Laboratory of Catalysis and Gas Electrochemistry
Physical Chemistry Chair

Chemistry Department
M.V. Lomonosov Moscow State University

Edited by V.V. Lunin and S.V. Savilov

Current Achievements 2018/19

LETTER FROM THE PRESIDENT

Valery Lunin
President of the Chemistry Department, Full Member of the Russian Academy of Sciences, Dr. habil., Professor

Laboratory of Catalysis and Gas Electrochemistry has a long history of about seven decades. From the beginning all the steps to the frontier of science to move it forward were made by a strong team of world-recognized scientists inspired by their researches.
The background that was set here makes it possible to unite fundamental knowledge and valuable technologies in laboratory's daily routine.
It seems a good moment to reflect upon whether we have fulfilled to the mission given to us by society, nature, technologies and economy.
First, our scientific contribution and impact have grown rapidly. The level of publications and reports on international conferences have risen.
Second, we remain committed to bring the best research to our laboratory which is best-equipped.
Third, we still continue to build new scientists to unite our path and continue our work.
Fourth, we strive to transfer accumulated experience to industry and perform collaboration projects.
At the basis of our research - the fundamental knowledge of the professors N.I. Kobosev, Fillipov Yu.V., Maltsev A.N. and others outstanding Soviet scientists.
Thus, our aim is to bring up the world class specialists integrated in the world scientific community.
Our lab is represented by 54 members of stuff, from whom 16 are young researches.

We thank to Ms. Yulia Tambovtseva and Mr. Denis Grachev for the help in production of this issue.

CONTENT

STRUCTURE

The laboratory is currently organized into 7 research groups focused on different areas of chemistry, material science, gas electrochemistry and catalysis.
Research groups, as a key units of laboratory, develop research projects and collaborations to drive the frontier of science forward and transfer fundamental knowledge into valuable technologies.

Materials for Multifunctional Applications

- Synthesis and integration of carbon nanomaterials and polymer-based composites
- Development of new carbon 2D materials and 3D frameworks
- Development of new generation of carbon 2D materials and 3D frameworks

Ionistors, Li-ion and Na-ion batteries

- New 1D and 2D carbon nanomaterials for electrochemical energy storage
- Design and application of new high-voltage ionic liquids
- Multiscale characterization of electrode materials and processes

Catalysis and Green Chemistry

- Applications of functionalized carbon nanomaterials for Fischer-Tropsch synthesis
- Catalysts regeneration processes
- Composite materials in gas-phase hydrodechlorination
- Potassium-promoted iron catalysts

Ozone applications and atmospheric studies

- Study of biomass delignification by ozone and lignin extraction
- Physicochemistry of ozone reactions with halogen-containing compounds
- Technologies of water purification by ozone
- Atmospheric aerosols study

Advanced Ceramics

- New generations of ceramic materials
- Elaboration of new ways of synthesis
- Systematic studies of obtained ceramics
- Application of ceramics in functional devices

Supercritical Fluids

- Studies of compounds and mixtures in supercritical state
- Discoveries of new processes
- Development of new laboratory and manufacture practice

Biological tissues

- Laser technologies for tissues treatment
- Magnetic materials for anticancer therapy

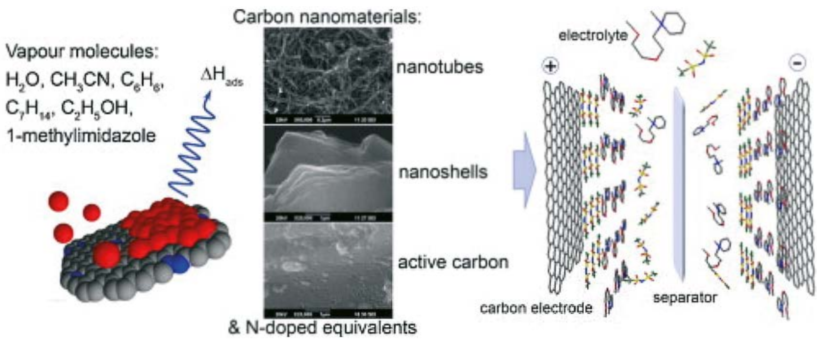
INTERNATIONAL COOPERATION

PROJECTS IN FOCUS

NEW EFFECTIVE EXPERIMENTAL APPROACHES TO SYNTHESIS OF HETEROSUBSTITUTED GRAPHENE MATERIALS WITH ADJUSTABLE PROPERTIES

Funding: Russian Foundation for Basic Research
Project period: 2017-2019
Principal Investigator: S.V. Savilov

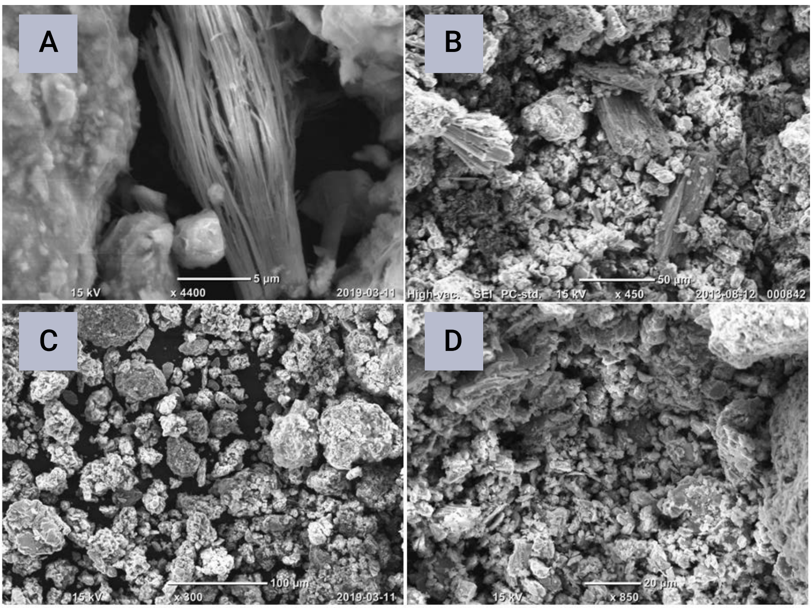
The aim of this project is to develop the new approaches for synthesis of boron- , silicon- and nitrogen-doped carbon nanomaterials, scale these processes for mass manufacturing. Heterosubstitution and surface functionalization with different types of oxygen-containing groups, metal and oxide nanoparticles are applied to adjust electrical, catalytic and mechanical properties of carbon nanomaterials and composites based on them. Application of modified materials in catalysis, new materials design and energy storage devices was studied.



NEW 2D MATERIALS AS ELECTRODES FOR ENERGY STORAGE SYSTEMS

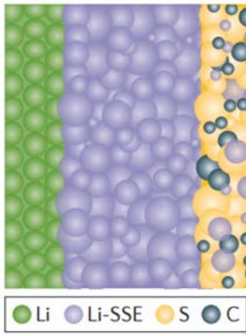
Funding: Russian Foundation for Basic Research
Project period: 2019-2021
Principal Investigators: V.V. Lunin, E.V. Suslova

The project is aimed at development of highly efficient devices for energy storage and assumes a comprehensive approach for their implementation based on various types of two-dimensional materials. The project participants have the experience in both - synthesis and characterization of electrode materials and electrolytes, as well as in the assembling and testing of electrochemical devices. Project is focused on the use of the 2D layered materials – graphene derivatives and MXenes - for supercapacitors with non-aqueous electrolytes, as well as lithium and sodium-ion batteries. The implementation of this project will allow to develop new energy storage devices with high capacitance, power and performance characteristics, increase their safety. It is important that there are a lot of young participants in the team, and getting the project will allow them to gain a foothold in the scientific community, establish close international relations.

