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Removal of uranium from contaminated drinking water: a mini review of available treatment methods

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ABSTRACT

In the present article, the major treatment methods applied for uranium removal from groundwater, with specific applications in drinking water treatment, are reviewed. These include *pump-and-treat* technologies, such as membrane filtration methods, anion exchange, and the use of adsorbents, such as iron oxides, or titanium dioxide, as well as the application of coagulation processes with the addition of Fe/Al salts, or by lime softening. In all cases, uranium removal is mainly dependent on its speciation, which is greatly affected by the (usually coexisting) carbonate ions in the contaminated water. Under circumneutral pH values, uranium forms anionic complexes with carbonate of the type $UO_2(CO_3)_2^{2-}$, or $UO_2(CO_3)_3^{4-}$. *In situ* treatment technologies comprise mainly the use of permeable reactive barriers. These contain reactive materials, such as zero valent iron or hydroxyapatite, and uranium is usually removed by reduction to the respective insoluble products of U(IV); reducing bacteria, when present, can play a supplementary role.

Keywords: Uranium; Drinking water; Speciation; Anion exchange; Reverse osmosis; Adsorption

1. Introduction

Uranium is introduced into groundwater mainly from the processing of uranium ores. Uranium mining, milling, processing, and disposal, all have the potential to contaminate groundwater. In addition, natural uranium mobilization occurs in areas with high natural uranium background concentrations [1].

Uranium is a toxic and radioactive element. The principal health effects from orally ingested natural uranium can be attributed to its high chemical toxicity [2]. Uranyl compounds have a high affinity for phosphate, carboxyl, and hydroxyl groups and readily combine with proteins and nucleotides to form stable complexes [2]. The skeleton and kidney are the primary sites of uranium accumulation; little can be found in the liver. The absorbed uranium rapidly enters the bloodstream and forms a diffusible ionic uranyl hydrogen carbonate complex in equilibrium with a nondiffusible uranyl albumin complex. In the skeleton, the uranyl ion replaces calcium in the

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hydroxyapatite complex of the bone crystal [3]. Once equilibrium is attained in the skeleton, uranium is excreted by urine and feces. Under alkaline conditions, the uranyl hydrogen carbonate complex is stable and is also excreted. However, when the pH value drops, the complex dissociates and binds to the cellular proteins in the tubular wall. The half-life of uranium in the rat kidney was found to be around 15 days, although it is considerably longer (300–5,000 days) in the skeleton [2,3].

The contamination of groundwater with uranium is a matter of concern in several countries around the world, such as in the USA [4], Canada [5], Germany [6], Finland [7], Norway [8], and Greece [9–11]. In the USA, the EPA estimates that more than 500 water systems of (smaller) communities nationwide contain uranium levels in the water greater than 30 μg/L [4]. Uranium concentrations up to 700 μg/L have been found in certain private groundwater supplies in Canada [5]. In Germany, and particularly in Bavaria, from the 1,100 samples of groundwater and drinking water that were tested, 17.1% contained uranium in concentrations 5–20 μg/L, whereas 2.8% contained more than 20 μg/L [6]. In Greece, in several areas, in smaller public water supplies, uranium has been found up to 48 μg/L [10] and in some regions in Finland, the median concentration was found to be 28 μg/L [7].

To date, the European Commission has not established a maximum admissible concentration (MAC) for uranium in drinking water. However, the recommended MAC value from the WHO for uranium is 15 μg/L. The EPA has set a maximum contaminant level (MCL) for uranium of 30 μg/L [2].

2. Uranium speciation

Uranium is principally present in aerated, oxidized groundwaters. In a survey performed in Northern Greece, in reducing and oxidizing groundwaters, uranium was present in considerable concentrations mainly in the oxidizing groundwaters and showed a positive correlation with the redox potential of water [9]. In oxic groundwater, uranium is present in its hexavalent form. The hexavalent uranium is mainly present as the uranyl cation (UO_2^{2+}), or with its hydroxy complexes, but in the presence of other anions—such as carbonate, phosphate, or sulfate—uranium forms complexes and, therefore, its speciation is greatly affected by the respective ionic water composition [12,13]. Fig. 1 shows the speciation diagrams of uranium in water in the absence and presence of typical carbonate concentrations to depict the representative uranium speciation in groundwater.

