

Nanomaterials in Nuclear Engineering and Radioecology

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Abstract—Key results concerning large-scale application of nanomaterials in nuclear engineering are reviewed. The data on redox reactions of uranium and actinides in solutions and solid-phase transformations for the development of modern technologies for fuel reprocessing and handling of alkaline radioactive waste are discussed. The information concerning various methods for treatment of liquid radioactive waste with nanostructured sorption materials (carbon materials included) is also presented.

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INTRODUCTION

Russia is one of the leading energy powers of the world and has such energy reserves that do not only cover its own demands but also allow for export to other countries. In fifty years of studies in the field of nuclear power engineering in Russia, the research community has justified the viability of nuclear power and determined the best development options with allowances made for its latent potential and environmental considerations.

The prospects for long-term development of nuclear power engineering are associated with a distinct possibility of recovery and reprocessing of nuclear fuel without the loss of competitive ability and safety of nuclear engineering. The industrial policy envisions gradual introduction of the fourth-generation nuclear energy technology with fast reactors, a closed nuclear fuel cycle, and uranium–plutonium fuel in 2010–2030. This will help remove the constraints regarding fuel material in the foreseeable future.

A closed nuclear fuel and energy cycle implies the development of modern nuclear technologies applied throughout the entire complex of technological operations at facilities in the nuclear power industry: the mining of uranium ore; the production of uranium concentrate; the conversion of uranium, production of uranium hexafluoride, and enrichment of U-235; the production of uranium oxide and mixed nuclear fuel and fuel elements (FEs); the reprocessing of spent nuclear fuel (SNF), extraction of uranium and plutonium for recycling, and production of certain radionuclides that find scientific, industrial, and medicinal

applications; the management, storage, and transmutation of radioactive waste; the decommissioning of nuclear reactors; and the decontamination and remediation of soil contaminated with radionuclides.

In our view, wide application of nanomaterials may become one of the most significant trends in the development of new nuclear technologies and the modernization of existing ones. The results of studies concerned with the most promising directions of development of nuclear technologies which make use of nanomaterials and have already given an impetus to progress are discussed below.

HETEROGENEOUS CATALYSIS

The refinement of technological processes that leads to an increase in productivity or cost saving is highly sought in any industry. Heterogeneous catalysts are used in chemical engineering to address the aforementioned issues. Researchers at the Frumkin Institute of Physical Chemistry and Electrochemistry (IPCE) were the first to propose the use of Pt, Ru, and Pd nanoparticles applied to porous surfaces as heterogeneous catalysts in various redox reactions of actinides and in the processes of decomposition of organic components of liquid radioactive waste (LRW). They took part in the development of certain flow diagrams for the reprocessing of irradiated nuclear fuel that were concerned primarily with stabilizing the given oxidation states of uranium, neptunium, plutonium, and americium [1–5]. The reactions of catalytic reduction of uranium(VI) in nitric and sulfuric solutions to the tetravalent state with H₂, N₂H₄, HCOOH, H₂CO, and C₂H₅OH used as reduc-

ing agents and Pt/Al₂O₃, Pt/SiO₂, Pt/C, Pd/SiO₂, and Re₂S₇ applied as catalysts; reduction of plutonium(IV) to (III), neptunium(VI) and (V) to (IV) (with H₂, N₂H₄, and HCOOH used as reducing agents and Pt/Al₂O₃, Pt/SiO₂, and activated carbon serving as catalysts); and oxidation of neptunium(IV) (HNO₃ and Pt/SiO₂ were used as the oxidizing agent and the catalyst, respectively) were investigated in the course of systematic multiyear research. Rate constants of these reactions and activation energies were calculated as functions of the catalyst type, the ratio of its amount to the volume of actinide solution, and the metal content of the carrier. The establishment of industrial production of U(IV), the scientific foundation of which was laid by IPCE, at the Siberian Integrated Chemical Plant (SICP) [6] attests to the operational reliability of catalytic techniques in radiochemical technology. The average daily throughput of the SICP setup was no less than 130 kg of uranium; the SICP demand for reducing agents was thus fully satisfied. The methods for catalytic sorption partitioning of transuranium elements, including the extraction of a high-energy radionuclide (plutonium-238) for the production of radiation current sources with the use of anion-exchange catalysts, were studied, patented, and introduced into practice at IPCE [7, 8].

