Fe-silicalite-1 inside the pores of nafion membrane. The effects of the supercritical CO_2 activation and the size of embedded zeolite particles on the proton conductivity and methanol permeability of composite membranes are observed. In selected cases, the membranes exhibit extremely low methanol permeability and high selectivity (defined as proton conductivity/methanol permeability ratio).

FULL PHENOL PEROXIDE OXIDATION OVER Fe-MMM-2 CATALYSTS WITH ENHANCED HYDROTHERMAL STABILITY

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V.B. Fenelonov

Appl. Catal. B.,
75(3-4) (2007) pp. 290-297.

Iron-containing mesoporous mesophase materials Fe-MMM-2 have been synthesized by a sol-mesophase route under mild acidic conditions and characterized by DRS-UV-vis, XRD, and N_2 adsorption measurements. It was found that pH of the synthesis solution and iron content in the samples affect both the textural characteristics and the state of iron atoms. Isolated iron species predominate in silica framework under Fe < 2 wt. % and pH ~ 1.0 or Fe ~ 1 wt. % and pH < 2.0. These species are stable to leaching and highly active in full H_2O_2-based phenol oxidation. The increase in iron loading and pH of the synthesis solution lead to the agglomeration and formation of oligomeric iron species, which, in turn, results in the reduction of the catalytic activity of Fe-MMM-2 and the increase of iron leaching.

PREPARATION AND CHARACTERISATION OF PALLADIUM-LOADED POLYPROPYLENE POROUS HOLLOW FIBRE MEMBRANES FOR HYDROGENATION OF DISSOLVED OXYGEN IN WATER

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J. Membr. Sci., 229(1-2) (2007) pp. 38-44.

Dissolved oxygen is one of the major components to be removed for the production of ultra-pure water. For the removal of oxygen to very low levels (less than 1 ppb for rinse water for the semiconductor industry), the catalytic reduction with hydrogen is an attractive method. The objective of this study was to integrate the palladium catalyst with the shell side of hydrophobic porous polypropylene membrane hollow fibre membranes to obtain a compact and efficient technique that preferably can be based on commercially available materials and modules. It was demonstrated that palladium could be deposited on a hydrophobic porous membrane hollow fibre, while maintaining its hydrophobic nature. Palladium loadings as low as 0.36% (w/w) were sufficient to catalyse the hydrogenation of dissolved oxygen while maintaining diffusion limited kinetics. Hence, a fast oxygen removal system is obtained that has the potential of maintaining removal rate, even at very low oxygen concentrations.

REACTIONS OF ELEMENTAL PHOSHORUS AND PHOSPHINE WITH ELECTROPHILES IN SUPERBASIC SYSTEMS. XIX. FORMATION OF THE C-P BOND WITH PARTICIPATION OF ELEMENTAL PHOSPHORUS UNDER MICROWAVE ASSISTANCE

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Russ. J. General Chem.,
77(3) (2007) pp. 415-420.

Microwave irradiation facilitates phosphorylation of aryl methyl chlorides and styrene with red phosphorus in the presence of strong bases and increases the yield of the main products, tertiary phosphine oxides.

CHEMICAL AND OPTICAL PROPERTIES OF THE TITANIUM DIOXIDE PRODUCED FROM COMBUSTION OF TITANIUM MICROPARTICLES IN AIR

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Atmos. Oceanic Opt.,
20(6) (2007) pp. 486-491.

It is shown that the solid titanium dioxide aerosol, obtained from combustion in air of a pyrotechnic mixture containing titanium microparticles, has optic, chemical, and photocatalytic properties similar to those of titanium dioxide produced in other ways. An aerosol cloud produced from such titanium dioxide immediately in the place of abrupt emission of pollution favors purification of the gas-phase atmosphere nearby the pollution source

SYNTHESIS AND CHARACTERIZATION OF MESOPOROUS SILICA THIN FILMS AS A CATALYST SUPPORT ON A TITANIUM SUBSTRATE

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Thin Solid Films,
515(16) (2007) pp. 6391-6394.

Mesoporous silica films with a thickness of 500–900 nm were synthesized on a titanium substrate by the evaporation-induced self-assembly method (with 900–1200 rpm for 90 s) using cetyltrimethylammonium bromide (CTAB) as structure-directing agent and tetraethyl orthosilicate as the silica source. Prior to coating deposition, the titanium substrate was oxidized to increase the surface roughness up to 500 nm and to produce a thin titania layer. Just before the synthesis, the titania layer was made super hydrophilic by an UV treatment for 2 h to provide a better adhesion of the silica film to the substrate. Films with hexagonal and cubic mesostructures with a uniform pore size of 2.8 nm and a surface area of 1080 m^2/g were obtained and characterized by different methods. An alternative approach for surfactant removal by gradual heating up to 250°C in vacuum was applied. Complete removal of CTAB from the as-synthesized silica films was confirmed by infrared spectroscopy.

Carbon and Carbon Related Processes and Materials

INFLUENCE OF CATALYSTS’ ACTIVATION ON THEIR ACTIVITY AND SELECTIVITY IN CARBON NANOTUBES SYNTHESIS

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Phys. Status Solidi B,
244(11) (2007) pp. 3920-3924.

In this paper the influence of catalysts' activation conditions on their selectivity in carbon nanotubes CVD synthesis by means of methane decomposition reaction using Fe- and FeCo-catalysts synthesized via polymerized complex rout providing homogeneous distribution of catalytic components have been investigated. It was shown that variation of catalysts' reduction conditions results in formation of different types of NTs (MWNTs or SWNTs). The most effective catalyst activation leading to formation of SWNTs consists in catalyst reduction by reaction mixture at high temperature. That can be explained in terms of carbon deposits nucleation on metals.

EVIDENCE OF AN EQUIMOLAR C_2H_2-CO_2 REACTION IN THE SYNTHESIS OF CARBON NANOTUBES

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Angewandte Chemie – Intern. Edition,
46(3) (2007) pp. 441-444.

Chemical vapor deposition (CVD) is considered to be the most viable process for the in situ production of nanotubes integrated into a device. Researchers have successfully attempted to control accurately the physical form of the carbon nanotubes produced. However, the method still suffers from low yields with respect to the carbon source and from high temperatures required for this conversion. A huge effort has been devoted to enhance the production efficiency at lower temperatures by modifying the catalyst (pregrowth chemical activation or by avoiding catalyst poisoning (e.g., by introducing an etching agent that prevents encapsulation by the precipitated amorphous carbon).

INFLUENCE OF HELIUM, HYDROGEN, OXYGEN, AIR AND METHANE ON CONDUCTIVITY OF MULTIWALLED CARBON NANOTUBES

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Sens. Actuators, A,
138(2) (2007) pp. 350-354.

Temperature dependences of the conductivity of catalytic multiwalled carbon nanotubes (MWNTs) were measured in various gas environments: helium, hydrogen, oxygen, air and methane. It has been found that in the vicinity of the melting and vaporization temperatures of hydrogen, oxygen, air and methane the conductivity sharply decreases by (2–12%). The observed reversible change of the conductivity is connected with the adsorption–desorption of gases on the surface of nanotubes and breakup-recovery of contacts between nanotubes as a result of melting and vaporization of hydrogen, oxygen, air and methane.

SEPARATING WEAK-LOCALIZATION AND ELECTRON-ELECTRON-INTERACTION CONTRIBUTIONS TO THE CONDUCTIVITY OF CARBON NANOSTRUCTURES

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O.B. Anikeeva*,, T.I. Buryakov*,**,**
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Bull. NSU, ser. Physics, 2(3) (2007) pp. 76-80.

Effect of modification of curvilinear carbon nanostructures on the electrophysical properties was studied. A special synthesis technique was used to obtain multiwall catalytic carbon nanotubes (MWNTs) and expanded carbon that are practically free of amorphous carbon impurities. It makes possible to observe quadratic growth in the positive magnetoconductivity in the fields of up to B ~ 1 T, so called electron–electron interaction effects. They were unnoticeable in previously synthesized nanotubes.

SEPARATING WEAK-LOCALIZATION AND ELECTRON-ELECTRON-INTERACTION CONTRIBUTIONS TO THE CONDUCTIVITY OF CARBON NANOSTRUCTURES

E.N. Tkachev*,, A.I. Romanenko*,**,**
O.B. Anikeeva*,, T.I. Buryakov*,**,**
V.E. Fedorov*, A.S. Nazarov*,
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J. Exper. Theor. Phys.,
105(1) (2007) pp. 223-226.

The effect of the modification of curvilinear carbon nanostructures (nanotubes) on their electrical properties has been studied. The samples were prepared using a special method of synthesis, which excluded the formation of amorphous carbon particles in multiwalled carbon nanotubes and in expanded graphite. Such materials exhibit a quadratic growth in the positive magnetoconductivity in the fields of up to B ~ 1 T, which is not observed in the samples synthesized by usual methods.

ELECTRON–ELECTRON INTERACTION EFFECTS IN MULTIWALL CARBON NANOTUBES

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O.B. Anikeeva*,, V.L. Kuznetsov,**
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Low Temp. Phys.,
33(2) (2007) pp. 272-275.

The temperature dependence of the electrical conductivity σ and the field dependence of the magnetoresistivity ρ of samples of multiwall catalytic carbon nanotubes containing a small amount of amorphous carbon impurity are studied. Below a temperature of 20 K the observed temperature dependence of the conductivity of the nanotubes shows a contribution from two-dimensional quantum corrections to the conductivity. At a temperature of 4.2 K one observes negative magnetoresistance on the ρ (B) curves in the field interval 0–10 kG, due to the dominance of the contribution to the magnetoresistance from the quantum corrections for interacting electrons. A special synthesis technique is used to obtain multiwall catalytic carbon nanotubes that are practically free of amorphous carbon impurities, making it possible to observe electron–electron interaction effects unnoticeable in previously synthesized nanotubes of this kind.

EFFECT OF GASES ON THE TEMPERATURE DEPENDENCE OF THE ELECTRIC CONDUCTIVITY OF CVD MULTIWALLED CARBON NANOTUBES

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J. Exper. Theor. Phys.,
105(1) (2007) pp. 155-159.

The influence of various gaseous media on the temperature dependence of the electric conductivity σ of multiwalled carbon nanotubes (MWNTs) synthesized using the method of catalytical chemical vapor deposition (CVD) has been studied. The $\sigma(T)$ curves were measured in a temperature range from 4.2 to 300 K in helium and its mixtures with air, methane, oxygen, and hydrogen. The introduction of various gaseous components into a helium atmosphere leads to a significant decrease in the conductivity of MWNTs in the interval between the temperatures of condensation and melting of the corresponding gas.

Upon a heating-cooling cycle, the conductivity restores on the initial level. It is concluded that a decrease in σ is caused by the adsorption of gases on the surface of nanotubes.

SOFT X-RAY SPECTROSCOPY AND QUANTUM CHEMISTRY CHARACTERIZATION OF DEFECTS IN ONION-LIKE CARBON PRODUCED BY NANODIAMOND ANNEALING

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Diamond Relat. Mater.,
16(4-7) (2007) pp. 1222-1226.

The electronic structure of onion-like carbon (OLC) consisted of quasi-spherical and polyhedral nanoparticles and produced as a result of nanodiamond annealing at 1800 K and 2140 K has been probed by soft X-ray emission and X-ray absorption spectroscopy. The enhanced density of states in the vicinity of the Fermi level was revealed for OLC prepared at moderate temperature. *Ab initio* calculation on carbon models showed the only unpaired electrons at the zigzag edges of graphitic fragments can provide localized states observed in the X-ray spectra. With the increase of the annealing temperature the defects, which likely constitute the boundaries of incompletely closed onion shells, are healing.

ENCAPSULATION OF MOLECULAR NITROGEN IN MULTIWALL CN_X NANOTUBES

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Phys. Stat. Sol. B,
244(11) (2007) pp. 4078-4081.

Multiwall CN_x nanotubes have been grown in the result of acetonitrile decomposition over Fe catalyst (CVD method). The random nanotubes samples have been produced by classical technique using catalyst nanoparticles formed by iron bimaleate thermolysis. The aligned nanotubes have been synthesized by aerosol assisted CVD method where ferrocene was used as a catalyst source. X-ray photoelectron spectroscopy (XPS) showed the random sample contains two kinds of nitrogen, while additional high-energy peak was detected in the N 1s-spectrum of the aligned CN_x nanotubes. X-ray absorption spectra measured near the N K-edge (NEXAFS) of the samples exhibited three peaks for both type of CN_x nanotubes. The relative intensity of the high-energy peak was found to be much higher in the spectrum of aligned nanotubes. Assignment of the NEXAFS peaks was made using results of quantum-chemical calculations on carbon tube model incorporating pyridinic, three-fold coordinated and molecule nitrogen. The high energy peak was shown to correspond to N_2 molecules and its absence in the XPS spectrum of random CN_x nanotubes indicates the gaseous nitrogen is mainly concentrated in the interior region of nanotubes. The larger encapsulation of molecules in the aligned CN_x nanotubes could be due to the higher diffusion of nitrogen atoms through the catalytic particles formed directly in the CVD process.

ATTENUATION OF ELECTROMAGNETIC WAVES IN ONION-LIKE CARBON COMPOSITES

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Diamond Relat. Mater.,
16(4-7) (2007) pp. 1231-1235.

The first results on measurements of electromagnetic wave attenuation of onion-like carbon (OLC) powders and OLC-based polymer films on a substrate are reported. The measurements cover a wide frequency range 2–38 GHz and demonstrate promising high potential of OLC-based composites as basic components for wideband electromagnetic wave absorbing materials. A description of the measurement technique is presented. Possibility and mechanisms of the absorption efficiency tuning are discussed.

SURFACE ELECTRONIC STRUCTURE OF DETONATION NANODIAMONDS AFTER OXIDATIVE TREATMENT

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Diamond Related Mater.,
16 (2007) pp. 2090-2092.

X-ray absorption spectroscopy has been used for comparative study of electronic structure of detonation nanodiamonds (ND) purified using different oxidative treatments. The treatment of detonation soot with a mixture of nitric and sulphuric acids followed by ion exchange and ultrafiltration of hydrosol obtained was found to result in developing of ND surface coverage consisting of oxidized carbon species, which electronic state is close to that of strongly oxidized graphite. The deeper purification of ND was demonstrated to allow cleaning of ND particles from most of the oxidized carbon contaminations.

NANODIAMOND AND ONION-LIKE CARBON POLYMER NANOCOMPOSITES

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Diamond Relat. Mater.,
16(4-7) (2007) pp. 1213-1217.

The current work demonstrates that nanodiamond (ND) of detonation origin and onion-like carbon (OLC) are valuable additives in multifunctional polymer composites, particularly for polymers used in microelectronic applications. It is demonstrated that addition of ND to a polyimide matrix increases the thermal degradation temperature of the composites up to 30°C and also improves adhesion. The addition of 2 wt.% of ND increases thermal conductivity of PDMS up to 15%. Finally, it was also demonstrated that the addition of OLC to polydimethylsiloxane and polyurethane matrices increases the loss tangent of the composites.

THERMODYNAMIC ANALYSIS OF NUCLEATION OF BORON NITRIDE NANOTUBES ON METAL PARTICLES

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Phys. Status Solidi B,
244(11) (2007) pp. 4165-4169.

Boron nitride nanotubes nucleation on the catalyst's surface after the formation of metal particle alloys oversaturated with boron and nitrogen atoms is the most important step for any type of the catalytic processes of BN nanotubes formation. A thermodynamic analysis of the boron-nitride nanotubes nucleation on the catalysts surface was performed. The master equations for the dependence of critical size of BN nucleus on reaction parameters, such as reaction temperature, supersaturation degree of catalyst particles with B and N, work of adhesion of catalyst to BN were obtained. These equations combined with the phase diagram approach can be used for the description of different scenarios of BN deposits formation and for the development of the main principles of catalysts design for BN nanotube production.

OXIDATIVE DEHYDROGENATION OF ETHYLBENZENE TO STYRENE OVER ULTRA-DISPERSED DIAMOND AND ONION-LIKE CARBON

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Carbon,
45(11) (2007) pp. 2145-2151.

The catalytic properties of sp^3-hybridized ultra-dispersed diamond and sp^2-hybridized onion-like carbon in the oxidative dehydrogenation of ethylbenzene to styrene were investigated, highlighting the structure sensitivity of the reaction. The sp^3-carbon led initially to C–C cleavage and benzene formation, while a switchover of the main reaction pathway into the styrene formation occurred with time on stream due to the formation of surface sp^2 carbon, required for the selective styrene formation. This was confirmed by the behavior and the high stable styrene selectivity shown by onion-like carbons. High temperature oxygen pre-treatment created catalytically active species at the sp^2 carbon surface, confirming that a high thermal stability carbon–oxygen complex was the active surface site for forming styrene.

STUDY OF THE ELECTRONIC STRUCTURE AND PROPERTIES OF ^{13}C-ISOTOPE-BASED COMPOSITES

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L.G. Bulusheva*, O.B. Anikeeva******,**
Yu.V. Lavskaya*, A.V. Okotrub***,**
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J. Surf. Invest. X-Ray, Synchrotron Neutron Tech., 1(6) (2007) pp. 645-651.

The morphology and electronic structure of ^{13}C-isotope-based graphite composites were studied by transmission electron microscopy (TEM), X-ray diffraction, and X-ray fluorescence spectroscopy. High-resolution TEM images showed that composites contain several forms of carbon particles. According to an x-ray diffraction analysis, the size of graphene stacks of graphite particles is 20 and 40 Å. The CK_α X-ray fluorescence spectra of the initial ^{13}C isotope powder and composites based on it detected an increase in the density of high-energy occupied states in comparison with the graphite spectrum. *Ab initio* quantum chemical calculation of the structure of C_{150} graphene showed that the increase in the density of states stems from the electrons of dangling bonds of boundary carbon atoms of particles ~20 Å in size. Electrical properties of ^{13}C-isotope-based samples were studied by analyzing the temperature dependence of the conductivity. It was assumed that the change in the logarithmic dependence of the conductivity observed at liquid-helium temperatures to the linear dependence as the temperature increases is caused by carrier transfers between the disordered graphene layers forming a nanocomposite.

DIRECT SYNTHESIS OF NITROGEN-CONTAINING FILAMENTOUS CARBON ON A HIGH-PERCENTAGE Ni-Cu CATALYST

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V.B. Fenelonov, V.I. Zaikovskii, A.V. Ishchenko

Kinet. Catal., 48(1) (2007) pp. 103-115.

Nitrogen-containing catalytic filamentous carbon (N-CFC) of chemical composition NC_{18}-NC_{104} has been synthesized by the decomposition of pyridine (Py) from gaseous mixtures with argon or H_2 at 550-800°C on Ni/Al_2O_3 (Ni) and $Ni\text{-}Cu/Al_2O_3$ (Ni–Cu) high-percentage catalysts. The activity of the Ni–Cu catalyst in Py decomposition in mixtures with H_2 is about one order of magnitude higher than its activity in Py/Ar mixtures (more than 70 g N-CFC per metal gram in 4.5 h at 750°C), which is interpreted as arising from the nickel-catalyzed hydrogenation of Py. The formation and growth of carbon fibers occurs through the decomposition of Py (from Ar/Py mixtures) and/or Py hydrodenitrification products (from H_2/Py mixtures). The carbon material has been characterized by elemental analysis, low-temperature nitrogen adsorption, high-resolution transmission electron microscopy (HRTEM), and X-ray photoelectron spectroscopy (XPS). The effect of the noncatalytic reactions of Py and its transformation products on the composition and texture of N-CFC is discussed.

SYNTHESIS OF NITROGEN-CONTAINING CARBON MATERIALS FOR SOLID POLYMER FUEL CELL CATHODES

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O.Yu. Podyacheva, Ch.N. Barnakov*,
A.P. Kozlov*, R.I. Kvon, I.Z. Ismagilov,
M.A. Kerzhentsev (**Institute of Carbon and Carbon Chemistry, Kemerovo, Russia*)

Kinet. Catal., 48(4) (2007) pp. 581-588.

The following nitrogen-containing supports with various nitrogen contents and structure and texture properties were synthesized: carbon nanofibers (N-CNFs) and amorphous microporous carbon materials (N-AMCMs). It was found that the above characteristics can be regulated by varying synthesis conditions: precursor compositions and reaction temperature and time. Mesoporous nitrogen-containing CNFs with a specific surface area of 30–350 m^2/g and a pore volume of 0.10–0.83 cm^3/g were formed by the catalytic decomposition of a mixture of ethylene with ammonia at 450–675°C. Microporous materials (N-AMCMs) with a specific surface area of 472–3436 m^2/g and a micropore

volume of 0.22–1.88 cm^3/g were prepared by the carbonization of nitrogen-containing organic compounds at 700–900°C. An increase in the carbonization temperature and reaction time resulted in an increase in the specific surface area and microporosity of N-AMCMs, whereas lower temperatures of 450–550°C and reaction times of 1–3 h were optimal for the preparation of N-CNFs with a developed texture. It was found that milder synthesis conditions and higher nitrogen contents of precursors were required for obtaining high nitrogen concentrations in both N-CNFs and N-AMCMs. The synthetic method developed allowed to prepare carbon supports with nitrogen contents up to 8 wt %.

SYNTHESIS OF NITROGEN-CONTAINING CARBON NANOFIBERS BY CATALYTIC DECOMPOSITION OF ETHYLENE/AMMONIA MIXTURE

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Carbon,
45(9) (2007) pp. 1808-1820.

The formation of carbon nanofibers (CNFs) doped with nitrogen was investigated during decomposition of C_2H_4/NH_3 mixtures at 450–675°C over metal catalysts: $90Ni–Al_2O_3$, $82Ni–8Cu–Al_2O_3$, $65Ni–25Cu–Al_2O_3$, $45Ni–45Cu–Al_2O_3$, $90Fe–Al_2O_3$, $85Fe–5Co–Al_2O_3$, $62Fe–8Co–Al_2O_3$, $62Fe–8Ni–Al_2O_3$. It was found that the yield of CNFs, their structural and textural properties, as well as nitrogen content in CNFs are strongly dependent on the synthesis conditions such as: catalyst used, feed composition, temperature and duration. The $65Ni–25Cu–Al_2O_3$ was proved to be the most efficient catalyst for the production of nitrogen-containing carbon nanofibers (N-CNFs) with nitrogen content up to 7 wt.%. Ammonia concentration in the feed equal 75 vol.%, temperature 550°C and duration 1 h were found to be the optimum reaction parameters to reach the maximum nitrogen content in N-CNFs. TEM studies revealed that the nanofibers have a helical morphology and a “herringbone” structure composed of graphite sheets. According to the XPS data, the nitrogen incorporation in the N-CNF structure leads to the formation of two types of nitrogen coordination: pyridinic and quaternary, and their abundance depends on the reaction conditions.