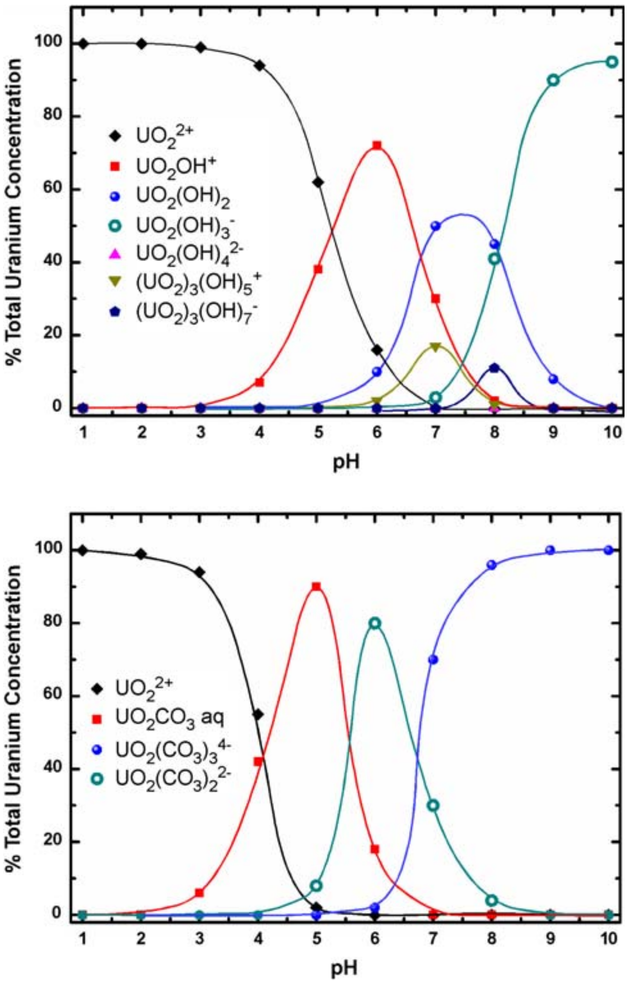

Fig. 1. U(VI) speciation in the system UO_2^{2+}–H_2O; (a) [U(VI)]= 4.10^{-7} M and (b) U(VI) speciation in water containing carbonate (2 mM) (generated with MineqL + software program).

Table 1
Predominant species of uranium in carbonate-containing groundwaters as a function of respective pH values [12]

pH range	Predominant species
<5	UO_2^{2+}
5–6.5	UO_2CO_3
6.5–7.6	$UO_2(CO_3)_2^{2-}$
7.6–10.5	$UO_2(CO_3)_3^{4-}$
>10.5	$(UO_2)_3(OH)_5^+$

The speciation of uranium in the presence of carbonates is the most common case, because almost all waters contain significant concentrations of alkalinity, and drives in most cases the mechanisms of removal by the applied treatment methods. Table 1 shows a summary of the predominant uranium species, as affected by pH values in carbonate containing (ground) water.

3. Options for decreasing uranium concentrations in water supplies

Community water supplies have mainly five options for the removal or reduction of uranium concentrations from drinking water sources: (i) locating and developing a new source of drinking water, (ii) purchasing drinking water from another system, (iii) blending water from a contaminated source with water from an uncontaminated source, (iv) building and operating an appropriate drinking water treatment plant to remove uranium (*Point-of-Entry* [POE]), and (v) installing and maintaining *Point-of-Use* (POU) treatment devices at each drinking water tap [14]. For the latter cases, the major treatment options for uranium removal include: anion exchange [15], reverse osmosis (RO) [16], coagulation [15], lime softening [15], and the use of specific adsorbent media [17].

In private water supplies, the most practical treatment alternative to remove uranium is the POU system [14]. Such a system is usually placed under or near the faucet and treats only the water coming out from this tap for drinking or cooking purposes. RO, special adsorbent media (such as titanium dioxide), and anion exchange can remove uranium and also a variety of other contaminants [14].

Another important pathway for the removal of uranium is the reductive precipitation, in which hexavalent uranium is reduced to tetravalent, which is insoluble and thus can be removed from water by precipitation. Aqueous uranium can be reduced in anaerobic environments, either by reaction in subsurface environments with zero valent iron [18] and other reactive materials or by a variety of micro-organisms, including iron- and sulfate-reducing bacteria and, in some cases, even by denitrifying bacteria [19]. The product of uranium reduction is uraninite, UO_2, which is a highly insoluble mineral.

