

## Removal of the Am-241 from aqueous solutions using different sorbents

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This review is devoted to the problem of extracting americium from aqueous media of various origins by the sorption method based on the materials of modern world scientific studies published during two last decades. The methods for quantitative assessments of the effectiveness of sorbents are considered. The selection of data for Am(III) was substantiated taking into account the fact that trivalent lanthanides, especially Eu, are often taken as a chemical analogue of trivalent actinides Am and Cm. Various types of sorbents and materials used for binding americium radionuclides, methods of their preparation and sorption mechanisms are considered in detail. In addition, this review comprehensively considers sorption processes involving americium occurring in radioactive storages.

**Keywords:** Americium, sorption method

**Видалення Ам-241 із водних розчинів із використанням різних сорбентів.**  
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Представлений огляд, який присвячений проблемі вилучення америцію з водних середовищ різного походження сорбційним методом, базується на матеріалах сучасних світових наукових досліджень, що були опубліковані протягом двох останніх десятиліть. Розглянуто методи кількісної оцінки ефективності сорбентів. Вибірка даних для Am(III) була складена з урахуванням того, що тривалентні лантаноїди, особливо Eu, часто використовують як хімічні аналоги тривалентних актинідів Am і Cm. Детально розглянуті різні типи сорбентів і матеріалів, які використовують для зв'язування радіонуклідів америцію, способи їх отримання та механізми сорбції. Крім того, в огляді детально розглянуті сорбційні процеси за участю америцію, що відбуваються в радіоактивних сховищах.

### 1. Introduction

Radionuclides can be released into the environment through a variety of vectors, such as mining activities, leaks or spills of nuclear waste, explosion of a nuclear weapon, or terrorist activities (e.g., "dirty bombs") or accidental processes for peaceful commercial uses of the isotopes [1–4]. Spent nuclear fuel (SNF) reprocessing produces high level radioactive wastes (HLW) that contain significant amounts of the minor actinides (MA) which cause most of the ra-

diotoxicity of SNF and HLW in long and medium-term periods [5, 6]. Moreover, various activities in nuclear installations, all over the world, generate different varieties of waste streams other than High Level Liquid Waste (HLLW) produced in spent fuel reprocessing [7]. Among the minor actinides <sup>241</sup>Am, is one of the most serious concern due to its long half-life and its high radiation toxicity with  $\alpha$ -particles emission. Its maximum permissible amount in human body is 11.1 kBq and maximum permissible concentration in water is 1.48 Bq/mL.

Hence, the secure management of liquid radioactive wastes is an important, increasing problem world-wide [8] and indeed considerable attention is paid for treatment of waste water containing  $^{241}\text{Am}$  or its disposal [6, 9]. The recovery or removal of radionuclides from sea waters and waste waters is a challenging problem. According to the fundamental principles of radioactive waste management, radionuclide waste minimization is required to be done in an effective manner [3, 10]. The most commonly used methods for the removal and recovery of radionuclides includes adsorption, membrane filtration, solvent extraction, reductive precipitation, bioprecipitation, biosorption, dialysis, chromatography, and ion exchange methods [3, 4, 8, 11]. Methods based on extraction and sorption are the most widely used for this purpose [4, 12]. Among these different techniques, adsorption is the economically feasible technique and it has been developed as the guaranteed procedure for water and wastewater treatment [3, 8, 11]. In addition, the sorption data can help in predicting the partitioning as well as the migration ability of the radionuclides [13], since different sorbents are used as components of the engineering barriers in the radioactive waste repositories [8].

The problems of americium sorption are very relevant and are widely considered in modern publications [14]. Thus, the compilation of a review based on the materials of modern world scientific publications on the problem of extracting americium from aqueous media of various origins seems to be quite reasonable and relevant.

## 2. Data selection for Am(III)

Trivalent lanthanides, especially Eu, are frequently taken as a chemical analog for the trivalent actinides Am and Cm [7, 15–19]. For example, the pH dependent Eu sorption data obtained in batch experiments are consistent with spectroscopic data on Eu and Cm within experimental uncertainties thus demonstrating the validity of Eu as a homologue for trivalent actinides [15]. The results of the sorption studies with Eu and Am in water and brines using precorroded glass as substrate show clearly that the sorption behaviour of Eu is representative for trivalent actinides [18]. Considering the above all sorption data of Am, Eu and Cm were treated as representing Am sorption, i.e., no separate parameterization for Eu and Cm was done on the basis of the above-mentioned chemical analogs [16].

## 3. Quantitative characteristics of sorption

The metal sorption performance is often evaluated in terms of the distribution coefficient (*mass distribution coefficient*) ( $K_d$  and rare  $D_w$  [20], mL/g), which is a mass-weighted partition coefficient of the metal ion between solid phase and liquid supernatant phase [1, 16, 21]. The distribution coefficient is a useful measure of a sorbent's binding affinity for a specific analyte, typically in a diluted condition [1]. Commonly, the sorption distribution coefficient  $K_d$  is used to describe the partition of constituent  $i$  in solid and solution and is defined as:

$$K_d = S_i / C_i, \quad (1)$$

where  $S_i$  is the concentration of constituent  $i$  sorbed on solid,  $C_i$  is the concentration of constituent  $i$  in solution [22]. Traditionally,  $K_d$  tends to be determined in laboratory batch type sorption tests carried out at a fixed liquid volume to solid mass ratio. Based on its definition (Eq. (2)), experimentally derived  $K_d$  can be expressed as:

$$K_d = \frac{C_{init.} - C_{eq.}}{C_{eq.}} \cdot \frac{V}{M}, \quad (2)$$

where  $V$  and  $M$  are the volume of solution and the mass of solid used in the test, respectively.  $C_{init.}$  is the initial concentration/activity of sorbate in the solution, and  $C_{eq.}$  is the final (equilibrium) concentration/activity of sorbate [1, 8, 22–27]. The experimentally derived  $K_d$  value is a function of the system chemistry. It is sensitive to spatial variability in the composition of solid phases or for variable chemical conditions, such as pH, ionic strength, alkalinity, or concentrations of complexing ligands that are encountered along a groundwater flow path [22]. In transport models including radionuclide migration and simulation in performance assessment for nuclear waste repository, the distribution coefficient  $K_d$  is considered a property of the geological medium [22]. The higher the  $K_d$  value, the more effective the sorbent is for capturing that metal ion. Generally speaking, a  $K_d$  value of 5,000 and above is considered very good, and a  $K_d$  value greater than 50,000 is considered outstanding [1].

