

Current state of investigations in surface chemistry, adsorption and ion exchange in Ukraine

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1. INTRODUCTION

In the history of modern Ukrainian science, the studies of surface phenomena always were of high priority. It can be mentioned, for example, that yet at the beginning of the century, the interaction of various electrolytes, oxy-acids and urea with colloid hydroxide of iron and hydroxides of other heavy metals were extensively studied in Kyiv Polytechnic Institute by Academician A.V.Dumansky [1,2]. B.A.Szyszkowski, who was a lecturer in Kyiv University, had in 1908 proposed the well-known equation, which describes the decrease of surface tension in water solutions of surfactants with the increase of the concentration [3]. This equation, expressed in the differential form, can be used to transform the Gibbs' adsorption equation into the Langmuir equation, which describes the monomolecular adsorption [4].

In 1920s, one of the founders and managers of Nosivka Agricultural Experimental Station (1913-1928), Chernigiv province of Ukraine, Academician K.K.Gedroitz, had performed a series of fundamental investigations in the ion-exchange properties of soils [5], which formed a basis for the branch of science which deals with the colloid chemistry of soils. In early 1930s, M.V.Polyakov, employed as a scientific worker (subsequently the professor) in the Institute of Physical Chemistry of Ukrainian National Academy of Sciences (UNAS) had undertaken basic studies concerning the formation of silica gel porous structure in the presence of vapours of organic substances - benzene, toluene, and xylene, and the adsorption properties of such substances [6]. In particular, the so-called memory effect was discovered, that is, the increased selectivity of adsorbents and ion exchangers with respect to the molecules and ions, which were present in the process of the structure formation. It is now clear that M.V. Polyakov had

anticipated the concept of template synthesis of selective adsorbents and catalysts which is currently developed.

In 1950s, the method of immersion heats had become widely used by Academician A.V.Dumansky and his followers in the Institute of General and Inorganic Chemistry (IGIC) UNAS for the estimation of energy characteristics of the interaction of water with various disperse materials - clays, cellulose, gelatine, starch etc. These studies, which were summarised in the monograph [7], parallel to the works performed by A.C.Zettlemoyer in the USA [8], had laid the foundations of the thermochemical method used to investigate the interaction of the molecules of various liquids with the surface of disperse solids.

The new stage in the Ukrainian studies of adsorption and ion exchange is related to the works performed by Academician F.D.Ovcharenko (IGIC), Professors I.E.Neymark and D.N.Strazhesko (Institute of Physical Chemistry). F.D.Ovcharenko had studied the adsorption properties of clays and clay minerals possessing various structure and composition, envisaged the methods for the control of properties of such materials and the areas of their technological applications [9].

The methods for the synthesis of silica gels and other inorganic gels with predefined porous structure were developed in the laboratory headed by I.E.Neymark; these pioneering works had enabled him and his co-workers to synthesise (for the first time in Ukraine and in the former Soviet Union) various zeolites, combined zeolite/oxide systems, organosilicone adsorbents. Studies performed by Prof. I.E.Neymark and his scientific school are summarised in the publications [10-13].

In the studies performed by D.N.Strazhesko, the electrochemical theory of adsorption of ions by active coals was successfully developed [14]. In his works [15,16], which were performed in the co-operation with I.A.Tarkowska and other co-workers, resulted in the development of methods for the preparation of oxidised coals, investigation of their adsorption and ion exchange properties, and their application for the efficient cleaning of various substances, in particular, the salt systems. Also, in the laboratory headed by D.N.Strazhesko, the cation exchange on silica at relatively low pH values (2-5) was studied [17], which formed a basis for the practical application of silica gel in the radiochemistry [18]. In the high pH values range (> 9) the inversion of the selectivity series for the alkali cations at silica gel surface was discovered [18,19]; the series was shown to be: $K^+ < Na^+ < Li^+$.

The scientific schools founded by these scientists had succeeded in the development of the adsorption science in the Institute of Physical Chemistry, Institute of Surface Chemistry, Institute of Colloid Chemistry and Chemistry of Water, Institute of Sorption and Problems of Endoecology, Institute of Biocolloid Chemistry UNAS. In addition to these institutions supervised by the Ukrainian National Academy of Sciences, the studies in various fields of adsorption, surface chemistry and ion exchange are currently performed at chemi-

cal faculties in a number of Ukrainian universities, and by several applied chemical science institutions supervised by the Ukrainian Ministry of Industrial Policy. Summarised below are the most important results obtained by Ukrainian scientists, with respect to the particular direction of studies.

