

Nanostructured Materials for Uranium Removal

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Groundwater is commonly used as a drinking water resource all over the world. Therefore, groundwater contamination by toxic metals is an important issue of utmost concern for public health, and several technologies are applied for their effective removal, such as coagulation, ion exchange, adsorption, and membrane applications like reverse osmosis. Adsorption is acknowledged as a simple, effective and economic technology, which has received increased interest recently, despite certain limitations regarding operational applications. The respective scientific efforts have been specifically focused on the development and implementation of novel nano-structured adsorbent materials, which may offer extensive specific surface areas, much higher than the conventional adsorbents, and hence, are expected to present higher removal efficiencies of pollutants. In this paper, the recent developments of nanomaterial applications for uranium removal from groundwaters are critically reviewed. Particularly, the use of novel composite materials, based mainly on hybrid metallic oxide nanoparticles and on composites based on graphene oxide (GO) (i.e., graphene-based hybrids), showed promising evidences to achieve efficient removal of toxic metals from water sources, even in full scale applications.

Keywords: nanostructured adsorbents ; graphene ; uranium

I Uranium (U)

Water pollution by toxic metals and metalloids (e.g., arsenic, copper, mercury, cadmium, lead, uranium, chromium) is nowadays considered a serious global environmental problem, especially for groundwaters. Even in concentrations in the $\mu\text{g L}^{-1}$ range, their presence in various water bodies is considered particularly dangerous for human health and hence, their maximum allowable concentration in waters is strictly regulated by international organizations (World Health Organization (WHO), US Environmental Protection Agency (EPA), EU). Therefore, the efficient treatment of contaminated waters for the removal of toxic contaminants is necessary to provide people with safe drinking water. In order to detoxify the polluted waters, various treatment techniques, such as coagulation–flocculation, lime softening, photocatalytic, chemical or biological oxidation, bioremediation, ion-exchange, reverse osmosis, and adsorption have been employed with different efficiency degrees, depending upon the specific applicable condition ^[1]. All the relevant compounds of the examined toxic metals have the characteristic of being present in waters as oxyanions, in pH relevant to ground/waters, i.e., 6.5–8.5. U(VI) is present mainly as the uranyl cation (UO_2^{2+}), which, in waters containing bicarbonate anions however, is most probably complexed and presented as $\text{UO}_2(\text{CO}_3)_2^{2-}$ ^[2].

These oxyanions can usually be removed from waters by the application of different technologies, due to differences in their aquatic chemistry. U(VI) is mostly removed by lime softening or adsorption onto iron oxides ^[1]. In several cases, these oxyanions have been simultaneously present in natural ground/waters and their adsorption on efficient adsorbents with extended surface areas could be a perfect solution, when considering their simultaneous removal.

Adsorption is a conventional technology which can have applications in the removal of all these toxic oxyanions and therefore, in this review, several novel nanostructured adsorbents are critically evaluated, in order to present their ability regarding the removal of these metals from ground/water sources.

Considerable interest has attracted the preparation and use of novel nanostructured adsorbent materials with average particle sizes below 100 nm, expecting that their properties will be superior to those of conventional materials with structures of bigger size. The application of nano-scaled adsorbents comprises a novel and promising technology, especially for toxic metals removal from water sources, because of their respective substantially higher surface areas, which may provoke higher reactivity and increased affinity, capacity and selectivity for different metals ^[2].

Identified as a harmful element, uranium (U) is plentiful in nuclear waste disposal facilities (and testing sites), as well as in uranium mining, processing and milling sites. In groundwaters, it is usually present in the form of U(VI). Uranium contamination poses a threat to both surface and groundwaters ^{[3][4]}. Uranium disposed into the environment can

ultimately reach the top of food chain and be consumed by humans, initiating severe kidney or liver damage and even death [5]. Thus, the WHO and US EPA recognized U(VI) as a human carcinogen and recommended $30 \mu\text{g L}^{-1}$ as the temporary guideline level for its presence in drinking water, after the 2012 relevant revision [6]. The contamination of groundwaters with uranium is a subject of concern in several countries around the world, such as in the USA, Canada, Germany, Finland, Norway, and Greece [6]. Therefore, it is very important to choose a suitable and effective method to remove uranium from water sources.