A method for decomposing mother oxalate solutions, which are left as radioactive waste at the end of the most important phase of extraction and refinement of valuable components of radiochemical technology (neptunium and plutonium) by oxalate treatment, with platinized catalysts was developed in joint investigations conducted by IPCE and the Mayak Production Association (MPA) [9, 10]. The method consisted in passing the initial model mixture of nitric and oxalic acids, which contained iron (0.83 g/L), chromium (0.17 g/L), manganese (0.23 g/L), and uranium (0.17 g/L), through a column filled with 0.85 wt % Pt on silicagel MSKG at a temperature of 50–80°C. It was demonstrated that a catalytic reaction of decomposition of oxalic acid was initiated in the column, and the H₂C₂O₄ content at the outlet was stabilized as soon as six column volumes of the indicated solution were passed through the column. It was noted that the concentration of oxalic acid at the outlet was reduced from 10 to ≤0.5 g/L when the initial solution (5 M HNO₃ and 17 g/L of H₂C₂O₄) was passed through the column at an elevated (raised from 50 to 80°C) temperature. A change in the concentration of nitric acid from 5 to 2.5 M did not significantly alter the process yield. On the contrary, the catalytic decomposition of oxalic acid slowed down when the rate of flow of solution through the column (an equivalent of the time of contact between the catalyst and the initial mixture) was increased. The results revealed the following advantages of the above-described method: high efficiency, reduced process temperature, and shorter process completion time. The alter-

native (and currently used) industrial method for decomposing oxalic acid in nitric acid solutions consists in heating a mixture of H₂C₂O₄ and HNO₃ to 90°C for 48 h [11–13].

Heterogeneous catalytic processes were also proposed to be used for denitration of technological solutions at radiochemical facilities [14–16]. If the common method for denitration of nitric acid solutions by HCOOH with no heterogeneous catalysts is used, the reaction has an incubation period, and the process proceeds via the autocatalytic mechanism. As a result, the reacting mass is heated and may even explode. Experiments showed that the use of heterogeneous catalysts in the aforementioned processes helps increase the reaction rate by a factor of 10–100 and maintain readily controlled reaction conditions. The technological process yielding reaction products of the required composition may thus be completed in a short time. The developed technology was based on the method of catalytic denitration of nitric acid solutions with formic acid. This method ensures the initiation of a denitration reaction with no induction period and thus makes the process much safer and guarantees a high degree of denitration of processed solutions. The key parameters of denitration processes in stationary conditions (a fluidized bed reactor) were determined experimentally: the effect of stoichiometry of the denitration process (the HCOOH/HNO₃ ratio), the process completion time, and the temperature on the efficiency of decomposition of ammonium nitrate was evaluated. The use of non-platinum catalytic materials (Ni/SiO₂, activated carbon (BAU and SKT), and stainless steel Kh18N9T in the form of fine chip) for catalytic denitration of technological products of MPA was also proposed.

A method for decomposing carbamide was developed in order to devise techniques for the treatment of radioactive waste with certain organic components. It was found that organic components are decomposed instantly when HCOOH is added to a nitric acid solution of carbamide over a platinized catalyst. The kinetics of catalytic decomposition of 0.2–0.4 M [(NH₂)₂CO] in the presence of 1% Pt/SiO₂ were studied in the following conditions: [HNO₃] = 0.7–7 M, [HCOOH] = 0.2–1.3 M, *t* = 50–80°C, and the ratio of the solid phase to the liquid one (S : L) = 0.02–0.1 g/mL. It was demonstrated that the reaction of catalytic decomposition of (NH₂)₂CO in these conditions was initiated at 60°C when [HNO₃] ≥ 2.5 M with no induction period in accordance with the zero-order law with respect to [(NH₂)₂CO] with an activation energy of 58.7 ± 1.6 kJ/mol. An increase in the concentration of formic acid and the concentration of catalyst in the reaction mixture (S : L) raised the rate of carbamide decomposition [17–18].

