Radionuclides Removal from Aqueous Solutions: A Mini Review on Using Different Sorbents

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Radionuclides Removal from Aqueous Solutions: A Mini Review on Using Different Sorbents

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Abstract: *In this study, mini review is presented on the use of different sorbents for radionuclides removal of from water. Four sorbents categories were considered: carbon-based, nanomaterials, bio-sorbents, and miscellaneous sorbents. Carbon-based sorbents showed excellent removal performance towards radionuclides, owing to the high specific surface area and abundant oxygen-containing functional groups. While Fe^0 and Fe^0-based composites was found to exhibit rapid sorption rate, high removal capacity, and strong redox performance for radionuclides, indicating that such nanomaterials can be very promising for in-situ removal of radionuclides, once their drawbacks (aggregation and poor mobility) are overcome. The use of bio-sorbents showed high potential of such materials to remediate radioactive contaminated water under specific environmental conditions. The critical review in this study shall represent a great contribution to the potential researchers and decision makers choosing the highly efficient, feasible, and environmentally friendly sorbents for the removal of radionuclides from aqueous solutions.*

Keywords: radionuclides; sorption; mini review; nanomaterials; bio-sorbents.

1. INTRODUCTION

Water is the source of life on earth, and it is and will be the major challenge for our existence to have a drinkable and uncontaminated water. The main sources of water contamination are, discharging wastewater from industrial activities, municipal wastewater, human agricultural activities, and environmental/global changes [1].

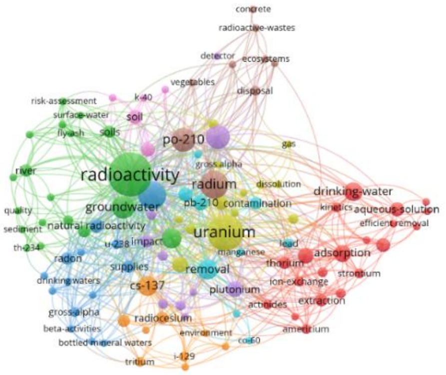

Fig. 1. Bibliometric map of radionuclides-related keywords occurrence in the literature (Scopus database 2011-2021).

Owing to the current grown concern with the development of nuclear power, water contamination with radionuclides has become another great environmental concern. Radionuclides are chemical elements emitting either α-, β-, or γ-rays, or neutrons, which can be classified as naturally occurring and anthropogenic [2]. The naturally occurring radionuclides include uranium (^{238}U, and ^{235}U), thorium (^{232}Th), tritium (^{3}H), etc. While the anthropogenic radionuclides mostly include isotopes of cesium (^{137}Cs), iodine (^{129}I), strontium (^{90}Sr), technetium (^{99}Tc), plutonium (^{239}Pu), cobalt (^{60}Co), radium (^{226}Ra, and ^{228}Ra), radon (^{222}Rn), and americium (^{241}Am) [3]. Figure 1 shows the bibliometric map of radionuclides-related keywords occurrence in the literature based on Scopus database within the period from 2011-2021. Such bibliographic map reflected the emerging concern towards most of the formerly mentioned radionuclides and the clear connection with their presence in water, monitoring, and removal technologies.

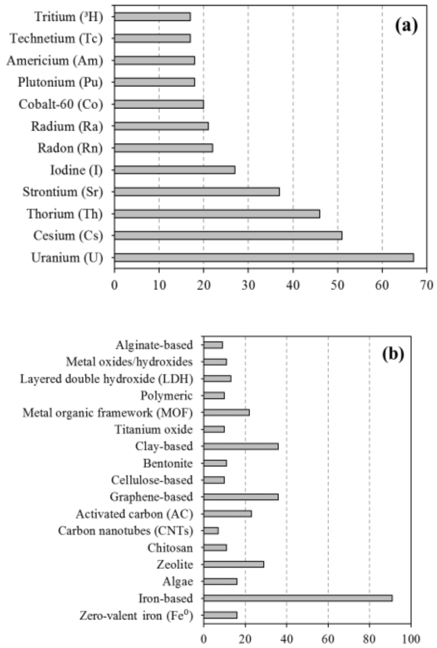

Fig. 2. Number of publications of (a) radionuclides removal from water, and (b) sorbents used for radionuclides removal from water (Scopus database 2011-2021).