For considering of the effectiveness of a given sorbent, the efficiency of the solution

purification is often used, ( $E_r$ , %), that is the percentage of sorption of the target ions [10, 13, 24, 28], which is calculated with the formula (3) [8]:

$$E_r = \frac{C_{init.} - C_{eq.}}{C_{init.}} \cdot 100. \quad (3)$$

The metal recovery efficiency depends on the sorbent selectivity and specificity in the presence of associated and impurity inorganic and organic compounds contained in the process and waste solutions [29].

As the  $K_d$  values and sorption percentages are not always presented in literature, therefore they can be calculated using the information provided in the respective publications [23].

Another important performance characteristic of a sorbent is adsorption capacity ( $q_e$ , mg/g or Bq/g) which is show the amount of the radionuclide absorbed per 1 g of sorbent [10, 8, 24]:

$$q_{max.} = (C_{init.} - C_{eq.}) \cdot \frac{V}{M}. \quad (4)$$

To assess the efficiency of separating one metal ion ( $M_1$ ) from another ( $M_2$ ) by a sorbent, the separation factor ( $SF$ ) is used (the ratio of  $K_d$  of these ions) [20, 26, 30, 31]:

$$SF = \frac{K_d M_1}{K_d M_2}. \quad (5)$$

#### **4. Sorbents for the extraction of radionuclides of americium from aqueous media**

Materials that are capable of binding radionuclides in liquid media due to the sorption process are usually used in several directions:

- \* remediation of sea and ground waters contaminated with radioactive elements, especially, the long lived actinides [3, 32];

- \* removal of long-lived radionuclides from nuclear waste solutions [33, 34] to reduce long-term radiotoxicity of nuclear waste [35];

- \* solidification and volume reduction of the nuclear liquid wastes to obtain suitable waste forms for final disposal [36–38];

- \* to extract valuable radionuclides from liquid nuclear industrial waste [39];

- \* for multi-barrier systems which prevent migration of long-lived radionuclides from nuclear waste repositories to the environment [40].

To solve this problem different synthetic or natural sorbent materials have been studied, including polymer resins, porous silica, polymer membranes, metal oxide particles, clays, carbon-based materials or magnetic nanoparticles etc. [41, 42].

Among the inorganic sorbents for the extraction of americium radionuclides, the following prevail: materials whose solid matrices contain silicon in the form of silica gel [1, 29, 30, 41, 43–45], clay minerals such as kaolinite [46–49], bentonite [6, 16, 19, 46, 50–54], smectite/montmorillonite [46], illite [46], red clay [46, 50], vermiculite [55], Triassic clay [56], Opalinus Clay (OPA) [57], clay-salt slimes [8, 58], quartz [22, 59], nanoporous silica self-assembled with monolayer (SAMMS) [1], attapulgite (a kind of aluminum-magnesium silicate mineral) [60], silicic volcanic tuffs [22, 27, 61], natural [14, 62] and synthetic [62] zeolites, silica in the compositions of natural sediments [13].

Carbon based sorbents possess a number of properties that make them very convenient for application [63]. Activated carbon [63], graphene oxide (GO) [4, 24, 28, 33, 42, 64–66], carbonaceous nanofibers (CNFs) [10], carbon nanotubes [4, 67, 68] have been reported to be efficient sorbents of americium.

Magnetic solid-phase separation (MSPS) methodology based on Fe (hydr)oxides (FHOs; hematite, magnetite, goethite and ferrihydrite) [23, 26, 69, 70] is emerging as a promising technique for the separation of toxic and radiotoxic metal ions including americium from various aqueous streams [26]. Potassium ferrate (VI) can effectively remove americium from alkaline radioactive liquid waste since it interacts with water resulting in formation of iron(III) hydroxides without any additional reagents [71]. Mn (hydr)oxides (MnOOH; MnO<sub>2</sub>; Mn<sub>3</sub>O<sub>4</sub>) have been found to be very effective in separation of Am(III) from low level waste solutions having pH above 1.0 [34, 72]. Group IV metal oxides such as titania and zirconia [41] as well as inorganic ion exchanger based on nano-cerium vanadate [39] and thorium oxalate [73] successfully used for americium separation from spent nuclear fuel. Metal sulfides materials with a robust acid-stable layered metal sulfide structure (KInSn<sub>2</sub>S<sub>6</sub>) with the strong affinity toward trivalent minor actinides were reported for simultaneously separating trivalent <sup>241</sup>Am(III) and <sup>152</sup>Eu(III) [4].

Some synthetic polymers [35, 74–76] as well as natural biopolymers [2, 3, 9, 37, 77–82] are also used as solid matrices to develop new materials having effective sorption characteristics with respect to americium.

Apatite II, a biogenic hydroxyapatite, obtained from a natural source (fish bones), has been determined to be a good adsorbent for radionuclides of americium from neutral aqueous solutions containing common chelating agents and inorganic salts at moderate concentrations [40].