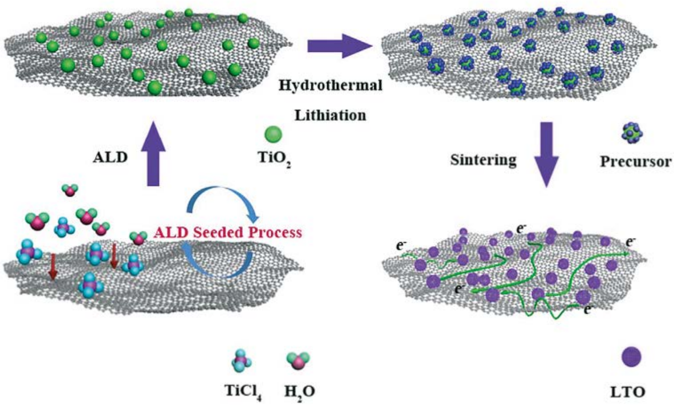


ALL-SOLID-STATE THIN FILM LITHIUM BATTERIES BASED ON ELECTRODE/ELECTROLYTE INTERFACE MODIFICATION

Funding: Russian Foundation for Basic Research
Project period: 2018-2020
Principal Investigators: V.V. Lunin, S.V. Savilov
Partners: Prof. Xia Hui, Nanjing University of Science and Technology

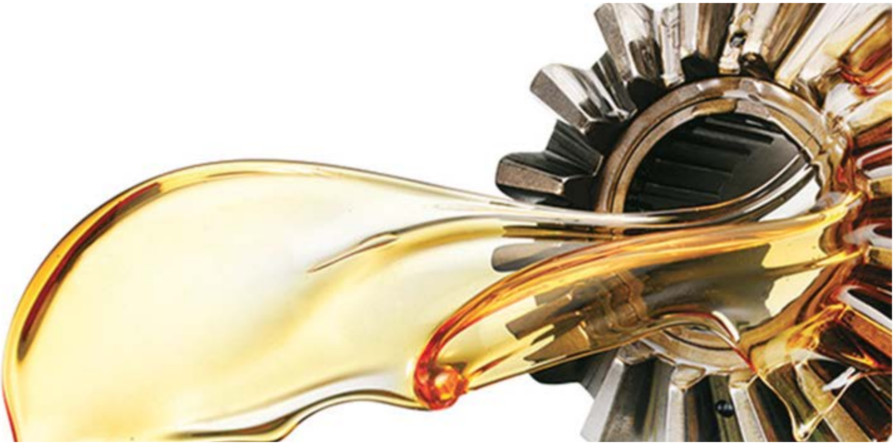


Rapid development of microelectronics influences on building up new small-sized energy storage devices with high exploitation performances. Solid-state thick Li-ion batteries have high values of energy density, safety, mechanical strength and a wide range of working temperatures from -40 to 160 °C. 3D structure of such systems promotes high values of specific surface area, high energy density and optimize Li+ transport. Combination of small size and unique properties of solid-state Li-ion batteries makes them perspective. Composition of solid batteries consists of layered cathode, solid electrolyte and anode with conductive substrate. The main difficulties in production appears with uniform layer-by-layer cathodes forming. This projects aims to develop a new approach of thick cathode films forming, study the mechanism of ionic conductivity and to underestimate an influence of buffer layers on electrode-electrolyte borders resistance. The additional goal is to optimize total cell construction for enhancing productivity and efficiency of solid-state Li-ion batteries.



DEVELOPMENT OF THE HIGH-PERFORMANCE LUBRICANTS CONTAINING CARBON NANOSTRUCTURES

Funding: Russian Foundation for Basic Research
Project period: 2018-2020
Principal Investigators: S.V. Savilov, S.Y. Kupreenko
Partner: Prof. N.V. Usol'tseva, Ivanovo State University.

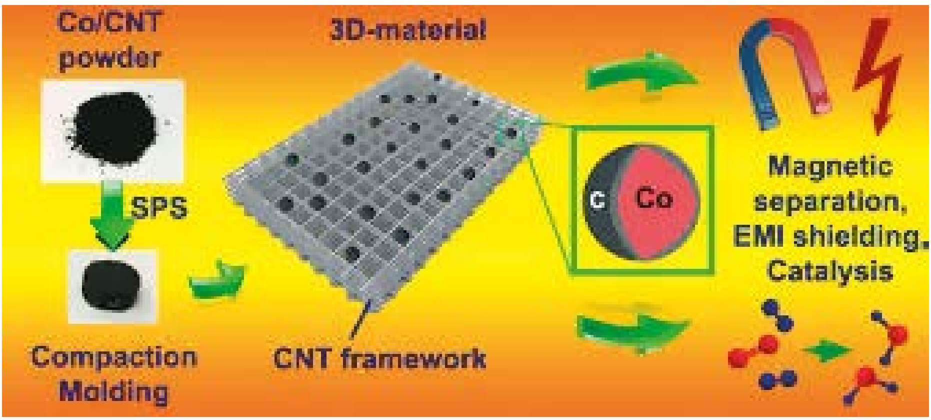


The properties of plastic lubricants highly depend on the nature of the fillers. It is important to study the influence of the structural features of natural and synthetic carbon nanostructures on the structure, rheological and tribological characteristics of lubrication systems with the aim to develop the physico-chemical basis. Since the influence of carbon nanostructures on the plastic lubricant properties depends on their interaction with other components of the system, the scientific novelty and significance of this study are determined by a comprehensive approach and use of a wide range of nanoparticles differing in their structures.
The results obtained will allow to recommend new "smart" compositions of lubricants. Taking into account the interdisciplinary nature of the project's tasks, the team includes specialists in the field of synthesis and characterization of carbon nanostructures, as well as methods for studying the physicochemical and tribological properties of plastic lubricants, and also for modeling tribological processes.

MATERIALS BASED ON NANOCARBON FRAMEWORK STRUCTURES FOR CATALYTIC INDUSTRY AND ENERGY STORAGE

Funding: Russian Science Foundation
Project period: 2018-2020
Principal Investigator: S.V. Savilov

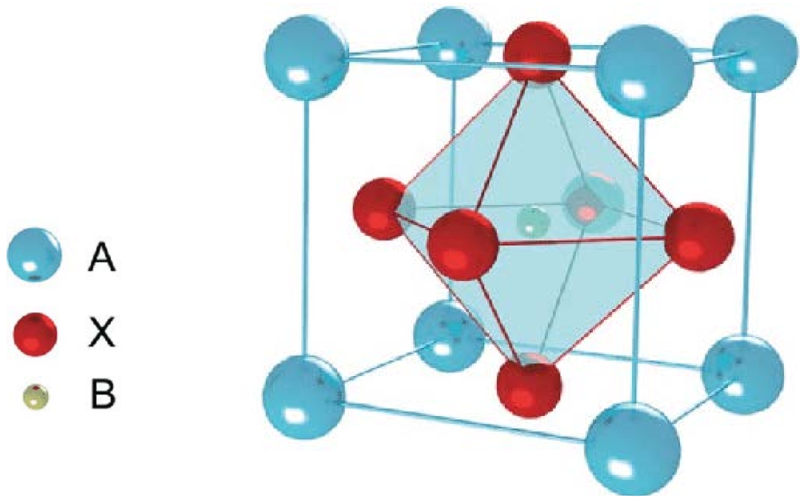
The project is aimed at the synthesis of a new type of materials, based on the framework structure with variable micro- and mesoporosity, from carbon nanotubes of different morphology and their composites with metals, oxides and intermetalides. The scientific achievements of the project team allow to claim that it is highly probable that 3D frameworks of CNTs will allow to stabilize catalytically active systems containing transition metals due to controlled defectiveness, porosity variation, preliminary oxidative conversion with further carbonisation, as well as rigid structure and chemical stability to oxidation and reduction agents. Moreover, compaction of CNTs allows solving the important technological issue which prevents practical application of CNM – the demand for the sharp increase in the CNM density and its granulation without the deterioration of the unique properties of nanotubes and few layers graphite fragments. Cooperation with the Blokhin Russian Oncology Centre allows finding new and important application of the functionilized framework materials. Their modification with oxide systems, e.g. with manganite $La_{0.7}Ca_{0.3-x}Sr_xMn_{O3-\delta}$ may leads to the synthesis of highly perspective composite material for hyperthermia. Modern synthetic approaches will be used to solve the problems of the project, such as pyrolytic synthesis of CNTs of different morphology with and without heterosubstitution, their spark plasma sintering, including their preliminary self-assembling by the cross-linking.