The main source of the presence and release of the anthropogenic radionuclides into water bodies is the radioactive waste generated by nuclear power plants, nuclear weapon production and/or testing, and nuclear fuel processing units. The exposure to contaminated water with radionuclides may have more detrimental health concerns because of the chemical as well as radiological toxicities, including cancer at different organs (lungs, breast, colon esophagus, thyroid, and ovary), birth defects, neurological disorders, and infertility [4,5]. Owing to such multi-toxic nature in addition to the long half-life time of the radionuclides, the world health organization (WHO), and U.S. environmental protection agency (USEPA) recommended maximum contamination levels of radionuclides in water. Hence, several reported research studies have been concerned with developing different technologies for the removal of radionuclides from aqueous solutions. As presented in Fig. 2(a), and based on Scopus database, around 361 studies were reported in the literature within the last decade on the removal of the most concerning naturally occurring and anthropogenic radionuclides from water. Uranium and cesium topped this list with a total publications number of 67 and 51, respectively. Followed by thorium, strontium, iodine, and then the others. To date, various conventional and modern removal technologies have been considered in the literature for the removal of radionuclides from aqueous solutions, including chemical precipitation, ion exchange, reverse osmosis (RO), electrodialysis, solvent extraction, and evaporation [6–11]. Precipitation is relatively simple and cost effective in converting the aqueous radionuclides into hydroxides, sulfides, and carbonate precipitates. While ion exchange and RO are the most widely used in the ex-situ removal of radionuclides from contaminated groundwater. All of these approaches have been experimentally and theoretically evaluated for the separation of radionuclides from aqueous solutions. However, most of these methods still present limitations in terms of the high installation costs, secondary pollution, and complex operation. Impressively, sorption technology has recently attracted great attention for its merits as a highly efficient, versatile, and easy-to-operate technique for eliminating trace levels of radionuclides from water [12]. Within the past 10 years, different kinds of sorbents have been extensively investigated for the removal of radionuclides from aqueous solutions, including nanoscale zerovalent iron (Fe^0), graphene oxide (GO), zeolite, carbon nanotubes (CNTs), activated carbon (AC), etc., as shown in Fig. 2(b). All of these reported sorbents have their pros and cons, in terms of the reactivity, applicability, and feasibility.

Hence, in this study, a mini review is presented on the removal of radionuclides from water via sorption technique considering several reported sorbents in the literature, categorized as 4 major types, including carbon-based sorbents, nanomaterials, bio-sorbents, and miscellaneous sorbents. The main aim of this study is to present a critical review on the performance of such sorbents in radionuclides removal from water to help the potential researchers and decision makers choosing the coming path for research and development towards highly efficient, feasible, and environmentally friendly sorbents for the removal of radionuclides from aqueous solutions.

2. SORPTION of RADIONUCLIDES

A wide range of sorbents have been reported in the literature for the removal of radionuclides from aqueous solutions. For the simplicity, sorbents have been classified into 4 different categories, as follows.

2.1 Carbon-based sorbents

Several carbonaceous materials have been reported in the literature for the sorption of radionuclides from aqueous solutions, such as activated carbon (AC), graphene oxide (GO), reduced graphene oxide (rGO), and carbon nanotubes (CNTs). These carbon-based materials have great abilities as sorbents owing to their higher acid-base stability than other inorganic sorbents, in addition to the better thermal resistance comparing with organic exchanger resins [13,14]. Table 1 summarizes some of the reported studies on radionuclides removal by carbon-based materials.

2.1.1. Activated carbon (AC)

AC is advantaged by its remarkable specific surface area (500–1500 m^2/g) in addition to the microporous structure [15]. Generally, several carbonaceous materials were utilized in the preparation of AC, including agricultural byproducts such as wood, coal, coconut husks, bamboo, plum kernels, lignin, etc. [16–18]. It has been reported before that pure AC has shown good removal performance towards U(VI), especially at acidic conditions, with maximum sorption capacity of 28.30 mg/g [13]. While Chen et al. reported that combining AC with GO in one composite resulted in a higher U(VI) sorption capacity of 298 mg/g at less acidic conditions (pH 5.5) [19]. Meanwhile, Moloukhia et al. found that modified AC synthesized from coconut shells was able to remove Cs(I) and Sr(II) with saturation capacities of 69.85 and 60.00 mg/g, respectively [20]. The sorption mechanism that mainly governs the removal of radionuclides by AC is the electrostatic interaction, via repulsion or attraction). Hence, modifying AC’s surface with different functional groups (e.g., hydroxyl, carbonyl, carboxyl, methyl, etc.) could be one of the techniques that enhances the sorption capacity towards radionuclides [21]. In such regard, it was reported that maximum ^{60}Co sorption capacity of 121 mg/g was achieved by AC prepared from doum stone (activated with H_3PO_4), with the involvement of sorption and rate-controlling removal mechanisms [22].