In order to develop a sorbent material for nuclear separations, several factors must be considered. For example, sorbents for selective separation of targeted elements from highly acidic aqueous solutions of irradiated nuclear fuel should preferably exhibit favourable hydrolytic and radiolytic stability, as well as chemical stability. It is considered preferable for solid phase sorbents to have open, porous structures in order to provide high surface areas which result in more active sites per gram of material (high capacity) and facilitate rapid mass transport (fast kinetics). In order for a solid phase sorbent to be practically useful in a flow-through column setup, it should also have a granular physical form with a particle size large enough to prevent issues with loss of pressure and clogging [41]. The adsorption process can be affected by some factors, such as (1) the adsorbent surface area — the larger the available surface area for adsorption, the greater the adsorptive capacity; (2) the porosity — pore size, shape, and distribution determine the adsorption capacity and velocity; (3) the physical-chemical properties of the adsorbate-solubility, polarity and the presence of substituent groups influence the species adsorptive capacity; (4) pH — the solvent and the adsorbate natures may change according to the pH of the medium (species can attract or repel themselves by electrostatic forces in an acid or basic environment) [83]. Consequently, with the objective of developing a remediation process for nuclear waste management which is of importance, the improvement and refinement of sorption material is continued [9].

Composite sorbents of natural or synthetic origin as well as functionalized solid materials prepared by surface modification of the promising sorption materials, as a rule, exhibit stronger binding for americium and have a better mechanical strength,

chemical stability, ideal size and porosity [9, 74].

Composite materials are anisotropic and inhomogeneous materials and usually present unique properties. Composite material is made by combining a minimum of two or more materials, often with different properties [84]. They are formed from the main substance, one way or another distributed in the volume of the second substance, called the matrix. Composite materials achieve the majority of their beneficial properties from a strong bond between the strong, stiff reinforcement — usually fibers (filaments) or reinforcements with other geometrical shapes, for example, particles, platelets — and the weaker, less stiff matrix [85]. Generally speaking, any material consisting of two or more components with different properties and distinct boundaries between the components can be referred to as a composite material. Correspondingly, the majority of natural materials that have emerged as a result of a prolonged evolution process can be treated as composite materials [86].

Multibarrier systems which prevent migration of long-lived radionuclides from nuclear waste repositories to the environment usually include various inorganic ion exchangers which strongly adsorb ions of radioactive metals [40]. The usage of these barriers requires a large amount of natural materials exhibiting specific physicochemical and sorption properties [8]. Different clays of complex composition [46, 54, 56, 57] and rock formations [14, 22, 27, 61, 87], as well as sediments of different geological ages and lithological type [8, 13, 62, 88] and Apatite II [40] being natural composite sorbents, are chemically active minerals capable of inhibiting the migration of radionuclides from a radioactive waste storage as part of engineering and natural barriers [56].

Synthetic composites with enhanced sorption properties in respect to positively charged Am(III) ions are produced by a sol-gel technique [11, 44, 89], chemical precipitation methods [65, 90], heterogeneous precipitation in an aqueous solution [69] and co-precipitation [65, 66, 91], chemical oxidation [65], solvothermal [4] and hydrothermal synthesis [89, 92], polymerization [65, 89], by plasma-induced polymerization technique [65], by ionotropic gelation [38, 93], by a phase inversion technique [32, 89], by immobilization of impregnated mesoporous silica in a biopolymer matrix [93], a method that includes successive stages such as se-

quential crushing, grinding, mixing, incineration and grinding [17], alkali-induced hydrolysis of hexafluorotitanic acid ( $H_2TiF_6$ ) under near-ambient conditions [94].

Much attention is given to the development of various types of solid phase extractants, i.e., of polymeric or mineral matrices on which organic compounds used in liquid extraction are immobilized by covalent or noncovalent binding [12]. Generally the surfaces of porous materials are functionalized with polymer chains either chemically (through covalent bonding) or physically (by physisorption) [95]. For immobilization on solid supports, the following reagents (organic extractants) for impregnation are widely used: diglycolamic acid [26, 30]; diglycolamides (DGAs) such as N,N,N',N'-tetraoctyl diglycolamide (TODGA) [12, 96] and tripodal diglycolamide [97]; benzene-centered tripodal diglycolamide ligands [96]; diglycolamide-functionalized calix[4]arene, in cone conformation [97]; tetrahydroxythiacalixarene tetraphosponates [76]; 2-ethylhexyl phosphonic acid mono-2-ethylhexyl ester,  $C_{16}H_{35}O_3P$ , (commercially known as PC88A) [7, 29]; picolinamide [41]; poly(amidamine) dendrimer [67]; imidazolium bis(2-ethylhexyl) phosphate (ionic liquid) [43]; N,N'-dimethyl-N,N'-dibutylmalonamide [74]; pyridine-2,6-dicarboxamide [98]; 2,6-bis(5,6-dialkyl-1,2,4-triazine-3-yl)-pyridine (R-BTP) [29]; polyazoles (1,2,3-triazole, 1,2,4-triazole and tetrazole) [75]; partially hydrogenated heterocyclic compounds (PHHC), containing nitrogen as donor atoms [45]; 8-hydroxyquinoline [35]; hydroxypyridinones [1]; amides of pyridine-2,6-dicarboxylic acid [5]; tributyl phosphate (TBP) [12]; diaryl(dialkylcarbamoylmethyl)phosphine oxides (CMPO) [12]; CMPO + TBP [12]; octyl(phenyl)-N,N-diisobutylcarbamoylmethylphosphine oxide (CMPO) [29]; 2-ethylhexyl (N,N-diethylcarbamoylmethyl)phenylphosphinate [12]; ethylenediamine (EDA) with N,N'-dicyclohexylcarbodiimide (DCC) [68]; sodium dodecyl sulfate [82]; polyacrylamide (PAM) [65]; formaldehyde [3, 9]; sodium alginate [9]; N,N'-Diethyl-N,N'-di(p-tolyl)-dipicolinamide [12]; Di(2-ethylhexyl) dihydrogen methanedi-phosphonic acid ( $H_2DEH[MDP]$ ) [12]; N-Benzoylphenylhydroxylamine (BPHA) [12]; diphenyl(dibutylcarbamoylmethyl)phosphine oxide ( $Ph_2Bu_2CMPO$ ) [12]; pyridoxal-5'-phosphate [3]; carboxylic acids [12, 68]. As solid carriers for preparing solidphase extractants for Am(III) ions removing, the fol-