In the present article, the most important and widely applied treatment methods used for the removal of uranium from contaminated groundwaters are reviewed, setting specific criteria for their applicability. This article focuses on the methods which are of practical importance for drinking water treatment and have been proven to meet the MCL of uranium for drinking water (i.e. 15 μg/L). The methods or materials which are still in the lab-scale investigation phase are only briefly mentioned, to depict the research trends towards uranium removal.

4. Treatment methods for uranium removal

4.1. *Removal of uranium by ion exchange*

Removal of uranium by anion exchange is probably the most widely applied technique for POE systems. Its principle is based on uranium speciation, as affected by the groundwater composition and the pH value of water to be treated. At pH values above 6, uranium exists in aqueous solutions primarily as the anionic uranyl carbonate complex [12]. This form of uranium presents high affinity for strong base anion resins [15,20]. The relative order of affinity of strong base anion resins for some common ions found in drinking water shows that uranium is at the top of the respective list [20,21]: Uranium/Perchlorate >> Sulphate/Chromium > Selenium/Arsenate > Nitrate > Chloride > Bicarbonate > Fluoride.

Therefore, the theoretical removal capacity is generally very high, although the actual operating capacity will depend on the concentration of competing ions, such as sulfate and chloride, which typically occur in (ground) water at much higher concentrations.

Cationic resins in the hydrogen form have been observed to remove uranium, probably by converting the uranium complex to the uranyl cation [20]. Removal rates are in the 90–95% range, but the pH of the effluent has to be low (about 2.5–3.5), and the resin used in this method is not selective, removing all the other cations as well. Obviously, cationic resins are not a viable ion exchange removal method, because of the pH requirements and the low efficiency compared to the strong base anion resins [15,20].

Anionic resins in the chloride form can easily reduce uranium levels by over 90% [2,18]. A mixed bed of cationic and anionic resins can be also considered for certain applications that are used to remove both radium and uranium. In this case, the percentage of cationic resins is usually limited to 10% and the rest is completed with the anionic ones [21].

Strong base anionic resins have been used in South Africa at Vaal River Operation for the last two decades. It is also used in Key Lake in Canada, in Olympic Dam in Australia, in the USA, and in Germany for the treatment of contaminated groundwaters near Berlin and also in Bayern [22]. For example, the results of a campaign in South Germany were as follows: for the treatment of groundwater, the anionic resin

LEWATT S 6838 of the sulfate form was used [22]. The inlet water contained uranium 12 μg/L, the pH was 6.5, sulfate concentration was 65 mg/L and nitrate 16 mg/L; after the treatment of 40,000 bed volumes (BV), the outlet uranium concentration was below 0.07 μg/L (i.e. 99% removal was achieved) and the breakthrough was not still observed [22].

4.1.1. Factors affecting the resin capacity

The uranyl carbonate complex presents high affinity for strongly basic anion exchange resins that is over 100 times greater than any other coexisting common ions, including the divalent ones such as carbonates and sulfates [21], at usual pH values in natural waters (i.e. between 6 and 8). Higher pH values could result in uranium precipitation, thus reducing the efficiency of the resin, but increasing the overall removal of uranium. On the other hand, at lower pH values, the speciation of uranium changes to nonionic and/or cationic forms, which would prevent the exchange reactions from operating effectively. Therefore, it is essential to control the inlet water pH above 6 at all times [21]. It has also been shown that sudden changes in pH of the influent water to values below 5.6 can result in dumping of previously removed uranium. In cases where the pH value cannot be maintained above 5.6, other treatment methods should be considered [21].

4.1.2. Regeneration

Sodium chloride is the most common regenerant. The chloride ion is the most effective ion for the regeneration of resins commonly applied for potable water treatment. Neutral salts (sodium chloride is by far the most common) are usually preferred because of environmental considerations [21].

Resin loading is a matter of concern in the treatment of radionuclides. In practice, consideration must also be given to the radioactive nature of uranium and the extent to which it can be loaded onto the resin for safe handling, transportation, and disposal. Practitioners resort to limiting the loading to no more than 2,000 picoCuries/g. Converting uranium concentration expressed as picoCuries/L to μg/L gives: picoCuries/L divided by 0.69 = μg of uranium/L. Thus, for typical anion resins, a loading of 3 mg/g is acceptable [21].