2. SURFACE CHEMISTRY AND CHEMICAL MODIFICATION OF DISPERSE SOLIDS

The studies performed in the Institute of Surface Chemistry (ISC UNAS) headed by Academician A.A.Chujko had resulted in an extensive experimental material concerning the structure of surface layer of silicone dioxide, its active centres, the peculiar features and mechanisms of the chemisorption processes which take place at silica [20-23].

The analysis of the electrostatic potential spatial distribution profile created by SiO_2 structure had shown that the areas with high positive values of this potential exist in the vicinity of silicon atoms bearing isolated (terminal) hydroxyl groups [24]. These atoms, according to the views of this scientific school, are co-ordinately unsaturated, and act as the primary adsorption centres for small polar molecules.

It was shown that the physicochemical characteristics of the surface of SiO_2 and other oxides are determined not only by the properties of local active centres, but also by the co-operative behaviour of the ensemble formed by the terminal functional OH-groups. The theory is developed for the rotational mobility of such groups, which takes into account their mutual interaction and the interaction with the substrate [25].

High-disperse silicas are synthesised which contain oxides of boron, aluminium, phosphorus, 3d-metals, see e.g. [26]. Also, another approach to the preparation of multicomponent oxide systems, namely, the lay-up of the oxides of other elements at the SiO_2 surface [20,27] is developed in the Institute.

The scientists working in the ISC had performed comprehensive studies related with the kinetics and mechanism of the reactions of various organic and element-organic compounds with the active centres existing at the surface of disperse silica, which had led to the revealing of main principles of the chemisorption processes that take place in the surface layer [28]. These studies had identified the main factors which affect the rate of electrophilic substitution of proton in the structural silanol groups. It was shown that in the transition complex, which incorporates the surface centre and the attacking molecule, the withdrawal of leaving group can be promoted by external factors, related to electron-donor or proton-donor compounds. In the Institute, for the first time ever, the studies were performed of a number of chemisorption processes involving the oxy-chlorides and chlorides of some elements and silica structural OH-groups, accompanied by the restructurisation of forming surface compounds.

A rather generalised methods for the activation of surface functional organo-silica were proposed, capable for the soft-condition binding of various organic ligands and ligand-based metal complexes, important organic reagents, ferments and other biologically active compounds [29]. The adsorption, ion exchange and catalytic properties of inorganic matrices possessing chemically-fixed compounds containing oxygen, nitrogen, sulphur and some macrocycles were studied [30]. The solids modified in such a way were used as the basis for the specific adsorbents, promising supports for the chromatographic separation of ions and molecules, and efficient heterogeneous metal-complex catalysts. The studies of the equilibrium for the adsorption from solutions at modified silica surface lead to the evaluation of optimum conditions with respect to various practical application of these materials [31].

Studies performed in the Institute in co-operation with scientists from the Chemical Department of M. Curie-Skłodowska University (Lublin) had shown that the simultaneous processes of geometrical and chemical modification of silica can take place at increased temperature and pressure. The comprehensive studies of the course of chemical reactions and mass transfer of silica under such conditions were directed towards the preparation of the matrices possessing predefined geometric structure and surface properties [33,34].

It was shown [35] that the catalytic and thermal hydrosilanisation can be performed involving the inoculated silicon-hydride or olephinic groups of modified silica; this opens the way to the preparation of hydrolytically stable compounds with Si—C bonding at the surface of the matrix, contributing essentially to the technology of the synthesis of functional silica.

A branch of surface science, the study of synthesis and structure of the modifying layer, surface chemistry and analytical applications of complex-forming silica is under development at Chemical department of Kyiv National University, under the supervision of Academician V.V.Skopenko. The schemes of synthesis are developed and the preparation is performed for the silica possessing covalent-bonded complex-forming groups: polydentate aliphatic and heterocyclic (dipipidyl, phenantrolyne, chinolyne) amines, phosphorus-organic complexones, thiourea, phtalocyanines, porphirines etc., see e.g. [36-38]. It was shown that to describe the properties of silica which possess complex-forming groups fixed at their surface, some additional parameters should be known, in particular, the topography of the fixed groups. Most compounds of this type are characterised by cluster-like distribution of the fixed groups, with mean distance of 0.7 nm between them. The immobilised layer possesses the collapse-geometry. This affects the complex-forming processes: the complexes with the composition MeL_2 prevail at the surface [39]. To study the composition and structure of fixed complexes, spectroscopic methods are extensively used.