NOVEL CARBON NANOFIBERS WITH Ni-Mo AND Co-Mo NANOPARTICLES FOR HYDRODESULFURIZATION OF FUELS

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Intern. Sci. J. Alternative Energy Ecology,
3(47) (2007) pp. 150-158.

Methane decomposition over $Ni–Mo–Al_2O_3$ (10 wt. % Al_2O_3) and $Co–Mo–Al_2O_3$ (25 wt. % Al_2O_3) catalysts was studied at 550 and 500°C. TEM, EDX and XRD investigations revealed localization of parent metal catalyst nanoparticles on the tips of the resulting carbon nanofibers (CNFs). The composition of these particles and the state of Ni, Co and Mo were examined by EXAFS spectroscopy. The catalytic activity of the isolated Ni–Mo and Co–Mo nanoparticles located on the tips of separate CNFs was tested in thiophene hydrodesulfurization (HDS) at 300°C. The HDS activities of the CNF catalysts were found to be comparable with those of highly dispersed bimetallic sulfide catalysts supported on Sibunit carbon.

PREPARATION AND INVESTIGATION OF NANOSTRUCTURED CARBONACEOUS COMPOSITES FROM THE HIGH-ASH BIOMASS

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Chem. Eng. J.,
134(1-3) (2007) pp. 246-255.

The various pathways of high-ash biomass processing to the valuable carbonaceous composites are considered. From the rice husk carbon–silica composites and supermicroporous carbon materials (SMC) were prepared. In the new SMCs, the specific surface area A_{BET} reaches 3460 m^2/g while micropore volume V_μ is up to 1.9 cm^3/g at the total of pore volume, V_Σ, as large as 3.0 cm^3/g. These carbons were shown to absorb up to 41 wt% of methane at room temperature and 60 atm and more than 6 wt% of hydrogen under mild cryogenic conditions (at 77 K). Also methods for the synthesis of carbon–silica composites with A_{BET} up to 710 m^2/g and with average size of SiO_2 particles less 5 nm are proposed. The expected practical application of the obtained carbon–silica composites may be first of all reinforcing rubber extenders and bi-functional sorbents for the gas and liquid purification.

CARBON ADSORBENTS AS HYDROGEN FUEL STORAGE MEDIA FOR VEHICULAR APPLICATIONS

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Kinet. Catal., 48(4) (2007) pp. 599-602.

The prospects of carbon adsorbents as vehicular hydrogen storage media are briefly discussed.

NANOSCALE CARBON FORMATION FROM VARIOUS HYDROCARBONS OVER NANOCRYSTALLINE Co/MgO CATALYST

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J. Phys. Chem. C,
111(22) (2007) pp. 7868-7874.

A series of nanocrystalline MgO samples containing 1.2-15.9% carbon were prepared by MgO carbonization at 500°C in 1,3-butadiene flow diluted with argon in a 1:75 ratio. For the sample with 15.9% C almost all the MgO surface was covered with carbon. An increase of the MgO carbonization rate from 500 to 750°C resulted in the growth of the carbon deposition rate. Still, the carbon deposits covered the MgO surface in a fairly uniform way. This is proved by the very high surface area (2000-2500 m^2/g) of porous carbon obtained by dissolving MgO from the carbonized samples with hydrochloric acid. Nanocrystalline MgO was used as a support for synthesis of Co/MgO catalysts. The concentration of supported cobalt was varied from 2.5 to 30%. The nature and formation kinetics of carbon deposits from butadiene over Co/MgO catalysts were studied. High-resolution transmission electron microscopic (HRTEM) studies showed that carbonization of the Co/MgO catalyst in butadiene diluted with argon in the temperature range of 600-750°C resulted in the formation of carbon nanotubes and a carbon film decorating the MgO surface. Unlike butadiene, methane was not found to carbonize the MgO surface at temperatures below 800°C. Carbonization of the Co/MgO catalysts with methane in the temperature range of 600-800°C resulted in the formation of carbon nanotubes. As reduction of the catalysts with hydrogen was found to yield highly dispersed cobalt nanoparticles with a narrow particle size distribution, the resulting carbon nanotubes are characterized by high uniformity. The number of layers in the nanotubes varied from one layer to six to eight layers depending on the reduction conditions and carbonization temperature. In the case of methane, a carbonization temperature increase from 600 to 800°C led to a decrease of the carbon nanotube diameter from 4-5 to 1-2 nm and the number of layers from one to three layers to one layer. The observed decrease of the nanotube diameter with the temperature increase was explained assuming the key role of the graphite phase nuclei in the formation of carbon nanotubes.

PREPARATION AND CHARACTERIZATION OF ALUMINOSILICATE SUPPORTS WITH A SYNTHESIZED LAYER OF CATALYTIC FILAMENTOUS CARBON. I. SYNTHESIS OF CARBON NANOFIBERS ON A SUPPORTED NICKEL CATALYST

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Kinet. Catal., 48(5) (2007) pp. 749-756.

The synthesis of catalytic filamentous carbon (CFC) on a Ni catalyst supported by homogeneous precipitation onto the surface of aluminosilicate ceramic supports (honeycomb monoliths, ceramic foam, glass foam, and haydite) was studied. The effects of CFC synthesis conditions (the catalyst concentration on a support, the pyrolysis temperature of a propane–butane mixture, and the composition of the gas mixture) on the specific surface areas of supports, the yield of carbon, and the morphology of a surface CFC layer were examined. As found by scanning electron microscopy, the uniformity of distribution and the size of carbon nanofibers synthesized depended on the conditions of their synthesis. The resulting CFC-containing supports were tested as adsorbents for the immobilization of enzymatically active substances (individual enzymes, cell membranes, and microorganisms) in order to prepare highly stable heterogeneous catalysts for biotechnology and biocatalysis.

PREPARATION AND CHARACTERIZATION OF ALUMINOSILICATE SUPPORTS WITH A SYNTHESIZED LAYER OF CATALYTIC FILAMENTOUS CARBON. II. SYNTHESIS OF CARBON NANOFIBERS ON A SUPPORTED COBALT CATALYST

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Kinet. Catal., 48(5) (2007) pp. 757-764.

The synthesis of a layer of catalytic filamentous carbon (CFC) on a Co catalyst supported by homogeneous precipitation onto the surface of aluminosilicate supports (ceramic foam and vermiculite) was studied. The effects of CFC layer synthesis conditions (the catalyst concentration on a support, the pyrolysis temperature of a propane–butane mixture, and the composition of the gas mixture) on the specific surface areas of supports, the yield of carbon ((g C)/(g Co)), and the morphology of a surface CFC layer were examined. It was found that, in the case of ceramic foam, the concentration of cobalt hydroxide precipitated on the surface was lower by a factor of 15 and the yield of carbon was higher by a factor of 20–40 than those in vermiculite. The specific surface areas of supports, the yield of carbon, and the amount of synthesized carbon increased as the pyrolysis temperature of a propane–butane mixture was increased from 500 to 600°C. As found by scanning electron microscopy, the carbon content increased with pyrolysis temperature because of an increase in the length of carbon nanofibers. The properties (activity and stability) of biocatalysts prepared by the adsorption immobilization of a recombinant protein having glucose isomerase activity on CFC–Co-containing supports (ceramic foam and vermiculite) were studied.

CATALYTIC PROPERTIES OF GLUCOAMYLASE IMMOBILIZED ON SYNTHETIC CARBON MATERIAL SIBUNIT

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Appl. Biochem. Microbiol., 43(4) (2007) pp. 374-378.

Glucoamylase (commercial preparation Glucavamorin) was immobilized by sorption on a carbon support Sibunit. Starch saccharification by the resulting biocatalyst (dextrin hydrolysis) was studied. Investigation of the effect of adsorptional immobilization on kinetic parameters of glucoamylase, including the rate constant of thermal inactivation, showed that immobilization of Glucavamorin on Sibunit resulted in a thousand fold increase in glucoamylase stability in comparison with the dissolved enzyme. Presence of the substrate (dextrins) in the reaction mixture had a considerable stabilizing effect. Increase in dextrin concentration increases the thermostability of the immobilized enzyme. The overall factor of glucoamylase stabilization adsorbed on Sibunit with the presence of 53% dextrin solutions in comparison with the dissolved enzyme approximated 10^5. The biocatalyst for starch saccharification made on the base of Subunit-adsorbed Glucavamorin had a high operational stability. Its half-inactivation time at 60°C exceeded 30 days.

SYNTHESIS OF MULTIWALL CARBON NANOTUBES OVER COBALT CATALYSTS

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Theor. Found. Chem. Eng., 41(5) (2007) pp. 634-638.

CH_4 pyrolysis over Co/Al_2O_3 (60 wt %) catalysts, prepared by different methods and doped with tervalent-metal oxides, was studied gravimetrically under isothermal conditions at 600–750°C and the atmospheric pressure. The process starts at the maximum rate and decays with time. The pyrolysis rate is a function of linear CH_4 rate peaks at 40 cm/min. With the provision of the maximum process rate, the activation energy of CH_4 pyrolysis over various catalysts falls within a range of 150–240 kJ/mol. For catalysts with identical compositions, the activity depends on their preparation. Transmission electron microscopy showed that the product was multiwall carbon nanotubes with diameters ranging from 6 to 30 nm. Mathematical equations were proposed to describe the process kinetics. A productivity of 12 g/h is achieved on a laboratory continuous reactor.

FORMATION OF A NICKEL CATALYST ON THE SURFACE OF ALUMINOSILICATE SUPPORTS FOR THE SYNTHESIS OF CATALYTIC FIBROUS CARBON

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Kinet. Catal.,
48(6) (2007) pp. 803-811.

Conditions for the homogeneous precipitation of nickel hydroxide in the presence of urea onto the surface of aluminosilicate honeycomb monoliths, which were prepared based on clay, talc, and amorphous aluminum hydroxide, were examined. Factors affecting the concentration of supported nickel (synthesis time, starting solution concentrations, loaded amount of the support, and support calcination temperature) were studied. The possibility of supporting nickel hydroxide onto the surface of cellular ceramic foam, glass foam, and haydite was demonstrated. The morphology of nickel hydroxide particles, nickel metal particles on support surfaces, and carbon coatings synthesized in the course of the catalytic pyrolysis of a propane–butane mixture was studied by scanning electron microscopy.

MODELING OF HETEROGENEOUS SURFACES AND CHARACTERIZATION OF POROUS MATERIALS BY EXTENDING DENSITY FUNCTIONAL THEORY FOR THE CASE OF AMORPHOUS SOLIDS

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Appl. Surf. Sci.,
253(13) (2007) pp. 5610-5615.

A method of characterization of carbonaceous materials using nongraphitized carbon black as a reference is considered. The Tarazona density functional theory was applied to amorphous solids to describe nitrogen adsorption on nongraphitized carbon black. This allows to describe energetic heterogeneity without the need to invoke any energy distribution functions. To derive the pore size distribution (PSD) of porous carbon whose pore walls are non-graphitized, the entropy concept in the regularization method was used. With this approach PSD is more well-behaved than that obtained with the usual means. This new theory was applied to study the effects of technological parameters on porous structure of a series of activated carbon.

CHARACTERIZATION OF THE POROUS STRUCTURE OF CARBON MATERIALS BY MEANS OF DENSITY FUNCTIONAL THEORY

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Kinet. Catal., 48(4) (2007) pp. 589-598.

Nitrogen adsorption isotherms were analyzed using density functional theory (DFT) to characterize active carbons. It is shown how the effect of technological parameters on the structure of active carbons can be revealed with the help of DFT. DFT is used to analyze the pore-size distribution and to determine the pore surface area for active carbons of different types.

PROPERTIES OF A GLASS SURFACE MODIFIED BY A NEW FROSTING PASTE. SURFACE MORPHOLOGY

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Glass Ceramics,
64(11-12) (2007) pp. 425-428.

A new paste is proposed for frosting glass surfaces. The action of the paste is based on the physical-chemical process of surface chemical ion exchange. The paste does not contain hydrofluoric acid or its derivatives. The frosting process occurs at 300°C and differs substantially from the existing frosting processes — chemical etching and sandblasting. Electron microscope investigations of the glass surface were performed with a JSM-6460RV scanning electron microscope (Jeol, Japan). The morphology of the surface layer of the frosted glass is characterized by a system of microblocks (linear dimensions 50–150 μm) and microcracks (width 100–200 nm). The microblocks have even faces and a smooth frontal surface. The surface morphology of the glass is the same after frosting and subsequent deposition of a metal by chemical reduction.

Composite Sorbents

NEW FAMILY OF MATERIALS FOR ADSORPTION COOLING: MATERIAL SCIENTIST APPROACH

Yu.I. Aristov

J. Eng. Thermophys., 16(2) (2007) pp. 63-72.

This paper summarises literature data concerning a new family of materials for adsorptive cooling. They are composites of a type "a salt confined to a porous host matrix" (so called Selective Water Sorbents or SWSs). These materials demonstrate an intermediate behaviour between solid adsorbents, salt hydrates and liquid absorbents. Thermodynamic equilibrium with water vapour has been measured for more than thirty SWSs based on halides, sulphates and nitrates of alkaline and alkaline earth metals, that are confined to various matrices (silica, alumina, porous carbons, clays, MCM-41, etc.). The important advantage of the SWSs is the possibility to controllably modify their water sorption properties in a wide range by varying a) chemical nature of the impregnated salt, b) porous structure of the host matrix, c) the amount of the confined salt, and d) preparation conditions. This, in principle, allows purposeful synthesis of new solid sorbents with predetermined properties which fit demands of particular adsorptive cycle. Appropriate examples are presented in the paper. Recent study of adsorption chillers with granulated and compact layers of SWS-1L ($CaCl_2$ in mesoporous silica) showed the experimental COP as high as 0.6 that is larger than reported for "silica gel/water" and "zeolite/water" systems at the same cycle conditions. The concept of matrix modification with an active salt can be also used for synthesis of efficient sorbents of carbon dioxide, methanol and ammonia. Other practical applications of SWSs are briefly discussed, such as heat protection, gas drying and fresh water production from the atmosphere.

NOVEL MATERIALS FOR ADSORPTIVE HEAT PUMPING AND STORAGE: SCREENING AND NANOTAILORING OF SORPTION PROPERTIES

Yu.I. Aristov

J. Chem. Eng. Japan,
40(13) (2007) pp. 1241-1251.

The brief review of innovative materials for adsorptive pumping and storage of low temperature heat as well as current trends in this field showed their great potential. To implement it in the near future it is reasonable to follow two complementary lines: (a) systematic testing for this application novel adsorbents initially developed for other targets (gas drying, separation, etc.), and (b) tailoring of new specific porous materials adapted just to variety of adsorptive cooling, heating and storage cycles under different climatic and boundary conditions. The main idea of the latter approach is that for each particular adsorptive cycle there is an optimal adsorbent, the thermodynamic characteristics of which could allow perfect performance of this cycle. The first step of the analysis is the formulation of requirements to this desirable (ideal) adsorbent. Then, the final step is to design and synthesize a new adsorbent with sorption properties close or even equal to those determined before as perfectly fitting the cycle. Particular examples of nanotailoring of adsorption properties are given for two phase materials, composites "a salt in a porous host matrix", which can be used as efficient adsorbents of water, methanol and ammonia.

AN ADVANCED SOLID SORPTION CHILLER USING SWS-1L

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Appl. Therm. Eng.,
27(13) (2007) pp. 2200-2204.

In this paper, the experimental testing on an advanced solid sorption chiller based on a heat exchanger coated with a compact layer of SWS-1L ($CaCl_2$ in mesoporous silica gel) is presented. The experimental results showed a specific power of 150-200 W/kg of adsorbent and a cycle time of 10-20 minutes. These values are sensibly better (i.e. one order of magnitude different) than those measured in the same unit working with a SWS-1L pelletised bed. The cooling COP ranged between 0.15 and 0.3, depending on the operating conditions.

SOLAR DRIVEN ADSORPTIVE CHILLER: AT THE INTERFACES BETWEEN CHEMICAL AND THERMAL ENGINEERING

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Chem. Eng. J., 134(1-3) (2007) pp. 58-65.

Thermochemical systems for energy conversion are just at the interface between chemical and thermal engineering because the final aim is a transformation of energy, while the specific tool is a thermally driven chemical reaction. Designing the efficient systems calls for appropriate choice of chemical process, comprehensive analysis of its thermodynamics and kinetics, intensification of heat and mass transfer, intelligent integration of components into overall heat device, etc. This paper reviews the joint activity of the six Institutes from the four countries on applying chemical reactions in modern devices for production of cold, which are driven by solar heat.

SORPTION EQUILIBRIUM OF METHANOL ON NEW COMPOSITE SORBENT «$CaCl_2$/Silica Gel»

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Adsorption, 13(2) (2007) pp. 121-127.

This paper presents experimental data on methanol sorption on new composite sorbents which consist of mesoporous silica gels and calcium chloride confined to their pores. Sorption isobars and XRD analysis showed the formation of a solid crystalline solvate $CaCl_2 \cdot 2MeOH$ at low methanol uptake, while at higher uptake the formation of the $CaCl_2$-methanol solution occurred. The solution confined to the silica pores showed the sorption properties similar to those of the $CaCl_2$-methanol bulk solution. Calorimetric and isosteric analyses showed that the heat of methanol sorption depends on the methanol uptake, ranging from 38±2 kJ/mol for the solution to 81±4 kJ/mol for the solid crystalline phase $CaCl_2 \cdot 2MeOH$. The above mentioned characterizations allowed the evaluation of the methanol sorption and the energy storage capacities, clearly showing that the optimal applications of these new composite sorbents are the methanol removal from gaseous mixtures, heat storage and sorption cooling driven by low temperature heat.

MODELING OF THE LIMITING STEP OF WATER SORPTION BY COMPOSITE SORBENTS OF THE “CALCIUM CHLORIDE IN POROUS MATRIX” TYPE

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Theor. Found. Chem. Eng., 41(1) (2007) pp. 83-90.

A nonstationary model is proposed to describe the water sorption from air by composite sorbents of the “salt in a porous matrix” type via the interaction of water with the salt to form a salt solution in pores. It is shown that the dynamics of the moisture content at the adsorber outlet cannot be described under the assumption of constancy of the effective sorption rate constant. The limiting step of water sorption is analyzed, and it is supposed that the limiting step is the water diffusion through the solution layer forming near the external surface of a granule. An expression for the sorption rate constant as a function of the moisture content of the sorbent is proposed that takes into account the monotonic decrease in this constant with an increase in the amount of water sorbed. The effective diffusion coefficient at the limiting step of sorption is determined, which turns out to be close to the effective diffusion coefficient in an aqueous calcium chloride solution.

EFFECT OF CAPILLARY CONDENSATION ON WATER SORPTION BY COMPOSITE CALCIUM CHLORIDE IN A POROUS MATRIX SORBENTS

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Theor. Found. Chem. Eng., 41(2) (2007) pp. 200-204.

To develop a model of water sorption from air by composite salt in a porous matrix sorbents, the effect of capillary condensation was studied. It was shown that, at high humidity of incoming air, it is necessary to take into account the capillary condensation of water in small pores of the matrix because the capillary condensation significantly affects the concentration and temperature profiles in the sorbent bed and leads to intense moisture accumulation at the bed inlet. The model was used to study the kinetics of water vapor sorption in a flow adsorber with a fixed bed of calcium chloride in porous aluminum oxide and calcium chloride in a porous carbon–carbon composite Sibunit sorbents.

ADSORPTIVE AIR CONDITIONING SYSTEMS DRIVEN BY LOW TEMPERATURE ENERGY SOURCES: CHOICE OF WORKING PAIRS

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J. Chem. Eng. Japan,
40(13) (2007) pp. 1287-1291.

The performance of an adsorptive air-conditioning cycle driven by low temperature heat (80–85°C) was studied. The analysis rested on the Polanyi potential theory and took into account literature and author's experimental data on sorption equilibrium of various conventional and innovative adsorbents with water and methanol as working fluids. This adsorbent screening is aimed at evaluating the optimal working pairs for this application. The composite sorbent of methanol Lithium Chloride in mesoporous silica gel and the adsorbent of water FAM-Z02 were selected as challenging pairs for severe conditions of heat rejection typical for solar cooling in hot countries and air conditioning in cars.

INFLUENCE OF CHARACTERISTICS OF METHANOL SORBENTS "SALTS IN MESOPOROUS SILICA" ON THE PERFORMANCE OF ADSORPTIVE AIR CONDITIONING CYCLE

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Ind. Eng. Chem. Res.,
46(9) (2007) pp. 2747-2752.

In this paper a new family of methanol sorbents "salts in mesoporous silica" is proposed for utilization in an adsorptive air conditioner driven by a low-temperature heat (T = 338-373 K). The methanol sorption capacity of composites, under typical cooling cycle conditions, was measured by an express method based on the Polanyi principle of temperature invariance. An estimation of the cooling coefficient of performance (COP) was carried out in order to compare the performance of such sorbents with other methanol and water sorbents. Results obtained showed that most of the synthesized composites present methanol sorption ability higher than that of common methanol sorbents (e.g., active carbons). The composite LiCl (30.6 wt %)/SiO_2 shows the highest sorption capacity, w_{ads} = 0.8 g/g, and uptake variation per cycle, Δw = 0.71 g/g. The corresponding cooling COP is 0.74 (maximum T_{des} = 363 K, single-bed cycle), which is comparable to typical COP values for the best water sorbents.

ISOTHERMAL SORPTION CHARACTERISTICS OF THE $BaCl_2$–NH_3 PAIR IN A VERMICULITE HOST MATRIX

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Appl. Therm. Eng.,
27(14-15) (2007) pp. 2455-2462.

Chemisorption could be useful in adsorption systems due to the large concentration change compared with physisorption. Equilibrium concentration characteristics of ammonia with a composite adsorbent material ($BaCl_2$ impregnated into a vermiculite matrix) are investigated: the maximum concentration is about 0.4 kg ammonia/ kg adsorbent. Hysteresis was observed between the synthesis and the decomposition reaction. The analysis of the data suggests that the hysteresis could be due to the dimensional changes of the solid during the reactions. The bivariant behaviour observed was contrary to the monovariant behaviour anticipated and the reasons are discussed. The COP of a basic adsorption cycle for typical ice-making and air-conditioning applications utilizing ammonia and the composite material were calculated. The results show that the material could be used for air conditioning or other refrigeration applications. The COP could reach up to 0.6 at typical conditions (T_{ev}=15°C, T_{con}=35 °C).