FUNDAMENTAL PRINCIPLES FOR THE CREATION OF THE EFFECTIVE HIGH SENSITIVE DEVICES BASED ON THE PEROVSKITE-LIKE MANGANITES

Funding: Russian Foundation for Basic Research
Project period: 2018-2020
Principal Investigator: A.N.Ulyanov

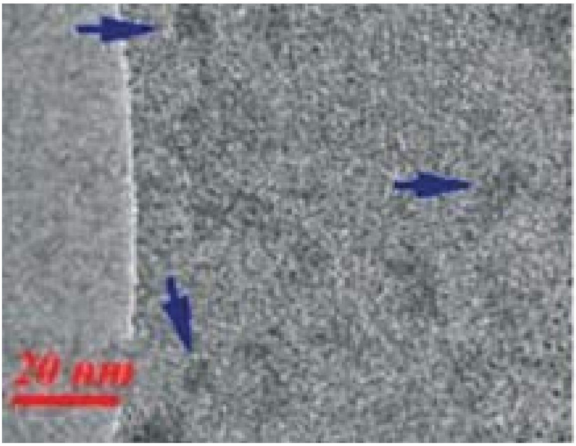
Aim of the project is to study the fundamentals of creation of high sensitive sensors based on manganites - materials showing the colossal magnetoresistance effect. Partial substitution of atoms in parent perovskite-like $LaMnO_3$ manganites causes the significant changes of properties of doped materials. The changes take place with lanthanum (A-position) and manganese (B-position) substitutions in perovskite ABO_3 cell, oxygen non-stoichiometry and with the creation of vacancies in A- and B-position in the cell. The substitutions leads to a change of phase equilibrium and phase transitions in the materials. It causes a colossal magnetoresistivity and giant magnetocaloric effect, and anomalous magnetic susceptibility at second order phase transitions as well as at the stability boundary of metastable phase at first order phase transitions.



NEW MATERIALS WITH TUNABLE PROPERTIES BASED ON UNDOPED, DOPED AND FUNCTIONALIZED CARBON QUANTUM DOTS

Funding: Russian Science Foundation
Project period: 2018-2020
Principal Investigator: S.A. Chernyak

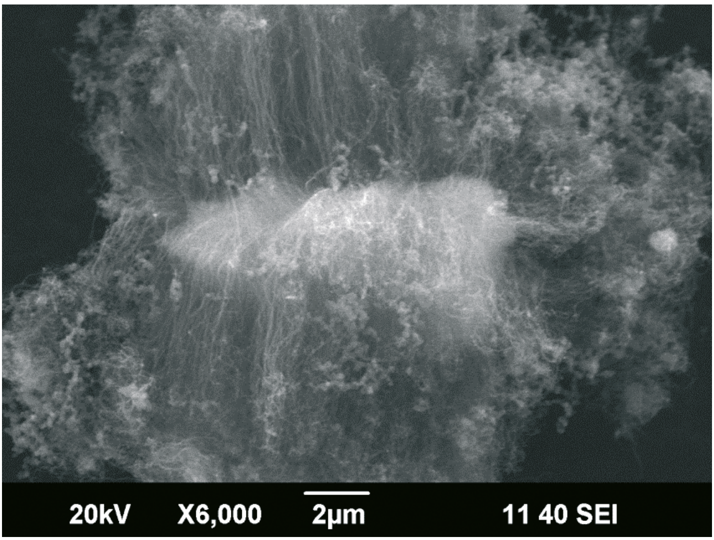
Carbon quantum dots, especially those having a graphene structure, demonstrate pronounced fluorescent properties. They are suitable for aqueous dispersions, chemically stable, biocompatible and resistant to bleaching. These features make them promising both for the use in sensors and biomarkers and for the application in photocatalysis, solar energy devices and medicine. As a result, this project should develop the synthesis technology of carbon dots with the specified optical and physico-chemical characteristics. State-of-the-art scientific equipment is used for the project needs: high-resolution transmission electron microscope, scanning electron microscope, X-ray photoelectron and Raman spectrometers, thermal analyzer with evolved gas analysis mass-spectrometer, spectrofluorimeter, and spectrophotometer.



STABILITY AND REGENERATION OF COBALT-BASED FISHER-TROPSCH CATALYSTS SUPPORTED ON CARBON NANOTUBES

Funding: Russian Foundation for Basic Research
Project period: 2018-2019
Principal Investigator: S.A. Chernyak

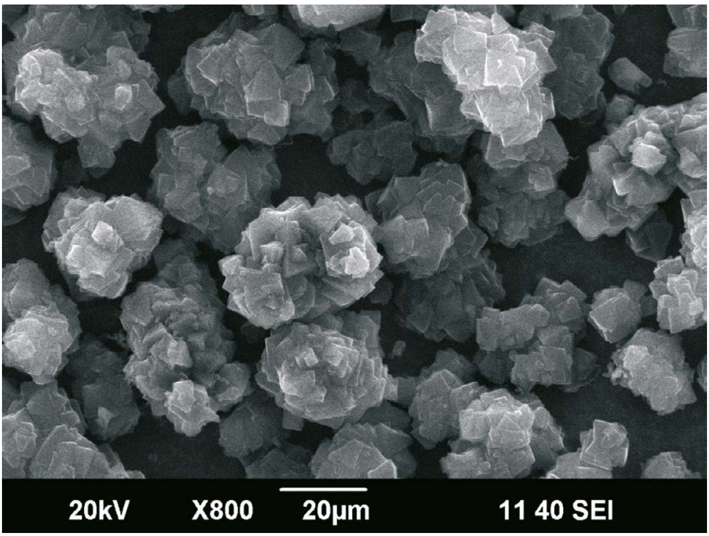
The project is aimed at studying the changes in the structure and catalytic properties of cobalt – carbon nanotubes (Co/CNT) system in the Fischer-Tropsch process. Inside it long-term stability tests of catalysts for monitoring their state by a complex of modern physicochemical approaches: transmission electron microscopy, X-ray photoelectron spectroscopy, Raman spectroscopy, XRD, magnetometry, low-temperature nitrogen sorption as well as thermogravimetric analysis with mass-spectral control of the gases. It is also planned to select the optimal conditions for the regeneration of such catalysts, taking into account the individual characteristics of the support: low stability in aggressive environments at temperatures above 400°C. Based on the data obtained, models of transformation of the Co/CNT structure during the catalytic cycle will be proposed. The results of the work should substantially clarify the possibility of using Co/CNT system in industry. They will also allow gaining the maximum benefits from the useful properties of CNT such as chemical inertness, good thermal and electrical conductivity, which determine the advantage of such supports over traditional oxide materials.



MODIFIED CARBON NANOMATERIALS FOR TOXIC ORGANIC COMPOUNDS REMOVAL

Funding: Russian Foundation for Basic Research
Project period: 2018-2019
Principal Investigator: N.E. Strokova

The principal aim of this project is the development of the modified carbon nanomaterials (CNM) for the effective adsorption of the toxic organic compounds from air. Two types of carbon structures were selected: carbon nanotubes (CNT) and carbon nanoflakes (CNF). Modification supposed to be done in two directions: implementation of nitrogen atoms to the graphene layers and surface oxidation. Acetone, ethylacetate, acetic acid, toluene and butylamine will be tested as toxic volatile organic compounds as far as their acidic and structural properties vary significantly and they are widely used in the laboratory techniques. The heat of sorption values for all adsorbent-adsorbate combinations calculated during the project realisation allow to make a conclusion about the effect of nitrogen and oxygen atoms presence in the structure of CNM onto efficiency of the absorption of organic compounds with varying structure and polarity.