The complex-forming adsorbents were employed in the methods of the analysis of heavy and noble metals, see e.g. [40,41]. It was shown that modified silica can be used as the chromatographic phases [42].

The studies of the preparation of complex-forming adsorbents based at pyrogenic silica, the aerosil, and their application in the analytical chemistry, are performed also in the Odesa State University. In particular, the methods are proposed for the adsorptional and chemical immobilisation of o-dioxychromenols at the silica surface; this complex-forming adsorbent is used in the methods developed for the adsorption-spectrophotometric determination of microquantities of Mo(VI) and Ti(IV) in natural specimens possessing complicated composition [43,44].

In the Institute of Colloid Chemistry and Chemistry of Water (ICCCW UNAS), the method of liquid-phase modification of the surface of non-porous and wide-porous silica was developed [45,46]. This method is based on the impregnation of silica by toluene solution or water emulsion of organic oligomer, e.g., polymethylhydrosiloxane, with subsequent thermal processing of the system at 200-250°C. The main advantage of the method is the formation of continuous film of the modifier, consisting of the organosilicon oligomer molecules 'sewed together', at the surface. This film is 'inoculated' to the active centres at the silica surface by chemical bonds, and makes the residual (non-reacted) active centres, which exist at the surface of the matrix, inaccessible for the adsorption. This method of the modification of silica and perlite was employed in the process of the preparation of inert chromatographic supports [46], and the industrial adsorbent 'Zhemchug' used for the removal of floating and emulsified petroleum products from water [47].

In the ICCCW, extensive studies of surface chemistry of layer silicates were performed, which employed the combination of spectral and adsorption-analytical methods. This had led to the collecting of large scope of experimental knowledge regarding these materials. The types and number of active centres which exist at the surface of montmorillonite, vermiculite, kaolinite, hydromica, palygorskite were determined. It was shown that the adsorption characteristics of layer silicates which possess swelling structural cell (montmorillonite and vermiculite) essentially depend on the exchange cations. The concept of the formation of surface co-ordination compounds during the hydration (solvation) of these minerals is developed [48]. The properties of palygorskite surface are shown to depend essentially on the co-ordinately unsaturated (Mg^{2+}) ions, acidic (Si-OH) and basic (Al-OH, Mg-OH) hydroxyl groups. The application of conductometric method for the determination of the concentration of acidic and basic hydroxyl groups is summarised in [49], where other problems related to the surface chemistry of clay surface are also considered.

The monograph [50] deals with the interrelation between fine structural characteristics of layer silicates, and the nature of active centres which exist at the surface of these minerals. The method of structural modelling is developed and shown to be promising in the studies of surface chemistry of layer silicates [50,51]. The acidic properties of water molecules co-ordinated by multicharged

inorganic cations are studied [50,52]. The IR spectroscopy is applied for the determination of effective charges at surface oxygen atoms in layer silicates [53].

The structure and properties of layer silicates organic derivatives, prepared by the substitution of natural exchange complex for long-chained organic cations, were studied [46,54]. These substances were used as the basis for the development of promising adsorbents for gas chromatography [46] and the fillers for special rubbers [49]. Also, layer silicates possessing rigid structural cell (kaolinite, pyrophyllite) modified by thin layer of inoculated polymer were employed as basic materials in the development of fillers for polymer media whose polymer matrix is well compatible with such inoculated layer [55].

3. FUNDAMENTAL STUDIES OF ADSORPTION FROM GASEOUS AND LIQUID MEDIA ON SOLID SURFACE

In the Institute of Colloid Chemistry and Chemistry of Water (ICCCW UNAS), the theory of adsorption of organic molecules from aqueous solutions at carbon adsorbents was developed by Prof. A.M.Koganovsky and co-workers, based on extensive experimental studies. This theory is relied on the assumption that the standard Gibbs' energy of adsorption for dissolved substances is the additive function of the increments of Gibbs' energy corresponding to the adsorption of particular structural elements from water [56]. This enables one to determine equilibrium constants for the adsorption from water solutions for many organic compounds with complicated structure, without reference to the experimental data, but using only the tabulated values of the increments. This theory was further extended to the adsorption of the mixtures of organic components from water solutions, where it become possible to calculate adsorption isotherms for each component of the mixture from their individual adsorption characteristics, and to determine the value of total adsorption of all components in the multicomponent mixture [57].