lowing materials (including composites) are the most widely used: silica gel [29, 30, 43, 45]; Diapak C16 [12]; nanoporous silica [1, 12]; magnetic silica particles [41]; macroreticular styrene-divinylbenzene matrix immobilized in porous silica gel particles [12, 29]; silica-based polymeric composite material (CMPO/SiO<sub>2</sub>-P) [29]; GO [65]; multi-walled carbon nanotubes (MWCNTs) and carboxylic acid-functionalized MWCNTs [12, 67, 68]; Taunit? (carbon nanostructural material) [12]; furane resin [63]; chromatographic resins (Chromosorb-W) [97, 96]; chloromethylated Merrifield resin [74-76]; phenol based resins [35]; polymethacrylate (PMA) and polystyrene-divinylbenzene (PS-DVB) resins [5]; the commercial resin Diphonix (styrene-divinylbenzene sulfonated ion-exchange resin) [41]; Amberlite XAD-4 and XAD-7 [12]; Amberchrom CG-71 [12]; macroporous polymeric beads from polyethersulfone,  $[C_{12}H_8O_3S]_n$  (PES) [7]; polypropylene fiber [12]; polyacrylonitrile fiber [12]; titania [98]; iron oxide particles [26]; Punica granatum peel [82]; nonviable Rhizopus arrhizus biomass [3, 9]; chitosan cryogels [3].

Chemical modification of the surface of a promising sorption material is usually carried out using such chemical reactions as the amidation reaction [67, 74], functionalization with amido-amine group [68]; soft oxidation treatment [63]; condensation reaction [30], silanization reaction [1, 26, 43, 98], azid-alkyne cycloaddition [75], alkaline polycondensation [35]; Williamson reaction [76]; cross-linked polymerization [3, 9]; plasma-induced polymerization technique [65, 89]; by ionotropic elation [9].

Materials with a mobile phase of extractants are impregnates and TVEXs [29]. So-called "dry" and "wet" procedures are used for preparing solid-phase extractants by impregnation [12]. A definite amount of a reagent is dissolved in a solvent, and the resulting solution is brought in contact with the pretreated support for a definite time with stirring, in some cases under ultrasonic treatment. The "dry" procedure, which is more widely used, involves solvent removal after keeping the reagent solution with the support and drying of the material at moderate temperatures in air or in a vacuum. In the "wet" procedure, solid-phase extractants are not dried and the solvent for impregnation is taken in a definite amount required for swelling of the solid phase. At dynamic procedure a support is placed in a column and washed, after which

a reagent solution is passed directly before performing preconcentration [12]. Alternatively, materials which are called TVEXs, or Levetrel resins can be prepared by introducing an extractant into the matrix during its synthesis e.g., by suspension copolymerization of styrene with divinylbenzene in the presence of a radical polymerization initiator. Though prepared by fundamentally different methods, impregnates and TVEXs are similar in structures and properties [29]. These are materials combining the properties of sorbents and extractants and their distinctive feature is that the extractant is not chemically bonded to the solid support, which is responsible for high kinetic characteristics of sorption processes involving these materials, favorably comparing with conventional sorbents [29]. In terms of density, bulk density, and mechanical strength, impregnates and TVEXs resemble sorbents, while the recovery mechanism by which they work is similar to solvent extraction [29]. An advantage offered by impregnates is the possibility of using practically water-insoluble solvents and complexing agents with long hydrocarbon chains, whose high viscosity and surface activity make them unsuitable for recovery of aqueous phase components [29]. Unlike ion exchangers, these sorption materials do not swell in aqueous solutions; they retain mechanical strength in both wet and air-dry states, which makes them suitable for application in modern high performance equipment [29]. At the same time, it is necessary to take into account that the non-covalent modification is the reason of slow leaching of the sorbed extractant from the pores of the support material during the sorption what poses a serious limitation to reusability and, hence, the economic viability of such sorbents [74]. Specific features of this process are determined by the solubility of the extractants in the aqueous systems used [29]. Covalent grafting techniques are preferred to maximize a stable interfacial compatibility between the two phases [74, 95].

Table 1 lists some of the values of  $K_d$  of the various sorbents reported for the extraction of radionuclides of americium from aqueous media.

### **5. Types of sorption and adsorption mechanism of americium radionuclides on various sorbents**

Adsorption is a process of spontaneous redistribution of system components be-

tween the surface layer and the bulk phase. Adsorption occurs on the surface of the condensed phase, and absorption occurs in its bulk. Absorption, like adsorption, can be physical or chemical. In some situations, the differences are small or the absorption mechanism is unclear; in such cases, the more general term *sorption* is used as a synonym for absorption without specifying a detailed mechanism. In physical adsorption, the adsorbate adheres to the surface only through weak intermolecular Van der Waals interactions in an environment of low temperature and under appropriate pH conditions. In contrast, in chemical adsorption, the adsorbate molecules are held to a solid by newly created chemical bonds (covalent, ionic) due to strong interactions between adsorbent and adsorbate [99]. The main mechanisms for rare earth ions<sup>3+</sup> adsorption are: (i) surface complexation, (ii) ion exchange, (iii) precipitation, (iv) electrostatic attraction, and (v) physical adsorption. Surface complexation, ion exchange, and precipitation belong to chemisorption, which has a more significant role in the removal of rare earth ions<sup>3+</sup> from aqueous solution than physisorption (electrostatic attraction, physical adsorption). In general, the specific role of each mechanism in rare earth ions<sup>3+</sup> adsorption varies depending on the adsorbate, ionic environment of aqueous solution, and adsorbents (their surface, functional groups, etc.) [99]. So, for example, in the solutions of lower pH, ion exchange process appears to be dominant compared with complexation by the surface binding groups [8].