DESIGN AND PROBE METHODS FOR THE DIAGNOSTICS OF POLYMERIC BIOMEDICAL MATERIALS FORMED IN SUPERCRITICAL CARBON DIOXIDE

Funding: Russian Foundation for Basic Research
Project period: 2018-2019
Principal Investigator: V.V. Lunin

The project is aimed at solving the actual problem of creating structures of a given architectonics based on biocompatible and bioresorbable polymers for tissue engineering and other biomedical purposes. To solve this problem, the approaches of several scientific disciplines: physical chemistry, chemistry of high-molecular compounds, spectroscopy, materials science, regenerative medicine, were applied. Obtaining of biocompatible matrixes basing on foaming polylactides and polylactoglycolides in supercritical carbon dioxide with simultaneous impregnation of polymers by molecules of spin and luminescent probes is expected. Using of probe methods allows to detemine mean and local concentrations, as well as distribution of probe molecules in polymer structures at macro and micro levels will be determined. The parameters of the mobility of probe molecules will be determined and the relaxation processes in the obtained matrixes will be analyzed by spin probe method. Kinetic characteristics of the degradation of polymer structures as well as transport of probe molecules from polymer matrices during their swelling and hydrolysis in vitro will be established using EPR spectroscopy and confocal fluorescence microscopy. Cytotoxic effects of the matrix with encapsulated probes will be revealed under in vitro conditions, and a morphological analysis of the bioresorption and probe distribution at the site of implantation in small laboratory animals will be performed. As a result of the project, the main principles of formation of matrixes for tissue engineering with the use of supercritical carbon dioxide will be developed.

TRANSFORMATION OF THE NASAL SEPTUM CARTILAGE INTO THE TISSUE ENGINEERING CONSTRUCTION FOR ORTHOPEDICS USING INFRARED LASER RADIATION

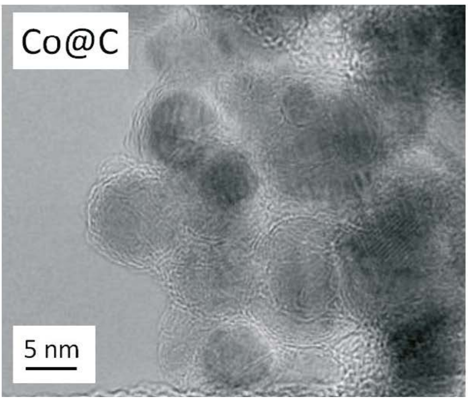
Funding: Russian Foundation for Basic Research
Project period: 2019-2021
Principal Investigator: N.Y. Ignatyeva

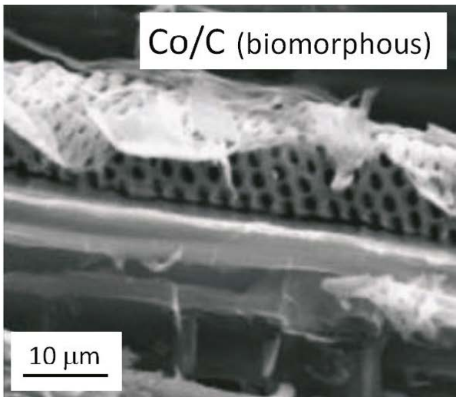
The project is aimed at developing a new approach to human tissue repair, which consists of creating a biomimic supporting structure (scaffold) based on animal tissue. We propose using cartilage of the nasal septum, including hyaline cartilage, transitional zone and perichondrium. After a complex treatment with moderate intensity IR radiation and chemical reagents, the non-immunogenic and multilayer scaffold will have the necessary elastic-mechanical properties and will provide the necessary differentiation of the cells in different parts. The effect of photothermal and photomechanical effects of IR laser radiation of moderate intensity on the damage of cellular structures in a solid matrix and on the change in the matrix of a complex composition will be evaluated.

INTERACTION OF COMPONENTS IN NANOCOMPOSITES WITH COBALT-CONTAINING CORE AND CARBON SHELL: THE INFLUENCE ON CATALYTIC PROPERTIES IN REACTIONS INVOLVING HYDROGEN

Funding: Russian Foundation for Basic Research
Project period: 2018-2020
Principal Investigator: S.V. Klokov

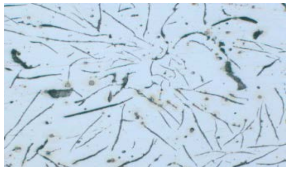
The aim of this work is to clarify possibilities and features of the catalytic use of carbon-covered Co particles in hydrodechlorination of chlorobenzene. To achieve this purpose, the comparison of two types of nanocomposite materials including Co and C will be made. In the first system Co nanoparticles are covered with layered carbon shell, in the second one they are immersed into carbon matrix. We suppose that carbon shell activated by the presence of defects and Co in subsurface layer will not prohibit catalytic action, but will be able to mitigate deactivation under the influence of hydrogen chloride which is by-product of hydrodechlorination reaction. New data about the influence of Co-C interaction on physical-chemical and catalytic properties will be obtained from the detailed study of nanocomposites using electronic microscopy, adsorption and magnetic methods, XRD, XPS and Raman spectroscopy, as well as catalytic tests in chlorobenzene hydrodechlorination and in limited scope in Fischer-Tropsch synthesis. The influence of preparation condition and modification by Pd on the Co-C interaction will be demonstrated. As a result new fundamental data on the mechanism of catalytic action of carbon-coated cobalt particles will be obtained. It will constitute the basement for future development of effective, not-expensive and stable catalysts for hydrodechlorination, hydrogenation, Fischer-Tropsch synthesis etc.





PRIVATELY-FUNDED R&D PROJECTS

INVESTIGATION OF THE CRACKS APPEARANCE AND GROWTH IN THE SHIP'S BASEMENT



Funding: SOGAZ
Project period: 2018-2019
Principal Investigator: S.V. Savilov

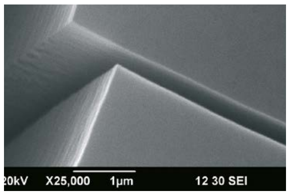
EXTRACTION SEPARATION OF LITHIUM ISOTOPS: DEVELOPMENT OF NEW TYPE OF MACROCYCLIC LIGANDS AND EXTRACTION SYSTEMS

Funding: Rosatom
Project period: 2018-2019
Principal Investigator: V.V. Lunin

COPPER WIRES CUTTING TIME DETERMINATION

Funding: Sukhoi
Project period: 2018-2019
Principal Investigator: S.V. Savilov

CHARACTERIZATION OF COMPOUNDS AND MATERIALS



Funding: Skoltech
Project period: 2019
Principal Investigator: S.V. Savilov

ADVANCED ELECTROCHEMICAL MATERIALS CHARACTERIZATION

Funding: Global SO
Project period: 2012 - 2020
Principal Investigator: S.V. Savilov

IMPROVEMENT OF THE PLASTIC LUBRICANTS PROPERTIES

Funding: Intesmo (Lukoil group)
Project period: 2018 - 2019
Principal Investigator: S.V. Savilov

OZONE DEPLETION MECHANISMS AND EARTH'S POLLUTION PROCESSES

Ozone is known as one of the strongest oxidizing agents in nature. The main areas of Its application are cleaning industrial emissions, water treatment, metal extraction from ores, oxidizing processes in chemical technology, disinfection, semiconductor manufacturing, therapy and sterilization of medical instruments, agricultural production and storage of products.

It is known that ozone can be destroyed in various ways. Photolysis of ozone is well studied and applies to reactions with non-thermal activation method.

However, the behavior of ozone at low temperatures not considered in sufficient detail, especially when adsorbed on various types of surfaces.