2.1.2. Graphene-based

Graphene oxide (GO) is one of the suitable materials for radionuclides removal because of the availability of oxygen functional groups on its surface. It has been reported before that the sorption mechanism of U(VI) on GO surface was inner-sphere surface complexation, and the high sorption capacity was achieved owing to the good dispersive features of GO which provided more available sorption sites [23]. While in case of Cs(I) and Sr(II) sorption by GO, ion exchange was the main removal mechanism involved [24]. Manipulating the microstructure and self-assembly of GO can enhance its colloidal behavior within radionuclides removal from aqueous solutions.

Table 1. Radionuclides sorption by carbon-based materials

Sorbent	Nuclide	Initial conc. (mg/L)	Sorbent dosage (g/L)	pH	Maximum sorption capacity (mg/g)	Ref.
AC	U(VI)	100	1.0	3.0	28.30	[13]
	U(VI)	200	1.2	4.5	82.00	[14]
	U(VI)	50	4.0	5.0	8.20	[25]
	Cs(I)	100	5.0	6.5	67.90	[26]
	Sr(II)				29.60	
	Co(II)	100-250	1.2	6.0	121.0	[22]
GO	U(VI)	—	0.4	4.0	299.0	[27]
	Sr(II)	20	0.5	7.0	702.18	[28]
GO-AC	U(VI)	50	0.2	5.5	298.0	[19]
MWCNTs-Fe_3O_4	Th(IV)	20	0.2	4.0	32.60	[29]
	U(VI)	20	0.2	4.0	36.3	
	Sr(II)	0.018	0.6	7.0	—	[30]
Ca/Al-LDH-MWCNTs	U(VI)	30	0.1	6.0	115.0	[31]

2.1.3. Carbon nanotubes (CNTs)

CNTS have unique structural, physico-chemical, mechanical, and electronic properties. They can be classified according to their structure into single-wall carbon nanotubes (SWCNTs), and multi-wall carbon nanotubes (MWCNTs). MWCNTs are much stronger than SWCNTs, related to the involvement of several tubes set in concentric layers within the cylindrical shape. Moreover, MWCNTs can exhibit higher efficiency than SWCNTs, especially in the highly corrosive environment, owing to the low oxygen reduction when used as a catalyst. It has been reported that doping the surface of MWCNTs with other efficient materials can enhance its performance towards radionuclides removal. Magnetic MWCNTs, as a composition with Fe_3O_4, showed good sorption performance towards U(VI), Th(IV) and Sr(II), with a clear dependency on pH and ionic strength [29,30]. While decorating MWCNTs with Ca/Al layered double hydroxides resulted in enhanced sorption capacity of U(VI) [31].

2.2 Nanomaterial sorbents

2.2.1. Iron-based

Iron is the fourth most abundant element on the Earth. The main two aqueous forms of iron are the relatively water soluble Fe(II) and the highly water insoluble Fe(III) [32–37]. Zero valent iron (Fe^0) is one of the iron forms that can be found under specific environmental conditions [38–41]. Fe^0 is one of the most widely used reactive nanomaterials in water treatment applications within the last decade. The unique features of Fe^0, represented in the nano size, high redox potential, core-shell structure, and high specific surface area, enables it to remove most of the soluble contaminants via several removal mechanisms (e.g., sorption, precipitation, co-precipitation, and reduction) [42–46]. As presented in Table 2, the use of iron-based sorbents in the removal of radionuclides from water has been intensively reported in the literature, owing to the great sorption abilities of such materials [47]. It has been reported before that Fe^0 could efficiently remove Tc(VII) with maximum sorption capacity of 27.5 mg/g [48]. Doping the surface of Fe^0 with Cu resulted in better anti-aggregation effect and hence enhanced removal of ^{137}Cs [49]. Furthermore, supporting Fe^0 on LDH improved its sorption capacity towards U(VI) to reach 176 mg/g at pH 5.0 [50].

2.2.2. Metal oxides/hydroxides

Different metal oxides/and hydroxides have been investigated in the literature for radionuclides removal. For instance, titanium oxide (TiO_2) which has high chemical stability within a wide pH range in aqueous systems. It has been reported that the presence of HCOONa would enhance the removal effect of U(VI) on the surface of TiO_2 with improved sorption effect from 178 mg/g to 221 mg/g [51]. While, the 3D Ag_2O-Ag/TiO_2 composite yielded maximum sorption capacity of radioactive I(I) of 212.77 mg/g [52]. Furthermore, the use of environmentally-friendly metal hydroxides, such as magnesium hydroxide colloids, have proven their efficiency in the removal of Am and Pu from alkaline aqueous solutions towards further applications within nuclear industry processes [53].