A NEW APPROACH TO REGENERATING HEAT AND MOISTURE IN VENTILATION SYSTEMS

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Energy and Buildings, 40(3) (2008) pp. 204-208.

For countries with a cold climate the large difference (30-60°C) in winter between indoor and outdoor temperatures leads to a) large heat losses in ventilation systems; b) moisture freezing at the systems exit and c) great reduction in the indoor humidity. Here a new approach is presented for regenerating heat and moisture in ventilation systems in cold climates which allows resolution of these problems. The method has been tested under climatic conditions of West Siberia (winter 2005-2006). The prototype system requires very little maintenance, has a low capital cost, is compact and energy efficient. Technical, economic and social aspects of this method are discussed.

CHANGE OF CO_2 CARRYING CAPACITY OF CaO IN ISOTHERMAL RECARBONATION-DECOMPOSITION CYCLES

A.I. Lysikov, A.N. Salanov, A.G. Okunev

Ind. Eng. Chem. Res.,
46(13) (2007) pp. 4633-4638.

The change of CO_2 carrying capacity of CaO sorbents prepared from different precursors has been studied using thermogravimetric analysis in a long series of isothermal recarbonation-decomposition cycles in the temperature range of 750-850°C. The residual capacity of the CaO sorbents after a large number of cycles was found to depend on the precursor type, the experimental temperature, and the duration of the recarbonation stage. The residual capacities of the CaO derived from the powdered calcium carbonates were much higher than that of the CaO produced from the crystalline $CaCO_3$. A simple tentative model has been suggested, according to which recarbonation-decomposition cycles result in formation of the interconnected CaO network that acts as a refractory support and determines sorption properties of the material. By using a new model, a simple synthesis procedure has been suggested that produces CaO sorbents with high residual CO_2 carrying capacities.

Electrocatalysis and Electrochemical Processes

ON THE INFLUENCE OF THE METAL LOADING ON THE STRUCTURE OF CARBON-SUPPORTED PTRU CATALYSTS AND THEIR ELECTROCATALYTIC ACTIVITIES IN CO AND METHANOL ELECTROOXIDATION

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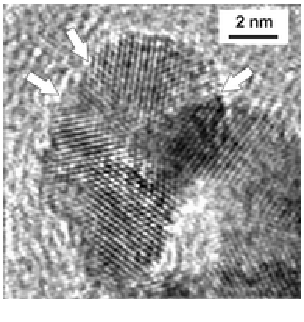
Phys. Chem. Chem. Phys.,
9(40) (2007) pp. 5476-5489.

PtRu (1 : 1) catalysts supported on low surface area carbon of the Sibunit family (S_{BET} = 72 m^2 g^{-1}) with a metal percentage ranging from 5 to 60% are prepared and tested in a CO monolayer and for methanol oxidation in H_2SO_4 electrolyte. At low metal percentage small (<2 nm) alloy nanoparticles, uniformly distributed on the carbon surface, are formed. As the amount of metal per unit surface area of carbon increases, particles start coalescing and form first quasi two-dimensional, and then three-dimensional metal nanostructures. This results in a strong enhancement of specific catalytic activity in methanol oxidation and a decrease of the overpotential for CO monolayer oxidation. It is suggested that intergrain boundaries connecting crystalline domains in nanostructured PtRu catalysts produced at high metal-on-carbon loadings provide active sites for electrocatalytic processes.



THERMAL CONDUCTIVITY OF COMPOSITE CATALYSTS CONTAINING METALLIC COPPER AS A REINFORCING COMPONENT

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Theor. Found. Chem. Eng.,
41(2) (2007) pp. 184-192.

The dependences of the electrical and thermal conductivities of porous composite materials containing a metallic component (copper) on the volume copper content are investigated experimentally. The measured thermal conductivities of samples prepared according to the proposed technique indicate that the thermal conductivity of monolithic catalysts with a copper content of no less than 15 vol % exceeds 1 W m^{-1} K^{-1}. This corresponds to the formation of a connected cluster consisting of conducting spheres in a random packing of conducting and insulating spheres. A comparative analysis of the thermal and electrical conductivities of the composites demonstrates that, at a copper content of higher than 20 vol %, the thermal conduction through a percolation cluster formed by copper particles makes the dominant contribution. In employing composite materials containing a catalytically active component in exothermic catalytic processes (Fischer–Tropsch synthesis, steam conversion of carbon monoxide CO, etc.), their high thermal conductivity is an important advantage that makes it possible to decrease the temperature gradient across the porous composite catalyst bed. A semiempirical method for calculating the thermal conductivity of composites is developed. The results of the calculations performed using the proposed method are in good agreement with experimental data.

MODEL Pd-BASED BIMETALLIC SUPPORTED CATALYSTS FOR NITRATE ELECTROREDUCTION

S.N. Pronkin, P.A. Simonov, V.I. Zaikovskii, E.R. Savinova

J. Mol. Catal. A: Chem.,
265(1-2) (2007) pp. 141-147.

An approach for *in situ* preparation of bimetallic Pd–Cu catalysts active in nitrate electroreduction is described. The catalysts are prepared by modification of the surface of Pd carbon-supported nanoparticles by monolayer of Cu. The morphology of Pd/C nanoparticles is characterized by HR-TEM and by their electrochemical properties in the adsorption of hydrogen and Cu adatoms. Modification of Pd surface by Cu adlayer results in significant increase of the activity in nitrate electroreduction. Dependencies of the reaction rate for Cu/Pd/C catalysts on the electrode potential, nitrate concentration and anion of supporting electrolyte are discussed.

ELECTRICAL PROPERTIES OF NANOCOMPOSITES BASED ON COMB-SHAPED NEMATIC POLYMER AND SILVER NANOPARTICLES

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J. Phys. Chem. C,
111(24) (2007) pp. 8451-8458.

A nematic comb-shaped copolymer and its nanocomposites containing 0.063-0.54 in vol % of silver nanoparticles were studied by broadband dielectric spectroscopy. The frequency dependence of specific alternating current (ac) conductivity was used to estimate the temperature-frequency intervals of charge transfer by long and short distances, respectively. With increasing the concentration of nanoparticles, specific ac conductivity increases. The concentration dependence of dielectric permittivity suggests that distribution of nanoparticles is homogeneous, and conducting channels are not formed. With increasing the concentration of silver nanoparticles, the glass transition temperature of the nanocomposites, described in terms of the strength/fragility concept, increases, whereas the strength parameter *D* decreases (i.e., "fragility" increases).

INFLUENCE OF CARBON SUPPORT ON THE PERFORMANCE OF PLATINUM BASED OXYGEN REDUCTION CATALYSTS IN A POLYMER ELECTROLYTE FUEL CELL

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J. Appl. Electrochem.,
37(12) (2007) pp. 1429-1437.

Novel carbons from the Sibunit family prepared via pyrolysis of hydrocarbons [Yermakov YI, Surovikin VF, Plaksin GV, Semikolenov VA, Likholobov VA, Chuvilin AL, Bogdanov SV (1987) React Kinet Catal Lett 33:435] possess a number of attractive properties for fuel cell applications. In this work Sibunit carbons with BET surface areas ranging from ca. 20 to 420 $m^2\ g^{-1}$ were used as supports for platinum and the obtained catalysts were tested as cathodes in a polymer electrolyte fuel cell. The metal loading per unit surface area of carbon support was kept constant in order to maintain similar metal dispersions (~0.3). Full cell tests revealed a strong influence of the carbon support texture on cell performance. The highest mass specific activities at 0.85 V were achieved for the 40 and 30 wt.% Pt catalysts prepared on the basis of Sibunit carbons with BET surface areas of 415 and 292 $m^2\ g^{-1}$. These exceeded the mass specific activities of conventional 20 wt.% Pt/Vulcan XC-72 catalyst by a factor of ca. 4 in oxygen and 6 in air feed. Analysis of the I–U curves revealed that the improved cell performance was related to the improved mass transport in the cathode layers. The mass transport overvoltages were found to depend strongly on the specific surface area and the texture of the support.

Catalysis by Heteropoly Compounds and Polyoxometallates

STABILIZATION OF PALLADIUM IN HOMOGENEOUS CHLORIDELESS CATALYSTS FOR OLEFINS C_2-C_4 OXIDATION INTO CARBONYL COMPOUNDS WITH OXYGEN

V.F. Odyakov, E.G. Zhizhina, K.I. Matveev

Catal. Ind., 4 (2007) pp. 19-25.

The first step of homogeneous oxidation of olefins C_2–C_4 by dioxygen in the presence of catalyst Pd + HPA-x, where HPA-x is molybdovanadophosphoric heteropoly acid $H_{3+x}PV^V_xMo_{12-x}O_{40}$, is reduction of HPA-x with olefin in the presence of Pd complexes into H_mHPA-x at 30–60°C. Here $m = [V^{IV}]_\Sigma / [HPA\text{-}x]$ is a degree of reduction of the catalyst. In this case, the redox-system Pd^I_2/Pd^0_2 works, but it converts into Pd^{2+}/Pd_{met} system with higher redox potential with increasing of temperature. During this transformation, interval of *m* values where the catalyst remains homogeneous diminishes, and metallic Pd begins to precipitate from the reduced catalyst. To prevent Pd precipitation, it is necessary to introduce stabilizers of Pd. As stabilizers, 2,6-pyridinecarboxylic (dipicolinic) acid (**I**) and cheaper 3-pyridinesulfonic acid are proposed. In the presence of these stabilizers, activity of the catalyst diminishes, but an area of its homogeneity expands. Complexes of Pd with **I** are stable at temperatures of regeneration of the catalyst (150–160°C). Therefore, **I** is considered as rather perspective stabilizer of Pd in catalysts Pd + HPA-x and can be used in industrial homogeneous processes of olefins C_2–C_4 oxidation by oxygen.

STABILITY OF PALLADIUM IN HOMOGENEOUS CHLORIDE-FREE CATALYSTS Pd + HETEROPOLY ACID FOR OXIDATION OF C_2-C_4 ALKENES BY DIOXYGEN

E.G. Zhizhina, V.F. Odyakov, K.I. Matveev

React. Kinet. Catal. Lett.,
91(2) (2007) pp. 325-332.

A possibility of the use of polycarboxylic acids as stabilizers of Pd in homogeneous catalysts for the oxidation of C_2–C_4 alkenes is studied. Pyromellitic acid is shown to have the stabilizing properties.

HOMOGENEOUS CATALYTIC OXIDATION OF PROPENE TO ACETONE AND BUTENE-1 TO BUTANONE IN THE PRESENCE OF PALLADIUM AND MOLYBDOVANADOPHOSPHORIC HETEROPOLY ACID

E.G. Zhizhina, M.V. Simonova, V.F. Odyakov, K.I. Matveev

Appl. Catal., A, 319(1) (2007) pp. 91-97.

Oxidation of propene to acetone and butene-1 to butanone in aqueous solutions was studied in the presence of homogeneous catalysts Pd + HPA-*x* (HPA-*x* = $H_{3+x}PV_xMo_{12-x}O_{40}$ having Keggin composition, *x* = 1–4). The reaction order with respect to alkenes is 1, to palladium is 0.5 for propene and 0.66 for butene-1, respectively. The rate of alkene oxidation does not depend on concentration of HPA-*x* and acidity of catalyst solution. Alkenes are also successfully oxidized in the presence of new catalysts Pd + HPA-x′ containing modified high-vanadium non-Keggin HPA-*x*′ (*x*′ > 4). The HPA-*x*′ solutions have improved thermal stability and are very promising for oxidation of the lower alkenes. Kinetic peculiarities of oxidation of both alkenes in the presence of solutions of HPA-*x*′ and HPA-*x* are similar.

HYDROGEN PEROXIDE AND OXYGEN-HYDROGEN OXIDATION OF AROMATIC COMPOUNDS IN CATALYTIC SYSTEMS CONTAINING HETEROPOLY COMPOUNDS

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J. Hazard. Mater., 146(3) (2007) pp. 569-576.

Hydrogen peroxide and Pt activated mixture of gaseous O_2 and H_2 have been applied to oxidation of aromatic compounds in the presence of red-ox active heteropoly compounds in the form of acid $H_4PMo_{11}VO_{40}$ and tetrabuthylammonium (TBA) salts $TBA_4PMo_{11}VO_{40}$ and $TBA_4HPW_{11}Fe(OH)O_{39}$. Benzene, toluene and phenol were subjected to hydroxylation of the ring, that was accompanied by secondary oxidation in reaction with hydrogen peroxide. Oxygenation of toluene was equally directed to the ring and to methyl group. The total reactivity of substrates was increased in order of benzene < toluene < phenol in oxidation by both O_2/H_2 and H_2O_2, that indicated identical nature of active intermediates for both oxidants. It was suggested HPC bonded radical species to be responsible for oxidation of hydrocarbons.

AEROBIC OXIDATIONS OF α-PINENE OVER COBALT-SUBSTITUTED POLYOXOMETALATE SUPPORTED ON AMINO-MODIFIED MESOPOROUS SILICATES

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J. Catal.,
246(2) (2007) pp. 241-248.

Co-containing polyoxometalate $[Bu_4N]_4H[PW_{11}Co(H_2O)O_{39}]$ (Co-POM) was supported on various NH_2-modified mesoporous silicate matrixes (SBA-15, MCF, and SiO_2-xerogel). The catalysts were characterized by elemental analysis, N_2 adsorption, DRS–UV, and FTIR spectroscopy. α-Pinene autoxidation and its co-oxidation with isobutyraldehyde (IBA) over the supported Co-POM catalysts have been studied and compared with the corresponding processes in the presence of the homogeneous Co-POM. The autoxidation process affords allylic oxidation products, the selectivity to verbenol/verbenone decreases with alkene conversion and attains 70% at 20% conversion and 40% at 46% conversion. The catalysts can be used repeatedly without loss of the activity and selectivity during several catalytic cycles. Co-oxidation of α-pinene and IBA produces selectively α-pinene epoxide with up to 94% selectivity at 96% alkene conversion. The catalysts can be regenerated by evacuation.

DITITANIUM-CONTAINING 19-TUNGSTODIARSENATE(III) $[Ti_2(OH)_2As_2W_{19}O_{67}(H_2O)]^{8-}$: SYNTHESIS, STRUCTURE, ELECTROCHEMISTRY, AND OXIDATION CATALYSIS

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Chem. Eur. J.,
13(17) (2007) pp. 4733-4742.

The dititanium-containing 19-tungstodiarsenate(**III**) $Ti_2(OH)_2As_2W_{19}O_{67}(H_2O)]^{8-}$ (**1**) has been synthesized and characterized by IR, TGA, elemental analysis, electrochemistry, and catalytic studies. Single-crystal X-ray analysis was carried out on $Cs_8[Ti_2(OH)_2As_2W_{19}O_{67}(H_2O)]\cdot 2\,CsCl\cdot 12\,H_2O$ (Cs-**1**), which crystallizes in the monoclinic system, space group $P2_1/m$, with a=12.7764(19), b=19.425(3), c=18.149(3) Å, β=110.234(3)°, and Z=2. Polyanion **1** comprises two (B-α-$As^{III}W_9O_{33}$) Keggin moieties linked through an octahedral $\{WO_5(H_2O)\}$ fragment and two unprecedented square-pyramidal $\{TiO_4(OH)\}$ groups, leading to a sandwich-type structure with nominal C_{2v} symmetry. Synthesis of **1** was accomplished by reaction of $TiOSO_4$ and $K_{14}[As_2W_{19}O_{67}(H_2O)]$ in a 2:1 molar ratio in aqueous, acidic medium (pH 2). Polyanion **1** could also be isolated as a tetra-n-butyl ammonium (TBA) salt, $\{(n\text{-}C_4H_9)_4N\}_5H_3[Ti_2(OH)_2As_2W_{19}O_{67}(H_2O)]$ (TBA-**1**). TBA-**1** was studied by cyclic voltammetry in acetonitrile (MeCN) solutions containing 0.1 M $LiClO_4$ and compared with the results obtained with Cs-**1** in aqueous media. In MeCN, the Ti^{IV} and W^{VI} waves could not be separated distinctly. An important adsorption phenomenon on the glassy carbon working electrode was encountered both in cyclic voltammetry and in controlled potential electrolysis and was confirmed by Electrochemical Quartz Crystal Microbalance (EQCM) studies on a carbon film. TBA-**1**, dissolved in MeCN, reacts with H_2O_2 to give peroxo complexes stable enough for characterization by UV-visible spectroscopy, cyclic voltammetry, and EQCM. TBA-**1** shows high catalytic activity (TOF=11.3 h^{-1}) in cyclohexene oxidation with aqueous H_2O_2 producing products typical of a heterolytic oxidation mechanism. The stability of TBA-**1** under turnover conditions was confirmed by using IR, UV-visible spectroscopy as well as cyclic voltammetry.

Nanocomposite Nanostructured Materials: Synthesis, Characterization, Application

STUDY OF SYNTHESIS GAS PRODUCTION OVER STRUCTURED CATALYSTS BASED ON LaNi(Pt)O_x- AND Pt(LaPt)-CeO_2-ZrO_2 SUPPORTED ON CORUNDUM

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Stud. Surf. Sci. Catal.,
167 (2007) pp. 343-348.

Study of partial oxidation of methane (POM), steam (SR), authothermal (AR) and dry (DR) reforming of methane over catalysts containing $LaNiO_3$ (pure or promoted by Pt) or $LaPtO_x/CeO_2$-ZrO_2 supported on the fragments of corundum monolith reveals that the most effective catalysts contain $LaNiO_3$ and $LaPtO_x$ with La excess. The reduction pretreatment as well as addition of Pt to $LaNiO_x$ facilitates the formation of syngas at a lower temperature, furthermore, Pt favors a high catalyst stability preventing catalyst coking.

PERFORMANCE OF MONOLITHIC CATALYSTS WITH COMPLEX ACTIVE COMPONENT IN PARTIAL OXIDATION OF METHANE INTO SYNGAS: EXPERIMENTAL STUDIES AND MODELING

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Stud. Surf. Sci. Catal.,
167 (2007) pp. 361-366.

Experimental studies and modeling demonstrated importance of the heat and mass transfer for the process of the natural gas partial oxidation into syngas at short contact times on monolithic catalysts with a complex active component Pt/$LaNiO_3$/Ce-Zr-La-O. A simplified approach for modeling based upon using rate constants for the reactions of methane transformation into syngas estimated for separate structural elements of monolithic catalysts in nearly isothermal conditions was successfully verified. This approach opens the way for optimization of performance of syngas generators equipped with catalysts containing active components of any complexity.

DESIGN OF STRUCTURED CATALYSTS BASED ON METALLIC MONOLITHS FOR SYNGAS PRODUCTION VIA PARTIAL OXIDATION OF NATURAL GAS

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Stud. Surf. Sci. Catal.,
172 (2007) pp. 241-244.

Structured catalysts based on metallic supports of different types with a high thermal conductivity were designed. The metal monolithic supports were made from a heat-resistant foil or gauze coated with a protective layer of Al_2O_3 or ZrO_2 using blast dusting. To prepare catalysts, Ce-Zr-La-O and $LaNiO_x$ promoted with Pt were successively supported on monoliths. A high and stable performance of these catalysts in the partial oxidation of methane (POM) at short contact times in the autothermal mode was demonstrated. In the reactor with a heat exchanger these catalysts operate without any heat preheat at very high gas velocities in the autothermal mode.

Pt-SUPPORTED NANOCRYSTALLINE CERIA-ZIRCONIA DOPED WITH La, Pr OR Gd: FACTORS CONTROLLING SYNGAS GENERATION IN PARTIAL OXIDATION/AUTOTHERMAL REFORMING OF METHANE OR OXYGENATES

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Solid State Phenomena,
128 (2007) pp. 239-248.

Nanocrystalline CeO_2-ZrO_2 (Ce:Zr 1:1) samples doped with La, Pr or Gd cations (containing up to 30 at.%) were prepared via the Pechini route. Pt (1.4 wt.%) was supported via impregnation with H_2PtCl_6 solution followed by drying and calcination. The samples' surface features were studied by SIMS and FTIRS of adsorbed CO. The oxygen mobility was characterized by the dynamic oxygen isotope exchange and H_2 TPR. Catalytic activity was studied in the flow installation using diluted feeds (0.7% CH_4 +0.5% O_2 or 1% C_3H_6O + 0.5% O_2 +0.5% H_2O in He). In the selective oxidation of methane (POM), the catalytic activity correlates with Pt dispersion controlled by the oxidized sample's ability to stabilize Pt^{2+} cations as precursors of small reactive Pt clusters formed under reaction conditions. This is favoured by a larger doping cation (La) and a developed network of nanodomain boundaries. At comparable Pt dispersion, the highest performance was demonstrated by a La-doped system, which correlates with the highest surface/near-surface oxygen mobility controlled by the strength of Ce-O bonds in the surface layer. In the autothermal reforming of acetone, the activity trends differ from those in POM because of the more prominent role of the oxygen mobility required to prevent surface coking.

NANOCRYSTALLINE DOPED CERIA-ZIRCONIA FLUORITE-LIKE SOLID SOLUTIONS PROMOTED BY Pt: STRUCTURE, SURFACE PROPERTIES AND CATALYTIC PERFORMANCE IN SYNGAS GENERATION

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Mater. Res. Soc. Symp. Proc.,
988 (2007) QQ06-04.

Nanocrystalline ceria-zirconia samples doped with rare-earth (Gd, Pr, Sm, La) cations were prepared via modified Pechini route. Effect of their real structure and surface composition characterized by a combination of sophisticated physical methods (XRD, TEM+EDX, EXAFS, WAXS, UV-Vis, XPS, SIMS) on the mobility and reactivity of the lattice oxygen estimated by oxygen isotope exchange, H_2, CH_4 and CO TPR was analyzed. For the reaction of acetone autothermal reforming into syngas, catalytic activity correlates rather well with the oxygen mobility controlled by the type and content of a dopant.

DESIGN OF ANODES FOR IT SOFC: EFFECT OF COMPLEX OXIDE PROMOTERS AND Pd ON ACTIVITY AND STABILITY IN METHANE STEAM REFORMING OF Ni/YSZ (ScSZ) CERMETS

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Mater. Res. Soc. Symp. Proc.,
972 (2007) AA03-06.