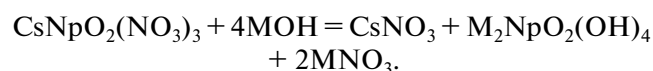
The catalytic decomposition of hydrazoic acid for its removal from aqueous tail solutions (fuel-processing waste) was also investigated [19]. This operation is

of particular importance owing to the fact that HN_3 -containing systems are highly flammable. The experiments were carried out over a catalyst (1% Pt/SiO₂) at $[\text{HN}_3] = 0.025 \text{ M}$, $[\text{HNO}_3] = 0.5\text{--}5 \text{ M}$, and a temperature of 30–60°C in the dynamic regime. It was demonstrated that an increase in temperature from 30 to 45°C at $[\text{HNO}_3] = 1 \text{ M}$ resulted in a 2.4-fold increase in the rate of decomposition of HN_3 . This corresponded to an activation energy of ~ 67 kJ/mol. The presence of NaNO_3 (20 g/L) and uranium (0.1 g/L) in the solution did not affect the efficiency of catalytic decomposition of hydrazoic acid. The rate of decomposition of HN_3 was reduced in 0.5 M HNO_3 saturated with tributyl phosphate (TBP). This was attributed to the adsorption of organic molecules onto the catalyst surface. It was found that the method for catalytic decomposition of hydrazoic acid in the dynamic regime is fairly efficient and is suited for industrial application.

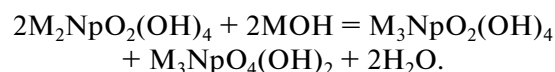
SOLID-PHASE TRANSFORMATIONS OF HYDROXIDES OF Np(V) AND Np(VI) IN ALKALINE MEDIA

Significant amounts of high-activity alkaline waste (slurry) that contains hydroxides of Fe, Al, Ni, and Cr; sulfides of Fe and Ni; and ferrocyanides of Na, K, and Ti with long-lived radionuclides (neptunium, plutonium, and americium included) are still being stored at various industrial facilities throughout the world. The storage of high-activity slurries in tanks is not radiologically safe; therefore, this waste has to be utilized. Large-scale studies into solid-phase transformations of actinide compounds occurring at various storage times were carried out at IPCE in order to prevent accidents and reprocess the solid phase of slurry. The determination of the distribution of uranium and transuranium elements between liquid and solid phases and the estimation of criticality were among the objectives of this research. Transuranium elements (neptunium included) are concentrated in alkaline radiochemical waste in the form of hydroxide salts. It was of interest to study the reactivity of hydroxide actinide compounds (with Np taken as an example) with various inorganic substances accumulated in industrial waste. With this aim in view, the interaction of certain compounds of pentavalent neptunium with various reagents was investigated [20, 21]. The interaction was established by grinding the compounds with subsequent compaction in a NaCl matrix under a pressure of 600 MPa. It was demonstrated that the interaction of solid neptunium(V) hydroxide $\text{NpO}_2\text{OH} \cdot n\text{H}_2\text{O}$ (I) or $\text{NpO}_2\text{NO}_3 \cdot \text{H}_2\text{O}$ with MOH (M = Li, Na, K, Rb, or Cs) in a box within which air was cleaned from water vapor and CO₂ yielded (depending on molar ratios of the initial substances) hydroxide compounds $\text{MNpO}_2(\text{OH})_2 \cdot n\text{H}_2\text{O}$ (II) and $\text{M}_2\text{NpO}_2(\text{OH})_3 \cdot n\text{H}_2\text{O}$ (III). When (II) interacted

with MOH, salts of (III) were produced. The studies of solid-phase transformations of neptunium(V) compounds in alkaline and carbonate media showed that the interaction of $\text{NaNpO}_2\text{CO}_3$ with MOH (M = Na or K) with molar ratios (μ) $[\text{MOH}] : [\text{NaNpO}_2\text{CO}_3]$ varying from 3 : 1 to 5 : 1 resulted in the accumulation of (II). When μ was higher than 5 : 1, the fraction of compound (III) was increased. This compound was not formed completely in LiOH. It was noted that the reaction of (I) with MHCO_3 (M = Li, Na, K, or NH_4^+) yielded MNpO_2CO_3 . The latter compound was also produced when (II) and (III) interacted with M_2CO_3 or MHCO_3 (M = Li, Na, K, or NH_4^+). It was also demonstrated that (I) produced $(\text{NpO}_2)_2\text{C}_2\text{O}_4$ or $\text{NpO}_2\text{HC}_2\text{O}_4$ (depending on the conditions) in a reaction with solid oxalic acid. Compounds of the $\text{MNpO}_2\text{C}_2\text{O}_4$ type were obtained in solid-phase interaction of (II) or (III) with $\text{H}_2\text{C}_2\text{O}_4$. Such compounds were produced in reactions of (I) not only with oxalic acid but also with oxalates of alkaline metals. When $\text{M}(\text{OH})_2 \cdot x\text{H}_2\text{O}$ (M = Mg, Ca, Sr, or Ba) was ground with (I) or $\text{NpO}_2\text{NO}_3 \cdot \text{H}_2\text{O}$, mixed hydroxide compounds of Np(V) with alkaline-earth metals were produced. Mixed hydroxide compounds of Np(V) with lanthanum and thorium were obtained in solid-phase interaction of (I) with $\text{La}(\text{OH})_3$ or $\text{Th}(\text{OH})_4$. This was confirmed by a change in the IR spectra of products after grinding. Rapid sorption of neptunyl cations from solutions by lanthanum and thorium hydroxides was noted. The obtained products retained neptunium after being washed with hot distillate for a considerable time. The process of disproportionation of Np(VI) in the solid phase was studied [22]. In order to do that, crystalline $\text{CsNp}^{\text{VI}}\text{O}_2(\text{NO}_3)_3$ salt was ground with solid caustic alkalis (MOH). The molar ratio of the initial compounds was varied; Np(VII) was found in reaction products already at $\mu = 1 : 5$. Np(VII) was identified by electron absorption spectroscopy and Mössbauer spectroscopy. Using the data on forms of penta-, hexa-, and heptavalent actinides in alkaline media, one may devise the following representation of the indicated reaction. At the first stage, a hydroxide compound of Np(VI) is formed:



At the second stage, neptunium(VI) disproportionates into Np(V) and Np(VII):



The interaction of $\text{Na}_2\text{NpO}_2(\text{OH})_3$ and $\text{Na}_3\text{NpO}_4(\text{OH})_2$ in a NaOH medium results in the reproporationation of Np(V) and Np(VII). This was confirmed by the presence of Np(VI) in reaction products. The obtained data suggest that solid neptunium(V) hydroxide may interact with certain inor-

ganic compounds present within the storage tank. Since their solubility in alkalis is considerably higher, its concentration in the liquid phase of slurries is high.

The interaction of U(IV) salts with noble-gas derivatives is another demonstrative example of solid-phase interaction of actinide compounds with inorganic substances. This reaction is chemiluminescent and thus allows one to devise a high-sensitivity method for determination of uranium in concentrations below 10^{-10} mol/L. The chemiluminescence of uranium and transuranium elements was discovered and studied by Russian researchers at the Institute of Organic Chemistry of the Ufa Scientific Center of the Russian Academy of Sciences and IPCE [23–26].

REMOVAL OF RADIONUCLIDES FROM RADIOACTIVE WASTE

The researchers at IPCE have addressed the issue of handling of alkaline LRW of various activity levels by developing efficient techniques for extracting long-lived radionuclides (actinide elements, radiocesium, radiostrontium, technetium, etc.) from such waste. It was demonstrated that the introduction of water-soluble compounds of transition metals subjected to thermal treatment into LRW resulted in the emergence of nanostructured insoluble carriers throughout LRW and the coprecipitation of radionuclides onto them (the method of “emerging reagents”). It was found that the efficiency of removal of actinides from 0.5–1.0 M alkaline solutions by coprecipitation at 80°C is maximized when the following insoluble carriers are used: Cr(OH)₃, which is produced in the presence of Cr(CH₃COO)₃ or Cr(NO₃)₃, for Np(IV) (partition coefficient $K_p = 370$ and 440, respectively); Co(OH)₃, which is produced in the presence of Co(NH₃)₆Cl₃ or [Co(NH₃)₅Cl]Cl, for Pu(IV) ($K_p = 500$ and 130); MnO₂ or Mn(OH)₂, which are produced in the presence of KMnO₄, for Pu(IV) ($K_p = 25$ and 220); and Fe(OH)₃, which is produced in the presence of KMnO₄, for Pu(IV) NaFe(CN)₅NO ($K_p > 150$). It was demonstrated that iron(II) hydroxide is an efficient carrier for extracting Tc from alkaline waste. The procedure is as follows. (NH₄)₂Fe(SO₄)₂ (up to 0.1 M Fe(II)) is introduced into a solution with an alkali concentration from 0.5 to 4M. When the mixture is heated to 60°C, technetium(VII) is reduced to the tetravalent state, and insoluble complex salt (NH₄)₂Fe(SO₄)₂ is transformed into Fe(OH)₂ with subsequent coprecipitation with 96–98% Tc(IV). The same result was obtained in similar conditions when a 0.15M N₂H₅OH + 0.15M FeCl₃ mixture was replaced with (NH₄)₂Fe(SO₄)₂ [27–30].