U(VI) is the most ubiquitous species of uranium in aerated waters, whereas in anoxic groundwaters, uranium is usually absent, because the respective reduced form (U(IV)) is insoluble in water [7]. The major uranium species in water supplies are the anionic carbonate complexes, i.e., the at pH values lower than 7 and the at pH greater than 8, whereas at pH values between 5 and 6.5 the neutral UO_2CO_3 species may also make up an important part, which depending on pH, could vary between 20%–90% [7].

The creation of the aforementioned complexes with carbonates determines its removal from water, depending on the application of specific treatment processes. For example, conventional methods, such as coagulation or lime softening, can remove uranium from water, but they are very sensitive to pH changes and water composition [1]. On the contrary, ion-exchange [8][9] is the most efficient removal method, because it can remove about 98% of uranium from water, mainly through the removal of anionic uranium carbonate species; the ion exchange resins used in this case are usually selective strong base (anionic) [1]. Membrane treatment methods, such as nano-filtration [10] or reverse osmosis, were also found to be efficient for the removal of uranium, removing the respective carbonate complexes by more than 90%, but their application requires experienced personnel and their use is quite expensive, especially when designed for the treatment of small volumes of contaminated water [1][10][11].

Application of $\text{Fe}_3\text{O}_4@\text{SiO}_2$ Composite Nanoparticles for U(VI) Removal

Das et al. (2010) [12] studied the sorption of U(VI) on magnetite (Fe_3O_4) nanoparticles, but the sorption capacity was relatively small. Silica has been known as one of the most ideal coating layers for the magnetic Fe_3O_4 nanoparticles, due to its reliable chemical stability, biocompatibility, and facile surface modification. Magnetic $\text{Fe}_3\text{O}_4@\text{SiO}_2$ composite particles were successfully applied by Fan et al. (2012) [3] as a novel and effective adsorbent material for the removal of U(VI) from aqueous solutions. The sorption of U(VI) onto magnetic $\text{Fe}_3\text{O}_4@\text{SiO}_2$ composite particles was strongly dependent on pH values. With increasing pH (i.e., from 2 to 6), the adsorption capacity was also increased. When the initial pH value varied from 2 to 4, the sorption capacity of U(VI) increased from 0 to 8.5 mg g^{-1} , while when the initial pH was further increased from 4 to 6, the sorption capacity of uranium increased substantially (i.e., from 8 to 20 mg g^{-1}). However, when increasing the pH value from 6 to 8, the sorption capacity started to decrease. The maximum sorption value for U(VI) onto $\text{Fe}_3\text{O}_4@\text{SiO}_2$ magnetic composites was about 20 mg g^{-1} , when the initial concentration of U was 50 mg L^{-1} . Therefore, the optimum (initial) pH value was 6, when applying this treatment technique, which is in agreement with other relevant studies regarding the sorption of U(VI) onto iron oxides or hydroxides, i.e., they present a maximum sorption capacity in the pH range between 6–7 [13][14][15].

The aforementioned magnetic $\text{Fe}_3\text{O}_4@\text{SiO}_2$ composite particles showed an excellent ability to remove uranium from aqueous solutions and the maximum U(VI) sorption capacity was about 52 mg g^{-1} at 25°C . Hence, this adsorbent material could be a potential candidate to remove the toxic U(VI) forms from aqueous solutions and the results of this study can also provide a technique for the removal or recovery of other heavy/toxic metal ions from aqueous solution [13].

Application of a Novel Graphene Oxide-Activated Carbon Felt Composite for U(VI) Removal

Chen et al. (2013) [4], prepared a composite material, Graphene oxide (GO)-activated carbon felt (ACF)(GO-ACF) and tested for the removal of U(VI) from aqueous solution, comparing the simple ACF and the composite GO-ACF materials. The adsorption of U(VI) on ACF is remarkably improved by the presence of GO, covalently bonding with ACF. The maximum sorption capacity of GO-ACF for U(VI) was evaluated to be 298 mg g^{-1} at pH 5.5, i.e., much higher than that of ACF (173 mg g^{-1}), suggesting that the carboxyl functional groups of GO-ACF can play an important role in the sorption.