The sorption equilibrium is usually depicted by isotherm models whose parameters explicit the sorbent surface properties and affinity at a constant temperature [82]. The analysis of the isotherm data is important to describe how the adsorbates interact with adsorbents, affording the most important parameter for designing a desired adsorption system [24]. Adsorption isotherms reflect the influence of adsorbate initial concentration on the adsorption process [99]. In general, a sorption isotherm is an invaluable curve describing the phenomenon governing the retention (or release) or mobility of a substance from the aqueous porous media or aquatic environments to a solid-phase at a constant temperature and pH. Its physicochemical parameters, together with the underlying thermodynamic assumptions, provide an insight into the sorption mechanism and surface properties as well as the

Table 1. Comparison of  $K_d$  values of the various sorbents reported for the extraction of radionuclides of americium from aqueous media

Sorbent material	Max $K_d$ value, mL/g	pH or acid concentration
Apatite II	$2.8 \cdot 10^5$	6
Mn-doped $\text{Fe}_3\text{O}_4$	$1.5 \cdot 10^5$	8
Rhizopus arrhizus dead biomass NCIM 997	$1.3 \cdot 10^5$	2
Super paramagnetic graphene oxide — $\text{Fe}_3\text{O}_4$ nanoparticle composites	$10^5$	3
Zeolitic tuff	$\cdot 10^5$	10
Nanoporous silica self-assembled with monolayer (SAMMS) of hydroxypyridinone derivative (3,4-HOPO-SAMMS)	$\sim 10^5$	$\sim 7$
Resorcinol substituted phenol-formaldehyde resins	$8.4 \cdot 10^4$	4
Thorium oxalate-silica gel sorbent	$7.8 \cdot 10^4$	1 M $\text{HNO}_3$ + 0.1 M $\text{H}_2\text{C}_2\text{O}_4$
Hematite ( $\alpha\text{-Fe}_2\text{O}_3$ )	$7 \cdot 10^4$	9.7
Opalinus Clay	$6.5 \cdot 10^4$	7.6
Multi-podant diglycolamide-functionalized calix[4]arene (C4DGA)	$>5 \cdot 10^4$	0.01–4 M $\text{HNO}_3$
The imidazolium bis(2-ethylhexyl) phosphate modified silica gel	$4 \cdot 10^4$	0.001 M $\text{HNO}_3$
Magnetite	$4 \cdot 10^4$	7.8
Extraction chromatographic resins containing benzene-centered tripodal diglycolamide ligands	$3.6 \cdot 10^4$	3 M $\text{HNO}_3$
Tripodal diglycolamide (T-DGA) resin	$3.1 \cdot 10^4$	3 M $\text{HNO}_3$
Diglycolamic acid-functionalized multiwalled carbon nanotubes (DGA-CNTs)	$2.6 \cdot 10^4$	3 M $\text{HNO}_3$
Lime-silt sandstones	$2.4 \cdot 10^4$	7.9
Layered hydrazinium titanate LHT-9, $(\text{N}_2\text{H}_5)_{1/2}\text{Ti}_{1.87}\text{O}_4$	$2.1 \cdot 10^4$	3–6
Oven-dried biomass of <i>R. arrhizus</i> NCIM 997	$1.5 \cdot 10^4$	2
Iron oxide ( $\text{Fe}_3\text{O}_4$ ) particles functionalized with diglycolamic acid (Fe-DGAH)	$10^4$	3
Devitrified tuff	$10^4$	10
Extraction chromatographic resins containing benzene-centered tripodal diglycolamide ligands	$9.9 \cdot 10^3$	3 M $\text{HNO}_3$
Dipicolinic acid functionalized titania ( $\text{TiO}_2$ -DPA)	$8 \cdot 10^3$	1 M $\text{HNO}_3$
Triassic clay	$7.8 \cdot 10^3$	6.9
Poly(amidoamine) dendrimer functionalized carbon nano tubes	$6.4 \cdot 10^3$	0.5 M $\text{HNO}_3$

degree of affinity of the adsorbents [65]. The most frequently used adsorption isotherms for rare earth ions<sup>3+</sup> adsorption from aqueous solution are Langmuir, Freundlich, BET, Temkin, Dubinin-Radushkevitch, Hill, Redlich-Peterson, and Sips isotherms [99]. Langmuir isotherm was found to be predominately operative [98] in many cases of adsorption of americium ions from aqueous media [2, 4, 7, 10, 11, 17, 24, 26, 30, 33, 34, 36, 39, 43, 50, 60, 67, 72, 76, 81, 82, 91–93, 96–100]. The Langmuir

adsorption isotherm describes chemical adsorption and assumes: (i) a limited number of localized adsorption-binding sites on the surface of the adsorbent; (ii) binding of one molecule to each binding site (monolayer adsorption) and (iii) a homogeneous surface of adsorbent from the energy point of view (uniform affinity of all binding sites to adsorbate without interactions between adsorbate molecules) [99].

As a rule, sorption of metal ions proceeds in several stages. In the first, the

transfer of metal ions from the solution onto the sorbent takes place through a diffusion process. Then, these metal ions diffuse throughout the outer surface of the sorbent (film diffusion) or migrate inside through pores (pore diffusion) and interact with active sites of the sorbent. The overall rate of sorption is dominated by the slowest of these steps or, if the rates of several steps are comparable, is formed by a combination of them [8]. In studies on adsorption behavior, kinetics, which reflects the influence of adsorption time on the adsorption process, has generally been the primary focus and challenge [54, 99]. Researchers attempted to describe the kinetic and thermodynamic properties of adsorption by using different adsorption models, and got many valuable results. Based on experimental data, these models are obtained by combining with mathematics or computer technology [54]. Models which consider adsorption as the slowest process are the pseudo-first order, pseudo-second order, and Elovich models; adsorption is usually the slowest process when the adsorbate uptake on the adsorbent is a chemical nature. The pseudo-first order model, governed by the non-reversible kinetic equation, rests on five assumptions : (i) sorption only occurs on localized sites and involves no interaction between the adsorbed ions, (ii) energy of adsorption is not dependent on surface coverage, (iii) maximum adsorption corresponds to a saturated monolayer of adsorbates on the adsorbent surface, (iv) concentration of adsorbate is considered to be constant, and (v) metal ion uptake on the adsorbent is governed by a first-order rate equation. The assumptions for the pseudo-second order model are almost the same as for the pseudo-first order model, except that the ion uptake is governed by a second-order rate equation [99]. Pseudo-first order models rely on mass transfer from the bulk to the adsorption sites and are based on the assumption that the driving force is expressed as a concentration difference. For pseudo-second order models, chemical reaction/chemisorption is assumed to be the rate-controlling step [24]. Experimental data indicate that the pseudo-second-order model fits the best sorption of americium ions on most various sorbents [7, 8, 10, 11, 24, 29, 34, 36, 37, 43, 44, 50, 60, 67, 68, 76, 78, 96–99]. Consequently, it may be postulated that the rate determining step in sorption of  $\text{Am}^{3+}$  on these materials is chemical sorption (chemisorption), which in-

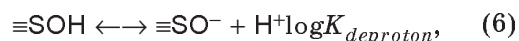
volves valency forces through mutual membership or replacing electrons between sorbent and sorbate [8].