The studies in the adsorption kinetics and dynamics of dissolved compounds, along with the adsorption static phenomena, are important with respect to the development of the adsorption technology applied in the cleaning of waste waters. In this regard, in the Institute the numerical criteria were worked out to determine the mass transfer coefficients for the adsorption of dissolved species in either dense or pseudoliquefied layer of granulated adsorbents. These results were used to develop the methods for the calculation of the technological parameters of adsorbents incorporated in the complex scheme of industrial waste water cleaning [58,59].

In the ICCCW, in the department headed by Corresponding Member of UNAS Yu.I.Tarasevich, the combined theoretical and experimental studies of the adsorption of gases and vapours by various types of adsorbents are per-

formed. The comprehensive structural-chemical, chromatographic and molecular approach was developed for the study of adsorption and separating properties of silica and layer silicates hydrophobised by cationic surface active and oligomeric organosilicone compounds [47,60]. The molecular statistical theory of the adsorption of hydrocarbons in Henry region is generalised for the adsorbents possessing regularly-inhomogeneous surface [47,61,62]. It was shown that for the adsorbents like palygorskite, which possess slit-like micropores, the significant contribution (ca. 50%) into the total adsorption energy of hydrocarbons comes from the induction interaction of adsorbed molecules with polar SiOH-groups and molecules of coordinately-bound water [47,62].

To estimate the size of surface micropores, the chromatographic version of molecular probe method was developed. This method was used to determine the parameters of surface micropores which exist at the surface of the surface-porous adsorbent prepared by the thermal oxidation of the modifying organosilicon layer imposed at the surface of diatomite and perlite [63].

The experimental data were used to build the dependencies of thermal coefficients of contact angle of wetting by water $d\theta/dT$ on the value of contact angle θ , and thermal coefficient of surface pressure of water film $d\pi/dT$ at the solid surface on surface pressure π [64,65]. This makes it possible to employ Bingham-Razouk's rigorous equation [66] which relates the values of π , θ and specific heat of wetting q , for the analysis of the data obtained for the adsorption of water at various surfaces. The criterial values of π , θ and q were determined which enables one to discriminate between hydrophilic and hydrophobic surfaces.

The adsorption calorimetric method was used to study the adsorption of water and benzene vapours at wide series of hydrophilic and hydrophobic adsorbents: cation-substituted forms of kaolinite and vermiculite, octadecylammonium kaolinite, modified silochrom, pillared Al_{13} -montmorillonite, zeolite ZSM-5, deeply de-aluminated ultrastable zeolite Y-US-Ex. The generalisation of the adsorption-calorimetric data, those obtained in ICCCW, and those which were published in the scientific literature, related to a wide range of adsorbate-adsorbent systems, formed a basis for the proposed classification of thermal diagrams for various adsorption systems [67].

From the analysis of the differential adsorption entropy, six types of adsorption are shown to be distinguished with respect to the character of the motion of the molecules adsorbed at solid surface. These are the localised adsorption; the localised adsorption with the contribution of vibrational degrees of freedom; pseudolocalised adsorption where the translational degrees of freedom are replaced by the vibrational ones; intermediate non-localised within the cells; non-localised and supermobile adsorption. The criterial values of the adsorption heats and entropies are determined which can be used to distinguish between these types of adsorption [67].

The studies of interaction of water molecules with hydrophilic centres existing at the surface of hydrophobic adsorbents are under way now in ICCCW; in these studies the combination of chromatographic, calorimetric and quantum chemical methods is used [68].

In the Institute of Bio-organic Chemistry and Petroleum Chemistry UNAS, in the course of EPR studies of adsorption of 2,2,6,6-tetramethyl-4-oxypiperidine-1-oxyle from hydrocarbon solutions at NaY zeolite surface, the phenomena of vibrational adsorption was discovered by Prof. K. I. Patryliak[69]. The essence of the phenomena is the initial saturation of the zeolite by the radical, followed by partial (to 70%) spontaneous desorption of the initially adsorbed radical, with subsequent repeated adsorption and so on; this cyclic process was observed during 1400 hours (maximum duration of the experiment) in the thermodynamically closed system.

In the Kharkiv Physico-Technical Institute of Low Temperatures UNAS, the studies of temperature dependence of the cryo-adsorption of individual gases (nitrogen, hydrogen, krypton, xenon, neon and their mixtures) on the NaA zeolites are performed [70,71]. The data obtained were used to calculate the adsorption heats of the gases, and the separation coefficients for the mixtures of these gases at various degrees of coverage. These laborious studies are aimed at the development of the adsorption-based technology for the preparation of extremely pure gases.