Application of a Three-Dimensional Layered Double Hydroxide-Graphene Hybrid Material for U(VI) Removal

Graphene has recently attracted attention for its U(VI) sorption and recent studies have reported a substantially high maximum sorption capacity of 299 mg g^{-1} at the pH value 4 [16], mainly due to the formation of inner-sphere surface complexes of U(VI) on GO. In the past few years, the layered double hydroxides (LDHs) containing transition metals, have also been employed as effective adsorption materials. Their large interlayer space and the high concentration of active sites have allowed the preparation of several multi-functional LDH materials to be used as anion exchangers, adsorbents, or magnetic materials. Tan et al. (2015) [17] synthesized a 3-D hierarchical composite with graphene sandwiched between two layers of NiAl-LDH nanosheets, by using a simple and cost effective in situ growth procedure; during the in situ crystallization process NiAl-LDH nanosheets grew on the surface of GO@AlOOH sheets, obtained by mixing boehmite AlOOH primer sol with GO solution, under specific conditions [17].

When examined as an adsorbent for U(VI) removal, several advantages of this NiAl-LDH composite material make it specifically attractive, because: (1) it is manufactured using an easy, non-toxic synthesis procedure; (2) the larger specific surface area of $257 \text{ m}^2 \text{ g}^{-1}$ can provide more adsorptive sites; (3) it can be used for the effective adsorption/removal of uranium(VI) ions.

The adsorption of U(VI) onto rGO/LDH was carried out by varying the pH values (2–12); it was found that the adsorption capacity was highly dependent on pH value. At pH less than 4, U(VI) is present in solution predominantly in the form of UO_2^{2+} with a lower sorption capacity, mainly due to the competition of H^+ ions for the binding sites of adsorbent. At the pH range 4–8, the hydrolysis of uranyl ions occurs, producing several uranium species that include $\text{UO}_2(\text{OH})^+$, $(\text{UO}_2)(\text{OH})_2$, $(\text{UO}_2)_3(\text{OH})_5^+$ and $(\text{UO}_2)_2(\text{OH})_2^{2+}$, which are available for adsorption onto rGO/LDH, and the sorption reaches a maximum value, especially in the absence of carbonates. In pH values > 8 , the fraction of $(\text{UO}_2)_3(\text{OH})_7^-$ anions (known for their generally low sorption affinity) increases, leading to a decrease of uranium(VI) uptake. As a consequence, the pH value 4 is considered optimum for the adsorption of U(VI) onto rGO/LDH with maximum sorption capacity of the rGO/LDH composite for uranium (VI) 278 mg g^{-1} . Therefore, rGO/LDH is a novel adsorbent, exhibiting a bright future for practical application, regarding the removal of U(VI) from aqueous solutions [17].

Application of Graphene Oxide and Its Amine-Functionalized Composite (GO-NH₂) for U(VI) Removal

A new amine-functionalized graphene oxide (GO-NH₂) nanosheet was prepared via covalently grafting reaction by Liu et al. (2016) [18]. The adsorption capacities of GO and of GO-NH₂ were found to be 97 and 215 mg g^{-1} at 298 K, respectively. These results showed that the adsorption capacity of GO was significantly improved by amine functionalization. The adsorption of UO_2^{2+} ions depends upon the uranium species distribution in solution, which mostly depends on the respective pH value. Uranium can be hydrolyzed into different mononuclear and polynuclear hydrolysis products in the form of $(\text{UO}_2)_m(\text{OH})_n]^{(2m-n)+}$ [19]. As the pH value is lower than 4, the UO_2^{2+} ions exist predominantly in the aqueous solution as the monomeric species. However, with the increase of pH, the hydrolysis of U(VI) ions will occur. Both materials achieved their maximum sorption capacity at the pH value 5.5.

Application of a Novel Graphene Oxide-Immobilized Saccharomyces Cerevisiae Gel Beads for U(VI) Removal

According to a recent study by Chen and Wang (2016) [20], the waste biomass of *Saccharomyces cerevisiae* was immobilized using several agents, including Ca-alginate (Ca-SA), Ca-alginate plus graphene oxide (Ca-SA-GO), or in combination with polyvinyl alcohol (PVA, 5% or 10%, w/v), i.e., PVA-Ca-SA-GO, using a CaCl_2 -boric acid solution, in order to evaluate their ability to adsorb dissolved U(VI). The obtained experimental results showed that graphene oxide at 0.01% (w/v) could enhance the performance of immobilized cells.