The importance of O_3 for atmospheric processes is also important, that determines a great interest to the fluctuations in its concentration associated with the processes of formation and destruction. Being an optically active gas, stratospheric ozone absorbs the ultraviolet radiation of the Sun, which determines thermal regime of the stratosphere, prevents the penetration of biologically active ultraviolet radiation on the surface of the planet. Photochemistry of ozone and basic atmospheric components is good studied and applied in numerical models of this Earth's gas shell. However, some halogen-containing pollutants can interact with ozone not only in gaseous, but in the condensed state. Experimental study of non-photolytic heterogeneous chemical reactions is complicated by the need for a number of approximations. Some of them are avoided due to the use of the original technology for recording IR spectra by low temperature grazing angle reflection. Its application allow to evener such experimental features of heterogeneous reactions asreagent layer thickness and its uniform distribution.

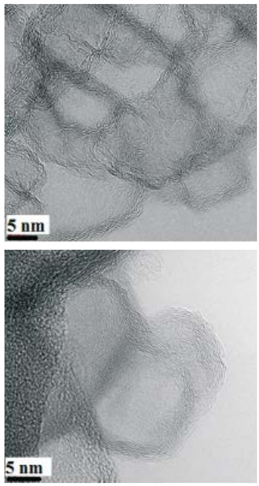
Thu support the experimental data ab initio quantum-chemical calculations are performed by group members. Based on the data obtained, a number of significant additions to classic theory of the stratospheric ozone depletion are proposed.

Scientists involved to the topic:
Lunin V.V., Savilov S.V., Zosimov A.V., Emelyanova G.I., Gorlenko L.E., Lazareva T.S., Strokova N.E., Ivanov A.S., Kupreenko S.Yu.

Open ceremony of the «Ecology hospital» in **Yuxin** (China)

CARBON NANOMATERIALS SYNTHESIS, INVESTIGATION AND APPLICATIONS

A scientific team of the leading researcher Dr. habil. Serguei V. Savilov provides a wide range of researches connected with carbon nanomaterials and their applications. Besides he takes part in various collaboration projects with universities all over the world. The continuous growth of the scientific interest to carbon nanomaterials (CNMs) deals with their application perspectives in different spheres on industry. Numerous of studies are devoted to their synthesis and investigation of physico-chemical properties as well as experimental search for the materials with the optimal parameters for certain areas. Nevertheless, only small part of researches is devoted to their fundamental characteristics. That is why the data on correlations between structure, composition and thermophysical parameters of different types of CNMs sometimes are contradictory. Group members summarize them as well as provide original experimental results demonstrating the dependence of burning temperatures, heats of combustion and enthalpies of formation on structure, type and amount of functional groups of CNMs. Original experimental results on thermochemical and structural features as well as surface chemistry of different types of carbon-based nanostructures were obtained by adiabatic bomb calorimetry, thermal analysis, XRD, NMR, EPR, HRTEM, XPS and Raman spectroscopy. It was demonstrated for the first time that heats of formation and burning temperatures are extremely sensitive to the structure, specific surface area, heterosubstitution as well as presence of functional groups and adsorbed species. They also provide the wide practical use of CNMs. Group members applied them as an effective fillers in polymer composites and plastic lubricants, component of metal alloys, catalysts, electrode materials for supercapacitors, ionistors and metal-ion batteries.



Scientists involved to the topic:
Lunin V.V., Savilov S.V., Suslova E.V., Chernyak S.A., Ivanov A.S., Maximov S.V., Egorov A.V., Burtsev A.A., Efremova O.S., Egorova T.B., Kupreenko S.Yu., Strokova N.E., Novotortsev R. Yu, Dvoryak S.V., Arkhipova E.A., Kuznetsova N.N.

CARBON NANOTUBES AND GRAPHENE NANOFLAKES FOR CATALYSIS APPLICATIONS

Research interests of Senoir Researcher Dr. Sergei Chernyak coud be divided in three areas:

I. Long-term study of Co-based Fischer-Tropsch catalysts supported on oxidized carbon nanotubes (CNTs).

High stability of this system was observed: CO conversion did not decrease during 3 weeks of catalytic tests. TEM monitoring of Co particle size during the experiment revealed that metal crystallite diameter grew at the induction period of 2-3 days from less than 4 nm to optimal value of 8-15 nm. Such increase promotes the high activity and selectivity of the catalyst.

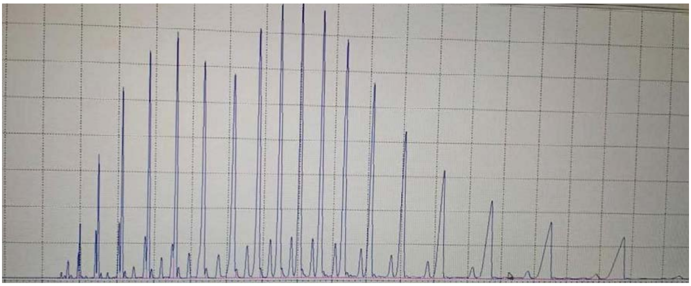


Fig.1 shows the chromatogram of liquid fraction obtained during the Fischer-Tropsch synthesis (FTS).

II. Effect of nitrogen doping of graphene nanoflakes (GNFs) on the structure and catalytic properties of Co/GNF FTS catalysts.

It was found that N-doping of carbon support by pyrrolic, pyridinic, and graphitic nitrogen increase catalyst activity by 2-3 times. XPS studies of initial N-GNFs, annealed and reduced catalysts allowed us to determine the mechanism of thermal transformations of nitrogen functionalities.

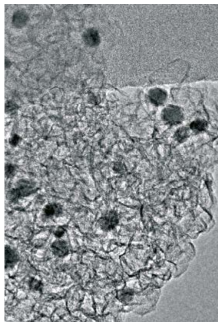


Fig.2. TEM image of Co/N-GNF catalyst.

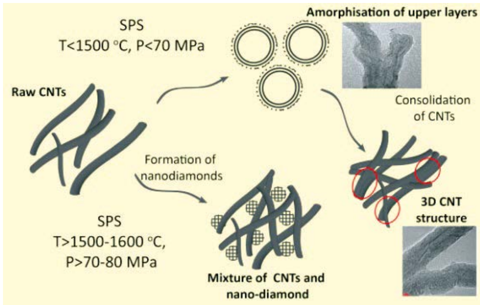
III. Synthesis of catalytically active materials using spark plasma sintering technique (SPS).

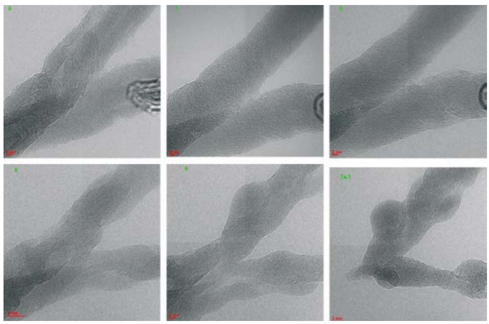
Metal-CNT frameworks contained Co, Fe, Ni, and Cu were obtained by SPS treatment of CNTs decorated with nanoparticles of appropriate metal oxides. Metallic phase formed during the synthesis because of oxide reduction by carbon and graphitic shells and were observed on the nanoparticle surface after the experiments. It was shown that metal addition increases the electrical conductivity of CNTs. Moreover, resulted samples demonstrated ferromagnetic properties. Co-based structures exhibited noticeable catalytic activity in FTS and carbon shells around the metal particles allowed us to skip the catalyst reduction stage.

3D FRAMEWORKS OF CARBON NANOMATERIALS

Senior Researcher Dr. E.V. Suslova works on creation and investigation of 3D frameworks of carbon nanomaterials as well as study of the thermochemical properties. She adapts synthesis conditions and concentrations of frameworks' precursors.