Sorption of metal cations is pH-dependent and is characterized by a narrow pH range where sorption increases to nearly 100 %, traditionally known as an adsorption edge. The pH position of the adsorption edge for a particular metal cation is related to its hydrolysis or acid-base characteristics. In addition to pH, sorption of metals is dependent on sorptive concentration, surface coverage, and the type of the sorbent [8].

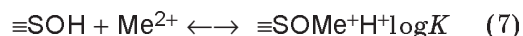
Americium in aqueous solutions is well known to exist in the III, IV, V, and VI oxidation states. The hydrated ions  $\text{Am}^{3+}$  (aq),  $\text{AmO}^{2+}$  (aq), and  $\text{AmO}_2^{2+}$  (aq) occur in the absence of complexing agents. Am(III) ion is precipitated by hydroxide, fluoride, phosphate, and oxalate ions [2]. Americium (III) forms strong hydroxo- and carbonato-aqueous complexes. As solids, it forms mixed hydroxo-carbonato species [61]. In seawater, Am(III) exists as Am(III) as a predominant species at acidic pH,  $\text{AmSO}_4^+$  at pH 7.0,  $\text{Am}(\text{CO}_3)^+$  at pH 7.8, and  $\text{Am}(\text{CO}_3)^{2-}$  and  $\text{Am}(\text{OH})_2^+$  at pH 9.0 [1].

The surface complexation constants allow prediction of sorption of americium in a natural complex system. The surface reactions involve an americium sulfate and an americium carbonate complex [22]. The generalized surface complexation sorption model for sorption of americium in a natural complex system involves 1) surface proton exchange, and 2) cation sorption on to a neutral surface hydroxyl species  $\equiv\text{SOH}$ . These reactions can be described as:

1) surface proton exchange:



2) cation sorption



[22].  $K_d$  of americium on volcanic tuffs reaches a maximum at a pH between 6 and 7. At a pH above 7,  $K_d$ 's of americium on volcanic tuffs decrease with increasing pH, likely due to the repulsion between the negatively charged americium carbonate complex and rock surfaces in the system [22]. The sorption of Am on natural sediment consisting of mineral oxides (such as silica) [13] increased with pH of the aqueous medium [10 % (pH 2) to ~ 100 % (pH 10)], which was explained in terms of the



increased negative surface charge on the sediment particles the fraction of which is increasing with the increasing pH. There is negligible effect of varying ionic strength on Am(III) sorption which suggests inner-sphere complexation/sorption process and negligible amount of the ion-exchange sites. Similar observations were made during the sorption of Am(III) onto silica and kaolinite (aluminosilicate) as a function of pH and ionic strength [13]. GO has a large surface area and a number of oxygen containing groups such as epoxy (C–O–C), hydroxyl (–OH), carboxyl (–COOH), and carbonyl (–C=O) capable of binding metals and radionuclides via coordination and electrostatic interaction [66]. The removal rate of Eu(III) by GO is connected with the relative distribution of Eu(III) species in solution and the surface properties of the GO. The Eu(III) sorption increased between pH 2.0 to 6.0 due to electrostatic attraction, and then retained this level at pH 6.0–9.0 due to surface complexation [65]. At pH < 3.9 the surface charge of GO is positive and pH > 3.9 it is negative due to protonation and deprotonation reactions, respectively. Therefore at pH > 3.9 the GO surface is charged negatively and the electrostatic attractions between metals in positive species and GO are stronger and as a result the adsorption values close to 100 % for both metals [28]. The value of Eu–C bond distance confirmed the formation of inner-sphere surface complexes on graphene oxide nanosheets (GONS). Surface complexation modeling gave an excellent fit with the predominant mononuclear monodentate  $>SOEu^{2+}$  and binuclear bidentate  $(>SO)_2Eu_2(OH)_2^{2+}$  complexes at pH < 5.0 and at pH > 8.0 accordingly [33]. The adsorption of Eu(III) on GO increases with pH increasing from 2.0 to 7.5, and then maintains high level at pH > 7.5 [24]. The adsorption of Eu(III) is mainly dominated by inner-sphere surface complexation at low pH values, whereas the adsorption is accomplished by precipitation and inner-sphere surface complexation at high pH values. Therefore, the adsorption of Eu(III) on GO is independent of ionic strength in the wide pH range [24, 33]. The effects of coexistent electrolyte ions on Eu(III) removal from solution to GO can be negligible. However, the