2.3 Natural and bio-sorbents

2.3.1. Zeolite

Zeolites are one of the most effective sorptive materials for the removal of radionuclides from water, due to their high thermal, radiation and mechanical stability, high sorption capacity, and selectivity. Still, they suffer from poor stability due to Si and Al dissolution at alkaline pH range in addition to the separation problems of the spent material [54]. Hence, combining zeolites with another materials, such as Fe^0, resulted in better Cs(I) and Sr(II) removal and enhanced applicability by the magnetic separation [55,56].

2.3.2. Bio-sorbents

Bioremediation of radioactive contaminated water has been intensively studies in the literature, considering different sorbents such as algae-strains and biomass, owing to their biocompatibility and high sorption abilities [57]. Fukuda et al. reported that *Galdieria. Sulphuraria* cells recovered 52 ± 15% of 30 µg/L Cs(I) within potassium-deficient medium in 10 days [58]. While the accumulation of water-soluble radioiodine was confirmed by the novel strain of microalgae *Parachlorella* sp. *binos* (Binos), owing to the thick alginate-containing extracellular matrix and abundant chloroplasts [59]. Such findings give a clear indication of the high potential of using bio-sorbents to remediate radioactive contaminated water under specific environmental conditions.

Table 2. Radionuclides sorption by iron-based nanomaterials

Sorbent	Nuclide	Initial conc. (mg/L)	Sorbent dosage (g/L)	pH	Maximum sorption capacity (mg/g)	Ref.
Fe^0	Tc(VII)	0.32-32	—	6.8	27.5	[48]
	^{137}Cs	100	20	8.0	2.60	[49]
	U(VI)	10	0.1	5.0	27.0	[50]
Fe^0/Cu	^{137}Cs	100	20	8.0	3.10	[49]
LDH@Fe^0	U(VI)	10	0.1	5.0	176.0	[50]
Fe^0/biochar	U(VI)	20	2.0	7.0	55.14	[60]
Fe^0/carbon nanofiber	U(VI)	20	0.5	3.5	39.53	[61]
Fe^0/CMC	U(VI)	60	3.0	5.0	322.5	[62]
Fe_3O_4	U(VI)	21.7	0.1	9.0	70.7	[63]
Fe_3O_4/SiO_2	Eu(III)	0.0001 M	—	7.0	37.9	[64]
Fe_3O_4/magnesium silicate	U(VI)	0.00002 M	1.0	5.5	242.5	[65]
Prussian blue/Fe_3O_4	Cs(I)	—	—	5.5	16.2	[66]
Magnetic chitosan beads	Sr(II)	5-50	1.3	8.2	11.6	[67]
Fungus/ Fe_3O_4	Th(IV)	20	0.2	3.0	101	[68]

2.4 Miscellaneous sorbents

2.4.1. Metal organic frameworks (MOFs)

The inorganic-organic hybrid porous materials, known as metal organic frameworks (MOFs), are unique with their ultrahigh/permanent porosity, flexibility in structure tuning, high surface area, and water stability [69]. Hence, several studies have reported the use of MOFs for radionuclides removal from aqueous solutions via several removal mechanisms, with high sorption capacity values (ranged from 100 to 350 mg/g, up to date), towards U(VI), Cs(I), Sr(II), and Th(IV) [70–73]. The highest sorption capacity was recorded for the sorption of U(VI) by the amino-functionalized acid resistant MOFs [74].

2.4.2. Cellulose-based

Cellulose is the most broadly distributed and regenerative biopolymer in nature, a very promising and low-cost raw material for preparing various functional materials. Cellulose-based materials have been greatly utilized in the immobilization of radioactive wastes from aqueous solutions. Abdel-Galil et al. reported that using cellulose/HO_7Sb_3 nanocomposite for Cs(I) and Co(II) removal yielded maximum sorption capacity of around 13.0 and 22.0 mg/g, respectively, owing to the presence of bifunctional ion-exchange groups (C–OH and Sb–OH) [75]. Developing bacterial cellulose membrane modified with EDTA provided a new route for integrating the cellulose-based sorbents in an efficient membrane for the removal of Sr(II) with maximum sorption capacity of 44.86 mg/g [76].