Effect of fluorite-like or perovskite-like complex oxide promoters and Pd on the performance of Ni/8YSZ and Ni/ScSZ anode materials in CH_4 steam reforming (SR) or selective oxidation (SO) by O_2 into syngas was studied. The spatial uniformity of dopants distribution in composites was controlled by TEM combined with EDX analysis, while the lattice oxygen mobility and reactivity were estimated by CH_4 and H_2 temperature-programmed reduction (TPR). The oxide promoters suppress the coke deposition even at stoichiometric H_2O/CH_4 ratio, while doping by Pd ensures a good performance at moderate (~550°C) temperatures required for the Intermediate–Temperature Solid Oxide Fuel Cells (IT SOFC) operation.

DOPED NANOCRYSTALLINE Pt-PROMOTED CERIA-ZIRCONIA AS ANODE CATALYSTS FOR IT SOFC: SYNTHESIS AND PROPERTIES

V.A. Sadykov, N.V. Mezentseva, G.M. Alikina, A.I. Lukashevich, V.S. Muzykantov, R.V. Bunina, A.I. Boronin, E.M. Pazhetnov, E.A. Paukshtis, V.V. Kriventsov, A. Smirnova*, O.D. Vasylyev, J. Irvine***, O.F. Bobrenok****, V.I. Voronin*****, I.F. Berger******* (**University of Connecticut, Connecticut, USA*; ***Institute of Problems of Material Science, Kiev, Ukraine; ***University of St. Andrews, St. Andrews, UK; ****Institute of Thermophysics, Novosibirsk, Russia; *****Ural State University, Ekaterinburg, Russia*)

Mater. Res. Soc. Symp. Proc.,
1023 (2007) JJ02-07.1-6.

Ceria-zirconia samples doped with Gd, Pr, Sm, or La cations were prepared via Pechini route and promoted by Pt. Effect of their real structure and surface properties (characterized by neutronography, EXAFS, XPS, FTIRS of adsorbed CO) on the mobility and reactivity of the lattice oxygen (by oxygen isotope exchange and CH_4 TPR) was analyzed. For the reaction of CH_4 steam reforming (SR), catalytic performance is determined both by Pt dispersion and lattice oxygen mobility. Ni-YSZ anodes promoted by these catalysts possess a stable and efficient performance in CH_4 SR in the 600-800°C range in stoichiometric feeds without coking.

EFFECT OF COMPLEX OXIDE PROMOTERS AND Pd ON ACTIVITY AND STABILITY OF Ni/YSZ (ScSZ) CERMETS AS ANODE MATERIALS FOR IT SOFC

V.A. Sadykov, N.V. Mezentseva, R.V. Bunina, G.M. Alikina, A.I. Lukashevich, T.S. Kharlamova, V.A. Rogov, V.I. Zaikovskii, A.V. Ishchenko, T.A. Krieger, O.F. Bobrenok*, A. Smirnova, J. Irvine***, O.D. Vasylyev****** (**Institute of Thermophysics, Novosibirsk, Russia; **University of Connecticut, Connecticut, USA; ***University of St. Andrews, St. Andrews, UK; ****Institute of Problems of Materials Science, Kiev, Ukraine*)

Catal. Today, 131(1-4) (2008) pp. 226-237.

Effect of fluorite-like or perovskite-like complex oxide promoters, Pd and Cu on the performance of Ni/8YSZ and Ni/ScCeSZ anode materials in CH_4 steam reforming (SR) or selective oxidation (SO) by O_2 into syngas was studied. The spatial distribution of dopants in composites before and after contact with the reaction feed, features of components mutual interaction and forms of deposited coke were controlled by TEM combined with EDX analysis. The lattice oxygen mobility and reactivity were estimated by CH_4 and H_2 temperature-programmed reduction (TPR), and the amount of deposited carbon after operation in the feed with stoichiometric H_2O/CH_4 ratio was estimated by the temperature-programmed oxidation. Promoters decrease the amount of deposited coke, while doping by Pd or Cu ensures also a good and stable performance at moderate (~550°C) temperatures required for the intermediate-temperature solid oxide fuel cells (IT SOFC) operation.

DOPED NANOCOMPOSITES CeO_2–$LnMeO_3$ (Ln = Ce, La, Nd, Pr, Sm; Me = Mn, Fe, Co) FOR MIDDLE-TEMPERATURE CATHODES OF FUEL CELLS: MECHANOCHEMICAL SYNTHESIS

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Chem. Sustain. Devel.,
15(2-1) (2007) pp. 65-71.

Nanostructured materials based on the oxide phases with perovskite and fluorite structures are prepared by mechanochemical synthesis from misch metal carbonate and transition metal oxide with stoichiometric composition of cations in initial mixture La : Me = 1 : 1. The reduction rate of the nanocomposites with hydrogen or methane is higher than the reduction rate of the individual phases. These nanostructured materials possess a high mobility of lattice oxygen. High thermal stability and high total electrical conductivity exceeding that of electrolyte (doped fluorites). Synthesized materials may be applied as cathodes in solid oxide fuel cells and membrane material in oxidation processes.

CERIA-ZIRCONIA NANOPARTICLES DOPED WITH La OR Gd: EFFECT OF THE DOPING CATION ON THE REAL STRUCTURE

V.A. Sadykov, V.V. Kriventsov, E.M. Moroz, Yu.V. Borchert*, D.A. Zyuzin, V.P. Kolko, T.G. Kuznetsova, V.P. Ivanov, A.I. Boronin, N.V. Mezentseva, E.B. Burgina, **J. Ross**** (**University of Bremen, Bremen, Germany; **University of Limerick, Limerick, Ireland*)

Solid State Phenomena,
128 (2007) pp. 81-88.

The real structure of nanocrystalline CeO_2-ZrO_2 (Ce:Zr=1:1) systems prepared via the polymerized polyester precursor (Pechini) route and doped with La^{3+} or Gd^{3+} cations, up to 30 at.%, was studied by X-ray powder diffraction, EXAFS and Raman spectroscopy and the surface features characterized by XPS and SIMS. Undoped CeO_2-ZrO_2 system revealed nanoscale heterogeneity, perhaps due to the co-existence of Zr- or Ce-enriched domains. With large La^{3+} dopant the system remains bi-phasic within the studied ranges of composition, incorporation of the smaller Gd^{3+} cation stabilizes the single-phase solid solution. For both systems, the increase of dopant content was accompanied by a decline of domain size and an increase of the average lattice parameter of fluorite-like phases. Depletion of the surface layer by smaller Zr^{4+} cations was observed, while the surface content of a doping cation is either, close to that in the bulk (La) or below it (Gd). Such a spatial distribution of components results in some ordering of cations within the lattice. It is reflected in different modes of rearrangement of oxygen coordination polyhedra with the Gd or La content (distances and coordination numbers by EXAFS), and specificity of XRD patterns not conforming to a simple model with statistical distribution of oxygen vacancies.

SYNTHESIS AND PROPERTIES OF NANOCOMPOSITES WITH MIXED IONIC–ELECTRONIC CONDUCTIVITY ON THE BASIS OF OXIDE PHASES WITH PEROVSKITE AND FLUORITE STRUCTURES

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Glass Phys. Chem.,
33(4) (2007) pp. 320-334.

Nanocomposites consisting of phases with fluorite (doped CeO_2) and perovskite ($LaMnO_3$, $GdMnO_3$) structures are synthesized using the method of ester polymeric precursors (the Pechini method) and two sources of rare-earth elements (*Ln*), such as pure cerium and gadolinium salts or a commercial mixture of rare-earth carbonates containing La, Ce, Pr, Nd, and Sm cations. The genesis of the nanocomposite structure as a function of the sintering temperature is investigated using X-ray diffraction and electron microscopy. It is revealed that the genesis of the nanocomposite structure is governed, in many respects, by the fact that the decomposition of the ester polymeric precursor leads to the formation of a metastable phase, namely, a fluorite-like solid solution based on ceria with an excess concentration of the cations Ln^{3+}(Ln^{3+}= La^{3+}, Pr^{3+}, Nd^{3+}, Sm^{3+}) as compared to the equilibrium concentration. As a result, the perovskite phase (identified by X-ray diffraction analysis) is formed only after the subsequent annealing at temperatures higher than 800°C, when Ln^{3+} cations escape from particles of the solid solution. It is demonstrated that, at annealing temperatures of up to 1100°C, particles of both phases have nanometer sizes and are characterized by a uniform spatial distribution necessary for percolation. The nanocomposites possess a high total electrical conductivity and a high mobility of lattice oxygen. The reduction rate of the nanocomposites with hydrogen or methane is higher than the reduction rate of the individual phases. The characteristics of the nanocomposites prepared from the commercial mixture of rare-earth carbonates are better than those of the samples synthesized from the pure salts.

SCANDIA–STABILIZED ZIRCONIA: EFFECT OF DOPANTS ON SURFACE/GRAIN BOUNDARY SEGREGATION AND TRANSPORT PROPERTIES

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Mater. Res. Soc. Symp. Proc.,
972 (2007) AA10-05.

The nanocrystalline samples of 10wt%Scandia stabilized Zirconia (10ScSZ) and 1wt%Ceria doped ScSZ (1Ce10ScSZ) prepared via co-precipitation route were characterized and compared to commercially available samples regarding their transport properties and electrical conductivity. The results of oxygen isotope experiments show that for Zr-based electrolytes, the rate of heteroexchange is lower than that for Sm-doped ceria. The results of Secondary Ions Mass Spectrometry (SIMS) indicate that all admixed components are present both in the surface layer and the bulk of the studied samples with pronounced segregation on the grain boundary. The highest total conductivity is observed for DKKK sample. In the range of 600-400°C the highest conductivity observed for synthesized nanocrystalline 1Ce10ScSZ sample is explained by the effect of segregated Scandia doped Ceria surface layers.

LOW-TEMPERATURE SYNTHESIS METHODS OF DOPED APATITE-TYPE LANTHANUM SILICATES

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J. Chem. Eng. Japan,
40(13) (2007) pp. 1187-1191.

Al- and Sr-doped apatite-type lanthanum silicates (ATLS) have been prepared using mechanochemical activation (MA) and Pechini (Pe) methods. MA of a stoichiometric mixture in the high-power planetary ball mill provides the formation of a high purity crystalline ATLS at room temperature after 20–35 min activation. The phase composition and local structure of doped ATLS are determined by milling time and the dopant content. 5 min MA of carbonate precursors obtained via Pe method results in the formation of a single-phase ATLS at 900°C.

MECHANOCHEMICAL SYNTHESIS OF SOLID SOLUTIONS BASED ON ZrO_2 AND THEIR ELECTRICAL CONDUCTIVITY

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Glass Phys. Chem.,
33(4) (2007) pp. 393-401.

Zirconia- and scandia-based complex solid solutions predominantly with a monoclinic structure are prepared by mechanochemical synthesis. The dense ceramic materials, which, for the most part, have a cubic structure with grain sizes of 250-400 nm and possess good mechanical properties, are produced by sintering submicron fractions of the powders under relatively mild conditions at temperatures of 1633-1653 K. It is revealed that the powders are characterized by nanostructuring due to the complex composition and the chemical inhomogeneity. This nanostructuring is partially retained upon rapid sintering of the ceramic powders. The nanostructured ceramic materials possess a high low-temperature conductivity, which decreases after annealing. Unlike conventional ceramic materials, the nanostructured ceramic materials have identical activation energies for bulk and grain-boundary electrical conduction. The high-temperature electrical conductivity of the nanostructured ceramic materials is rather low because of the small grain sizes and impurities of the monoclinic phase.

DESIGN OF MULTILAYER CERAMIC MIEC MEMBRANES

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Sep. Sci. Technol.,
42(13) (2007) pp. 2849-2861.

The development of multilayer mixed conducting oxide membranes on porous dead-end tubular mineral-based glass-ceramic supports is presented. Nanopowders of compatible complex perovskites and fluorites as membrane materials were obtained by mechanochemical synthesis. The deposition of oxide ceramic porous and dense layers was carried out by casting of slurries in organic medium, prepared from narrow fractions of agglomerated powders. A linear dependence between the sintering temperature T_s and calculated effective melting point T^*_m for complex perovskites and fluorites was revealed that allowed to propose a simple rule for the synthesis of complex oxides with required T_s. Dead-end tubular supports were produced by casting from selected kaolins with organic/inorganic additives. The misfit in sintering temperatures of porous substrate and MIEC ceramic layers was eliminated by modification of substrate and heavily doping of mixed oxides. The elimination of shrinkage misfit between the porous substrate and ceramic layers was achieved by optimization of operation conditions and compositions of both substrate and ceramics. Almost gas-tight cost efficient catalytic membrane reactors CMRs with large operation surface ~30 cm^2 and good prospects for up-scaling were produced and tested.

Catalysis for Organic Products Synthesis

PREPARATION OF 2-METHYL-1,4-NAPHTHOQUINONE (VITAMIN K_3) BY CATALYTIC OXIDATION OF 2-METHYL-1-NAPHTHOL IN THE PRESENCE OF IRON PHTHALOCYANINE SUPPORTED CATALYST

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Comptes Rendus Chim.,
10(7) (2007) pp. 598-603.

Iron tetrasulfophthalocyanine (FePcS) supported catalyst was prepared by covalent grafting onto amino-modified silica by a novel practical one-pot method using activation of sulfonate groups of FePcS by triphenylphosphine triflate. FePcS/SiO_2 in combination with tBuOOH behaved as an efficient catalyst for the oxidation of 2-methyl-1-naphthol to 2-methyl-1,4-naphthoquinone, vitamin K_3. To optimize the catalytic system, the influence of different reaction parameters on the efficiency of this oxidation has been studied. Vitamin K_3 was obtained with 59% selectivity at 96% conversion of 2-methyl-1-naphthol using only 0.5 mol% of catalyst. ^{18}O labelling experiments indicate a non-radical mechanism for this oxidation.

NEW ROUTES TO VITAMIN K_3

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Catal. Today,
121(1-2) (2007) pp. 58-64.

New clean and efficient methods for the production of 2-methyl-1,4-naphthoquinone (MNQ, menadione, vitamin K_3) based on the oxidation of 2-methyl-1-naphthol (MNL) with cheap and environmentally benign oxidants, aqueous hydrogen peroxide, tert-butyl hydroperoxide and molecular oxygen, using three types of true heterogeneous catalysts, such as hydrothermally stable mesoporous titanium-silicate Ti-MMM-2, silica supported iron phthalocyanine and supported gold nanoparticles, are reported. Advantages and drawbacks of the catalytic systems are discussed. Surprisingly, non catalytic oxidation of MNL with molecular oxygen shows superior selectivity and volume yield of MNQ compared to the known catalytic methods.

HIGHLY EFFICIENT PRODUCTION OF 2,3,5-TRIMETHYL-1,4-BENZOQUINONE USING AQUEOUS H_2O_2 AND GRAFTED Ti(IV)/SiO_2 CATALYST

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Green Chem.,
9 (2007) pp. 731-733.

The oxidation of 2,3,6-trimethylphenol (TMP) with aqueous H_2O_2 over titanium(IV) grafted on commercial mesoporous silica produces 2,3,5-trimethyl-1,4-benzoquinone (TMBQ, vitamin E precursor) with nearly quantitative yield. The grafted catalysts are prepared by a simple, affordable and cheap synthesis methodology, combine excellent activity, very high selectivity to the desired product TMBQ and good recyclability in the TMP oxidation by aqueous H_2O_2. All these allow considering them as perspective heterogeneous catalysts for the clean and sustainable synthesis of TMBQ.

EPR STUDY ON THE MECHANISM OF H_2O_2-BASED OXIDATION OF ALKYLPHENOLS OVER TITANIUM SINGLE-SITE CATALYSTS

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J. Mol. Catal. A: Chem.,
277(1-2) (2007) pp. 185-192.

The selective oxidation of 2,3,6-trimethylphenol (TMP) and 2-methyl-1-naphthol (MNL) with H_2O_2 catalyzed by titanium single-site catalysts, TiO_2–SiO_2 aerogel and mesostructured hydrothermally stable titanium-silicate, Ti-MMM-2, have been studied by means of EPR spectroscopic technique with spin traps. The formation of phenoxyl (naphthoxyl) and hydroxyl radical intermediates during the oxidation process have been detected using 3,5-dibromo-4-nitrosobenzene-sulfonic acid (DBNBS) and 5,5-dimethyl-1-pyrroline-*N*-oxide (DMPO) spin traps, respectively. The character of the EPR spectra of the DBNBS adducts strongly depends on the nature of the organic substrate and the reaction temperature. A compilation of the EPR and by-product studies strongly supports a homolytic oxidation mechanism. Study by DR-UV–vis spectroscopy has confirmed the previously suggested chemical adsorption of phenol on the titanium center.

ACCESS TO FUNCTIONALIZED QUINONES VIA THE AROMATIC OXIDATION OF PHENOLS BEARING AN ALCOHOL OR OLEFINIC FUNCTION CATALYZED BY SUPPORTED IRON PHTHALOCYANINE

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New J. Chem.,
30(12) (2006) pp. 1768-1773.

The controlled oxidation at only one position of compounds with several oxidizable sites, while keeping the other sites intact, has been demonstrated for phenols bearing alcohol or olefinic functional groups. Iron tetrasulfophthalocyanine supported on silica was found to be an efficient catalyst for the preparation of functionalized quinones under mild conditions, with tert-butylhydroperoxide as the oxidant. A novel rapid and mild one-pot procedure for the covalent grafting of iron tetrasulfophthalocyanine onto silica has been developed. The supported catalyst was characterized by chemical analysis, a specific surface study, UV-vis spectroscopy and XPS. A non-radical mechanism for this unusual selective oxidation has been revealed by ^{18}O labelling experiments.

ENANTIOSELECTIVE CHROMATOGRAPHIC RESOLUTION AND ONE-POT SYNTHESIS OF ENANTIOMERICALLY PURE SULFOXIDES OVER A HOMOCHIRAL Zn-ORGANIC FRAMEWORK

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J. Am. Chem. Soc.,
129(43) (2007) pp. 12958-12959.

(R)- and (S)- enantiomers of alkyl aryl sulfoxides can be obtained by chromatographic resolution of the racemic mixtures of the sulfoxides on a microporous homochiral Zn-organic polymer or by simultaneous catalytic oxidation of the corresponding sulfides with H_2O_2 and enantioselective chromatographic resolution of the resulting sulfoxides in a one-pot process.

PREPARATIVE PROCEDURE FOR THE SYNTHESIS OF 4-ALLYLOXYPYRIDINE-2,6-DICARBOXYLIC ACID

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Russ. J. Org. Chem., 43(1) (2007) pp. 156-158.

4-Allyloxypyridine-2,6-dicarboxylic acid (**I**) is used as starting compound in the synthesis of 4-allyloxy-2,6-diacetylpyridine; *N*-arylimines derived from the latter at both carbonyl groups attract considerable interest as ligands for immobilized iron(II) complexes as highly efficient catalysts in the polymerization of ethylene. Kim et al. described a procedure for the synthesis of dicarboxylic acid **I** starting from 4-hydroxypyridine-2,6-dicarboxylic acid monohydrate (**III**) in three steps. The authors have found that acid **I** can be synthesized in two steps and that its yield can be raised to 89% by carrying out alkylation of anhydrous dicarboxylic acid **III** with allyl bromide in the presence of anhydrous potassium carbonate in a mixture of acetone with DMF. The subsequent alkaline hydrolysis of diallyl 4-allyloxypyridine- 2,6-dicarboxylate (**IV**) gives the target product.

DESIGN OF SCHIFF BASE-LIKE POSTMETALLOCENE CATALYTIC SYSTEMS FOR POLYMERIZATION OF OLEFINS. IV. SYNTHESIS OF 2-(ARYLIMINOMETHYL)-PYRROLE AND 7-(ARYLIMINOMETHYL)INDOLE DERIVATIVES CONTAINING CYCLOALKYL SUBSTITUENTS

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Russ. J. Org. Chem.,
43(4) (2007) pp. 571-576.

Reactions of 4,6-substituted 2-cycloalkylanilines with 1*H*-pyrrole-2-carbaldehyde and 1*H*-indole-7-carbaldehyde in methanol in the presence of formic acid gave the corresponding Schiff bases which can be used as ligands for titanium and zirconium complexes.

DESIGN OF SCHIFF BASE-LIKE POSTMETALLOCENE CATALYTIC SYSTEMS FOR POLYMERIZATION OF OLEFINS. V. SYNTHESIS OF SALICYLALDEHYDE IMINE LIGANDS CONTAINING CYCLOALKYL SUBSTITUENTS

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Russ. J. Org. Chem., 43(11) (2007) pp. 1671-1676.

Reactions of substituted cycloalkylanilines with salicylaldehyde, 3-*tert*-butylsalicylaldehyde, and 3,5-di-*tert*-butylsalicylaldehyde in methanol in the presence of formic acid gave a series of the corresponding Schiff bases as ligands for titanium(IV) complexes.

SYNTHESIS OF SALICYLALDEHYDES BEARING BULKY SUBSTITUENTS IN THE POSITIONS 3 AND 5

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Russ. Chem. Bull.,
56(6) (2007) pp. 1125-1129.

Reaction of 2,4-disubstituted phenols with paraformaldehyde in the presence of $SnCl_4$ and 2,6-lutidine afforded a number of new salicylaldehydes, containing bulky substituents (*tert*-butyl, 1-phenylethyl, 1-(4-*tert*-butylphenyl)ethyl, α-cumyl, and trityl) in the positions 3 and 5.

SYNTHESIS OF DIIMINE LIGANDS WITH CYCLOALKYL SUBSTITUENTS BASED ON 4,6-DIBENZOFURAN-AND 4,6-DIBENZO-THIOPHENEDICARBOXALDEHYDES

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Russ. Chem. Bull.,
56(6) (2007) pp. 1174-1177.

A formic acid-catalyzed reaction of substituted cycloalkylanilines with 4,6-dibenzofuran- and 4,6-dibenzothiophenedicarboxaldehydes in methanol-dichloromethane mixture afforded a number of the corresponding diimines, which can be of interest as components of catalytic systems for polymerization of olefins.

HYDROGENOLYSIS OF DIMETHYL DISULFIDE IN THE PRESENCE OF BIMETALLIC SULFIDE CATALYSTS

A.V. Mashkina, L.N. Khairulina

Kinet. Catal., 48(1) (2007) pp. 125-131.