In the Physical Scientific and Research Institute at Odesa State University, the studies of static, kinetic and dynamic properties of adsorption of various acidic and basic vapours (NH_3 , amines, SO_2 , Cl_2 , HCl , HF , SiF_4) on mineral adsorbents and synthetic ionites of granular and fibrous type are under way [72-74]. New phenomenologic expression was proposed to describe adsorption system [75], and the method was developed to predict the value of Henry constant from the principle of corresponding states [76].

In the Odesa Technical University of Low-Temperature Technique and Energetics, the methods of numerical experiment are developed for the study of adsorption systems; these studies are performed by Prof. A.L.Tcykalo and co-workers, see e.g., [77].

In the Institute of Chemistry of High Molecular Compounds (ICHMC UNAS), the group led by Academician Yu.S.Lipatov applies a complex of methods to study the adsorption of polymers from various solvents at solid surfaces [78,79]. These investigations are performed in close co-operation with the research group in Lviv University, headed by Prof. M.M.Soltys. The adsorption of polymethyl acrylic acid from water solutions at zinc-sulphide fluorescers, aluminium oxide and titanium dioxide was studied, see e.g., [80]. These studies are expected to give insight in the role played by adsorption macromolecules in the stabilisation of colloid systems used in the technology. In particular, this scientific school had succeeded in the establishing of the dependence between the adsorption amount and the ratio of solid to liquid phase. It was shown that

the increase of the adsorbent content in the system often results in the decrease of the polymer adsorption due to the formation of disperse particle aggregates.

4. ACTIVITIES IN THE PRODUCTION OF INDUSTRIAL AND ENVIRONMENTALLY IMPORTANT ADSORBENTS AND THE DEVELOPMENT OF NEW ADSORPTION ACTIVE MATERIALS

In the former Soviet Union, Ukrainian scientists had played an active role in the works related to the development of industrial production of silica gels, synthetic zeolites, adsorbents based on raw clay materials. It was, however, a common practice to locate all vital industrial enterprises in Russia, and when the Soviet Union had collapsed, Ukraine was forfeited these industrially important materials, and had to import them from Russia and other countries. Much the same situation had arisen also in other branches of chemical industry. Therefore the Ukrainian government in 1993-1994 had considered and adopted for future 5 years the diversified scientific and technical program intended for the development of the limited-scale production of new chemical materials to replace the imported products. This program is supervised by Academician-Secretary for the Department of Chemistry UNAS V.D.Pochodenko (Vice-President UNAS since 1998); the branch of the program dealing with adsorbents and supports is managed by Corresponding member UNAS Yu.I.Tarasevich. And, while the economic problems which Ukraine is currently undergoes, had prevented this program from being performed in each particular, these activities had undoubtedly contributed greatly into the intensification of researches and development studies intended to organise the production of adsorbents and new adsorption active materials in Ukraine. The most important developments are summarised below.

At Sokirnitza deposit (Transcarpathian Ukraine, near Chust town), the construction of the plant which will produce a wide range of zeolite-derived materials is now at the final stage, with the first production line scheduled to be commissioned at the end of 1999. At the plant, in particular, the crushed clinoptilolite tuff of 1-3 and 3-5 mm fractions, containing not less than 70% of rock-forming material will be produced, to be used for the cleaning of drinking and waste water [47,81].

In the 1970s, the technology for the preparation of A-4 and A-4M adsorbents on the basis of the genetic mixture of palygorskite and montmorillonite from the Ukrainian Cherkasy deposit was developed with the participation of Yu.I.Tarasevich and I.I.Martsyn (ICCCW UNAS). These adsorbents were intended to be used for the cleaning of aromatic raw products from unsaturated hydrocarbons [50,82], and the workshop for the preparation of the A-4M adsorbent with the yearly production of 4000 tons was built at the petroleum processing plant in Ishimbay (Russia). The experience gained during the development of A-4 and A-4M adsorbents was used by I.I.Martsyn (Institute of Bio-

colloid Chemistry UNAS) for the organisation of the industrial production of granulated adsorbent based on Cherkasy palygorskite. First commercial parties of the adsorbent were produced at the workshop in Svitlovodsk (Kirovograd province) in 1997 and 1998; the planned yearly production is 2000 tons.

In the Institute of Physical Chemistry (IPC UNAS), the material-consuming technology for the preparation of several silica gel grades (accompanied by the utilisation of sodium sulphate as the by-product) was developed and engineering design was worked out (V.L.Struzhko).