4.2. Membrane methods

The main membrane methods capable of removing uranium from water are RO and nanofiltration (NF) [16,23] mainly because of the relatively larger size of uranium compounds than the pores of the membranes.

Huxstep and Sorg [16] have used RO for treating groundwater sources with uranium concentration of more than 300 μg/L and they reported removal efficiencies up to 99%. The application of NF has also shown promising results. Five NF membranes were tested for the removal of uranium from initial concentrations of 1 mg/L from synthetic solutions. Removal rates were in all cases higher than 90%. Especially, the divalent anion complex $UO_2(CO_3)_2^{2-}$ and the tetravalent anion complex $UO_2(CO_3)_3^{4-}$, which mainly occur in natural waters, were rejected at rates between 95 and 98% [23].

As already mentioned, removal of uranium by RO can be applied at a POU system as well. Fox and Sorg (1987) [24] tested RO on a domestic scale and reported removal efficiencies of more than 99% from initial concentrations of 69 and 183 μg/L. More recently, several systems have been developed for removing uranium from drinking water by RO at point-of-use level. The systems are usually placed under the kitchen sink for treating directly the water that is used for drinking, cooking, and making ice purposes [25]. Such systems are usually composed of a sediment filter, followed by a carbon filter, the RO system, and then a final carbon filter. A photograph of the installed system is shown in Fig. 2 [25]. This RO system was proven to be very effective in removing uranium. The average uranium concentration in the water of this application before the treatment was 3,240 μg/L. After the treatment, the average concentration is 1.95 μg/L. This is an overall removal of 99.9% of the uranium from the drinking water [25].

4.3. Removal of uranium by iron or alum coagulation and lime softening

Uranium removal can be accomplished by the application of coagulation using ferric sulfate, ferrous sulfate, or alum. Lee and Bondietti (1983) [15] investigated the treatment of uranium-containing waters by coagulant agents from 20 water treatment plants in the USA. Only in two of them, the removal of uranium was significant. They used aluminum and iron salts, and the flocks were removed by sedimentation and filtration. The efficiency of uranium removal was related to the pH value of water to be treated and the coagulant dose. They also investigated the pH effect on uranium removal. The results showed that the highest uranium removal took place at pH values 6 and 10, for an initial uranium concentration of 83 μg/L.

Fig. 2. Photograph showing a RO-based POU water treatment system for the removal of uranium from drinking water (taken with permission from http://www.scdhec.gov/environment/water/urro.htm, South California, Department of health and environmental control) [25].

While at pH 4, removal of uranium was negligible, accounting for approximately 20%; raising the pH value to 6 produced very good results and the removal of uranium was close to 90%. Further increase of pH value to 8 caused again a reduction in the efficiency to about 50% and only when the pH value was increased to around 10, the removal or uranium was again as high as required, to achieve the low concentrations required for meeting the stringent standards of drinking water [15]. The efficient uranium removal at pH 6 is attributable to the formation of the neutral UO_2CO_3 complex, which is readily able to precipitate. At pH 8, the coagulation media form negatively charged hydroxyl complexes, which adversely affect uranium removal efficiency. At pH 10 and higher, $CaCO_3$ is precipitating out of the solution, thus the concentration of carbonate decreases and, therefore, uranium speciation is dominated by the formation of $(UO_2)_3(OH)_5^+$ species, which react with metal hydroxides and precipitate very efficiently.

Very probably, a similar mechanism is responsible for the removal of uranium during lime softening in water treatment plants. Lime softening is a process which takes place at pH 10.5–11.5 and uranium removal can be accomplished by up to 95% [15]. The results of Lee and Bondietti [15] depicted that the application of lime softening at pH values below 9 showed very little efficiency for uranium removal. However, at pH values 9.7 and higher, uranium removal of more than 95% can be accomplished. These high pH values were achieved by the addition of lime in the coagulation basin.