The hydrogenolysis of dimethyl disulfide in the presence of Ni,Mo and Co,Mo bimetallic sulfide catalysts was studied at atmospheric pressure and T = 160-400°C. At $T \leq 200°C$, dimethyl disulfide undergoes hydrogenolysis at the S–S bond, yielding methanethiol in 95–100% yield. The selectivity of the reaction decreases with increasing residence time and temperature due to methanethiol undergoing condensation to dimethyl disulfide and hydrogenolysis at the C–S bond to yield methane and hydrogen sulfide. The specific activity of the $Co,Mo/Al_2O_3$ catalyst in hydrogenolysis at the S–S and C–S bonds is equal to or lower than the total activity of the monometallic catalysts. The $Ni,Mo/Al_2O_3$ catalyst is twice as active as the Ni/Al_2O_3 + Mo/Al_2O_3 or the cobalt–molybdenum bimetallic catalyst.

SUPERACIDIC ACTIVATION OF QUINOLINE AND ISOQUINOLINE; THEIR REACTIONS WITH CYCLOHEXANE AND BENZENE

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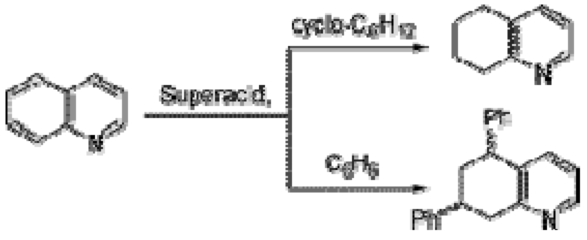
J. Org. Chem,
72(19) (2007) pp. 7394-7397.



Quinoline (**1**) and isoquinoline (**2**), upon activation by strong acids, lead to intermediate N,C-diprotonated dications, which are involved in reactions with weak nucleophiles. Thus, **1** and **2** undergo selective ionic hydrogenation with cyclohexane in CF_3SO_3H-SbF_5, HBr-$AlBr_3$-CH_2Br_2, or HCl-$AlCl_3$-CH_2Cl_2 acid systems to give their 5,6,7,8-tetrahydro derivatives. They also readily condense with benzene in the presence of HBr-$AlBr_3$ or HCl-$AlCl_3$ to provide 5,6,7,8-tetrahydro-5,7-diphenylquinoline and 5,6,7,8-tetrahydro-6,8-diphenylisoquinoline, respectively.

SUPERACIDIC AND HUSY-ZEOLITE ACTIVATION OF 1,3-INDANDIONE: REACTIONS WITH BENZENE AND CYCLOHEXANE

K.Yu. Koltunov

Tetrahedron Lett.,
48(32) (2007) pp. 5631-5634.

1,3-Indandione readily condenses with benzene and undergoes selective ionic hydrogenation with cyclohexane when activated by superacids, such as CF_3SO_3H, $AlCl_3$ and $AlBr_3$ to give 3,3-diphenyl-1-indanone and 1-indanone, respectively. Combination of these reactions in 'one-pot' yields 3-phenyl-1-indanone. In addition, similar reactions have been carried out using the regenerable solid acid, HUSY-zeolite, providing an effective excess of acidic sites. The mechanism of these reactions, with potential involvement of superelectrophilic dicationic intermediates, is discussed.

2,2′-BIPYRIDINE AND RELATED *N*-CHELANTS AS VERY EFFECTIVE PROMOTERS FOR Cu CATALYSTS IN THE DECARBOXYLATION

A.S. Lisitsyn

Appl. Catal., A, 332(1) (2007) pp. 166-170.

Effect of 2,2′-bipyridine (bipy) and analogous ligands on decarboxylation of 2,4-dimethylbenzoic (DMBA) and 4-hydroxybenzoic acids with Cu catalysts has been compared with that of quinoline. The chelating ligands were shown to exceed the traditional promoter in efficiency by two to three orders of magnitude per a mol basis. In combination with bipy-like promoters, Cu catalysts ensured complete conversion of even the low reactive DMBA for 15–30 min and gave expected product in >95% yield (S/C 40–100, Ph_2O as solvent, 250°C). In the decarboxylation of individual aromatic acids, the (Cu + bipy) system greatly surpassed carbon and Pd/C catalysts as well, but lost activity in solutions of natural acidic mixtures. The later was assigned to reduction of Cu ions by H-donor constituents of such mixtures.

POSSIBLE PREBIOTIC SYNTHESIS OF MONOSACCHARIDES FROM FORMALDEHYDE IN PRESENCE OF PHOSPHATES

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Adv. Space Res.,
40(11) (2007) pp. 1634-1640.

Condensation of formaldehyde and lower carbohydrates (glycolaldehyde, glyceraldehyde, and dihydroxyacetone) is effectively catalyzed by heterogeneous and homogeneous phosphates in neutral aqueous medium. The interaction of formaldehyde and dihydroxyacetone leads to the preferential formation of 3-pentulose and erythrulose with yields of 40% and 45%, respectively. In absence of formaldehyde, the condensation of glycolaldehyde and glyceraldehyde catalyzed by phosphates leads to the formation of ribose and fructose. The possibility of formation of higher monosaccharides from pure formaldehyde in the course of the combined photochemical and phosphate-catalyzed reactions in plausible prebiotic conditions was demonstrated.

ABIOGENIC SYNTHESIS OF PREBIOTIC MATTER FOR THE EARTH'S BIOSPHERE AS A STAGE OF SELF-ORGANIZATION ON AN ASTROPHYSICAL AND PALEONTOLOGICAL TIME SCALE

V.N. Snytnikov

Paleontological J.,
41(5) (2007) pp. 473-480.

Existing data suggest that an early circumstellar preplanetary disk was the most likely location for primary abiogenic synthesis of prebiotic organic matter from simple molecules along with the "RNA world" and the origin of life. This paper discusses the stages of self-organization that have resulted in the Earth's modern biosphere, and the relationships between astrophysical and paleontological events in evolution.

SELECTIVE SYNTHESIS OF ERYTHRULOSE AND 3-PENTULOSE FROM FORMALDEHYDE AND DIHYDROXYACETONE CATALYZED BY PHOSPHATES IN A NEUTRAL AQUEOUS MEDIUM

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Kinet. Catal., 48(4) (2007) pp. 550-555.

The aldol condensation of formaldehyde and the lower carbohydrate dihydroxyacetone in a neutral aqueous medium is effectively catalyzed by solid compounds (hydroxylapatite and calcium phosphate and carbonate), natural minerals (apatite and vivianite), and soluble phosphates. In excess formaldehyde, the decrease in the concentration of the lower carbohydrate is described by a first-order rate law with respect to dihydroxyacetone. The major products of the reaction between formaldehyde and dihydroxyacetone in the presence of the above catalysts are erythrulose (45–50% selectivity) and 3-pentulose (35–40% selectivity). Branched pentulose and hexulose are also identified among the reaction products.

Photocatalytic and Related Processes

PHOTOCATALYTIC OXIDATION OF 1,1-DIMETHYL HYDRAZINE VAPOURS ON TiO_2: FTIR *IN SITU* STUDIES

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Catal. Today, 122(1-2) (2007) pp. 178-185.

Gas-phase photocatalytic oxidation (PCO) of unsymmetrical dimethyl hydrazine (UDMH) in a batch reactor using TiO_2 as the photocatalyst was studied with *in situ* FTIR method. Carbon dioxide, water, nitric acid and nitrogen were detected as the ultimate PCO products of UDMH. Adsorbed N_2O species were detected as the main surface intermediates. The formation of the extremely toxic intermediate, nitrosodimethylamine $(CH_3)_2NNO$, was not observed. The reaction mechanism based on detected products distribution, kinetics analysis and previously published data is discussed.

Long-term experiments were carried out to determine the photocatalyst stability in PCO of UDMH: the catalyst exhibited stable performance; the rate of deactivation was low due to transformation of nitrogen mainly to N_2. Only 10% of UDMH nitrogen was transformed to HNO_3 in adsorbed form.

PHOTOCATALYTIC OXIDATION OF VX-SIMULATION SUBSTANCE

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Water Sci. Techn., 55(12) (2007) pp. 133-138.

Experimental studies of photocatalytic oxidation (PCO) by air oxygen of VX-gas simulation substance cysteamine-S-phosphate sodium salt ($NaHPO_3S-CH_2-CH_2-NH_2$, CPSS) at different initial concentrations and pH were undertaken. PCO ultimately resulted in complete mineralization of CPSS. The PCO by-products of CPSS include acetate, oxalate and trace amounts of formate ions. The formation rates of acetate and phosphate were equal to the rate of degradation of CPSS, which indicates easy breakage of P-S, C-S and C-N bonds. Sulphate was formed slower due to stepwise oxidation of reduced sulphur. Amino group, generally transformed to ammonia, was partially oxidised to nitrite and nitrate in alkaline media. The fastest mineralisation in terms of both TOC degradation and phosphate formation was observed at pH 9.5. Under neutral media conditions, the PCO rate increased linearly with the CPSS concentration increase. The maximum efficiency by TOC degradation was observed as big as 77 mg per Wh of incident UV flux.

PHOTOCATALYTIC AIR CLEANING FROM AUTOMOTIVE CONTAMINANTS

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Catal. Ind.,
6 (2006) pp. 19-28.

The photocatalytic method of air cleaning in internal spaces of motor vehicles from emissions is considered. A new photo-catalytic air cleaner is described. It consists of UV-lamp of 36 W in capacity and photocatalyst IK 12-31 applied on platelike packaged carriers. The photocatalyst contains photoactive TiO_2 (anatase, $S_{BET} = 340$ m^2/g) and high-dispersed platinum in amount of 0,2 wt %. The photocatalytic air cleaner was tested in the process of air cleaning from CO, NO_x, SO_2 as well as acetone, heptane, acetaldehyde, and toluene vapors in a closed chamber simulating the passenger compartment. It provides the air purification from indicated substances by oxidizing the organic substances and CO into CO_2, and NO_x and SO_2 into corresponding acids adsorbed on the catalyst surface. The quantum efficiency of oxidation varies within 2÷28 % for different substances at initial contaminant concentration 70-800 mg/m^3. As compared with the less efficient adsorption methods, it can reach deep cleaning from CO and light hydrocarbons. When operating with CO and hydrocarbons, the photocatalytic filter regeneration is not required and its time of operation is restricted by the operational life of the lamp (1-2 years). A conclusion is made on efficiency of photocatalytic filter application for air cleaning in passenger compartments of all types of vehicles. Implementation and efficient use of the photocatalytic filters requires integration of ventilation into the system, this can be obtained in joint developments with the manufacturers of vehicles.

OPPOSITE EFFECT OF GAS PHASE H_2O_2 ON PHOTOCATALYTIC OXIDATION OF ACETONE AND BENZENE VAPORS

A.V. Vorontsov

Catal. Commun.,
8 (2007) pp. 2100-2104.

The effect of additions of gas phase H_2O_2 was measured for gas phase photocatalytic oxidation of organic vapors. Photocatalytic oxidation of benzene vapor over TiO_2 in a flow reactor resulted in a quick catalyst deactivation. Additions of gas phase H_2O_2 into the reactor feed provided enhanced and sustained oxidation of benzene vapor. The increase of inlet H_2O_2 vapor concentration from 0 to about 1000 ppm led to the one order of magnitude growth of benzene vapor complete oxidation rate. The highest rate of 1.1 nmol/s was observed at C_6H_6 concentration 124 ppm and H_2O_2 concentration 1000 ppm. In the case of acetone vapor photocatalytic oxidation, the rate of complete oxidation in the flow reactor decreased with an increase of gas phase H_2O_2 inlet concentration. TiO_2 Degussa P25 provided higher oxidation rate in the presence of H_2O_2 than pure anatase TiO_2.

INFLUENCE OF MESOPOROUS AND PLATINUM MODIFIED TITANIUM DIOXIDE PREPARATION METHODS ON PHOTOCATALYTIC ACTIVITY IN LIQUID AND GAS PHASE

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77(1-2) (2007) pp. 35-45.

Mesoporous titanium dioxide photocatalysts have been synthesized by template method using widely used precursors and templates with the purpose of choosing the best method for liquid and gas phase photocatalytic oxidation. Titanyl sulphate (TS) and tetrabutyl titanate (TBT) were precursors and cetyl trimethylammonium bromide (CTAB) and dodecylamine (DDA) were templates. Hydrothermal treatment temperature was optimized for the best obtained photocatalyst. The catalysts were characterized by N_2 adsorption–desorption measurements, XRD and UV–vis diffuse reflectance spectroscopy. Samples obtained from TBT contained significant amounts of brookite. Activity was measured in liquid phase for dimethyl methylphosphonate (DMMP) and in gas phase for acetone complete oxidation. The highest activity in liquid phase was observed for catalyst prepared from TS and DDA and aged at 160°C. Activity was higher than that of TiO_2 Degussa P25 in the low concentration range. The highest activity in gas phase was observed for catalyst prepared from TBT and CTAB. Activity of mesoporous catalysts correlates with their surface area and pore volume for gas phase reaction, whereas in the case of liquid phase DMMP oxidation, there is no sufficient correlation. The effect of DMMP initial concentration on initial oxidation rate is well fitted by Langmuir–Hinshelwood competitive adsorption model. Mesoporous catalysts possess higher adsorption constant but lower reaction rate constant than TiO_2 P25. Platinum was deposited by impregnation and soft chemical reduction (SCR) or photodeposition methods (PD). Catalysts obtained by PD were 50% more active than by SCR and 4.5-fold more active than TiO_2 P25 in aqueous phase. The reasons for extraordinarily high activity of this mesoporous catalyst are discussed.

ACCELERATION OF ACETONE DESTRUCTION PROCESS UNDER SYNERGISTIC ACTION OF PHOTOCATALYTIC OXIDATION AND BARRIER DISCHARGE

A.S. Besov, A.V. Vorontsov

Plasma Chem. Plasma Process.,
27(5) (2007) pp. 624-634.

Separate and joint work of photocatalytic oxidation based on TiO_2 and surface barrier discharge as a source of ozone and UV radiation is investigated for acetone vapors destruction under ambient air conditions. Experiments were carried out in 404 l airtight plexiglas chamber and used five different combinations of photocatalyst, barrier discharge and ultraviolet irradiation under identical initial concentration (~ 170 ppm) of acetone. It is shown for synergistic action of photocatalytic method and barrier discharge that the initial rate of acetone decomposition per watt of input power increases in more than 1.5 times compared to the most effective case of photocatalytic oxidation alone. It has been determined that under operation of plasma of barrier discharge intermediate products such as CO, acetic acid, acetaldehyde and methane nitrate (CH_3NO_3) are formed. These products are not detected for photocatalytic oxidation. Some features of kinetics of acetone decomposition and formation of intermediate products are revealed.

EFFECT OF $TiOSO_4$ HYDROTHERMAL HYDROLYSIS CONDITIONS ON TiO_2 MORPHOLOGY AND GAS-PHASE OXIDATIVE ACTIVITY

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Res. Chem. Intermed.,
33(3-5) (2007) pp. 449-464.

The effect of synthesis conditions on morphology and catalytic activity has been studied for hydrothermal preparation of TiO_2 from acidified aqueous $TiOSO_4$ solution. It was found that the increase in $TiOSO_4$ and H_2SO_4 concentration results in the increase of photocatalytic activity of produced TiO_2, as it was revealed by steady-state gas-phase oxidation of acetone and ethanol vapors in a flow-circulating reactor. $TiOSO_4$ concentration exerts strong influence on the shape of the produced TiO_2 particles. At $TiOSO_4$ concentration less than 0.1 wt%, hydrolysis gives rise to hedgehog-like agglomerates consisting of spliced-blade TiO_2 nanocrystals. At higher concentration of $TiOSO_4$, the resultant TiO_2 consists of round agglomerates of 5-10 nm primary particles. The size of secondary particles depends mainly on the H_2SO_4 concentration. The increase of the time of hydrothermal treatment results in the enhancement of TiO_2 photocatalytic activity that reaches a maximum. Among different acids (HCl, $HClO_4$, HNO_3, H_3PO_4 and CH_3COOH) added during hydrolysis of $TiOSO_4$, sulfuric and acetic acids had the best effect on photocatalytic activity of TiO_2. The results obtained can help to finely tune this TiO_2 preparation method in order to obtain desirable particles size, shape and activity.

EXAFS STUDY AND PHOTOCATALYTIC PROPERTIES OF UN-DOPED AND IRON-DOPED ZrO_2-TiO_2 (PHOTO-) CATALYSTS

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Catal. Today,
128(3-4) (2007) pp. 245-250.

The local zirconium and iron arrangements of the iron-doped ZrO_2-TiO_2 system, prepared by sol–gel impregnation method, were studied by EXAFS spectroscopy. Only a tetragonal ZrO_2 structure is located on TiO_2 surface. For the iron-doped ZrO_2-TiO_2 system, the presence of the Fe-O-Fe species as well as and Fe-O-Zr species located on the surface/pre-surface region are shown; it seems that iron is heterogeneously distributed, forming small iron oxide nanoclusters and Fe_x/ZrO_2 (tetragonal) spots at the catalyst surface. The photocatalytic activity of the un-doped and iron-doped binary system ZrO_2-TiO_2 was investigated in two kind of photoreactions: the salicylic acid photooxidation and the photocatalytic reduction of Cr(VI). Different photocatalytic behaviour has been found for the un-doped and iron-doped ZrO_2-TiO_2 systems which have been explained in terms of the EXAFS study.

This study represents an example of attempt to prepare a new potential photoactive mixed oxide system, containing two ions (Ti^{4+} and Zr^{4+}) with good photocatalytic activity if it is compared with commercial TiO_2 (Degusssa P25) calcined at 600°C.

STRUCTURAL PECULIARITIES OF TiO_2 AND Pt/TiO_2 CATALYSTS FOR THE PHOTOCATALYTIC OXIDATION OF AQUEOUS SOLUTION OF ACID ORANGE 7 DYE UPON ULTRAVIOLET LIGHT

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Appl. Catal. B, 71(3-4) (2007) pp. 169-176.

TiO_2 (anatase) with different microstructure was synthesized by thermal hydrolysis of the titanyl sulfate and studied by X-ray powder diffraction, high resolution transmission electron microscopy and UV–vis diffuse reflectance spectroscopy. The effect of titanium dioxide structure, regular or distorted, on the photocatalytic degradation of Acid Orange 7 Dye (AO7) in water upon ultraviolet light was studied. It was found that synthesized TiO_2 possesses a relatively high reactivity when illuminated but also show different adsorption in the dark. The relationship between these behaviors depends on the real structure of the catalysts. Catalysts with a perfect structural ordering formed after heating at temperature higher than 500°C show better photocatalytic performance. Small amount of Pt added into the TiO_2 structure was found to improve further the catalyst reactivity. Pt-modified titania catalysts oxidize AO7 more efficiently than P-25 Degussa TiO_2. Doping effect of Pt on the structural and photocatalytic properties of the samples is discussed.

Polymerization Catalysts and Polymer Materials

POST-METALLOCENE CATALYSTS FOR OLEFIN POLYMERISATION

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Russ. Chem. Rev., 76(3) (2007) pp. 253-277.

The main types of post-metallocene catalysts for olefin polymerisation based on bis(imino), bis(imino)pyridyl, bis(phenoxyimino), bis(pyrrolylimino) and other complexes of transition metals developed in the last 10–15 years and having prospects for practical use are considered. Modern views on the mechanism of action of these catalysts are discussed.

ACTIVATION OF BIS(PYRROLYLALDIMINATO) AND (SALICYLALDIMINATO) (PYRROLYLALDIMINATO)TITANIUM POLYMERIZATION CATALYSTS WITH METHYLALUMOXANE

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Organomet., 26(2) (2007) pp. 288-293.

Cationic intermediates formed upon activation of olefin polymerization catalyst based on bis[N-phenyl-pyrrolylaldiminato]titanium(IV) dichloride (L_2TiCl_2, I) and [N-(3-tert-butylsalicylidene)-2,3,4,5,6-pentafluoroanilinato-N′-phenyl-pyrrolylaldiminato]titanium(IV) dichloride ($L'LTiCl_2$, II) with methylalumoxane (MAO) have been identified. Outer sphere ion pairs of the type $[L_2TiMe(S)]^+[MeMAO]^-$ and $[L'LTiMe(S)]^+[MeMAO]^-$ capable of ethene polymerization have been characterized by ^1H and ^{13}C NMR spectroscopy. Unlike methyl metallocenium cations, the barrier of the first ethene insertion into the Ti-Me bonds of these species is not significantly higher than that of subsequent insertions. Surprisingly, whereas homoligated catalyst precursors L_2TiCl_2 in the presence of MAO are prone to ligand transfer to aluminum, under the same conditions the heteroligated system $L'LTiCl_2$/MAO proved resistant to ligand scrambling.

ACTIVATION OF BIS(PHENOXYIMINO)ZIRCONIUM POLYMERIZATION CATALYSTS WITH METHYLALUMINOXANE AND $AlMe_3/[CPh_3]^+[B(C_6F_5)_4]^-$

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Organomet., 26(19) (2007) pp. 4810-4815.

The intermediates of olefin polymerization over homogeneous catalysts based on bis[*N*-(3-*tert*-butylsalicylidene)anilinato]zirconium(IV) dichloride $\{(L^{tBu})_2ZrCl_2\}$ (**1-^{t}Bu**), bis[*N*-(3-methylsalicylidene)anilinato]zirconium(IV) dichloride $\{(L^{Me})_2ZrCl_2\}$ (**1-Me**), and bis[*N*-(salicylidene)anilinato]zirconium(IV) dichloride $\{(L^H)_2ZrCl_2\}$ (**1-H**) with different activators {methylaluminoxane (MAO) and $AlMe_3/[CPh_3]^+[B(C_6F_5)_4]^-$} have been studied by ^1H and ^{13}C NMR spectroscopy. Heterobinuclear ion pairs

$[(L^{tBu})_2Zr(\mu\text{-}Me)_2AlMe_2]^+[Me\text{-}MAO]^-$ (**2-^{t}Bu**) and $[(L^{tBu})_2Zr(\mu\text{-}Me)_2AlMe_2]^+[B(C_6F_5)_4]^-$ (**2'-^{t}Bu**) are formed upon activation of **1-^{t}Bu** with MAO and $AlMe_3/[CPh_3]^+[B(C_6F_5)_4]^-$, respectively. These species are the precursors of the highly active intermediates of polymerization. In contrast, the activation of **1-Me** with MAO results mainly in the formation of the tight ion pair $[(L^{Me})_2ZrMe^+\cdots Me\text{-}MAO^-]$. This ion pair is the precursor to a poorly active intermediate of polymerization. In the catalytic systems **1-H**/MAO and **1-H**/$AlMe_3/[CPh_3]^+[B(C_6F_5)_4]^-$, the aluminum ion pairs $[L^HAl(\mu\text{-}Me)(\mu\text{-}Cl)AlMe_2]^+[Me\text{-}MAO]^-$ and $[L^HAl(\mu\text{-}Me)(\mu\text{-}Cl)AlMe_2]^+[B(C_6F_5)_4]^-$, which were inactive in ethylene polymerization, are predominantly formed. The isomeric composition of **2'-^{t}Bu** is close to that of the initial complex **1-^{t}Bu**. The main route of the **1-^{t}Bu**/MAO catalyst deactivation is the ligand transfer to aluminum with the formation of the complex $L^{tBu}AlMe_2$.