URANIUM OXIDE NANOTUBULENES AS POTENTIAL MATRICES FOR RADIONUCLIDE IMMOBILIZATION

The discovery of carbon nanotubes, which exhibit unique physical and physicochemical properties, was one of the most important advancements of the 1990s in materials science [31]. By 2005, nanotubes were synthesized for six families of inorganic compounds: chalcogenides; oxides and halogenides of transition metals; and mixed-composition systems based on boron, silicon, pure metals, etc. [32–38]. However, published data concerning the synthesis and studies of nanomaterials based on uranium and transuranium elements have so far been lacking. At the same time, the synthesis of such materials is essential, since they combine unique electronic properties of actinides with high specific surface energies of nanoparticles. Unique electronic properties of uranium and transuranium elements also add to the importance of fabrication of actinide-based nanomaterials. The researchers at IPCE together with their colleagues at the St. Petersburg State University have recently synthesized and studied two new compounds of hexavalent uranium ($K_5[(UO_2)_3(SeO_4)_5](NO_3)(H_2O)_{3.5}$ (**1**) and $(C_4H_{12}N)_{14}[(UO_2)_{10}(SeO_4)_{17}(H_2O)]$ (**2**)) that contain porous nanotubular structures [39–48]. The crystal-line structure of (**1**) has four independent U positions. Each of them is coordinated by two oxygen atoms that form a linear uranyl cation $[UO_2]^{2+}$. This uranyl cation is surrounded by five oxygen atoms, which belong to $[SeO_4]^{2-}$ groups, in the equatorial plane; as a result, pentagonal bipyramids $[UO_7]^{8-}$, which are typical of oxygen compounds of hexavalent uranium, are formed. The structure of (**1**) also incorporates five crystallographically independent Se atoms with tetrahedral coordination. Each $[SeO_4]^{2-}$ tetrahedron shares three vertices with neighboring pentagonal uranyl bipyramids, while the fourth one is “free.” The $[(UO_2)_3(SeO_4)_5]^{4-}$ tubes found in structure (**1**) are nanosized. For example, the outer tube diameter is 17 Å = 1.7 nm, and the inner diameter (the distance measured across a tube between the two closest oxygen atoms) is 7.4 Å = 0.74 nm. The latter value corresponds to a crystallographic free diameter of 4.7 Å, which is comparable to microporous zeolites and other molecular sieves such as titanosilicate ETS-4 (a synthetic equivalent of zorite). While all U and Se positions are filled completely in structure (**1**), structure (**2**) has both fully and partially filled positions. This, together with large unit cell parameters, complicated somewhat the interpretation of the X-ray results. The immediate coordination environment of U and Se positions with an occupation density in excess of 48% was localized, while the exact coordination of other positions could not be modeled. At the same time, there was no doubt that the key features of topology and geometry of the structure were determined. Just as

in structure (1), cations U^{6+} form pentagonal bipyramids $[UO_7]^{8-}$, and cations Se^{6+} form $[SeO_4]^{2-}$ tetrahedra. Selenate–uranyl nanotubes in structure (2) have an elliptic cross section with a size of $25 \times 23 \text{ \AA} = 2.5 \times 2.3 \text{ nm}$. Their inner diameter is 15.3 \AA , and the crystallographic free diameter is 12.6 \AA . The latter value makes compound (2) similar to highly porous zeolite structures and is close to the pore size in mesoporous materials. Thus, compounds (1) and (2) are the first examples of such materials, since their structures feature ordered packing of selenate–uranyl nanotubes. The above facts suggest that isolated nanotubes may also be obtained for structures based on uranium and transuranium elements.

NANOSTRUCTURED DIOXIDES OF URANIUM AND TRANSURANIUM ELEMENTS: SYNTHESIS, PROPERTIES, AND APPLICATION IN RADIOCHEMICAL TECHNOLOGY

The methods for synthesis and the physicochemical properties of dioxides of uranium and transuranium elements, which are of exceptional importance in radiochemical technology, are currently under investigation. These dioxides are used as weight forms in SNF reprocessing and as the initial substances for synthesizing the majority of other actinide compounds that find application in various scientific and engineering fields; in the production of fuel compositions in the nuclear industry; in nuclear synthesis of energy-intensive nuclides (^{238}Pu); and, possibly, as conserving matrices (UO_2) for fixing liquid high-activity waste into the solid phase. As an illustration, consider the following: a reduction in the consumption of natural uranium in energy production is needed in order for nuclear power engineering to progress. This reduction is brought about by an increase in the burnup fraction of nuclear fuel, which is achieved using nuclear materials with macrocrystalline structures with controlled porosities and densities. The researchers at MPA have cooperated with their colleagues at IPCE to determine the conditions for production of nanodisperse UO_2 powders and study their physicochemical properties and reactivities (for various powder fabrication and storage methods). It was demonstrated that the quality of mixed fuel is improved considerably when a nanostructured uranium dioxide phase is added to commercial UO_2 : the porosity is increased, and the formed solid solutions are more homogeneous [49, 50].