The yeast gel beads, prepared with 5% PVA, 1% SA, 2% yeast, 0.01% GO, 2% CaCl_2 and saturated boric acid, generally showed better physical-chemical properties, such as higher tolerance, when unfavorable environmental conditions were applied. Moreover, the gel beads exhibited more stable capacity for U(VI) sorption and desorption at various conditions, such as pH in the range of 3–9. The effects of initial pH at acidic (pH = 3), neutral (pH = 7) and alkaline conditions (pH = 9) on U(VI) adsorption were studied and the results suggested that the optimum pH value for U(VI) adsorption is between 2.6 and 5. The immobilized *Saccharomyces cerevisiae* biomass, using SA, PVA and/or GO substrate materials, showed particular changes in the molecular vibration of functional groups, such as carboxyl, amide and hydroxyl groups, which may be involved in the U(VI) binding, when compared with the raw yeast biomass.

Application of Carboxyl-Functionalized Graphene Oxide (COOH-GO) Material for U(VI) Removal

The effect of increasing the presence of specific chemical functional groups, such as the carboxyl groups, on the selectivity of uranium sorption was investigated by using a carboxyl-functionalized graphene oxide (COOH-GO) modified material, which was studied in comparison with the simple graphene oxide (GO) and with graphite [21]. According to Mohamud et al. (2018), the modified COOH-GO demonstrated superior performance as a sorbent material for the selective removal of uranyl ions from aqueous solution with distribution coefficient value, K , $3.72 \pm 0.19 \times 10^3 \text{ mL g}^{-1}$ in comparison to $3.97 \pm 0.5 \times 10^2$ and $2.68 \pm 0.2 \times 10^2 \text{ mL g}^{-1}$ values for GO and graphite, respectively. Moreover, COOH-GO presents a higher sorption capacity for U ($Q_{\text{max}} = 169 \text{ mg g}^{-1}$) and shows a greater selectivity towards U with $65.9 \pm 2.7\%$ retained in the presence of competing ions in comparison to $38.9 \pm 1.2\%$ value, observed for GO, at the optimum pH 4. These enhanced values are most probable due to the effect of selective surface groups presence, such as the carboxyl.

Application of a Composite Material (GO-DTPAA) for U(VI) Removal

A novel chelator diethyl-enetri-amine-penta-acetic phenyl-enedi-amine (DTPAA) was covalently bonded to a supporting matrix of graphene oxide (GO), and a composite material (GO-DTPAA) was obtained recently by Liu et al. (2018) [22]. The obtained results indicated that GO-DTPAA was a highly efficient absorbent for the removal of U(VI) from aqueous solutions at pH 6.5. The adsorption capacity of GO-DTPAA was as high as 485 mg g^{-1} at 298 K, which was far greater than that of pristine GO (97 mg g^{-1}) at the same temperature. The thermodynamic parameters revealed that the adsorption of uranium ions onto the pristine GO and by the GO-DTPAA composite material are feasible, spontaneous and endothermic.

Application of a Magnetic Reduced-Graphene Oxide/Tea Waste Composite for U(VI) Removal

Recently, Yang et al. (2019) [23] studied the preparation and application of relatively low-cost and highly efficient adsorptive materials for the removal of uranium from nuclear wastes, such as composites of graphene oxide (GO) and tea waste (TW). The composites GO-TW and the magnetic $\text{rGO/Fe}_3\text{O}_4/\text{TW}$ exhibited higher adsorption capacities and faster adsorption kinetics than the simple materials GO and TW ($Q_{\text{max (TW)}} = 92 \text{ mg g}^{-1}$, $Q_{\text{max (GOTW)}} = 112 \text{ mg g}^{-1}$ and $Q_{\text{max (rGO/Fe}_3\text{O}_4/\text{TW)}} = 105 \text{ mg g}^{-1}$), resulting to higher removal rates ($\sim 99\%$) for U(VI). As aforementioned, the solution pH substantially affects the speciation of uranium in the aqueous solutions, and hence, significantly influences the uranium adsorption process. The adsorption of U(VI) on TW, GOTW and $\text{rGO/Fe}_3\text{O}_4/\text{TW}$ significantly increased with increasing the pH value from 2.0 to 5.0. At the pH 5, the removal rate of uranium by TW, GOTW and $\text{rGO/Fe}_3\text{O}_4/\text{TW}$ materials reached the highest values, noting that at pH value < 4.0 , uranium exists mainly in the form of UO_2^{2+} . Furthermore, due to the advantageous magnetic properties, $\text{rGO/Fe}_3\text{O}_4/\text{TW}$ can be easily separated from the treated aqueous solutions, thus enhancing the post-treatment efficiency for further practical applications.