adsorption of Eu(III) is significantly affected by humic acids and fulvic acids (HA/FA). The adsorption is enhanced at low pH values and is reduced at high pH values [24]. Am(III)/GO coagulation occurs with the formation of nanoparticle aggregates of GO sheets, facilitating their removal. Coagulation is a sorption driven mechanism, and sorption is likely followed by bridge formation between different sheets of GO, thereby inducing the coagulation [64]. Magnetic graphene oxide (MGO2) sorbent which was uniformly covered with magnetite nanoparticles, has predominantly sorption sites typical of magnetite, and the mechanism of Am(III) sorption in this case should be similar to sorption on magnetite [66], when the exchangeable and carbonate sites play the principal role [56]. Magnetic graphene oxide (MGO1) which contained lower concentration of iron accordingly had a lower amount of magnetite nano-particles on the GO surface. In this case, both sorption sites characteristic of GO and magnetite were available for the uptake of radionuclides [66]. The maximum uptake of Am by GO was found at the initial pH values of 3–5 and at the initial pH value of 7 for MGO1, whereas an increase of Am adsorption from 60 to ~100 % was observed for MGO2 in the pH range from 3 to 9 [66]. The increasing adsorption of  $Eu^{3+}$  at pH 3.0–6.0 on the CNFs could be attributed to the presence of carboxylic groups which typically deprotonated at pH ~ 4.0 and made the CNFs negatively charged. They bond directly with cations or nanoparticles with opposite charges by electrostatic attractions. The high-level removal of Eu(III) on CNFs at pH > 6.0 was also ascribed to the precipitation of  $Eu(OH)_3$  [10]. A diglycolamic acid functionalized silica gel (Si-DGAH) [30] and iron oxide particles (Fe-DGAH) [26] contain acidic functional moieties on the surface. Therefore, these sorbents are cation exchangers. At the mutual separation of trivalent actinides and lanthanides using Fe-DGAH or Si-DGAN the distribution ratio of both Eu(III) and Am(III) decreases with an increase in the concentration of nitric acid, since this extraction was accompanied by the release of two moles of  $H^+$  ions from diglycolamic acid and co-extraction of one mole of  $NO_3^-$  ions from the aqueous phase by cation exchange mecha-

nism [26, 30]. Sorbent material created from nanoporous silica self-assembled with monolayer (SAMMS) of hydroxypyridinone derivative (3,4-HOPO) effectively captures  $^{241}\text{Am}(\text{III})$  in filtered seawater doped with competing metals at levels encountered in environmental or physiological samples, at pH 5–8 [1]. The  $K_d$  of 3,4-HOPO-SAMMS for Am(III) dropped sharply at pH 0 and at pH > 7 due to protonation of the 3,4-HOPO ligand-complex at very low pH and the poor solubility of actinides at very high pH, respectively. Actinides tend to form strong complexes with carbonate and hydroxide species under basic conditions reducing the effectiveness of all solid phase sorbent materials [1]. Bio-sorption in algae has been attributed mainly to the cell wall, where both electrostatic attraction and complexation can play a role [2]. The marine algal matrix of alginate consists of complex hetero-polysaccharides containing both OH and COOH binding sites; therefore the major binding groups are  $-\text{OH}$ ,  $\text{O}=\text{C}-\text{OH}$ ,  $\text{C}=\text{O}$ . The interaction of  $\text{Am}^{3+}$  with alginate is mostly ionic species of Am ( $\text{Am}^{3+}(\text{aq})$ ,  $\text{AmO}_2^+(\text{aq})$ , and  $\text{AmO}_2^{2+}(\text{aq})$ ) complexing with this groups. Carboxylic group exists in ionized form at pH ~ 4. As pH reaches lower values, the competition between  $\text{H}^+$  and cationic species in the solution affects the process, decreasing the metal uptake. In addition to this there is a definite probability that neutral species i.e.  $[\text{Am}(\text{OH})_3]$  may get adsorbed on the algal cell. The charge of the bio-sorbent does depend exclusively on the pH value. Covalent bonding of metals can "consume" negatively charged groups. Groups become large neutral that would otherwise have been negatively charged in metal free solution of the same pH [2].

### **6. Sorption processes involving americium, occurring in radioactive storages**

The safe disposal of radioactive wastes and specifically the need to protect humans and the environment in the far future is given particular attention in all countries engaged in nuclear power generation [9, 19]. According to the International Atomic Energy Agency (IAEA) guidelines, a geologic repository shall be designed based on the concept of a multibarrier system for radioactive waste isolation [6]. Key compo-

nents of such system that provides isolation of radionuclides from the environment are: a matrix (waste form) with incorporated radionuclides in metallic containers, an artificial anti-migration clay barrier (mostly smectite) and host rock [6, 19]. However, it is impossible to guarantee the long-term stability and integrity of the engineered barrier system [19]. Radionuclide migration from the repository would be impeded in the natural system by interactions among water, nuclides, and immobile mineral phases in the rock [61]. The steel in the engineered barriers (including rebar) for disposed waste or the carbon-steel tank liners holding the liquid waste do not sorb the radionuclides [23]. However, once the overpack comes into contact with groundwater higher concentrations of  $\text{CO}_3^{2-}$  ions, it will begin to corrode and, therefore, smectite could interact with dissolved iron, hydrogen gas and other corrosion products of the steel overpack such as magnetite ( $\text{Fe}_3\text{O}_4$ ), goethite  $[\text{FeO}(\text{OH})]$ , or siderite [19]. It was shown that in the aqueous systems, the sorption percentage or  $K_d$  of AM onto natural hematite increases with increasing pH from 2.5 to 5.7; at the same pH 5.7, the decrease in ion strength  $\text{NaNO}_3$  from 0.1 M to 0.001 M significantly increased Am sorption onto natural hematite. The increasing content of fulvic acid (FA) from 0 to 20 mg/L significantly increased the Am(III) sorption onto natural hematite at pH 5.7. Similarly, addition of HA ( $4.7 \cdot 10^{-5}$  M) to Am(III)-hematite systems also significantly increased the Am(III) sorption rates to near 100 % at the pH range of 4–6, independent of ion strength (0.05–0.5 M  $\text{NaClO}_4$ ). These results demonstrate that how strong FA, HA, and likely natural organic matter as complexing agents would influence Am(III) sorption to FHOs and other minerals. At pH 5.5 or 8.0, Am(III) sorbed onto ferrihydrite surfaces as a bidentate corner-sharing species which indicates that the bonding is strong and desorption is less likely to occur than in a monodentate system [23]. Moreover it was found that the steel container immobilized the high-radioactive waste, even during the corrosion process. Samarium formed of an insoluble phase of samarium silicate on the container surface. There was no evidence of samarium diffusion through the metallic container. Samarium remained adsorbed by the container also after desorption experiment with water. Therefore, steel canister is actively involved in the HRW immobilization [19]. Alumophosphate and