Coagulation has been investigated in several case studies and has given similar results with those reported from Lee and Bondietti [15]. The study of Gäfvert et al. [26] examined uranium removal at a waterworks with an average production rate of $1.3\,m^3\,s^{-1}$, providing with drinking water several large cities in the province of Scania. The raw water was surface water from a local lake Bolmen and the method used for purification was a combination of coagulation–flocculation and filtration in sand filters. Two different purification lines were in use, one using $Al_2(SO_4)_3$ as coagulant agent and the other using $FeCl_3$. The pH was adjusted at this stage to 6 and 5.2, respectively. After coagulation and flocculation, the precipitate was removed and the water was passed through two different sand filters (rapid and slow filtration, respectively). The results showed a high removal capacity for uranium (about 85%) and other radionuclides, in accordance with the aforementioned study of Lee and Bondietti [15].

Baeza et al. [27] compared the efficiency of two drinking water treatment processes in eliminating the uranium content from the source water. The examined procedure consisted of coagulation, flocculation, settling, filtration, and chlorination treatment stages, specifically designed to maximize the elimination of their natural radioactive content. The results showed that the uranium elimination efficiencies strongly depend on the water's carbonate, calcium, and magnesium ion concentrations. In particular, in the waters with increased concentrations of any of these ions, uranium elimination efficiency decreased from 90 to 60% at the optimal working pH value of 6.

4.4. Uranium removal by adsorption media

Sorption of uranium onto several mineral surfaces has been extensively studied over the last few decades. Sorption experiments have often involved solid materials typical of the field site under study as well as model minerals having well-defined surface properties. The most important and widely tested are iron oxides and titanium dioxide, for which we will analyze their applicability in more detail. Other materials have been tested up to laboratory scale with promising results. Some of them are granular activated carbon [28], bone charcoal and apatite [29], chitosan [30], zeolites [31], and some biosorbents such as *Saccharomyces cerevisiae* [32], *Sargassum* biomass [33], and calium alginate [34].

Iron oxides tested for uranium sorption and removal from water sources are hematite [35], goethite [36], ferrihydrite [37], and amorphous iron oxides [12,38]. In general, iron oxides adsorb uranium efficiently at pH values relevant to drinking water treatment, i.e. in the pH range between 5 and 9 [39]. Fig. 3 shows schematically the removal of uranium by iron oxides in the presence of carbonates, as predicted by surface complexation models.

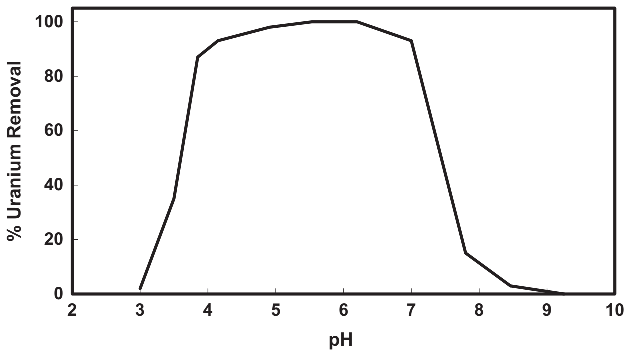

Fig. 3. Surface Complexation model fit of U(VI) adsorption onto 1 g/L hydrous ferric oxides (HFO) from groundwater. Aqueous complexation reactions and kinetic constants were taken from Barnett et al. [39]. Parameters used in the model: surface site density 0.875 mol of sites/ mol of Fe and specific surface area 600 m^2/g (Figure was generated using the MineqL + software program).

However, the increased concentration of carbonate was found to decrease the efficiency of uranium adsorption, because of the creation of uranyl carbonate complexes, which do not adsorb so strongly on iron oxides, due to the reduced affinity of carbonates for iron oxides. Wazne et al. [40] performed sorption isotherms for uranium removal onto iron oxy-hydroxides and found that approximately 0.125 mol of U(VI) was removed per mole of Fe(III) at equilibrium U(VI) concentration of 0.5 mg/L in the absence of carbonate. The amount of adsorbed U(VI) decreased substantially with increasing carbonate concentrations. For example, when total carbonate concentration was 1.68 mM, U(VI) removal was only 0.034 mol/mol of Fe(III) at the same as previously U(VI) equilibrium concentration of 0.5 mg/L.

Titanium dioxide is a very efficient adsorbent for uranium removal from water [41], since it presents strong affinity for this pollutant and because of its chemical stability, negligible solubility over a wide pH range, and near-neutral point-of-zero charge. There are already commercial products in the market based on titanium dioxide, with the most common being Degussa P-25 [41] and metSorb [42].