DEVELOPMENT OF GAS TREATMENT METHODS FOR RADIOCHEMICAL FACILITIES

Absorbent grains based on silicagel KSKG with nanometer particles of Ag compounds have recently

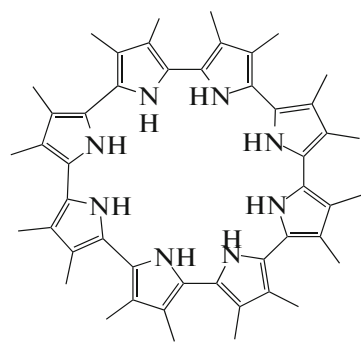
been introduced under the brand name Fizkhimin [51]. This sorption material allows one to remove various forms of radioactive iodine (inorganic forms and methyl iodide) from the steam–gas phase with a coefficient of purification of no less than 10^3 – 10^4 . The tests at a TUV Sudwest (Karlsruhe, Germany) stand showed that Fizkhimin is superior in its sorption characteristics to similar absorbent grains (e.g., Baylith (Bayer, Germany) and Termoksid (Russia)). On the basis of the results of various tests (including large-scale ones) of filter devices, Fizkhimin was chosen to be used as one of the primary elements of systems for beyond design basis accident management at NPPs with VVER-440/230 and VVER-1000 (NPP-92 project) and NPPs with VVER-1000 (NPP-2006 project) and VVER-1500 that are being constructed. A total of 720 kg of Fizkhimin were shipped in 2007 within the framework of the Russian–Indian contract in nuclear power engineering. This sorbent agent was to be used in filter modules of the passive filtering system in emergency filters to be installed at the first and the second units of the Kudankulam NPP (India). This was a joint effort between Atomstroyexport, OAO Krasnaya Zvezda, the Leipunsky Institute of Physics and Power Engineering (Obninsk), Atomenergoproekt, the Obninsk Center for Science and Technology, and several other Rosatom institutions. The behavior of $^{137}\text{Cs}^{131}\text{I}$ radioaerosols, formed in the process of evaporation of CsI from a Pt heater into the gas phase of argon or air, during their localization by aqueous solutions and various filtering materials (Petryanov filters, white ribbon filter paper, and metallic membrane filter TRUMEM) was studied. The dependence of the degree of localization of $^{137}\text{Cs}^{131}\text{I}$ radioaerosols on the amount of sublimated $^{137}\text{Cs}^{131}\text{I}$ and bubblers in the system, the nozzle size during bubbling, and the gas flow velocity was investigated. Data on the efficiency of sorption of $^{137}\text{Cs}^{131}\text{I}$ by a ten-layer bundle of Petryanov filters from the gas flow with and without a bubbler system were obtained. It was found that a bundle with a total thickness of 3 cm absorbed ~97% of $^{137}\text{Cs}^{131}\text{I}$, which was sublimated from a Pt heater, at a linear flow velocity of 2–3 cm/s. It was determined that the bubbler system did not affect the efficiency of localization of $^{137}\text{Cs}^{131}\text{I}$ radioaerosols by Petryanov filters but only contributed to a reduction in the aerosol load on these filters. Electron-microscopy studies of the size distribution of CsI aerosols permeating through aqueous solutions and various filtering materials (Petryanov filters and white ribbon filter paper) were conducted. The behavior of $^{137}\text{Cs}^{131}\text{I}$ radioaerosols, formed in the process of evaporation from a platinum substrate into argon or air, in the presence of UV radiation and without such radiation was studied. It was found that the degree of localization of radioaerosols in bubblers with an aqueous solution of $Na_2S_2O_3$ varied from 30% in argon to 60% in air when $^{137}\text{Cs}^{131}\text{I}$ was evaporated with no UV irradiation. The