The expedient technology for the preparation of X and Y zeolites possessing improved mechanical and diffusion kinetic properties on the base of Ukrainian kaolin was developed in the Institute of Bio-organic Chemistry and Petroleum Chemistry UNAS (K.I.Patryliak). Limited batches of such zeolites are produced at the experimental plant in the Institute; the engineering design is worked out for the yearly zeolite production of 1500 tons.

In the Kharkiv Research and Development Institute of General Chemistry (Ukrainian Ministry of Industrial Policy) the technology is developed for the preparation of the granulated zeolite CaA based on Ukrainian kaolin; the method developed by N.V.Timochina does not require the application of binders. The key stages of technological process were tested at the experimental plant in the Institute, and the experimental batch of the zeolite was prepared and found to be efficient for the extraction of n-paraffins from gas condensate. The production of limited commercial batches (yearly amount ca. 100 tons) is under way, and the line for the industrial production of CaA zeolite is currently under construction at Mykolaiv alumina plant with planned yearly yield of 200 tons. This zeolite is extremely important in Ukraine due to its properties with respect to fine dehydration of gases and liquids, and separation of gaseous and liquid mixtures.

The scientists from Physical Scientific and Research Institute at Odesa State University had taken an active part in the design and commissioning of an experimental production line for the preparation of cationite indicator adsorbent for the absorption of hydrogen sulphur. This line was constructed at Volnogradsk (Dnepropetrovsk province), at the premises of Ukrainian state consortium "Ecosorb"; the first commercial batch of the product (2.5 m^3) was produced for Russian Gazprom to be used in gas mask canisters at Astrakhan gas condensate deposit.

In the Research and Development Institute of Chemical Technologies "Chimtehnologia" at Siverodonetsk (Lugansk province) the technologic line was developed by M.A.Glikin for the preparation of coal adsorbents based on lignin, the by-product of cellulose and hydrolysis production. The batches of coal adsorbents with specific area of ca. $400 \text{ m}^2/\text{g}$ were found to be efficient in the cleaning of industrial waste waters at some enterprises in Siverodonetsk, Zaporizhzhia and other cities.

Active coals are very important adsorbents; the development of these materials is currently one of the priorities of Ukrainian scientists. In the laboratory headed by Corresponding member of UNAS K.E.Machorin (ICCCW UNAS) the technology is designed for the preparation of cheap and mechanically strong crushed active coal from the Ukrainian anthracite (Donbas coal deposit) [83,84]. The studies [85] had shown that the activated anthracite is characterised by more homogeneous microporous structure, more developed mesoporous structure and better kinetic properties as compared to many other brands of active coal.

The technology for the production of mesoporous coal "Acant-meso" which can be used for the high quality conditioning of drinking water was developed in ICCCW [86,87].

At the experimental plant of the Institute of Sorption and Problems of Endoecology (ISPE) UNAS the production of commercial batches of synthetic active coals used for medical purposes was organised [88]. In the ISC UNAS, the technology is developed and the production is under way of spherical carbon haemosorbent prepared on the basis of co-polymers of styrene and divinyl benzene [89].

The studies aimed at the preparation of extremely pure active coals are now performed in the ISPE UNAS. The prospective application of this materials is the extraction of noble metals from various solutions [90].

The technology for the production of mesophase pitch was developed in the Kharkiv Research and Development Institute of Coal Chemistry by V.I.Shustikov. This pitch is used as the raw material for the preparation of carbon fibre by the technology proposed by scientists working in the Institute of Materials Science Problems UNAS. The adsorption properties of such fibre are summarised in the publication [91].

Extensive studies of the technology for the preparation of bloated natural graphite and the application of the material for the removal of oil and petroleum products from water are performed in the Donetsk Institute of Physico-Organic Chemistry by A.F.Popov, M.V.Savoskin et al.

In Germany, the commercial production of a new class of adsorbents, Envi-sorb B+, which consists of the aluminosilicate matrix containing the incorporated microdisperse active carbon, was started by Engelhard Corporation in 1998. However, yet in 1980 the efficient method for the preparation of coal-mineral adsorbent based on the coked natural aluminosilicate was proposed in the ICCCW UNAS [92]; this material was shown to be applicable for the cleaning of waste waters [93]. This method was technologically developed and applied at the Ufa (Russia) oil refinery plant. Here the coked adsorbent is thermally activated at 500°C in absence of air; the resulting product is applied for the cleaning of waste water disposed by the plant from the petroleum-containing products [47]. Currently, the works aimed at the production and application of

coal-mineral adsorbent are performed in co-operation with M. Curie-Skłodowska University (Lublin) [94].