KEY INTERMEDIATES IN METALLOCENE- AND POST-METALLOCENE-CATALYZED POLYMERIZATION

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Kinet. Catal., 48(4) (2007) pp. 490-504.

The structures of intermediates formed upon the activation by methylaluminoxane (MAO) of a wide range of metallocene and post-metallocene catalysts of olefin polymerization were studied by ^{13}C, ^1H, and ^{19}F NMR. For all metallocenes considered (L_2ZrCl_2 and L_2TiCl_2), under conditions similar to real polymerization conditions (Al/Zr > 200), two types of intermediates were identified in the reaction solution, namely, hetero-dinuclear ion pairs $[L_2\mathbf{M}(\mu\text{-}Me)_2AlMe_2]^+[Me\text{-}MAO]^-$ (**III**) and zwitterionic intermediates $L_2\mathbf{M}Me^+\leftarrow Me\text{-}Al^-\equiv MAO$ (**IV**) (**M** = Zr, Ti). The relative concentration of **III** increases with an increase in the Al/Zr ratio. In the post-metallocene/MAO catalytic systems, the reaction solution can be dominated either by heterodinuclear pairs of type **III** (bis(imino)pyridyl iron complexes) or by zwitterionic intermediates of type **IV** (half-titanocenes, complexes with restricted geometry). Both species **III** and species **IV** catalyze olefin polymerization. Both the species initiating polymerization, $[L'_2TiMe(S)]^+[Me\text{-}MAO]^-$, and the species responsible for chain growth, $[L\ [L'_2TiP]^+[Me\text{-}MAO]^-$ (P is the polymer chain, and S is a solvent molecule), were characterized in the bis(phenoxyimine) titanium complex/MAO system.

FORMATION AND STRUCTURES OF CATIONIC ZIRCONIUM COMPLEXES IN TERNARY SYSTEMS Rac-(SBI)ZrX$_2$/AlBu$^i_3$/[CPh$_3$][B(C$_6$F$_5$)$_4$] (X= Cl, Me)

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J. Organomet. Chem., 692(4) (2007) pp. 859-868.

Using ^{13}C, ^1H and ^{19}F NMR spectroscopy, formation of cationic species was studied in ternary systems $(SBI)ZrX_2/AlBu^i_3/[CPh_3][B(C_6F_5)_4]$, where X = Cl, Me [(SBI) = *rac*-$Me_2Si(Ind)_2$]. In the first system (X = Cl), the ion pair $[(SBI)Zr(\mu\text{-}Cl)_2Zr(SBI)][B(C_6F_5)_4]_2$ (**IV**) predominates at low Al/Zr ratios (Al/Zr < 10), whereas at higher Al/Zr ratios (≥20) in the absence of monomer mainly $[(SBI)Zr(\mu\text{-}H)(\mu\text{-}C_4H_7)\ AlBu^i_2][B(C_6F_5)_4]$ (**V**) is formed. The binuclear complex $[(SBI)Zr(\mu\text{-}Cl)_2Zr(SBI)][B(C_6F_5)_4]_2$ has been characterized crystallographically. Species **V** is also formed in the system X = Me at high Al/Zr ratios. In the presence of $AlBu^i_3$, **IV** displays activity in propylene polymerization and is the most likely precursor of the polymerizing species. Consistent mechanisms have been proposed for the reactions in these catalytic systems.

STATE OF VARIOUS STEREOREGULATING ELECTRON-DONATING COMPOUNDS IN TITANIUM-MAGNESIUM CATALYSTS FOR PROPYLENE POLYMERIZATION: A DIFFUSE REFLECTANCE IR SPECTROSCOPIC STUDY

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Kinet. Catal., 48(3) (2007) pp. 403-408.

Propylene polymerization on $TiCl_4$/donor/$MgCl_2$ (donor = ethyl benzoate, dibutyl phthalate, diisobutyl phthalate, diethyl 2,3-diisopropylsuccinate) supported catalysts is considered. The states of the donors in the catalysts have been investigated by diffuse reflectance IR spectroscopy. Data characterizing the distribution of the donors and the active component ($TiCl_4$) on the support surface have been obtained. Molecular weight distribution data for polypropylene are presented. The molecular weight distribution of polypropylene depends on the location of the donor and $TiCl_4$ molecules.

Cu(II) COMPLEXES WITH 4,6-BIS (3,5-DIMETHYL-1*H*-PYRAZOLE-1-YL)PYRIMIDINE, 4-(3,5-DIMETHYL-1*H*-PYRAZOLE-1-YL)-6-(3,5-DIPHENYL-1*H*-PYRAZOLE-1-YL)PYRIMIDINE: SYNTHESIS AND CATALYTIC ACTIVITY IN ETHYLENE POLYMERIZATION REACTION

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Russ. J. Coord. Chem.,
33(8) (2007) pp. 601-607.

The Cu(II) complexes with 4,6-bis(3,5-dimethyl-1*H*-pyrazole-1-yl)pyrimidine (L^1) and 4-(3,5-dimethyl-1*H*-pyrazole-1-yl)-6-(3,5-diphenyl-1*H*-pyrazole-1-yl)pyrimidine (L^2) of the composition $Cu_2L^1Br_4$ and $Cu_2L^2A_4$ (A = Cl, Br), respectively, were synthesized and studied by IR and magnetochemical methods. The molecular structure of the complexes is likely to be binuclear. In the presence of cocatalysts methylalumi-noxane and triisobutylaluminium, the title complexes exhibit catalytic activity in the ethylene polymerization reaction.

COBALT(II) AND COPPER(II) COMPLEXES WITH CHIRAL PYRAZOLYLQUINOLINE, A DERIVATIVE OF TERPENOID (+)-3-CARENE. CATALYTIC ACTIVITY IN ETHYLENE POLYMERIZATION REACTION

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Russ. J. Coord. Chem.,
33(5) (2007) pp. 436-448.

Coordination compounds $[CoLCl_2]$ (**I**), $[CuLCl(NO_3)]$ (**II**), $CuL(NO_3)_2$ (**III**), and $CuLCl_2$ (**IV**) (where L is a chiral pyrazolylquinoline - a derivative of terpenoid (+)-3-carene) were synthesized. X-ray diffraction data showed that crystal structures **I** and **II** are built of mononuclear acentric molecules. In the molecule of complex I, the Co^{2+} ion coordinates two N atoms of bidentate cycle-forming ligand L and two Cl atoms. The coordination polyhedron of Cl_2N_2 is a distorted tetrahedron. For complex **I**, μ_{eff} = 4.50 μ_B, which corresponds to a high-spin configuration d^7. In the molecules of II(1), II(2) (which are diastereoisomers of complex II), each Cu^{2+} ion coordinates two N atoms of bidentate cycle-forming ligand L, the Cl atom, and two O atoms of bidentate cyclic NO_3^- ion. The ClN_2O_2 coordination polyhedra are tetragonal pyramids with different degrees of distortion. The structure of complex **II** consists of supramolecular clusters, i.e., isolated chains incorporating the molecules of **II**(1) and **II**(2). The values of μ_{eff} for **II**–**IV** correspond to the d^9 configuration. The results of EPR and IR study suggest that complex **III** contains the O_4N_2 polyhedron, whereas complex **IV** contains the Cl_2N_2 polyhedron. Complexes **I** and **IV** were found to show a high catalytic activity in ethylene polymerization reaction.

KINETIC FEATURES OF ETHYLENE POLYMERIZATION OVER SUPPORTED CATALYSTS [2,6-BIS(IMINO)PYRIDYL IRON DICHLORIDE/MAGNESIUM DICHLORIDE] WITH ALR$_3$ AS AN ACTIVATOR

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J. Polym. Sci., Part A: Polym. Chem.,
45(22) (2007) pp. 5057-5066.

The effects of polymerization temperature, polymerization time, ethylene and hydrogen concentration, and effect of comonomers (hexene-1, propylene) on the activity of supported catalyst of composition $LFeCl_2/MgCl_2$-$Al(i\text{-}Bu)_3$ (L=2,6-bis[1-(2,6-dimethylphenylimino)ethyl] pyridyl) and polymer characteristics (molecular weight (MW), molecular-weight distribution (MWD), molecular structure) have been studied. Effective activation energy of ethylene polymerization over $LFeCl_2/MgCl_2$-$Al(i\text{-}Bu)_3$ has a value typical of supported Ziegler-Natta catalysts (11.9 kcal/mol). The polymerization reaction is of the first order with respect to monomer at the ethylene concentration >0.2 mol/L. Addition of small amounts of hydrogen (9-17%) significantly increases the activity; however, further increase in hydrogen concentration decreases the activity. The IRS and DSC analysis of PE indicates that catalyst $LFeCl_2/MgCl_2$-$Al(i\text{-}Bu)_3$ has a very low copolymerizing ability toward propylene and hexene-1. MW and MWD of PE produced over these catalysts depend on the polymerization time, ethylene and hexene-1 concentration. The activation effect of hydrogen and other kinetic features of ethylene polymerization over supported catalysts based on the Fe (II) complexes are discussed.

ADVANCES IN THE DEVELOPMENT OF NEW CATALYSTS FOR ETHYLENE AND α-OLEFIN POLYMERIZATION

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Russ. Chem. Rev.,
76(7) (2007) pp. 617-637.

The role of catalyst systems in the perfection of polymerisation processes and the improvement of the performance characteristics of polyethylene and other polyolefins is considered. The scope of application of post-metallocene catalysts based on transition metal α-diimine, bis(imino)pyridine and phenoxy imine complexes designed in the last decade is analysed. The structures of complexes depending on the type of ligands are described systematically. The development of research into these catalyst systems is discussed, the optimal structures as regards the activity and selectivity are mentioned, and the mechanism of their action and the prospects for industrial use are demonstrated.

ORGANIC-INORGANIC CROSS-LINKED STRUCTURES PREPARED FROM REACTIVE *n*-BUTYL METHACRYLATE-3-(TRIMETHOXYSILYL)PROPYL METHACRYLATE COPOLYMERS

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Russ. J. Appl. Chem.,
80(1) (2007) pp. 93-101.

The features of formation of organic-inorganic cross-linked structures prepared by copolymerization of *n*-butyl methacrylate with 3-(trimethoxysilyl)propyl methacrylate, followed by hydrolysis of the trimethoxysilane groups of the copolymers and condensation of the resulting silanol groups, were studied. The quantitative composition of the functional groups of the cross-linked copolymers was determined. The physicomechanical and mechanical properties of the copolymers were studied in relation to the copolymer composition and conditions of hydrolytic condensation.

STYRENE-ACRYLATE COPOLYMER PLASTISOLS WITH STABLE COLLOIDAL PROPERTIES

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Polymer Sci., ser. A,
49(10) (2007) pp. 1086-1092.

Methods for the synthesis of emulsion-polymerized styrene–methyl methacrylate–methacrylic acid copolymers with different compositions and structures are proposed. Stable plastisols are obtained from these copolymers and phthalate plasticizers. Factors that affect the colloidal stability and rheological properties of the plastisols were studied. These are the polymer particle size, the type of surfactants used in emulsion copolymerization, the distribution of methacrylic acid units over the latex particle volume (uniform or gradient, with concentration rising from the particle center to periphery), the degree of carboxyl group neutralization with a base, and the nature of plasticizers. The plastisols obtained are characterized by a pseudoplastic flow. However, as the degree of carboxyl group neutralization in the copolymers is increased, the rheological properties of the plastisols approach those of Newtonian liquids.

CATALYTIC ACTIVITY OF SYSTEMS BASED ON TITANIUM BIS(PHENOXY IMINE) COMPLEXES: EFFECT OF THE LIGAND STRUCTURE

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Russ. J. Appl. Chem.,
80(9) (2007) pp. 1515-1522.

Polymerization of ethylene at 30–70°C in the presence of catalytic systems based on titanium bis(phenoxy imine) complexes of various structures, activated with methylalumoxane, was studied. An attempt was made to systematize the effect of substituents in ligands, to find correlations, to account for the results obtained, and to reveal conditions under which the reaction occurs by the living polymerization mechanism. Some properties and structural features of the polyethylenes obtained were determined.

FEATURES OF SELF-IMMOBILIZATION OF TITANIUM PHENOXYIMINE COMPLEXES IN ETHYLENE POLYMERIZATION

N.I. Ivancheva, M.Yu. Malinskaya, I.I. Oleinik*, S.Ya. Khaikin, G.A. Tolstikov* (**Vorozhtsov Novosibirsk Institute of Organic Chemistry, Novosibirsk, Russia*)

Doklady Phys. Chem.,
417(1-3) (2007) pp. 301-304.

Kinetic features of self-immobilization of catalytic complex over polyethylene obtained and kinetics of ethylene polymerization are illustrated by the example of methylaluminoxane-activated titanium bis(phenoxy imine) complexes modified with oxyallylic groups. The activity of catalyst self-immobilized over polyethylene is for the first time studied and compared with similar by composition homogeneous catalyst.

ETHYLENE POLYMERIZATION ON TITANIUM PHENOXYIMINE COMPLEXES WITH DIFFERENT STRUCTURES

N.I. Ivancheva, M.Yu. Malinskaya, S.S. Ivanchev, I.I. Oleinik*, A.I. Kochnev*, G.A. Tolstikov* (**Vorozhtsov Institute of Organic Chemistry, Novosibirsk, Russia*)

Kinet. Catal.,
48(6) (2007) pp. 829-834.

The kinetics of ethylene polymerization in the presence of catalytic systems based on methylaluminoxane-activated titanium bis(phenoxyimine) complexes with different structures has been investigated in the temperature range 30-70°C. The structures of the complexes have different substituents at the imine nitrogen atom and in the phenoxy group in the ligand, which affect the activity of the system and the molecular weight of polyethylene resulting from polymerization over at least 1 h. The polymerization kinetics is most sensitive to the structure of the substituent at the imine nitrogen atom and to bulky substituents in the *ortho* position of the phenoxy group. The results obtained are explained. An attempt is made to classify the influence of the substituents in the ligands. Process conditions ensuring living polymerization have been found. The physicochemical properties and structural features of the polyethylenes obtained have been determined.

A NEW POLYMERIC SILICONE HYDROGEL FOR MEDICAL APPLICATIONS: SYNTHESIS AND PROPERTIES

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Polym. Advanced Technol.,
17(11-12) (2006) pp. 872-877.

A novel silicone hydrogel polymeric material is developed. The preparation method is based on sequential interpenetrating network synthesis. A silicone network is obtained by the interaction between two siloxane oligomers comprising hydride and vinyl functional groups. A hydrophilic network is prepared by radical copolymerization of hydrophilic monomers (*N*-vinyl pyrrolidone, *N*,*N*-dimethylacrylamide) and crosslinking agent (ethylene glycol dimethacrylate). In the hydrated state the developed material has high mechanical properties, transparency, hydrophilicity, oxygen and water permeability. The developed new technology affords obtaining a silicone hydrogel material with a good wettability without additional chemical or plasma surface treatment.

EFFECT OF HYDROGEN BONDS ON ORIENTATIONAL ORDER IN LIQUID-CRYSTALLINE POLYMERS

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Polymer Sci., ser. B,
49(9-10) (2007) pp. 222-223.

The orientational behavior of nematic LC copolymers (4-{[6-(acryloyloxy)hexanoyl]oxy}phenyl-4-propoxybenzoate)-co-(4-(6-acryloyloxypropyl-1-oxy) benzoic acid) in a magnetic field has been studied by H^2 NMR spectroscopy. An increase in the content of carboxyl groups in the copolymer leads to an appreciable growth of the orientational order parameter S-zz.

OPTICALLY ISOTROPIC MESOPHASE IN COMB-SHAPED COPOLYMERS WITH SIDE MESOGENIC GROUPS

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Polymer Sci., ser. A,
49(2) (2007) pp. 148-157.

Two series of comb-shaped copolymers poly(4-(4-cyanobiphenyl-4'-yloxy)butyl acrylate)-*co*-(4-(6-

acryloyloxyhexyl-1-oxy)isophthalic acid)) and poly(4-(6-acryloyloxyhexyl-1-oxy)benzoic acid)-*co*-(acrylic acid)) have been prepared by free-radical copolymerization, and their phase diagrams have been constructed. Formation of the optically isotropic phase characterized by the absence of birefringence and high optical activity has been demonstrated for a number of copolymers. At the same time, the DSC curves of these compounds show a well-defined phase transition with the heat of melting equal to 2–5 J/g. Specific features of hydrogen bonding in the copolymers have been studied by IR spectroscopy. It has been speculated that there is correlation between microphase separation between hydrophobic and hydrophilic units in the copolymers and formation of the optically isotropic mesophase.

NEW POLYMER LIQUID-CRYSTALLINE CdS NANOCOMPOSITES FORMING A CHIRAL NEMATIC PHASE

E.B. Barmatov*, D.A. Pebalk, M.V. Barmatova** (**Novosibirsk Technological Center Shlumberger, Novosibirsk, Russia; **Moscow Lomonosov State University, Moscow, Russia*)

Polymer Sci., ser. B,
49(1-2) (2007) pp. 47-50.

A new approach has been developed for the design of liquid-crystalline polymer nanocomposites combining the unique properties of polymer cholesterics and quantum dots of CdS.

THE EFFECT OF SILVER NANOPARTICLES ON THE PHASE STATE OF COMB-SHAPED LIQUID CRYSTALLINE POLYMERS WITH CYANOBIPHENYL MESOGENIC GROUPS

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Polymer Sci., ser. A,
48(7) (2007) pp. 665-675.

A new approach is proposed for the preparation of a new class of hybrid polymer systems based on comb-shaped LC polymers with cyanobiphenyl mesogenic groups and silver nanoparticles with dimensions ranging from 5 to 54 nm. A correlation between copolymer composition and dimensions of the formed nanoparticles is established. As the concentration of nanoparticles in LC copolymer is increased, the resultant glass transition temperature increases, and the temperature interval of the existence of LC phase is reduced. This behavior is related to the adsorption of cyanobiphenyl and carboxylic polymer groups on the surface of silver nanoparticles. In this case, the conductivity and dielectric permittivity of the composites are also increased.

Chemical Engineering. Mathematical Simulation of Processes and Reactors

INNOVATIVE POTENTIAL OF CATALYTIC TECHNOLOGIES

V.N. Parmon, A.S. Noskov

Catal. Ind., 4 (2007) pp. 3-18.

The paper considers current trends in the area of catalytic technologies in Russian chemical and petrochemical industries caused by the ever increasing demands for catalysts and catalytic technologies for fine processing of hydrocarbons into valuable products, polymers, aromatic compounds, carbon nanostructured materials and other high-tech materials. The discussion is exemplified with developments of the Boreskov Institute of Catalysis in the framework of Russian State Innovative Projects. The Projects are aimed at development and commercialization of catalysts for synthesis of motor fuels, for large-scale production of polyolefins. Among recent BIC’s advances are the technologies for processing of oil associated gases into aromatic compounds, for hydrogenation of plant oils over palladium catalysts, for oxidation of benzene to phenol with nitrous oxide, for synthesis of formic and nicotinic acids, for gas cleaning from hydrogen sulfide by oxidation into elemental sulfur and many other ecologically friendly technologies. In conclusion, the authors assess the Russian market potentials regarding large-scale catalytic technologies for oil processing and petrochemistry.

FIXED BED REACTORS WITH GRADIENT CATALYST

V.M. Khanaev, E.S. Borisova, A.S. Noskov

In *“Chemical Engineering Research Trends”*,
Ed. Leon P. Berton , Nova Publishers, 2007,
Chapter 3, pp. 127-201.

High-energy catalytic processes operated in fixed bed reactors are widely used in many fields of Chemical Engineering, so the improvement of specific intensities of such processes to minimize the equipment size and related capital cost, to enhance the reliability is of great importance. The specific features of such processes are reactant concentration gradients through the bed length which may cause considerable temperature gradients in exo-endothermic reactions; therefore, the catalysts used for the existing and innovative high-energy catalytic processes must meet severe requirements. The catalytic processes in fixed bed reactors are rather complicated and usually include the chemical reactions, the processes of mass and heat transfer and reactant adsorption/desorption. As a result, a specially organized catalytic fixed bed with non-uniform properties (gradient catalysts) may appear more appropriate in some cases than the uniform one. At present, the ideas of gradient catalysts are used for creation the new type of afterburning catalysts, for microchannel reactors. In the theoretical investigation the attention is being increasingly focused on the complex approach based on the optimization of heat- and mass transfer and of the activity of the whole catalyst bed through the optimal bed packing, granule shape, distribution of the active component, heat conductivity etc., i.e. through creation of some optimal gradient medium for conducting the catalytic process. Fundamentals for optimization of the spatial bed structure, in particular for optimization of the active component distribution through the bed length, are developed intensively with regard to synthesis of new materials and creation of new methods for arranging reaction beds (for example, in microchannel reactors). The authors of the present paper are the first who proposed to use the variational approach for solving these problems, so they theoretically formulated the problem of a catalytic process optimization by means of non-uniform spatial active sites distribution and obtained the analytic solutions for several cases.

In the present paper, the following points will be discussed:

1. Setting the optimization problem for a catalytic fixed bed reactor, development of theoretical methods for analysis of the obtained mathematical problem with different reaction rate equations under conditions of intensive heat and mass transfer. Theoretical analysis of the influence of gradient catalysts on:
   - Economy of the total active component amount;
   - Improvement of the reactant conversion;
   - Optimization of the temperature profile;
   - Changes in the reaction selectivity.
2. Evaluation of influences of optimal active component distribution on the conversion and selectivity in real processes, in particular in the selective oxidation of methane.