In a recent relevant study, nanocrystalline titanium dioxide was used in batch tests to remove U(VI) from synthetic solutions as well as from contaminated water. The capacity of nanocrystalline titanium dioxide to remove U(VI) from water decreased in the presence of inorganic carbonate at $pH > 6.0$, i.e. neutral to alkaline pH values. Approximately, 0.02 mol of U(VI) was removed per mole of TiO_2 at an equilibrium U (VI) concentration of 0.45 mg/L and with no carbonate in the suspension ($CT = 0$ M). The amount of adsorbed U(VI) decreased substantially with increasing carbonate concentrations. When total carbonate concentration was 10.0 mM, U(VI) removal was only 0.01 mol per mole of TiO_2 at U(VI) equilibrium concentration of 0.45 mg/L. This substantial decrease was attributed to the aqueous complexation of U(VI) by the presence of carbonate [43].

4.5. Removal of uranium by permeable reactive barriers

Permeable reactive barriers (PRBs) are permeable walls that are installed across the flow path of a contaminant plume. This wall is designed to be at least as permeable as the surrounding aquifer material. The PRBs contain a zone of reactive material that is designed to act as a passive *in situ* treatment zone for a specific contaminant, as groundwater flows through it.

Several studies have been undertaken to investigate the removal of uranium by PRBs [18,44–46]. In one of these studies [18], a series of laboratory experiments were conducted on three classes of potential PRB materials, namely apatite, zero valent iron, and ferric iron-based materials [18]. In this study, the Fry Canyon site in southeastern Utah in 1996 was chosen to perform long-term investigations to assess the efficiency of the aforementioned materials. Zero valent iron (ZVI) PRB consistently lowered the input U concentration by more than 99.9% after the contaminated groundwater had travelled 0.5 m into the PRB. The percentage of U removed by the bone-char phosphate (PO_4) and amorphous ferric oxyhydroxide (AFO) PRBs exceeded 70% for most measurements made during the first year of operation.

Similar to this project, the long-term performance of PRBs has been examined by Simon et al. [44,46]. The laboratory and field studies showed that the most efficient materials for the removal of uranium from contaminated groundwater were elemental iron and hydroxyapatite. Initial experiments in columns showed that both elemental iron and hydroxyapatite were able to retain uranium from groundwater. In the case of elemental iron, the reductive precipitation of uranium was accompanied with an increase in pH values. In long-term column experiments, the pH was increased in the first 2–3 months by 2 pH units but after 7–8 months of operation, the effluent pH decreased and stabilized to around 8. According to the authors, this pH change could indicate a change in the uranium retention mechanism from reductive precipitation in the beginning to a sorptive removal on iron oxides later. In contrast to this, the pH of the effluent from hydroxyapatite columns remained nearly unchanged. The mechanism of uranium removal by hydroxyapatite is either sorption of uranium on apatite surface or precipitation as uranyl phosphates [46]. Field investigations were performed in Hungary in a site near the city of Pecs, by building a pilot-scale permeable reactive barrier. The reactive material consisted of elemental iron and good removal results were achieved for a period of more than 3 years [46].

4.6. Comparison of the major technologies for uranium removal from groundwater

In the previous sections, the major treatment technologies for uranium removal from contaminated (ground) water sources have been described and analyzed. In Table 2, a comparison of the major technologies, with regard to their efficiency, is performed.

The most efficient and widely used technologies applied specifically for uranium removal are anion exchange and RO. This is mainly attributed to the fact that the efficiency of these methods is not affected significantly by the presence of carbonate concentrations in the water to be treated and they are much easier to apply, compared for example to PRBs. Adsorption methods are intended for use mainly at the household level, however RO and anion exchange units have also been recently developed for application at POU systems.

4.7. Case studies—simultaneous removal of uranium and arsenic from groundwaters

Uranium is frequently present in groundwater simultaneously with arsenic, especially in oxidizing groundwaters, where arsenic is mainly in the pentava-

Table 2
Comparison of treatment technologies applied for the removal of uranium from contaminated (ground) waters