Group members study the application of spark plasma sintering (SPS) to multiwalled carbon nanotubes and nanoflakes, which was found to be an effective route to their compactisation for further use in catalysis and as electrode materials foe energy storage devices. It was found that the increase of temperature and pressure during SPS increased the density of the sintered samples and decreased their surface area. It is accompanied by appearance of the mesopores. Raman spectroscopy, TG and X-ray data show that the defectiveness of the CNTs decreased during SPS. Using a focused beam of the transmission electron microscope in-situ experimental simulation of the CNT consolidation and their crosslinking was successfully performed together with Dr. A.V. Egorov for the first time.

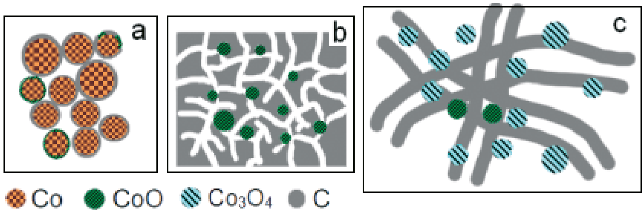




Together with colleagues she performed systematic investigations and stressed sufficient correlations between structure, composition and properties of carbon nanomaterials and their N-doped derivatives. The adiabatic bomb calorimetry technique was used for determination of these values, together with elemental and thermal analysis, electron microscopy, Raman and X-ray photoelectron spectroscopy applied for structure and composition analysis. The contributions of surface and bulk components in the values of enthalpies of formation for different CNMs were estimated for the first time. It is shown that the first one is highly influenced by the surface area while the latter — is defined by the number and homogeneity of inner layers, conformable to graphite structure. In the case of nitrogen-doped CNMs heat of formation is influenced not only by the nitrogen content but by coordination of heteroatom; substitutional nitrogen demonstrate higher effect comparing to pyrrolic and pyridine-like ones.

DEVELOPMENT OF COMPLEX CATALYSTS FOR HYDRODECHLORINATION OF CHLOROBENZENE AND CO OXIDATION

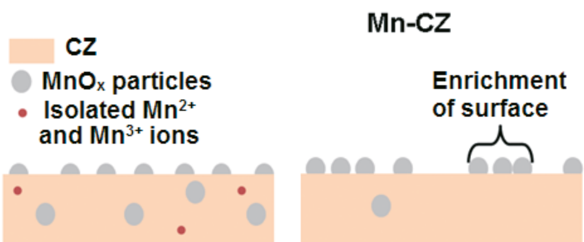
Leading researcher Prof. Ekaterina Lokteva and Associate professor Dr. Elena Golubina work with different composite materials including cobalt and carbon, comparing their properties in gas-phase hydrodechlorination of chlorobenzene in flow-type fixed-bed system. The role of different oxidized Co particles in catalytic activity was revealed.

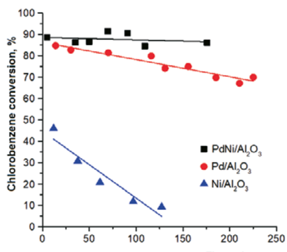


The scheme of structure of Co@C (a), Co/C (b) and Co/CNT (3) composites on the base of TEM, XPS and TPR data

Scientists involved to the topic:
Lokteva E.S., Golubina E.V., Klokov S.V., Kharlanov A.N., Fionov A.V., Voronova L.V.

For the first time bimetallic NiPd systems with average particles size of 1 nm synthesized by laser electrodispersion (LED) were tested in hydrodechlorination reaction. The NiPd/Al2O3 bimetallic catalyst at 150–350°C is superior in gas-phase hydrodechlorination of chlorobenzene not only to nickel, but also to palladium monometallic analogue, as a result of the formation of new bimetallic active centers at the contact of Pd0 and NiAlOx.
Oxide catalysts Ce0.8Zr0.2O2 (CZ), MnOx-Ce0.8Zr0.2O2 (Mn-CZ), CeO2 and MnOx were produced by co-precipitation from the corresponding salts solution using CTAB as the template, and tested in CO total oxidation. Both catalysts despite the big difference in SBET values are nearly equally effective in the wide temperature range in CO oxidation.



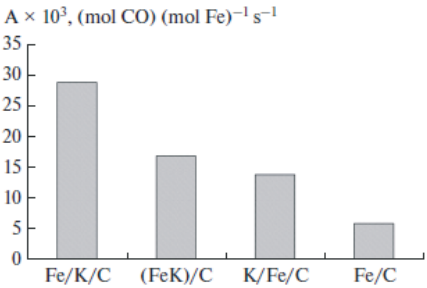


Chlorobenzene conversion vs reaction time at 200oC in the presence of mono- and bimetallic LED catalysts.

METAL-SUPPORTED FISCHER-TROPSCH SYNTHESIS CATALYSTS

During the last 5 years a research team under Prof. Peter Chernavkii supervision has been researching some of topochemical processes accompanying the synthesis and operation of metal-supported Fischer-Tropsch synthesis catalysts. A number of new results were obtained.

It was found out that the sequence of the supporting of active components onto potassium-promoted iron catalysts affected a number of their physicochemical and catalytic properties.

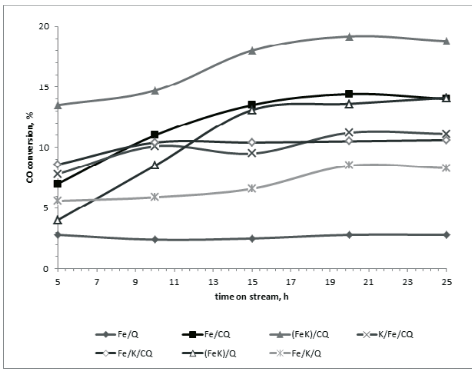


Dependence of the specific activity on the type of catalysts

Scientists involved to the topic:
Chernavsky P.A., Pankina G.V.

Moreover., this team is working on developing a new direction in the synthesis of composite materials based on traditional carriers and carbon. New carbon-silica composite support was prepared by soaking of silica in water solution of glucose followed by drying and calcinations in inert atmosphere. This material was successfully used for the preparation of iron Fischer–Tropsch synthesis catalysts.

Dr. Chernavskii suppose that for the composite support the extent of iron silicate formation is less since at least some of the iron oxides are initially located on the carbon substrate. Additionally, the presence of carbon in the support promotes conversion of iron oxides to carbides. Carbon-silica composite support has a beneficial effect on catalytic activity of iron in CO hydrogenation. Other advantages of the new catalysts are suppression of methane formation and higher value of chain growth parameter. In fig. below CQ means composite material C/SiO2.

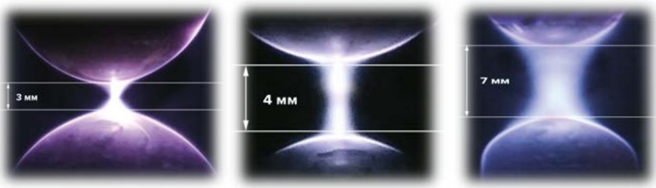


Carbon monoxide conversion as function of time on stream.

FILAMENTARY AND DIFFUSE BARRIER DISCHARGES IN NOBLE GASES WITH ADMIXTURES OF MOLECULAR GASES

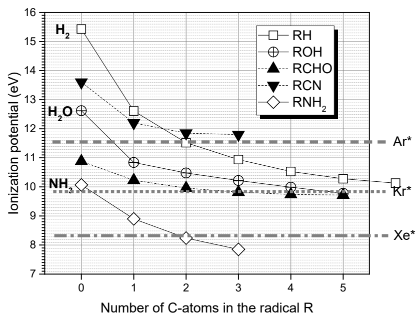
Recently Dr. Kozlov K.V. and Abramovskaia E.A. reported the results of the plasma diagnostics of the barrier discharges in argon with admixtures of acetone. These discharges were found to be diffuse only for a limited range of acetone concentration, the latter corresponding to the local minimum of the burning voltage. In the year 2018, they have proposed a semi-empirical physical model for the barrier discharges in inert gases with small admixtures of molecular gases, that accounts for the experimental findings mentioned above, and that provides a simple method to prognosticate the possibility of the diffuse mode appearance for any chosen combination “noble gas + molecular gas (admixture)”.