production of a considerable amount of nanosized particles which were not absorbed by aqueous bubbler solutions and were able to penetrate through a composite filter incorporating a Petryanov filter and white ribbon filter paper was also observed in the process of sublimation of $^{137}\text{Cs}^{131}\text{I}$ from a platinum heater. The variation of the Cs/I ratio in different fractions showed that the degree of conversion of CsI was minimized in an argon atmosphere; this was attributed to the presence of trace amounts of oxygen and water in the system. It was found that the amount of radionuclides in bubblers was reduced significantly in the UV radiation field both in argon and in air; along with that, the amount of ^{137}Cs penetrating through the composite filter was reduced by a factor of ~ 2 . It was concluded that nanosized particles grew larger in the process of photoactivation owing to their interaction with each other and with larger aggregates. The UV radiation affected primarily the kinetics of the process of aggregation of aerosols in the gas phase and exerted almost no influence on the rate of chemical processes, which involved aerosols, in the gas phase.

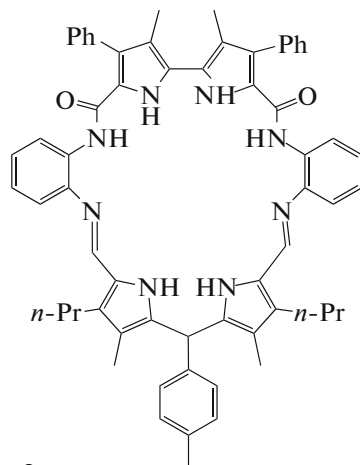
SYNTHESIS OF NANOSIZED LIGANDS FOR THE DEVELOPMENT OF RADIONUCLIDE IDENTIFICATION METHODS

The development of highly efficient systems capable of fixing anions selectively is the key to solving a

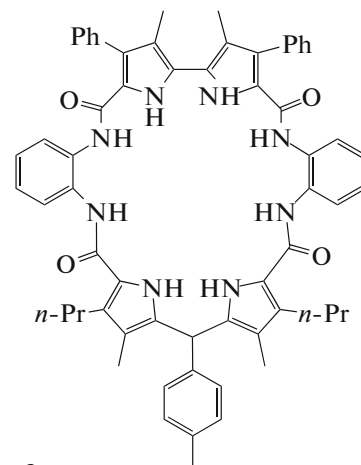
number of fundamental problems such as the identification, extraction, and separation of anions. The pertechnetate anion is the most widespread form of long-lived isotope ^{99}Tc ($t_{1/2} = 2.13 \times 10^5$ years) that is obtained from ^{235}U or ^{239}Pu . Since it is highly mobile and volatile, the research into its identification and extraction is relevant to environmental monitoring, spent nuclear fuel reprocessing, and nuclear medicine. It was found by UV-visible titration that bipyrrrole-based receptors **1**, **2**, and **3** fix selectively the perchlorate anion and the pertechnetate anion (its analog) with binding constants $K \approx 10^5 - 10^6 \text{ M}^{-1}$. The properties of ligands **1** and **2** were studied using their tetrafluoroborate salts, and ligand **3** was used as a neutral molecule. Extraction experiments revealed that receptors **1**• $(\text{HBF}_4)_2$ and **2**• $(\text{HBF}_4)_2$ may extract pertechnetate efficiently in a wide pH range even without a phase carrier. The **1**• $(\text{HBF}_4)_2$ receptor extracts 90% of pertechnetate in the interval of pH 2–13, and **2**• $(\text{HBF}_4)_2$ extracts 70% of pertechnetate at pH 3–7. Competitive extraction experiments were conducted for the most efficient ligands **1** and **2**. Their results revealed fine extraction of pertechnetate by **1** even in the presence of sulfate and perchlorate ions in a neutral medium [52–55].



1



2



3

FUNCTIONAL CARBON NANOMATERIALS

The development of highly efficient sorption materials is the basis for the advancement of sorption techniques for concentration and separation, including separation for radiochemical purposes. A method for the production of solid-phase extraction agents based on carbon nanotubes (CNTs) and organophosphorus

ligands (diphenyl(dibutyl)carbamoylmethylphosphine oxide (CMPO), TBP, trioctylphosphine oxide (TOPO), and ionic liquids of imidazole ($\text{C}_n\text{H}_{n+1}\text{MImPF}_6$) and phosphonium (Cyphos IL-101) varieties) by impregnation in a nitric acid solution was developed. The sorption properties of the indicated solid-phase extraction agents with respect to radionuclides were investigated, and the conditions of

sorption concentration and extraction of U(VI), Np(V), Pu(IV), Am(III), and Eu(III) from 3 M HNO₃ solutions were determined. It was demonstrated that Taunite CNTs with CMPO (partition coefficient $K_p \sim 5 \times 10^4$ mL/g) are preferable for group extraction of Am(III) and Eu(III), and CNTs with TOPO are best suited for extracting U(VI), Np(V), and Pu(IV) ($K_p \sim 2 \times 10^4$ mL/g). The data on the production and sorption properties of modified Taunite CNTs may be used to develop new techniques for concentration and separation of actinides and lanthanides in specific conditions [56–58].