The method of the preparation of another new class of adsorption-active materials, the so called surface-porous adsorbents was proposed in the ICCCW [63]; these materials were shown to be promising for the final cleaning of water from petroleum-containing products [47,95].

The innovative studies are under way during last 5 years in the Institute of Physical Chemistry UNAS, related to the development of the methods for the synthesis of various nanosize porous materials. In particular, the conditions were established for the synthesis of silica hydrosols with predefined degree of porosity. It was shown that these sols can be used as the starting materials for the preparation of submicron and nanosize powders, ceramic membranes and adsorbents [96]. In the Institute, the fundamentals of template synthesis of zeolites and various nanoporous structures are developed. The zeolite-like phosphates of Al, Ti are synthesised along with other metal containing adsorbents, mesoporous molecular sieves and a number of nanoporous compounds [97,98]. These zeolites and mesoporous molecular sieves were used as a basis for the preparation of materials which contain nanosize semiconductive molecular clusters CdS, PbI₂, Bi₂S₃, anionic hexacyanide complexes of transition metals etc. [99,100].

The studies of organosilicon adsorbents performed in the Institute of Physical Chemistry UNAS [13] had found their practical application with the development of efficient enterosorbent "Enterogel" [101], possessing general detoxicating action, which is now produced by Creoma-pharm enterprise in Kyiv.

5. STUDIES OF ION EXCHANGE AND PREPARATION OF NEW ION EXCHANGE MATERIALS

Systematic studies of ion-exchange equilibria and thermodynamics on natural layer silicates and zeolites are currently performed in the ICCCW UNAS. These investigations have led to the formulation of the crystal-chemical principle of the selectivity of zeolites with high silica contents with respect to large-size cations [47,102,103]: for the adsorption of large-size cations at natural zeolites with high silica contents, in addition to the thermodynamic factor, another, more significant crystal-chemical (geometric) factor becomes significant. This factor is ascribed to the localisation of these cations in the eight-member silicon-oxygen structural rings, with free section compatible with the adsorption cation.

It was discovered that natural zeolites are characterised by memory effect with respect to the cation in whose presence the crystallisation of zeolite had taken place, or to the cations which possess the size and charge similar to that of ion in the initial sample [47,104].

In the Institute, the calorimetric cell was developed for precise measurements of ion exchange heats. Using the Calvet microcalorimeter equipped with this cell, the heats of ion exchange at clinoptilolite were measured, which made it possible to calculate more precisely the thermodynamic functions of ion exchange involving uncharged cations [105].

In the studies of ion exchange equilibria at layer silicates, it was shown that the introduction of complex-forming reagents into the ion exchange system leads to the increase of layer silicates selectivity with respect to complex cations [106,107]. This feature of mineral ion exchangers was used, in particular, in the works related to the elimination of the consequences of Chernobyl nuclear disaster in 1986 [108].

The adsorption of alkali metal cations at basal faces of layer silicates (montmorillonite and kaolinite) was shown to obey the direct selectivity series ($\text{Cs}^+ > \text{Rb}^+ > \text{K}^+ > \text{Na}^+ > \text{Li}^+$), while for side faces of these minerals the inverse series takes place ($\text{Li}^+ > \text{K}^+ > \text{Rb}^+ > \text{Cs}^+$) [109]. The comparison of the results with the data concerning the adsorbability of alkali metal cations on silica gel for neutral and high pH values, and on aluminium and iron oxides for pH values above the corresponding iso-electric point shows, that the observance of direct or inverse cation adsorbability series depends on the concentration of exchange centres at the surface of mineral ion exchangers [109].

Systematic quantum chemical studies of the state of adsorbed cations in the structure of mineral ion exchangers, and the role of structural factor in the selectivity of layer silicates and zeolites with respect to various cations are also performed in the ICCCW [110,111].

Another group of the scientists in the Institute is engaged in the study of synthetic ionites in view of their application for water cleaning processes. The theory of sulphate-bisulphate equilibrium, which was developed in [112], had formed the background of the technology for the utilisation of sulphuric acid from the worked-out regenerative solutions of H-cationite filters from the equipment used in the water desalination. This technology enables one to extract 60-80% of the reagent from the worked-out regenerative solutions for its re-use in the regeneration of the cationite resin [113].