The model is based on the assumption of the Townsend mechanism of initial (pre-breakdown) phase of the microdischarge development and of the validity of the Paschen law for the breakdown voltage. Certain additional assumptions concerning electron energy distribution functions in the gas mixtures under consideration should be made. Then qualitatively, our model is able to explain the experimentally observed dependencies of the burning voltage upon the content of molecular admixture, as well as a transition of the discharge to the diffuse mode within a certain concentration range of molecular gases.



Transition from the filamentary mode to the diffuse mode of the barrier discharges in argon with admixtures of ethanol. Concentrations of ethanol: 0.005% (left), 0.06% (center), 0.5% (right).

To explain the latter exception, as well as an absence of the diffuse mode in the barrier discharges in humid argon, we assumed that the diffuse mode is impossible if ionization potential of the molecular admixture exceeds the energy of the metastable levels of argon, and included the corresponding limitation into the proposed physical model. Schematically, this hypothesis is presented in fig.2. For all the points located below the dashed line, small admixtures of the corresponding chemical compounds to argon can cause a transition of the filamentary barrier discharge to the diffuse mode.



Comparison of the ionization potentials of selected inorganic and organic compounds with the energies of the lowest metastable excited states of argon, krypton and xenon.

CATALYTIC METHOD FOR ASSESSING THE EFFECT OF THE CARBON MATRIX STRUCTURE ON THE FORMATION OF THE COMPOSITION OF THE SURFACE FUNCTIONAL GROUPS OF CARBON

Dr. Yu. N. Zhitnev and Dr. E. A. Tveritinova work on application of oxidized carbon nanomaterials in catalysis.

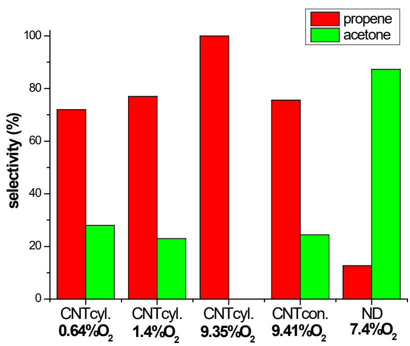
Using a pulsed microcatalytic method they study the influence of the CNM carbon matrix structure on the formation of the composition of surface functional groups formed during postsynthetic processing was studied. They test the conversion of alcohols among the catalytic reactions to idetify the content of acid and basic Lewis centers, which are the functional surface groups of CNM.

Dr. Yu. N. Zhitnev and Dr. E. A. Tveritinova focus on nanodiamonds (ND), carbon nanotubes (CNT): cylindrical CNT (CNT cyl) of various oxidation states and CNT conical (CNTcon) investigations.

They claim that the structure of the carbon matrix of the investigated CNM significantly affects the nature of the surface groups, which affects the composition of the products of the catalytic conversion of propanol-2.

The picture below shows the selectivity to propene and acetone for the conversion of propanol-2 at 270°C on ND, CNTcon and CNTcyl with different oxygen content. CNTcyl.

Thus, the use of a simple catalytic method allows not only to characterize the nature of the surface oxygen-containing groups, to regulate their composition, but also to establish the influence of the carbon structure on the formation of the surface composition of CNM due to the post-synthetic oxidative treatment.



FINE-CRYSTALLINE MATERIALS FOR CERAMICS PRODUCTION

Leading Researcher M.N.Danchevskaya, Dr. V.A.Kreisberg and Dr. Y.D.Ivakin study the quality of ceramics that could be obtained with original method of synthesis of fine-crystalline oxides in hydrothermal conditions in sub- and supercritical water fluids with the help of which fine-crystalline simple and complex oxides were obtained. The content of water and gases in the crystalline raw materials for the production of high-quality ceramics are the most important criteria for the quality of ceramics. Using the method of kinetic thermodesorption mass spectrometry, it was possible to quantify the gas content, gas release and diffusion characteristics of alumomagnesium spinel for the first time. This allows them to estimate the degassing time of the spinel with a certain degree of dispersion and ceramics made of it, and thereby promote the production of high-quality ceramics.

The kinetic curve of water evolution of the spinel sample, which is shown in **Fig. 1,** consists of the regions of a sharp increase in the water flow with a rapid increase in temperature and isothermal sections, when the flow of water released falls. The temperature dependence of the coefficient of water diffusion for spinel at temperatures of 500, 600 and 700 °C is represented in the Arrhenius coordinates in **Fig. 2.** The activation energy of water diffusion E is 146 ± 5 kJ/mol. Fig. 2 also presents the results of previously studied fine-crystalline samples of undoped corundum and quartz.

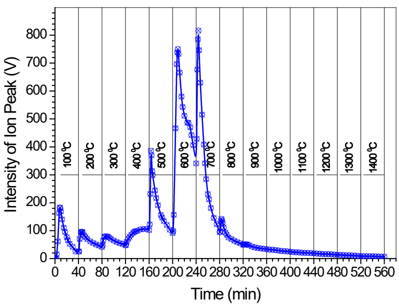


Fig. 1. Kinetics of the change in the intensity of molecular peak of water during stepwise vacuum heating of the weighted (0.1275 g) fine-crystalline spinel in mass spectrometric experiment at a sensitivity of 1 V = 2.03 · 10-4 μg H2O/s.

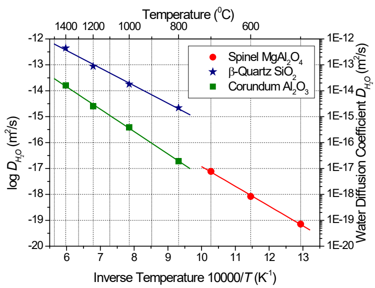


Fig. 2. Temperature dependence of the water diffusion coefficients for spinel, β-quarts and corundum.

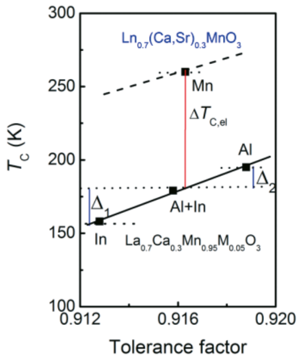
Scientists involved to the topic: M.N.Danchevskaya, Dr. V.A.Kreisberg, Dr. Y.D.Ivakin, Dr. habil. B.S. Lunin, Dr. A.V. Kholodkova, Dr. G.P. Muravieva

A- AND B – SITE SUBSTITUTED PEROVSKITES LIKE ABO_3 MANGANITES, AND SELF- (OR VACANCY) DOPED $Ln_{1-X}MnO_{3+\Delta}$ OXIDES

Doped $Ln_{1-x}D_xMnO_3$ (D= alkali-earth metal) and self-doped $Ln_{1-x}MnO_3$ manganites show

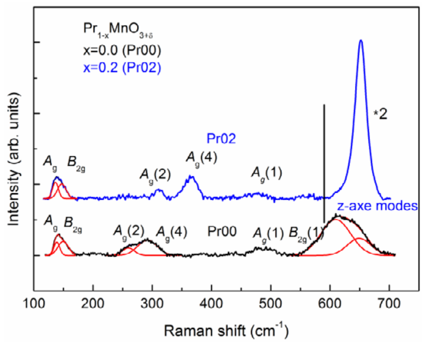
- Colossal magnetoresistivity effect;
- Large magnetocaloric effect.

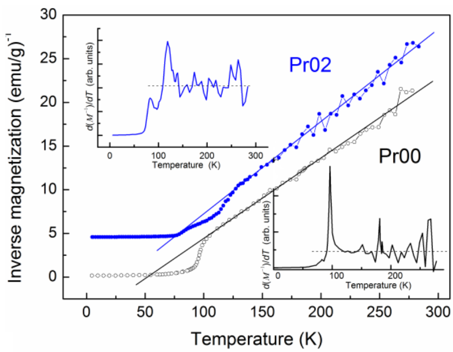
Unusual phonon modes and Griffiths singularity (ferromagnetic phase in paramagnetic matrix) are revealed in the materials. Here we describe the change of properties of manganites caused by the doping and vacancies.