Treatment method	Removal efficiency %	Comment
Coagulation with Fe/Al	50–90	Effective at pH values around 6 and 10
Lime softening	80–99	Effective at pH > 9.5
Anion exchange	>95	Regeneration with 2–4 M NaCl after 10,000–50,000 bed volumes. Recommended pH value > 6. Mainly for use in POE systems
Reverse osmosis	>99	Very effective, however rather expensive. Causes big changes in major ionic water composition. Applicable for POE and POU systems
Permeable reactive barriers using zero valent iron	>90	Difficulties in handling the materials after exhaustion. In situ treatment, directly in the groundwater aquifer
Adsorption media (iron oxides or titanium dioxide)	>90	Reduced efficiency in the presence of high carbonate concentration. Mainly application at POU systems

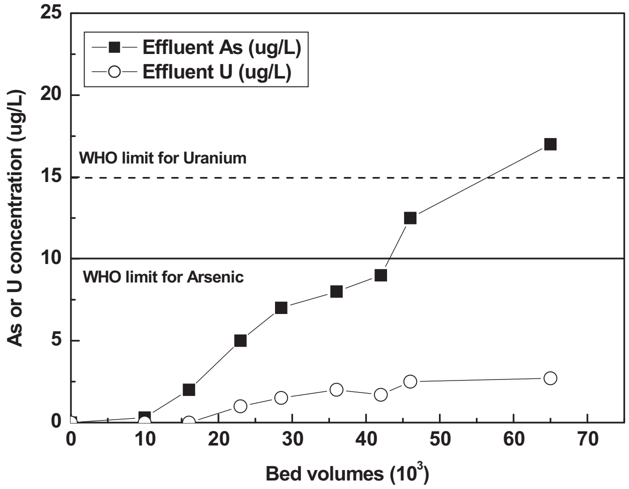

Fig. 4. Total arsenic and uranium breakthrough curves at a pilot-scale treatment system containing the MetSorb adsorbing material. Median inlet arsenic and uranium concentrations were 21 µg/L and uranium 38.8 µg/L, respectively (drawn with permission from data taken from [42]).

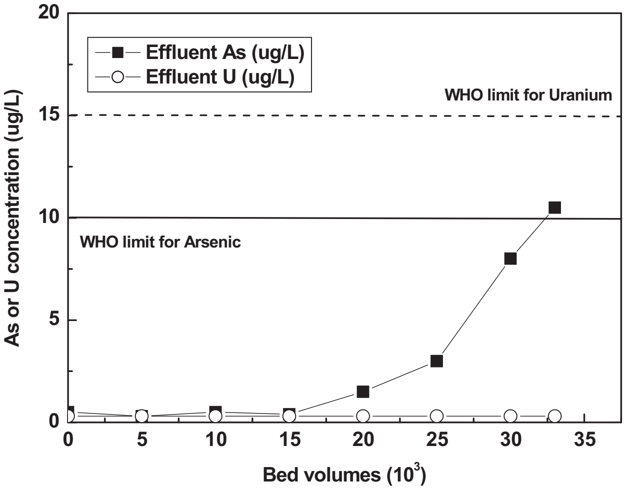

Fig. 5. Total arsenic and uranium breakthrough curves at a full-scale drinking water treatment system containing the HIX hybrid material. Average inlet arsenic and uranium concentrations were 40 µg/L and 33 µg/L, respectively (drawn with permission from data taken from [17]).

lent oxidized form and uranium is present as U(VI) [9]. For such cases, adsorptive or hybrid materials have been applied for the simultaneous removal of arsenic and uranium [17,42].

To define the treatment effectiveness of an adsorbent technology in source water, which simultaneously contains arsenic and uranium, a small water treatment system in Northern Arizona was selected to conduct on-site small-scale testing [42]. The water contained arsenic (21 µg/L) and uranium (38.8 µg/L) at pH 8.9. The total hardness was 28 mg/L and silica 42 mg/L. In this case, nanocrystalline titanium dioxide was applied with appropriate pretreatment for pH adjustment to circumneutral values. Following pH adjustment, the treatment unit was fed at a controlled flow rate of 10 L/min providing a total empty bed contact time (EBCT) of 3.0 min and allowed to operate 24 h per day, 7 days per week. The data concluded that the nano titanium oxide adsorbent media (MetSorb) effectively removed both arsenic and uranium to below the safe drinking water MCL for each contaminant. Breakthrough for arsenic (i.e. effluent concentration higher than 10 µg/L) was observed after the treatment of almost 40,000 BV of water. For uranium, effluent concentrations of over 30 µg/L were not reached after treatment of approximately 50,000 BV. The results are shown in Fig. 4, which shows the breakthrough curves for arsenic and uranium.