Table 1 shows the reviewed nanomaterials applied for U(VI) removal from waste/water sources, providing major information about their structural characteristics and their applications in water treatment.

Table 1. Novel nanostructured materials applied for the removal of U(VI) from waste/water sources.

Novel Nano-Material	$\text{Fe}_3\text{O}_4/\text{SiO}_2$	GO	GO-ACF	rGO/NiAl-LDH	GO- NH_2	SA-GO	COOH-GO	GO-DTPAA	$\text{rGO/Fe}_3\text{O}_4/\text{TW}$
Dose (g L^{-1})	2.5	0.4	0.2	0.5	0.2	0.1	1.0	0.1	0.5
C initial (mg L^{-1})	50	119	50	130	60	35.6	10	50	10
pH	6.0	4.0	5.5	4.0	5.5	5.0	4.0	6.5	5.0

Adsorption capacity (mg g ⁻¹)	52	299	298	278	215	162	169	485	105
References	Fan et al. (2012) [3]	Li et al. (2012) [16]	Chen et al. (2013) [4]	Tan et al. (2015) [17]	Liu et al. (2016) [18]	Chen and Wang (2016) [20]	Mohamud et al. (2018) [21]	Liu et al. (2018) [22]	Yang et al. (2019) [23]

As revealed from Table 1, the proposed materials seem to work more efficiently for the U(VI) removal at pH values between 4–6. The composite material (GO-DTPAA) exhibits maximum absorption capacity 485 mg g⁻¹. The other proposed materials with graphene, showed an average high adsorption capacity of 200–300 mg g⁻¹.

Table 2 provides the comparison of sorption capacity between different adsorbents, when applied for the removal of uranium. As can be observed by comparing the relevant data of Table 1 and 2, the sorption ability of the previously described nanostructured materials was generally substantially higher.

Table 2. Comparison of sorption capacities between various sorbents towards the removal of U(VI) ions.

Sorbent Material	pH	Sorption Capacity (mg g ⁻¹)		Reference
		Cr (VI)		
Manganese oxide coated zeolite	4.0	15		Han et al. (2007) ^[24]
Natural sepiolite	3.0	35		Donat (2009) ^[25]
Modified clays with titanium oxide	3.5	0.6		Humelnicu et al. (2009) ^[26]
Magnetite nanoparticles	7.0	5		Das et al. (2010) ^[12]
Ethylenediamine-modified chitosan	3.0	83		Wang et al. (2011) ^[27]
Goethite (α-FeOOH)	6.0	34		Yusan et al. (2011) ^[28]
Binary iron–manganese oxy-hydroxides (FMHO)	6.5	133		Dimiropoulos et al. (2015) ^[29]

Conclusions

The pollution of natural waters caused by toxic metals is a global issue, and various treatment technologies have been developed to remove these inorganic pollutants from water sources (mostly from groundwater). The application of most important novel nanostructured materials was summarized, regarding the removal of uranium (U(VI)) from aqueous systems. As it was shown, the use of nanostructured materials, exhibiting generally higher surface areas and average structural sizes below 100 nm, could be very promising in this direction. Uranium could likely be removed from aqueous solutions by the application of magnetic Fe₃O₄@SiO₂ composite particles, as a novel and effective adsorbent material, as well as by the application of several novel and optimum graphene modified composite materials, such as rGO/LDH, GO-ACF, GO-NH₂, COOH-GO, GO-DTPAA, GO-TW, and rGO/Fe₃O₄/TW, showing promising results. However, these materials need to be tested for the treatment of much lower initial concentrations, relevant to drinking water treatment.

Furthermore, these materials need to be applied in real natural ground/waters, containing all the commonly found co-existing anions, such as carbonate, phosphates and sulfates, because most of the presented results were obtained using deionized waters (i.e., performing model experiments).

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