Substitution of Mn3+ by Al3+, Ga3+, In3+ Sc3+ decreases Curie temperature (Tc) and increases or decreases Tc in dependence on radii of substituent ions.

Vacancies cause an appearance of unusually high Raman peaks and Griffiths singularity.





PHYSICOCHEMICAL STUDY OF PLANT BIOMASS DELIGNIFICATION BY OZONE

Present topic is developed by Senior researchers Dr. E.M.Benko, Dr. N.A.Mamleeva and leading researcher Dr. habil V.I. Bogdan. The aim of the study is to elaborate an effective and environmentally friendly method of delignification plant materials by ozone in relation to the subsequent fermentation in sugars and ethanol.

For these purpose the effect of various parameters on the kinetics of ozonation of lignin-cellulose material (LCM), structural changes in samples of ozonation, reactivity of ozonated samples in enzymatic hydrolysis processes under the cellulase complex are studied.

A summary of the obtained experimental data makes them to draw conclusions on the optimal ozone usage for plant raw materials:

• Moisture content in the sample is a key factor for the ozone consumption rate;
• Content of residual lignin and sugars yield are governed by ozone consumption;
• Ozonation of biomass causes a degradation of lignin and partially hemicelluloses and cellulose;
• Ozone treatment leads to noticeable structural changes of plant materials;
• The process of lignin removing in the course of ozonization is directed from the secondary to the primary cell shell.
• There are common patterns of ozone pretreatment for biomass of different types. This allows us to predict the optimum dose of ozone for biomass pretreatment. (2- 3 eq. O_3/PPU). At an average content of lignin in plant materials 25-35% the required amount of ozone (O_3 : LCM) corresponds to 10-15 wt. %.

STUDY OF OF OZONE REACTIONS WITH CRYSTALLINE HALOGENIDES

Associate professor Dr. A.V.Levanov and Senior Researcher Dr. O.Y.Isaikina work on the sources and mechanism of activation of bromine in the troposphere that can interact with ozone while being in dry or wet marine aerosol containing bromide ions. The purposes of this study are to investigate the interaction of gaseous ozone and O_3 + CO_2 mixtures with polycrystalline potassium bromide, determining the composition of products released into the gas phase and remaining in the solid phase, establishing quantitative kinetic patterns of their formation, and to identify a possible mechanism for this process. It was found that $KBrO_3$ was the only nonvolatile product of the interaction of solid KBr with gaseous ozone, and the kinetic laws of its formation were determined.

It was discovered for the first time, that $KHCO_3$ and $KBrO_3$ are formed in the solid phase, and molecular bromine Br2 is released into the gas phase, when solid KBr interacts with gaseous ozone in the presence of significant amounts of carbon dioxide.

$2KBr(cr.) + 2CO_2(g.) + H_2O(g.) + O_3(g.) \rightarrow 2KHCO_3(s.) + Br_2(g.) + O_2(g.)$.

A kinetic scheme has been proposed that satisfactorily explains all the kinetic patterns established in the experiments. The formation of Br_2 and bicarbonate is due to the interaction of hypobromite ion, the primary oxidation product of bromide, with carbon dioxide and water, according to the equation

$BrO^- + 2CO_2 + H_2O + Br– \rightarrow Br_2 + 2HCO_3^-$.

First reaction can be considered as the primary source of active bromine in the troposphere. In atmospheric conditions, its rate does not depend on the concentration of carbon dioxide (as it is in considerable excess), but directly proportional to the concentration of ozone.

OZONE APPLICATION FOR WATER TREATMENT

Scientific interests of Professor Sergey N. Tkachenko and Dr. Ilia S. Tkachenko are physical chemistry, kinetics and catalysis, ozone production and application, homogeneous and heterogeneous ozone decomposition, water treatment, ozone-catalytic oxidation, air treatment, ozone technologies.

New technologies have been developed in the group to clean water and air from man-made contaminants, which effectively use ozone in combination with other methods. Creation of systems for removal and destruction of residual ozone after various technological processes. Carrying out of physical and chemical studies of the developed series of high-efficiency catalysts for ozone decomposition and oxidation of toxic organic compounds (goptalyms of goptalyum catalysts GT and GTT grades with different content of oxides of transition metals of different compositions), including the reaction of complete oxidation of methane and oxidation of carbon monoxide. Investigation of ozone synthesis by surface barrier discharge by numerical simulation method. All research and developments take into account the application of «green chemistry» principles.

Recently developed goptalyum catalysts of GT and GTT brand have been put into operation and work effectively at more than 50 enterprises of the Russian Federation and abroad. Ozone-sorption station of water treatment for purification of underground waters of compounds of iron on JSC WIMM-BILL-DANN (PepsiCo); Residual ozone removal and destruction system at Zapadnay and Rublevskay water treatment stations in Moscow are designed, produced and operated. The goptalum catalyst of GTT grade works effectively at the ATLAS device in the Grand Hadron Collider, CERN, (Geneva).

In 2018 Lunin V.V., Tkachenko S. N., Tkachenko I.S. with colleagues were the laureates of the Russian Government Prize in science and technology.

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FACILITIES

Laboratory of Catalysis and Gas Electrochemistry has got all the necessary equipment and facilities to synthesize, characterize and test new advanced materials and devices based on them. We are also aimed to integrate them to lab-scale prototypes and experimental production lines.

Microstructural and chemical characterization	Synthesis	Catalysis tests	Functional properties
• Raman spectroscopy • X-rays photoelectron, Auger and secondary ion spectroscopy • High-resolution TEM with GIF • SEM • Elemental analysis • Atomic absorption and laser emission spectroscopy • IR spectroscopy • XRF microanalysis • X-Rays diffraction	• 0D, 1D, 2D and 3D carbon nanomaterials • Catalysts • Supercapacitors and metal-ion batteries • Non-aqueous electrolytes • Ionic liquids • Anti-cancer agents • Silicon based compounds from agricultural wastes • Hydrogels • Pectin and fibers • Amorphous SiO2	• Flow-type fixed bed reactor units • Batch-type reactor units • Pulse reactor units • Catalytic cell in-line with XPS spectrometer • High pressure catalytic sytem	• Thermal properties • Electrochemical characterization • Mechanical tests • NMR spectroscopy • EPR spectroscopy • Magnetic properties • Adsorption of gases and liquids • Glove-boxes lines

EQUIPMENT

IR Fourier Spectrometers

IR Fourier Spectrometers

Low-temperature differential scanning calorimeter

Elemental analyzer

Simultaneous thermal analysis (TG+DSC) system with quadruple mass and IR spectrometers for gases detection

EDX elemental analyzer

Confocal microscope with implemented video-capture system

Boilogic and NEWARE electrochemical systems

EQUIPMENT

Scanning Electron Microscope JEOL JSM-6480 LV

High-Resolution Transmission Electron Microscope JEOL JEM-2100F/Cs/GIF

Dynamic Vapour Sorption System SMS-UK DVS Advantage

Spectro-Systems Glove-Box lines

Kratos AXIS Ultra DLD X-ray photoelectron spectrometer

Testing Machine Trilogica TTM-50

Surface Analyzer Quantochrome Autosorb 1C/MS/TPR

Bruker ELEXSYS E500 Electron Paramagnetic Resonance Spectrometer

Powder diffractometer STOE STADI-P

Mettek gas analyser

High Perfomance Liquid Chromatograph Agilent 1100

Ion Chromatograph

Gas Chhromatograph with mass-spectrometer

Laser emission spectrometer

FOR NOTES

Laboratory of Catalysis and Gas Electrochemistry

Physical Chemistry Chair

Chemistry Department

M.V. Lomonosov Moscow State University

2018/19