2.4.3. Layered double hydroxides (LDHs)

LDHs were discovered in the 1840s as typical anionic layered structure compounds composed of positively charged host layers and guest interlayer anions, which are assembled by the non-covalent bond interaction [77,78]. Their environmental potential can be clearly observed redox reactions, and photocatalysis [79]. Generally, LDHs are expressed as the formula $[M^{2+}_{1-x}M^{3+}_x(OH)_2]^{x+}(A^{m-})_{x/m}\cdot nH_2O$, where M^{2+} and M^{3+} represent the divalent (Mg^{2+}, Zn^{2+}, Co^{2+}, Mn^{2+}, Ni^{2+}, and Ca^{2+}) or trivalent (Al^{3+}, Fe^{3+}, and Cr^{3+}) metal cations, A^{m-} is the interlayer charge-balancing anion (e.g., Cl^-, ClO_4^-, NO_3^-, CO_3^{2-}, and SO_4^{2-}), and x is regarded as the molar ratio of $M^{2+}/(M^{2+} + M^{3+})$ [80].

It has been previously reported that preparing MgCoAl-LDHs yielded high catalytic activation energy (60.5 kJ/mol) [81]. Using Ca/Al LDH@CNTs composite for efficient removal of ^{241}Am(III) and U(VI) from water was studied by Chen et al. [31]. Moreover, ion exchange was the main removal mechanism in the sorption of I^- and TcO_4^- onto Mg-Al-LDHs [82,83]. While intercalating Zn-Al-LDH with hexacyanoferrate (II) ions resulted in a maximum sorption capacity towards Cs(I) of 56.2 mg/g at pH 6.0 [84]. Several LDH-based sorbents; SiO_2@LDH, Fe_3O_4@polydopamine@LDH, graphitic-C_3N_4@LDH, and polydopamine@LDH, were utilized for U(VI) removal from aqueous solutions, where they showed maximum sorption capacities at pH 5.0 and 25 °C (considering different sorbent dosage) of 303.1, 344.4, 99.7, and 142.9 mg/g, respectively [85–88].

2.4.4. Alginate-based

Alginate is a natural polysaccharide that can be extracted from brown algae. It has several advantageous features to be non-toxic, polymeric structure, biodegradable, biocompatible, and rich with carboxyl and hydroxyl functional groups, which makes it one of the candidates for radionuclides removal from aqueous solutions. Hence, alginate-based sorbents; alginate/humic acid/Fe-amino clay, Prussian blue impregnated in alginate gel, graphite nano carbon/alginate, and graphene oxide encapsulated polyvinyl alcohol/sodium alginate hydrogel, have shown great removal performance towards several radioactive elements, including Sr(II), Cs(I), Co(II), and U(VI), with high values of maximum sorption capacity 45.65, 55.6, 11.63, 403.78 mg/g, respectively [89–92].

3. CONCLUSIONS & PERSPECTIVES

The environmental applications of different sorbents have been briefly reviewed in this study for the removal of radionuclides from aqueous solutions. Carbon-based sorbents showed excellent removal performance towards radionuclides, owing to the high specific surface area and abundant oxygen-containing functional groups. The defects of GO, represented in high cost and easy aggregation, were solved via considering different inorganic and organic modified carbon-based materials, aiming to design and prepare the more efficient and practical carbon-based sorbents. Fe^0 and Fe^0-based composites was found to exhibit rapid sorption rate, high removal capacity, and strong redox performance for radionuclides, indicating that such nanomaterials can be very promising for in-situ removal of radionuclides in environmental remediation once their drawbacks (aggregation and poor mobility) are overcome. Utilizing the water-stable MOFs in radionuclides removal from water can yield great sorption performance via several removal mechanisms, including ion exchange, combined sorption-reduction, electrostatic interaction, hydrogen binding, coordination interaction, and Lewis acid-base interaction. Integrating cellulose-based sorbents in a membrane structure can provide high hydrophilicity and easy separation for the sorption of radionuclides in aqueous solutions. The structural properties of LDHs yielded outstanding characteristics in terms of the existence of multiple alternative metal cations allowing chemical compositions of host layers to be precisely controlled, and the unique memory effect resulting in the restorability to their original layered structures. Such unique features enabled LDH-based nanomaterials to show great performance in radionuclides removal from water. The advantages of alginate-based sorbents have been depicted, in terms of the biocompatibility, biodegradability, non-toxicity, polymeric structure, abundancy and strong affinity to aqueous radionuclides. Hence, more attention should be paid on the development of alginate-based sorbents that are multipurpose, eco-friendly and potentially beneficial to wastewater treatment application. In term of cost-effectiveness, more work should be pursued to recycle the adsorbents using effective desorption agent and regeneration strategies.

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