borosilicate glasses are the most common matrixes for vitrification of HLW in the world due to economic advantages, the low requirements for initial radioactive waste conditioning, the relative simplicity of production, and their chemical durability that meets local legal regulations and guidelines of IAEA. The solubility of phosphate glass components and their crystallization during interim storage caused by elevated temperature and water vapour could result in formation of colloids in the aqueous phase. An increase in temperature caused an increase in the dissolution rate and alteration depth of phosphorous glass, and therefore an increased concentration of phosphate in water in the near field deep geological repository environment. It could affect on speciation of radionuclides and therefore their behavior in the near field environment, both in the liquid phase and for sorbed species. Americium can form insoluble phosphates at neutral and basic pH, and complex ions with  $\text{HPO}_4^{2-}$  and  $\text{H}_2\text{PO}_4^{2-}$  anions in acidic media. Am could also be associated with colloidal particles in solution or it could form its own colloids, designated radiocolloids. However, it was found that an increase in the Am water soluble mobile fraction is accompanied by an increase in the amount of strongly bound radionuclide species in presence of phosphate in the liquid phase. Taking into account the low diffusion rates in a geological environment and through a bentonite barrier, such a redistribution indicates that the clay barrier is an important and essential part of the safety system of a radioactive waste repository over the studied conditions [6]. Moreover the study [22] convincingly demonstrates that even in the worst case scenario, i.e. in the presence of very weakly adsorbing surfaces in geologic repository for the disposal of spent nuclear fuel and high-level radioactive waste, americium is virtually immobile as reflected by its very high  $K_d$  value.

Understanding the migration behavior of radioactive and non-radioactive toxic elements in geological environments is very important for the reliable long-term safety assessment of potential nuclear waste disposal sites and in nuclear waste management. Sorption and desorption of the hazardous elements onto mineral surfaces are important for the migration behavior of actinides in the natural aquifer system [47]. Geochemical processes, such as ion exchange, adsorption-desorption, dissolution-precipitation, redox processes, aqueous com-

plexation, surface complexation, colloid formation and migration, and microbiological processes, control the transport behavior of radionuclides [23]. The sorption data incorporated in the geochemical codes can help in predicting the partitioning as well as the migration ability of the radionuclides [13]. The geochemistry of the rocks and fluids in the natural system will influence the long-term behaviour of components in an engineered barrier system and the ability of any radionuclides that are eventually released to migrate away from a nuclear waste repository. The geochemistry also provides evidence of the way in which geologic and hydrologic processes operated in the past [61]. Field experiments and environmental monitoring observations have shown that Am transport can be enhanced by the presence of colloids [13, 53]. These colloids originate from the weathering of the surrounding rock structures, and comprise either various pure mineral oxides, silicates, carbonates or clay minerals (a composite mixture). Partitioning of the radionuclides in the aquatic body between the colloidal particles and the bulk aqueous medium depends on the chemical interaction-complexation/precipitation — between the radionuclides and surface sites of different minerals, and is influenced by the surrounding chemical environment [13]. Three criteria are important for colloids to increase the transport of radionuclides such as Am: (1) colloids must be mobilized, (2) mobile colloids must be transported long distances and (3) radionuclides must associate strongly with mobile colloids [53]. Radionuclide concentrations along flow paths from a repository could be limited by (1) low waste-form dissolution rates, (2) low radionuclide solubility, and (3) radionuclide sorption onto geological media [61]. It was shown that although bentonite colloids were highly mobile, the Am that was sorbed to them was readily desorbed and would not pose a significant transport risk if leaked from a repository with oxidizing pH = 8 groundwater and the absence of other competing radionuclides [53]. Colloid-mediated transport of Am in a bentonite-fracture fill material system is unlikely to result in transport over long distance scales because of the ability of the fracture materials to rapidly strip Am from the bentonite colloids and the apparent lack of a strong binding site that would keep a fraction of the Am strongly-associated with the colloids [53].

It has been established that in water with a high content of dissolved organic matter (DOM), which is the result of microbial degradation of mining debris (diesel, wood, etc.), the sorption of Am on an inorganic matrix is kinetically limited due to the aqueous Am-DOM complexation. This results in a higher Am transport rate. The slow disassociation of Am from soluble Am-DOM complexes and its subsequent sorption to mineral surfaces provides a mechanism for unretarded Am transport in the short term but also suggests that Am-DOM aqueous complex formation is reversible. Low diffusion rates of Am-DOM macromolecular complexes may focus Am transport into fractures and minimize retardation via matrix diffusion. The examination of Am sorption indicates that water chemistry, notably DOM, will play a major role in controlling Am mobility in ground waters [27].

### Conclusion

The presence of long-lived radionuclides in natural aquatic systems is of great environmental concern in view of their possible migration into biosphere of mankind [52]. This migration is significantly influenced by various factors such as pH, complexing ions present in aquatic environment and sorption of species involving radionuclides on the sediments around the water bodies.  $^{241/243}\text{Am}$  are two major radionuclides which can contribute a great deal to radioactivity for several thousand years [13]. The focus of this review is on sorption studies of the absorption of americium by various sorbents, carried out in recent decades. Methods for quantitative evaluation of the effectiveness of sorbents are considered. The selection of data for Am(III) is substantiated taking into account the fact that trivalent lanthanides, especially Eu, are often taken as a chemical analogue of trivalent actinides Am and Cm. Various types of sorbents and materials used for binding americium radionuclides, methods of their preparation and sorption mechanisms are considered in detail. In addition, this review comprehensively considers sorption processes involving americium occurring in radioactive storages. At the same time, it should be noted that the insufficient amount of literature on radiolytic degradation of the sorbent, stripping of metal ion from the loaded sorbent and interaction of sorbent functionality to the metal ions made the comparison very difficult [78]. In addition despite the large number of publications devoted to the

extraction of americium radionuclides from the aquatic environment, there are very few examples of solid phase materials used for nuclear separations at industrial or pilot scales [41].

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