An original electron-beam technique for the production of nanoporous charcoal from lignocellulose at the UELV-10-10T linear accelerator (8 MeV) under atmospheric pressure in the flow of a propane–butane mixture was developed. This method involves heating the initial material to $250 \pm 15^\circ\text{C}$ (for polysaccharides) or $300 \pm 20^\circ\text{C}$ (for polyphenols) with a radiation-chemical yield of polymer chain fragmentation of up to 30 cleavages/100 eV. The paths toward targeted correction (alkylation, hydration, hydroxylation, etc.) of obtained products, helping to increase their quality, were found. The electron-beam regime preserves the fibril structure and increases the free surface area and the sorption capacity of charcoal, thus making it a more efficient base material for the production of oil sorbents. The typical pore width is 1.7–2.3 nm, the specific pore surface area is up to 120 m²/g, and the maximum volume of sorbing pores is as high as 0.1 cm³/g. The sorption capacity of nanoporous charcoals (19.5 g/g with respect to oil VM-1) was determined. A method for immobilizing spent oil IS-20 in carbon materials (CNTs, activated charcoal, high-reactivity carbon mixture (HRCM), or CHP fly ash) with subsequent solidification into CaO and Portland cement was proposed and introduced at MPA. Preliminary studies of the properties (specific surface (the BET method for nitrogen absorption was used) and absorption, composition, etc.) of the indicated carbon adsorbing materials were carried out. The obtained blocks were cylindrical with a diameter of ~4.5 cm and a height of ~1.8 cm. Such a block had a volume of 30 ± 1 cm³ and a mass of 50 ± 2 g. Thus, the degree of incorporation of oil into the composition varied up to 18% by volume. It was demonstrated that the obtained composites had high holding capacities with respect to tritium (the coefficient of diffusion upon contact with water was 10^{-15} – 10^{-16} m²/s) [59–62]. The tests were carried out in accordance with GOST 29114-91. The concentration of oil in the leachate was determined by IR spectrometry.

The sorption behavior of actinides and primary fission products was studied in 2013 with a graphene oxide sample at various phase contact times, pH values, and sorbate concentrations. It was found that

equilibrium was established in the system within the first 5 min of interaction even at fairly low concentrations (<0.1 g/L). The dependences of sorption of Th(IV), Pu(IV), Am(III), Eu(III), U(VI), Sr(II), Tc(VII), and Np(V) over graphene oxide on pH were S-shaped. This shape is typical of cation sorption. The dependence of sorption of Tc(VII) on pH was an exception, since technetium was present in the solution in the form of pertechnetate anion TcO₄⁻. The sorption for the majority of cations reached quantitative values in a wide pH interval from 1.5 to 7. Selective sorption of radionuclides is feasible: for example, quantitative sorption of Th(IV) and Pu(IV) was observed at pH > 1.5, while that for Am(III) and Eu(III) was found at pH > 2.3. Quantitative desorption of radionuclides by dilute solutions of nitric acid was also demonstrated. The desorption kinetics suggests that a new dynamic equilibrium is established in the system in less than 10 min. Thus, the sorption of cations on the graphene oxide surface is reversible. The capacity of graphene oxide was calculated on the basis of isotherms of sorption of U(VI), Sr(II), and Eu(III). Although the graphene oxide concentrations were low (0.038 g/L), no saturation was observed in a number of systems. The Langmuir model ($C_{\text{sorb}} = Q_{\text{max}} \cdot K_{\text{La}} \cdot C_{\text{sol}} / (1 + K_{\text{La}} \cdot C_{\text{sol}})$) and the Freundlich model ($C_{\text{sorb}} = K_{\text{Fr}} \cdot C_{\text{sol}}^n$) were used to characterize the isotherms of sorption. Here C_{sorb} is the equilibrium concentration of a radionuclide sorbed on graphene oxide, C_{sol} is the equilibrium concentration of a radionuclide in the solution, Q_{max} is the sorption capacity, K_{La} and K_{Fr} are the Langmuir and the Freundlich constants, and n is an empirical coefficient [63].

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