The theory of ion exchange equilibrium was proposed for the quantitative description of the deviations of the properties of solutions in the adsorption phase from the ideal behaviour [114]. This theory results in the binomial distribution of the probabilities to find an exchange centre in any possible state determined by the composition of the solution in its nearest neighbourhood. This theory is capable for rather good description of experimental data obtained for systems characterised by strong deviations from the ideal behaviour, in particular, the potentiometric titration curves for carboxyl cationites.

The technologic solutions related to the desalination, softening and conditioning of water using the ion exchange resins are presented in [115]. In this regard the studies should be mentioned which were performed jointly by the

workers in ICCCW and Cherkasy Industrial Corporation "Azot", aimed at the development of the technology for the industrial production of the anionite CAB-990, which possesses high basic properties, and intended primarily for the removal of humine substances from water. This material is used as the load of pre-filters which protect the gel anionite filters from the intoxication. The characteristics of this material are described in [116].

The efficient complex-forming adsorbent which can be used for the extraction of heavy metal ions from water was developed in the ICCCW on the basis of natural mineral raw materials and polyphosphates [47,117]. This material was shown to be promising for the extraction of Ni^{2+} ions from the washing water at the galvanic production lines [47].

The scientists in Kyiv Polytechnic Institute had succeeded in the development of new adsorbent "Oxisorb", prepared by the modification of the vinyl-pyridinium anionite by chlorine derivatives of hydantoins, in particular, 1,3-dichlor-5-dimethylhydantion, DCDMH). This adsorbent, in addition to the enhanced affinity with respect to various mercury compounds, also possesses oxidation properties [118]. The inoculated modifier breaks C-Hg bonds which exist in mercury-containing organic compounds, and oxidises the metallic mercury, transforming it into the adsorbable form Hg(II).

In the Dnipropetrovsk State University of Chemical Technology, the organosilicon dipiridinium complexes, silacrown ethers and other organosilicon adsorbents were prepared; these materials possess high selectivity and exchange capacity with respect to the ions of heavy and noble metals, see e.g., [119].

The intensive works are performed by the Institute of Sorption and Problems of Endoecology and Institute of General and Inorganic Chemistry UNAS, aimed at the synthesis, study of properties and technologic applications of the zirconium and titanium phosphates, see e.g., [120,121]. The "crown-like" model is proposed for specific adsorption centres of large-size alkali cations in the ultrapores of these phosphates. This model forms the basis for the proposed technology of the synthesis of these adsorbents in the presence of the template ion, with respect to which the selectivity is required. It was shown that the templating cation, during the synthesis process, orients the functional groups of the cationite in its neighbourhood; this results in the formation of the increased number of ultrapores compatible by size with this cation, as compared to the pores formed when such cation is absent.

To summarise, the results presented above make evident that a wide variety of studies related to the surface chemistry, adsorption and ion exchange is performed in various regions of Ukraine by a number of institutes of Ukrainian National Academy of Sciences, universities and institutes concerned with applied studies supervised by the Ukrainian Ministry of Industrial Policy. These studies are aimed both at the solution of fundamental problems of solid surface science, and the practical application of traditional and innovative adsorbents and ion exchangers in the industry and environmental protection.

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CURRICULUM VITAE



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Area of scientific interests - colloid chemistry and physical chemistry of interface phenomena. Particular problems investigated: study of the state of molecules adsorbed on the surface of layer silicates using the vibrational and electron-optical spectroscopy; study of the adsorption and ion-exchange equilibria, the thermodynamics of adsorption and ion exchange on mineral ion exchangers - layer silicates, natural and synthetic zeolites, using the calorimetric, gas chromatographic and statistical thermodynamic methods; development of the methods for the preparation of new types of adsorbents, study of their structure using the electron paramagnetic resonance, optical spectroscopy,

X-ray methods and the application of these materials in adsorption and chromatography, and for environmental protection.

More than 400 scientific publications, including 3 monographs: Yu.I.Tarasevich, F.D.Ovcharenko, Adsorption sur des mineraux argileux (Rueil Malmaison, 1980); Yu.I.Tarasevich, Natural adsorbents in water cleaning processes (Naukova Dumka, 1981); Yu.I.Tarasevich, Structure and Surface Chemistry of Layer Silicates (Naukova Dumka, 1988). Lectures at international scientific meetings in France, Hungary, Italy, Japan, Poland, Slovakia and the USA.