CONVERSION OF CHEMICAL REACTION ENERGY INTO USEFUL WORK IN THE VAN'T HOFF EQUILIBRIUM BOX

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J. Chem. Educ.,
84(6) (2007) pp. 1053-1055.

The production of useful work by means of the ideal "van't Hoff equilibrium box" is considered in detail. It is shown that useful work arises according to the scheme “reaction energy → heat → useful work” without violation of the second law of thermodynamics, even at constant temperature, using the heat evolved by a reaction. Van't Hoff equilibrium box divided in two parts can produce simultaneously heat (in volume 100 % from theoretical one) and useful work (in volume 100 % from theoretical one) without violation of the first law of thermodynamics.

DIRECT CATALYTIC OXIDATION OF METHANE TO FORMALDEHYDE: NEW INVESTIGATION OPPORTUNITIES PROVIDED BY AN IMPROVED FLOW CIRCULATION METHOD

I.I. Bobrova, N.N. Bobrov, L.G. Simonova, V.N. Parmon

Kinet. Catal., 48(5) (2007) pp. 676-692.

An improved flow circulation reactor with reaction mixture quenching was used to study the direct oxidation of methane to formaldehyde at 600-770°C, a CH_4/O_2 volume ratio of 1 : 1 in the starting mixture, and gas mixture flow rates of 0.4-2.0 l/h. It was found that, in the presence of a surface containing SiO_2 (quartz reactor walls, the surface of silica packing materials, or the surface of SiO_2 as a catalyst constituent), the process occurred by a heterogeneous–homogeneous mechanism with chain continuation in the volume of a gas phase. The process was controlled by the size and shape of the free reaction volume, the contact surface area, and the residence time of a mixture in the reaction volume. The introduction of typical oxidation catalysts containing, for example, Pt or V_2O_5 as an active component along with SiO_2 resulted in a decrease in the yield of and selectivity for formaldehyde.

MICROCHANNEL CATALYTIC SYSTEMS FOR HYDROGEN ENERGETICS

L.L. Makarshin, V.N. Parmon

Russ. J. General Chem.,
77(4) (2007) pp. 676-684.

The concept of hydrogen energetics envisages economically reasonable hydrogen production from various organic compounds in stationary and mobile devices of low and medium performance, called "fuel processors". Fuel processors with a high specific performance in hydrogen can be developed with the aid of microchannel catalytic systems. The paper considers the present situation with microchannel catalytic reactors for fuel processors.

OVERALL PERFORMANCE OF THE CATALYST IN A MICROREACTOR FOR THE METHANE PARTIAL OXIDATION

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Intern. Sci. J. Alternative Energy Ecol.,
2 (2007) pp. 132-134.

The goal of this work is investigation of partial methane oxidation (POM) for syngas production in the microchannel system. The features of such systems are high specific productivity which allows to use their in various mobile devices. The method of firm catalyst deposition on the metal microchannel plates functioning at 900°C was developed which permits a repeated thermocycling of the plates.

At gas hourly space velocity 700000 h^{-1} the methane conversion was 90 % and carbon monoxide selectivity was 60 %. Hydrogen production was proportional to number of the microchannel plates loaded in the microreactor and was 34 l/h for one plate and 380 l/h for ten plates. Specific hydrogen production relative to the microreactor's volume was $1.7 \cdot 10^4$ l/(h·dm^3).

It is found that thermal load distributes non-uniformly along the microchannel plate due to the POM reaction exothermicity which results in corrosion of the front edge (about 1 mm) of the microchannel plate.

Additional experiments and theoretical calculations have shown that in the course of the POM process in front side of the microchannel plate the total oxidation of methane occurs following high heat evolution and H_2O and CO_2 formation. On the rest side of the plate endothermic steam and dry reforming of methane occurs with CO and H_2 formation.

EFFECT OF THE MICROCHANNEL PLATE DESIGN ON THE CAPACITY OF METHANOL STEAM REFORMERS

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Kinet. Catal., 48(5) (2007) pp. 765-771.

Methanol steam reforming in microreactors is considered, and the effects of the microreactor geometry (cylindrical and rectangular) and microchannel plate (MCP) design on the hydrogen capacity of the microreactor is analyzed. The MCPs were made from aluminum foil, stainless steel, and foamed nickel by laser engraving, electrochemical etching, and pressing. The amount of catalyst powder (CuO/ZnO = 40 : 60 mol/mol) fixed on one MCP was 0.04–2.5 g. The specific hydrogen capacity (U_w) of the cylindrical microreactor is more than 3 times as high as the U_w of the rectangular microreactor and is 6 times as high as the U_w of a conventional fixed-bed catalytic reactor. This gain in hydrogen capacity is due to the more efficient use of the catalyst in the microreactors. The MCP design, which determines the residence time of the reactants in the microreactor, also has a significant effect on the capacity of the microreactor.

INFLUENCE OF THE MICROCHANNEL PLATES DESIGN ON THE EFFICIENCY OF THE METHANOL STEAM REFORMING IN MICROREACTORS

**L.L. Makarshin, D.V. Andreev, A.G. Gribovskii,
V.N. Parmon**

Intern. J. Hydrogen Energy,
32(16) (2007) pp. 3864-3869.

The reaction of methanol steam reforming in microchannel microreactors was studied and the effect of catalyst structure and microreactor design on the reaction rate constant of methanol steam reforming was elucidated. Three types of microreactors were studied - rectangular, cylindrical and tubular with fixed catalyst bed. To analyze the experimental data a simple kinetic model with power-law rate equation with the first reaction order for the methanol was used. It was shown that the value of reaction rate constant and hydrogen production depends on structure of the catalyst loaded into the reaction volume. The highest reaction rate constant and specific hydrogen production equal $8.11/(hg_{cat})$was observed for the rectangular microreactor, with two metal plates with deposited catalyst. The cylindrical microreactors produced $6.21/(hg_{cat})$ of hydrogen. The tubular microreactor exhibited the lowest reaction rate constant and specific hydrogen production equal $4.91/(hg_{cat})$. This fact seems to be associated with the temperature gradient in the reactor due to the endothermic reaction of the methanol steam reforming. Using the Arrhenius equation the temperature gradient in the reaction volume was estimated - 6.7 and 13.6 K for the cylindrical and tubular microreactors, respectively. These results agreed with heat balance calculations.

COBALT FISCHER-TROPSCH SYNTHESIS: DEACTIVATION BY OXIDATION?

J. van de Loosdrecht*, B.S. Balzhinimaev,
J.-A. Dalmon, J.W. Niemantsverdriet***,**
S.V. Tsybulya, A.M. Saib*, P.J. van Berge*,
J.L. Visagie* (**Sasol Technology Ltd, South Africa; **Institute de Recerches sur la Catalyse, Villeurbanne Cedex, France; ***Eindhoven University of Technology, The Netherlands*)

Catal. Today, 123(1-4) (2007) 293-302.

Cobalt catalysts as used in the Fischer-Tropsch synthesis (FTS) are relatively expensive (as compared to iron) and need to have a high metal dispersion and long life to be able to offer a good balance between cost and performance. The oxidation of nano-sized metallic cobalt to cobalt oxide during Fischer-Tropsch synthesis has long been postulated as a major deactivation mechanism. However, to date there is no consistent picture. This paper presents an extensive overview of the literature on this topic of deactivation by means of oxidation for unsupported as well as silica-, alumina- and titania-supported cobalt catalysts. Furthermore, it presents results on the deactivation of an industrial Co/Al_2O_3 catalyst as obtained by pseudo *in situ* X-ray diffraction, magnetic measurements and X-ray absorption near-edge spectroscopy. These analyses were performed to study the oxidation state of spent industrial Co/Al_2O_3 catalyst samples withdrawn from a slurry reactor operating under realistic FTS conditions, and it was concluded that oxidation can be ruled out as a major deactivation mechanism. Finally, these data together with all relevant literature were used to create a common view on the oxidation behaviour of cobalt during FTS. The apparent discrepancies in literature on the oxidation behaviour of cobalt are most likely due to the lack of direct characterisation of the cobalt oxidation state and due to the comparison of catalysts with varying cobalt crystallites compared at different reactor partial pressures of hydrogen and water (P_{H2O}/P_{H2}). It was shown that the oxidation of cobalt can be prevented by selecting the correct combination of the reactor partial pressures of hydrogen and water (P_{H2O}/P_{H2}) and the cobalt crystallite size.

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Cobalt catalysts as used in the Fischer-Tropsch synthesis (FTS) are relatively expensive (as compared to iron) and need to have a high metal dispersion and long life to be able to offer a good balance between cost and performance. The oxidation of nano-sized metallic cobalt to cobalt oxide during Fischer-Tropsch synthesis has long been postulated as a major deactivation mechanism. However, to date there is no consistent picture. This paper presents an extensive overview of the literature on this topic of deactivation by means of oxidation for unsupported as well as silica-, alumina- and titania-supported cobalt catalysts. Furthermore, it presents results on the deactivation of an industrial Co/Al_2O_3 catalyst as obtained by pseudo *in situ* X-ray diffraction, magnetic measurements and X-ray absorption near-edge spectroscopy. These analyses were performed to study the oxidation state of spent industrial Co/Al_2O_3 catalyst samples withdrawn from a slurry reactor operating under realistic FTS conditions, and it was concluded that oxidation can be ruled out as a major deactivation mechanism. Finally, these data together with all relevant literature were used to create a common view on the oxidation behaviour of cobalt during FTS. The apparent discrepancies in literature on the oxidation behaviour of cobalt are most likely due to the lack of

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MODELING OF A HEAT-COUPLED CATALYTIC REACTOR WITH CO-CURRENT OXIDATION AND CONVERSION FLOWS

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Chem. Eng. J., 134(1-3) (2007) pp. 131-137.

Mathematical model and numerical method are developed to analyze the operation of a coupled methane steam reformer. The reformer operates in co-current mode of endothermic (steam reforming) and exothermic (fuel oxidation) departments. The exhaust anode gas of a fuel-cell battery containing hydrogen is used as fuel. Two-dimensional, two-temperature mathematical model of monolith is used to describe processes in exothermic and endothermic departments of the reformer. Calculation results were compared with experiments. The agreement between the modeling and the experimental results is rather good. The model analysis points out that the considered reactor design has high parametric sensitivity, there is a probability of arisen of a hysteresis with points of ignition and extinction.

ANALYSIS OF STABILITY OF THE INTERPHASE FRONT WITH EVAPORATION AND EXOTHERMIC REACTION IN THE POROUS CATALYST SLAB

A.B. Shigarov, A.A. Bocharov, V.A. Kirillov

Chem. Eng. Sci., 62(17) (2007) pp. 4770-4779.

Using a two-dimensional mathematical model and a number of simplifying assumptions, the analytical (linear) analysis of stability of the reaction–evaporation front inside the half-wetted porous catalyst slab was made. Based on the obtained spectral equation, the effect of different parameters on possible types of instability was studied.

n-HEXANE SKELETAL ISOMERIZATION OVER SULFATED ZIRCONIA CATALYSTS WITH DIFFERENT LEWIS ACIDITY

G.G. Volkova, S.I. Reshetnikov, L.N. Shkuratova, A.A. Budneva, E.A. Paukshtis

Chem. Eng. J., 134(1-3) (2007) pp. 106-110.

The *n*-hexane skeletal isomerization over $Pt/SO_4^{2-}/Al_2O_3/ZrO_2$ catalysts with different density of Lewis acid sites has been studied in a fixed-bed reactor. The acidity of the catalysts was characterized by FTIR spectroscopy of low temperature CO adsorption. The calculation of the rate constants, characterizing the catalyst activity and selectivity was performed on the base of mathematical model of plug flow reactor. It was shown that the rate of hexane isomerization is mainly determined by the density of Lewis acid sites on the surface of sulfated zirconia catalysts: rate constant of *n*-hexane isomerization for catalyst with Lewis acidity 250 $\mu mol\ g^{-1}$ is four-fold as that for catalyst with concentration of Lewis acid sites 150 $\mu mol\ g^{-1}$ and the rate constant of desired product (2,2-dimethylbutane) formation is also higher by two times.

UNSTEADY CATALYTIC PROCESSES AND SORPTION-CATALYTIC TECHNOLOGIES

A.N. Zagoruiko

Russ. Chem. Rev., 76(7) (2007) pp. 639-654.

Catalytic processes that occur under conditions of the targeted unsteady state of the catalyst are considered. The highest efficiency of catalytic processes was found to be ensured by a controlled combination of thermal non-stationarity and unsteady composition of the catalyst surface. The processes based on this principle are analysed, in particular, catalytic selective reduction of nitrogen oxides, deep oxidation of volatile organic impurities, production of sulfur by the Claus process and by hydrogen sulfide decomposition, oxidation of sulfur dioxide, methane steam reforming and anaerobic combustion, selective oxidation of hydrocarbons, *etc*.

SORPTION-ENHANCED STEAM REFORMING OF HYDROCARBONS WITH AUTOTHERMAL SORBENT REGENERATION IN A MOVING HEAT WAVE OF A CATALYTIC COMBUSTION REACTION

A.N. Zagoruiko, A.G. Okunev

React. Kinet. Catal. Lett.,
91(2) (2007) pp. 315-324.

A novel technological concept of sorption-enhanced steam reforming of hydrocarbons is suggested. The peculiarity of the concept is the autothermal regeneration of the carbon dioxide scavenger in the moving super-adiabatic heat wave of an exothermic catalytic combustion reaction performed directly inside the adsorption-catalytic bed. The capability and high efficiency of the proposed technological approach are confirmed by process simulation. The approach proposed is shown to open a way for the creation of an inexpensive, reliable and energy-saving adiabatic packed bed methane processors of unlimited processing capacity.

SIMULATION OF SELECTIVE REACTIONS' PERFORMANCE IN TRANSIENT REGIMES WITH PERIODICAL SEPARATE FEEDING OF REAGENTS. CASE STUDY: PROPANE OXIDATIVE DEHYDROGENATION IN ADIABATIC V–Ti CATALYST BED

A.N. Zagoruiko

Chem. Eng. J.,
134(1-3) (2007) pp. 117-122.

The model simulation study has shown that the anaerobic process of oxidative dehydrogenation of propane under periodic alteration of feeding between propane and air may be realized in adiabatic catalyst beds in stable continuous cyclic mode in two-reactor scheme. In case of appropriate choice of process parameters (cycle duration and residence times), the process appears to be autothermal, i.e. it does not require any inlet gas preheating for stable operation. Compared to similar steady-state adiabatic process, the proposed process is characterized with much lower maximum catalyst temperatures, giving the way to process pure propane without diluting it with inert gases, thus simplifying downstream procedure of product separation. Predicted propylene yield is competitive with one for the steady-state adiabatic process, while sufficient technological benefits of the new technology are expected (decrease of energy consumption and minimization of heat-exchange environment, simplification of product separation procedure, process safety improvement, suppression of coke formation and efficient coke incineration). Notably, the proposed process is characterized with much more degrees of freedom available for further development and optimization than conventional steady-state technologies.

KINETIC INSTABILITIES AND INTRA-THREAD DIFFUSION LIMITATIONS IN CO OXIDATION REACTION AT Pt/FIBER-GLASS CATALYSTS

**A.N. Zagoruiko, S.A. Veniaminov,
I.N. Veniaminova, B.S. Bal'zhinimaev**

Chem. Eng. J.,
134(1-3) (2007) pp. 111-116.

Experimental and mathematical simulation studies of CO oxidation reaction at Pt-based glass-fiber catalyst showed that reaction phenomena in fabric and threads of woven glass-fiber catalysts is rather complicated by transfer limitations, which may impose especially valuable influence on reactions with strongly non-linear kinetics. Such limitations are related to extra-thread and intra-thread heat and mass transfer. Additionally, in thin beds, consisting of one or few layers of woven catalyst, the significant role may be played by experimentally confirmed transient oscillations between CSTR and plug-flow regimes, therefore, the axial heat and mass dispersion is an important transfer factor for such beds as well.

Mathematical modelling of phenomena inside the catalyst thread on the basis of elaborated kinetic model showed possibility of existence of steady-state multiplicity as a self-sufficient result of intra-thread transfer limitations. In general, observed reaction rates at the catalyst thread and location of multiplicity region may depend upon many factors (thread diameter and porosity, surface CO concentration and temperature and even choice of reaction media balance gas), which should be taken into account for correct formulation of lab experimental procedure, interpretation of experimental data and process scale-up.

BENZENE HYDROGENATION IN THE THIOPHENE PRESENCE OVER THE SULFIDE Ni-Mo/γ-Al_2O_3 CATALYST UNDER PERIODIC OPERATION: KINETICS AND PROCESS MODELLING

S.I. Reshetnikov, E.A. Ivanov, A.N. Startsev

Chem. Eng. J.,
134(1-3) (2007) pp. 100-105.

An unsteady-state kinetic model of both benzene hydrogenation (HDA) and thiophene hydrogenolysis (HDS) on the sulfide hydrotreating catalyst Ni-Mo/Al_2O_3 has been developed. The model adequately describes experimental data obtained at the pressure 2 MPa, temperature 573 K and at various contact times and ratios of benzene/thiophene. The model is based on the assumption that the catalyst surface contains only one type of active sites, i.e., Ni atoms in the sulfide bimetallic species, which are responsible for both hydrogenolysis and hydrogenation reactions. On the base of the kinetic model, a theoretical analysis of the reactor performance under unsteady state conditions was carried out. The unsteady state conditions on the catalyst surface are supposed to be created by forced oscillations of thiophene concentration in the reactor inlet (periodic operation of reactor). The influence of various parameters like cycle split, length of period of forced oscillations in the reactor was investigated with respect to the conversion of the benzene. It is shown that for periodic reactor operation an average conversion was up to several times higher than a steady state value.

SYNGAS FORMATION FROM GASOLINE IN ADIABATIC REACTOR: THERMODYNAMIC APPROACH AND EXPERIMENTAL OBSERVATIONS

L.N. Bobrova, V.M. Korotkikh, V.A. Sadykov, V.N. Parmon

Chem. Eng. J.,
134(1-3) (2007) pp. 145-152.

To make viable the large-scale use, generation of hydrogen locally from the petroleum-derived fuels such as gasoline is an attractive way because of the existing production, distribution, and retailing infrastructure. The present work is centered on the syngas formation from gasoline. The thermodynamic equilibrium restrictions on the gasoline reforming process in comparison with isooctane and methane are discussed on the basis of multi-component calculations. To perform the thermodynamic modelling, the real fuel containing 191 hydrocarbon species was specified as an equilibrium distribution of 29 close-cut fractions C3–C10 with well-defined properties. The thermodynamic predictions are in a good correlation with the experimental data obtained in a nearly adiabatic monolith reactor.

HYDROGEN-RICH GAS PRODUCTION FROM GASOLINE IN A SHORT CONTACT TIME CATALYTIC REACTOR

L.N. Bobrova, I.A. Zolotarsky, V.A. Sadykov, V.A. Sobyanin

Int. J. Hydrogen Energy,
32(16) (2007) pp. 3698–3704.

Presented is a study of hydrogen-rich gas generation by selective catalytic oxidation of gasoline in a nearly adiabatic monolith reactor. Experiments were conducted using a gasoline mixture containing 191 hydrocarbon species, while corresponding thermodynamic models employed a surrogate mixture of 29 organics with well-defined properties. Based on the data obtained, it was demonstrated that a nearly equilibrium synthesis gas could be produced using catalytic monoliths. Also discussed are problems encountered with a short contact time reactor operating at a pilot-scale, issues that are largely independent of the catalysts employed. It was found that this particular reactor design is prone to pre-reformation of the fuel prior to catalytic reaction. In addition, the breakthrough of the feed mixture was found to arise near the reactor wall under certain circumstances. Feed composition, superficial velocity, and various reactor design factors were all found to affect these phenomena.

TRANSIENT BEHAVIOR OF THE METHANE PARTIAL OXIDATION IN A SHORT CONTACT TIME REACTOR: MODELING ON THE BASE OF CATALYST DETAILED CHEMISTRY

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Chem. Eng. J.,
134(1-3) (2007) pp. 180-189.

The objective of the paper is a numerical study of the partial oxidation of methane to synthesis gas in a monolith reactor with Pt/Ce-Zr-La catalyst. A dynamic one-dimensional two-phase reactor model of the processes with accounting for both transport limitations in the boundary layer of a fluid near the catalyst surface and detailed molecular unsteady-state kinetic model for surface reactions have been developed and verified with the transient experiments data. The mathematical model was used to explain a transient behavior of the process in the monolith reactor during start-up (ignition). Also studied is the influence on the process dynamics such parameters as linear velocity, equivalent diameter of triangular channel and effective thermal conductivity of the monolith. It was found that higher linear velocity and equivalent channel diameter as well as the worse axial conductivity of solid phase favor decreasing a time delay in syngas production in the Pt/Ce-Zr-La/α-Al_2O_3 honeycomb monolith with a triangular shape channels.

OPTIMUM DIMENSIONS OF SHAPED STEAM REFORMING CATALYSTS

A.P. Kagyrmanova, I.A. Zolotarskii, E.I. Smirnov, N.V. Vernikovskaya

Chem. Eng. J.,
134(1-3) (2007) pp. 228-234.

In this paper, a theoretical optimization of shaped catalyst dimensions with technologically imposed constraints is presented for operating conditions of a typical methanol reformer. A comprehensive two-dimensional mathematical model of a single reformer tube containing physically sound correlations for heat transfer parameters of holed catalyst particles is used. Three-hole pellets and Raschig rings were used as examples. A general conclusion is made that holed steam reforming catalysts should have maximum catalyst height and minimum diameter determined by requirements of uniform catalyst packing.

COMPACT REACTOR FOR WATER GAS SHIFT REACTION OVER THERMAL-CONDUCTING CATALYSTS

N.A. Baronskaya, T.P. Minyukova, A.G. Sipatrov, M.P. Demeshkina, A.A. Khassin, S.V. Dimov*, S.P. Kozlov*, V.V. Kuznetsov*, V.Ya. Terentiev, A.P. Khristolyubov**, O.F. Brizitskiy**, T.M. Yurieva** (**Kutateladze Institute of Thermophysics, Novosibirsk, Russia; **Russian Institute for Experimental Physics (VNIIEF), Sarov, Russia*)

Chem. Eng. J.,
134(1-3) (2007) pp. 195-199.

Thermal conductive catalyst plates (TCP) for compact WGSR reactors were prepared and studied. High thermal conductivity of 2.5–5 W $(m\ K)^{-1}$ can be achieved, while the effective catalytic activity of TCP is even higher than that of the commercial size catalyst grains. TCP can be efficiently used for temperature sensitive exothermic reactions in compact apparatuses with controlled temperature profile. Using the TCP reactor WGSR of the fuel gas containing 12% of CO down to 1% of residual CO can be performed in one stage at GSHV of above 6000 h^{-1}. The TCP shape and the way of arrangement are important for the reactor performance and should be in the focus of future studies.

PERMEABLE COMPOSITE MATERIAL FOR COMPACT APPARATUS FOR HYDROGEN-RICH GASES DEEP CLEANING FROM CO

T.P. Minyukova, I.Sh. Itenberg, A.A. Khassin, A.G. Sipatrov, E.V. Dokuchits, V.Ya. Terent'ev*, A.P. Khristolyubov*, O.F. Brizitskii*, T.M. Yurieva (**Russian Institute for Experimental Physics (VNIIEF), Sarov, Russia*)

Chem. Eng. J.,
134(1-3) (2007) pp. 235-238.

In order to provide a wide application of the proton exchange membrane fuel cell (PEMFC) technology, reduce cost and improve characteristics of PEMFC-based power plants, it is necessary to create a compact and economic apparatus for deep cleaning of converted fuel from carbon monoxide. The obstacle of preferential CO hydrogenation is the presence of about 20 vol.% of CO_2 and the same amount of H_2O along with 1 vol.% of CO. The compact apparatus for deep purification from CO can be only created basing on the highly active (providing 10–20 ppm CO in the outlet gas at GHSV above 6000 h^{-1}) and selective catalyst (at least 30% of CH_4 and higher hydrocarbons is to be formed by CO hydrogenation). The process of preferential CO hydrogenation over the nickel-chromium catalyst is studied, which was found earlier to be efficient in the CO hydrogenation over fine catalyst fraction: outlet CO content below 20 ppm and the selectivity not less than 0.3 at GHSV above 6000 h^{-1}. The effect of the catalyst grain size on the CO hydrogenation selectivity is reported and the possibility of using permeable composite materials for weakening the intraparticle diffusion constrains and for efficient performing the reaction under the study close to the kinetic region is considered.

MATHEMATICAL MODELING AND EXPERIMENTAL STUDY OF HIGH-PRESSURE ETHYLENE POLYMERIZATION REACTORS

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(**Central R&D Institute for Complex Automation, Moscow, Russia*)

Chem. Eng. J.,
134(1-3) (2007) pp. 175-179.

Mathematical models and experimental data are discussed for high-pressure ethylene polymerizations in stirred autoclaves and tubular reactors. The models are based on free radical mechanism of ethylene polymerization using material and energy balance equations as well as impact balance (for tubular reactors). A stirred autoclave reactor is modeled taking into account mixing modes in the reactor on the basis of the segregation approach.

The developed mathematical model was used to study the stability of stirred autoclave reactor and determine the range of the process parameters corresponding to their sustainable functioning.

AUTOWAVE PROCESSES IN A HETEROGENEOUS MEDIUM WITH CHEMICAL REACTIONS IN A GASEOUS PHASE AND IN A CATALYST BED

A.P. Gerasev

Theor. Found. Chem. Eng.,
41(2) (2007) pp. 116-123.

A mathematical model is constructed for autowave processes in a heterogeneous medium with chemical reactions in a gaseous phase and in a catalyst bed. The model accounts for the changes in the interphase heat and mass transfer and thermal conductivity coefficients of the catalyst bed as a function of the system parameters. The phase trajectories of the dynamic system are investigated using qualitative and numerical analyses, and an effective technique for searching for a physically comprehensive autowave solution of the problem is proposed. The mathematical model of hybrid autowaves is tested in a cycle of numerical calculations, and the influence of the system parameters on the main technological characteristics of the autowave processes is analyzed.

AUTOWAVES IN A HETEROGENEOUS MEDIUM WITH A CATALYTIC REACTION AND HEAT AND MASS TRANSFER

A.P. Gerasev

Combust., Explosion, Shock Waves,
43(2) (2007) pp. 162-169.

A mathematical model is developed to describe autowave processes in a catalytic fixed bed, which takes into account changes in coefficients of interphase heat and mass transfer and thermal conductivity of the catalytic bed, depending on the current values of system parameters. The behavior of the phase trajectories of the dynamic system is studied by methods of qualitative and numerical analysis, and an effective technique is proposed for searching for a phase trajectory corresponding to the autowave solution of the problem. Numerical studies of the mathematical model of autowave processes are performed, and the effect of system parameters on technological characteristics of the process, such as the maximum temperature and wave-propagation velocity, is considered.

APPLICATION OF CATALYTIC COMBUSTION CHAMBERS OF DECENTRALIZED CONTROL

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Herald Russ. Acad. Sci.,
77(9) (2007) pp. 819-826.

The objective of the paper is comprehensive analysis of catalytic combustion chambers for present-day gas-turbine units carried out at the BIC SB RAS and Baranov Central Institute of Aviation Motors. The results demonstrate the possibility of development of efficient combustion chambers using domestic high technologies.

CATALYTIC SO_2 REDUCTION FOR PURIFICATION OF WASTE GASES OF NONFERROUS-METALS INDUSTRY. TECHNOLOGIES AND CATALYSTS

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Herald of al-Farabi Kazakh National University, ser. Chem., 3(47) (2007) pp. 45-63.

The paper reviews the technologies for purification of waste gases of nonferrous-metals industry. Detailed description of catalytic technologies of SO_2 reduction to elemental sulfur is given. Catalytic systems for SO_2 reduction and methods of its preparation are reviewed

IMPROVEMENT AND DEVELOPMENT OF CATALYTIC PROCESSING OF WASTE GASES IN VANYUKOV AND FLASH SMELTING FURNACES

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Non-Ferrous Met., 7 (2007) pp. 80-85.

Presented are the results of laboratory and pilot tests of the processes of catalytic SO_2 reduction to elemental sulfur with methane and syn-gas. It is concluded that low-temperature process of SO_2 catalytic reduction with syn-gas has advantages over homogeneous and catalytic processes of sulfur dioxide reduction with methane.

ACTIVITY OF NEW DOMESTIC ALUMINA CATALYST OF CLAUS PROCESS

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Catal. Ind., 2 (2007) pp. 54-59.

Results of industrial tests (from 02.04.2003 to 26.06.2006) of new domestic alumina catalyst, AO-NKZ-2 (AO-MK-2), +6,6 mm fraction, in Claus reactor of coke gas purification line of Magnitogorsk metallurgical plant at 245–260°C and 2000 h–1 are generalized. It has been found that the activity of AO-NKZ-2 catalyst in Claus reaction matches that of best foreign analogs (DR «Rhone-Poulenc» and CR-31 «La Roche Chemicals»). Yearly irreversible aging rate of AO-NKZ-2 in hydrogen sulphide conversion does not exceed 2–3 %, which provides at least 7–10 year catalyst operation lifetime in Claus reactors. To increase the economical efficacy of Claus equipment, it is recommended to use specialized catalysts during the hydrolysis of organosulphur compounds and low-temperature Claus conversion steps.

REAL-TIME CONTROL OF SOLID-PROPELLANT COMBUSTION BY A CATALYTIC METHOD

N.N. Kundo, V.V. Romankov, V.I. Simagina, I.V. Eroshkina

Combust., Explosion, Shock Waves,
43(1) (2007) pp. 73-77.

Principles of contact catalysis of combustion are described. The essence of this process implies that a block catalyst contacting the surface of the burning propellant specimen increases the burning rate and allows controlling combustion at a temperature of 400 to 600°C with formation of a large volume of versatile gaseous products, i.e., is the basis for creating low-temperatures gas generators.

THE THERMODYNAMIC CHARACTERISTICS OF FORMATION OF NANOPARTICLES FROM SUPERCRITICAL SOLVENTS

V.I. Anikeev, A. Yermakova

Russ. J. Phys. Chem. A,
81(12)(2007) pp. 2024-2029.

A thermodynamic model for calculating equilibria and the phase states of fluid-solid systems over wide temperature, pressure, and composition ranges was suggested. Calculations were performed to determine the parameter regions where a substance was isolated in the form of a solid depending on the chemical composition of the initial mixture and the amounts of and ratio between the solvent and antisolvent in it. The calculations were performed for a model system containing ethanol, carbon dioxide, and phenanthrene. The rapid expansion of this supercritical fluid was simulated to model the conditions of the formation of a solid (phenanthrene) and nanoparticles based on it. It was shown that the selection of the state parameters of the supercritical mixture determined the optimum conditions of solid state formation as the fluid expanded.

THE THERMAL ISOMERIZATION OF TERPENE COMPOUNDS IN SUPERCRITICAL ALCOHOLS

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Russ. J. Phys. Chem. A,
81(5)(2007) pp. 711-716.

The experimental data obtained were used to construct a kinetic model of the isomerization of α-pinene in supercritical ethanol. The model took into account the influence of both temperature and pressure on the rate and selectivity of the reaction.

THERMAL ISOMERIZATION OF α-PINENE IN SUPERCRITICAL ETHANOL

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Chem. Eng. Sci.,
62(9) (2007) pp. 2414-2421.

The experimental study followed by mathematical processing of the data showed that supercritical ethanol is an effective reaction medium for thermal isomerization of *α*-pinene. Rate of the reaction in supercritical solvent is by several orders of magnitude greater than the rate observed under normal conditions, with selectivity to the target reaction products being retained. The study allowed for the first time to obtain the kinetic model of -pinene isomerization in supercritical ethanol that takes into account temperature and pressure effect on the reaction rate and selectivity.

COMPARATIVE THERMOLYSIS OF β-AND α-PINENES IN SUPERCRITICAL ETHANOL: THE REACTION CHARACTERIZATION AND ENANTIOMERIC RATIOS OF PRODUCTS

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Russ. Chem. Bull.,
56(6) (2007) pp. 1234-1238.

The thermolysis of β-pinene and the co-thermolysis of an equimolar mixture of β-and α-pinenes in supercritical ethanol were carried out. The reaction of β-pinene affords β-myrcene as the major product (>70%). Considerable differences in the temperature dependence of the reaction rate were revealed for the transformations of β-pinene into β-myrcene and of α-pinene into limonene. The pre-exponential factors and the activation energies were calculated. The enantiomeric ratios of the thermolysis products of β-and α-pinenes and limonene were estimated. The starting monoterpenes do not undergo racemization during thermolysis. The thermolysis of enantiomerically pure α-pinene affords racemic (±)-limonene, whereas (−)-β-pinene gives (−)-limonene. The enantiomeric ratio in the latter remains equal to the enantiomeric purity of the starting β-pinene.

COMBUSTION CHARACTERIZATION OF HYDRAZINIUM NITRATE/ENERGETIC BINDER/ALEX BASED MODEL PROPELLANTS

V.E. Zarko*, V.N. Simonenko*, G.I. Anisiforov, A.V. Aparin (**Institute of Chemical Kinetics and Combustion, Novosibirsk, Russia*)

Aerospace Sci. Technol.,
11(1) (2007) pp. 13-17.

The hydrazinium nitrate (N_2H_4*HNO_3-HN) can be considered as a candidate for using instead of ammonium perchlorate (AP) in eco-friendly solid propellant formulations. In combination with energetic binder it provides reasonably high energetic parameters of solid propellant. In addition, the substitution of AP by HN leads to decrease in the combustion temperature. The combustion of the HN based metallized propellants containing energetic binder has been investigated. The effect of partial substitution of commercial Al powder by Alex on the propellant combustion characteristics has been examined. The burning law was measured in the pressure range 20-80 atm, the ignition delay time and temperature profile were measured at atmospheric pressure. Some qualitative observations on the combustion behavior of the propellants under study have been made by analyzing the video camera and recoil force transducer records.

ON ONE APPROACH TO THE W-METHODS CONSTRUCTION FOR STIFF ODE SYSTEMS

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Comput. Technol.,
12(4) (2007) pp. 42-58.

A new approach for construction of W-methods for stiff system of ODE is presented. Using the proposed idea, a W-modification of the one-stage Rozenbrok scheme has been developed. Some computational features of the proposed new method are described.

APPLICATION OF THE CHEMPAK PACKAGE FOR MODELING OF GAS-DYNAMICS REACTOR

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O.P. Stoyanovskaya, A.V. Snytnikov* (**Institute of Computational Mathematics and Mathematical Geophysics, Novosibirsk, Russia*)

Comput. Technol.,
11(1) (2006) pp. 35-51.

The ChemPAK program package for solving the direct problems of chemical kinetics with arbitrary number of chemical reactions is suggested. The package contains an expandable library of computational modules and provides a function of data transfer to multiprocessor. The process of C_1-C_2 hydrocarbon's pyrolysis in gas-dynamics reactor with emission has been numerically simulated using the ChemPak software package.

THE PARALLEL ALGORITHM AND PROGRAM KEY7D FOR SOLUTION OF NON-STATIONARY 3D PROBLEMS OF GRAVITATIONAL PHYSICS

E.A. Kuksheva, V.N. Snytnikov

Comput. Technol., 12(1) (2007) pp. 35-44.

An effective parallel Key7D code for solution of 3D problems of collisionless gravitational physics in Cartesian coordinates is developed. The discrete analogue of fundamental solution of Poisson's Equation is used to determine boundary conditions during calculation of the gravitational potential. For this procedure an efficient parallel algorithm is developed. A quasistationary solution of the system is p resented using the Key7D program.

STRATEGIES FOR DEVELOPMENT OF A PARALLEL PROGRAM FOR PROTOPLANETARY DISC SIMULATION

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Lect. Notes Comp. Sci.,
"Parallel Comp. Technol.",
4671 (2007) pp. 128-139.

Protoplanetary disc simulation must be done first, with high precision, and second, with high speed. Some strategies to reach these goals are presented in the paper. They include: the reduction of the 3D protoplanetary disc model to quasi-3D, the use of fundamental Poisson equation solution, the simulation in the natural (cylindrical) coordinate system and computation domain decomposition. The domain decomposition strategy is shown to reach the simulation goals the best.

PRODUCTION OF NANOMATERIALS BY VAPORIZING CERAMIC TARGETS IRRADIATED BY A MODERATE-POWER CONTINUOUS-WAVE CO_2 LASER

V.N. Snytnikov, D.A. Dubov, Vl.N. Snytnikov,
V.I. Zaikovskii, A.S. Ivanova, V.O. Stoyanovskii,
V.N. Parmon

J. Appl. Mech. Techn. Phys.,
48(2) (2007) pp. 292-302.

The efficiency of utilization of CO_2 laser energy for vaporization of Al_2O_3 ceramics is evaluated using a mathematical model for the interaction of laser radiation with materials. It is shown that the calculated efficiency of radiation-energy utilization is not higher than 15% at a radiation power density of 10^5 W/cm ^{2} on the target. On the experimental facility designed for the synthesis of nanopowders, a vaporization rate of 1 g/h was achieved for Al_2O_3, which corresponds to a 3% efficiency of radiation-energy utilization. The dependence of the characteristic particle size of a zirconium oxide nanopowder on helium pressure in the range of 0.01–1.00 atm was studied. Results of experiments on vaporization of multicomponent materials ($LaNiO_3$ and the Tsarev meteorite) are given.

STUDY OF 3D DYNAMICS OF GRAVITATING SYSTEMS USING SUPERCOMPUTERS: METHODS AND APPLICATION

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(**Institute of Computational Mathematics and Mathematical Geophysics, Novosibirsk, Russia*)

Lect. Notes Comp. Sci.,
"Parallel Comp. Technol.",
4671 (2007) pp. 162-173.

Parallel numerical code for solving problems of stellar dynamics is described. The code is based on numerical solving of Poisson and Vlasov equations in cylindrical coordinates using particle-in-cells method. The code is designed for use on supercomputers with distributed memory. It is considered different possible strategies of parallelization according to initial technical parameters of numerical methods and physical conditions of the model. The results of numerical simulations for the following problems of stellar dynamics: investigation of influence of central potential on the vertical motions of thin gravitating disk; stability of uniform sphere with anisotropic distribution of velocity; numerical approximation of equilibrium states of gravitating systems are presented.

AN ALGORITHM FOR THE THREE-DIMENSIONAL MODELING OF ULTRARELATIVISTIC BEAMS

M.A. Boronina, V.A. Vshivkov*, E.B. Levichev, S.A. Nikitin**, V.N. Snytnikov** (**Institute of Computational Mathematics and Mathematical Geophysics, Novosibirsk, Russia; **Budker Institute of Nuclear Physics, Novosibirsk, Russia*)

Numer. Meth. Progr.,
8 (2007) pp. 352-359.

An algorithm for modeling the interaction charged particle beam effects in the three-dimensional ultrarelativistic case is considered. An algorithm to compute initial and boundary conditions when introducing an artificial potential is proposed. Some numerical test results for prescribing boundary conditions and for solving the problem of beam focusing at the region center are discussed.

REFORMING OF DIMETHYL ETHER TO HYDROGEN-RICH GAS FOR HT PEM FUEL CELLS

V.A. Sobyanin, V.A. Kirillov, S.D. Badmaev, V.D. Belyaev, V.D. Mescheryakov

Chem Eng. Trans.,
11 (2007) pp. 929-934.

Experimental results on the performance of the catalysts for DME steam reforming (DME SR) to hydrogen-rich gas and on HT PEMFC operation with pure hydrogen and reformate gas have been analyzed. Electric efficiency of the DME fuelled HT PEM FC system has been estimated. It has been found that expected electric efficiency of the system equals 38–41.5% (LHV).

CATALYTIC REFORMERS WITH MEMBRANE SEPARATION OF HYDROGEN

B.N. Lukyanov

Chem. Sustain. Devel.,
15(6) (2007) pp. 625-641.

Catalytic processes for hydrogen generation based on water-permeable membranes are described. Various types of solid membranes for hydrogen extraction from gas mixtures are considered. Design of catalytic membrane-integrated reformers is discussed to demonstrate their advantages against traditional fuel cell reactors. Relevant mathematical models are described and some modeling results on the reactors with membrane isolation of hydrogen are presented. Some prospects of employing these catalytic reactors for hydrogen energetics and for motor vehicles are analyzed.

A MULTICAPILLARY GAS CHROMATOGRAPHIC COLUMN WITH A SOL-GEL SORBENT BASED ON ALUMINUM OXIDE

Yu.V. Patrushev, Yu.G. Vervekin, V.I. Sidelnikov

Russ. J. Phys. Chem. A,
81(3) (2007) pp. 488-491.

A multicapillary gas chromatographic column with an adsorption layer based on alumina was prepared. The column allowed the high-speed separation of C1-C4 light hydrocarbons to be performed in 8÷12 s. The possibility of creating columns with various selectivities was demonstrated.

SCIENTIFIC PUBLICATIONS

JOURNAL PUBLICATIONS AND MONOGRAPHS

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   Novosibirsk, Academic Publishing House “Geo”, 2007, 206 pp.

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   Effect of $TiOSO_4$ Hydrothermal Hydrolysis Conditions on TiO_2 Morphology and Gas-Phase Oxidative Activity,
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Post-Metallocene Catalysts for Olefin Polymerisation,
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Modern Trends in the Development of Surface Science as Applied to Catalysis. The Elucidation of the Structure–Activity Relationships in Heterogeneous Catalysts,
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10. **V.I. Bukhtiyarov,**
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Catalytic SO_2 Reduction for Purification of Waste Gases of Nonferrous-Metals Industry. Technologies and Catalysts,
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Fixed Bed Reactors with Gradient Catalyst,
In *"Chemical Engineering Research Trends"*, Ed. Leon P. Berton, Nova Publishers, 2007, Chapter 3, pp. 127-201.

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Strategies for Development of a Parallel Program for Protoplanetary Disc Simulation,
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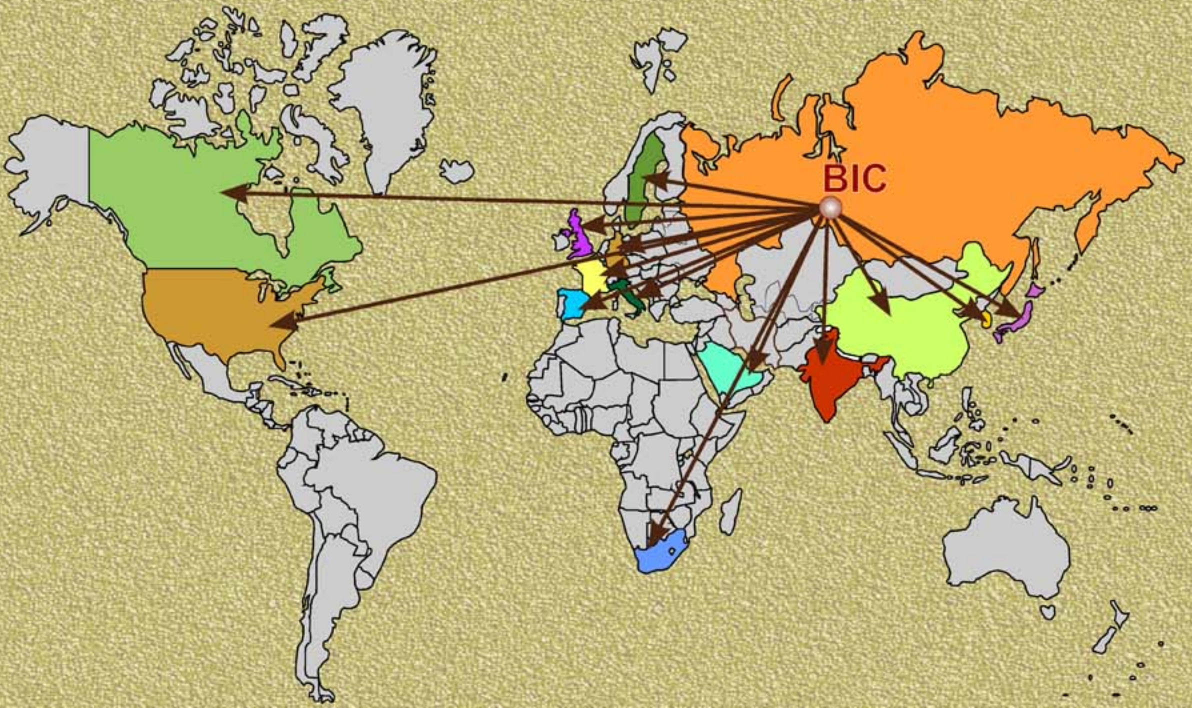

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