A second case study was performed for arsenic and uranium co-removal technology, which was demonstrated at Upper Bodfish in Lake Isabella, CA [17]. The main objective of the project was to evaluate the effectiveness of a hybrid ion exchange (HIX) technology in removing arsenic and uranium to meet the respective MCLs of 10 and 30 µg/L, respectively.

During the study period (from 13 October 2005 to 3 August 2006), the HIX treatment system operated for a total of 4,631 h, treating approximately 25,000 m^3 of water from the Upper Bodfish Well CH2-A. The average daily run time was 15.4 h/day and the average daily production was 83 m^3/day, corresponding to an hourly flow rate of around 5.5 m^3/h.

Source water from this well had near-neutral pH values (6.8–7.2), 88–145 mg/L of alkalinity (as CaCO3), 36–41 mg/L of sulfate, and 40–48 mg/L of silica. In addition, the well water contained 36.5–47.3 µg/L of total arsenic, with As(V) being the predominating species at an average concentration of 40.9 µg/L. The source water also contained 26.6–38.9 µg/L of total uranium, with concentrations exceeding the 30 µg/L MCL of EPA most of the time. The results of the tests are shown in Fig. 5.

During the first 10 months of system operation, the total arsenic concentrations in the treated water were reduced to <0.1 µg/L and gradually increased to 10.5 µg/L after 33,100 BV of throughput. This run length was 65% higher than the vendor-provided estimate of 20,000 BV. Meanwhile, uranium was completely removed to below the detection limit of

0.1 μg/L throughout the 10-month study period. The HIX system did not require backwashing because the head loss buildup across the adsorption vessel was insignificant and it did not influence any other water quality parameters in the distribution system.

5. Waste disposal issues

Water treatment systems that use source water containing uranium or other radionuclides produce wastes which must comply with specific rules in every country, for example in the USA with the Radionuclides Rule [47]. Because every technology produces different types of waste with varying contaminant concentrations and restrictions on disposal options, it is important that systems carefully investigate treatment and waste disposal options before installing new or upgrading existing treatment plants.

Anion exchange waste includes backwash water, regenerant brine, rinse water, and spent media. In coagulation/filtration, wastes include generated iron and alum sludge from the contact and settling basins, the supernatant from the sludge, filter backwash, and spent filter media. Additional liquid waste may be generated when the sludge is dewatered prior to landfill disposal. Lime softening wastes include a high volume of lime sludge, which precipitates with high uranium and radium concentrations; supernatant from sludge and filter backwash. In membrane technologies, waste comprises the concentrated waste stream and spent membranes, which may contain elevated levels of radionuclides [47].

The disposal options for the generated waste depend on the type of waste (liquid or solid), the concentrations of uranium or other radionuclides, the co-occurring contaminants, and the state or local regulations. In general, solid wastes must not contain free liquids and have to be dewatered prior to disposal. Depending on the characteristics of the solid waste, it can be disposed either in municipal and industrial solid waste landfill or in hazardous waste landfill and if it contains high concentrations of radionuclides, it might have to be disposed to a low-level radioactive waste landfill. In the USA, liquid wastes containing uranium at levels 60 pCi/L are considered as radioactive [48]. Therefore, they cannot be discharged to waters or injected to groundwater aquifers. Disposal options for such waste include the sanitary sewer system or solids drying beds [48].

6. Conclusions

The major treatment methods applied for the removal of uranium are the anion exchange, RO, NF, coagulation, lime softening, use of PRBs with appropriate reactive material often used in parallel with reducing bacteria, and adsorption on iron oxides or titanium dioxide. In the following, the major findings of this review article are summarized.

- Anion exchange is among the most efficient and widely applied methods, providing uranium removal of more than 95%. The removal is mainly based on uranium speciation, which is affected by pH and water composition. At circumneutral pH values in natural waters, containing carbonates, uranium forms complexes with carbonates and is present in the form of anionic complexes. Therefore, anion exchange removes uranium very efficiently at pH values above 6. This treatment method is mostly applied in POE systems.
- The removal of uranium by RO and NF is very efficient, with removal efficiencies reaching 99%. The removal is mainly based on the size of uranium complexes, which are larger than the membrane pores, hence are not allowed to pass through. It has the additional advantage of removing an array of other contaminants; however, it changes also the concentrations of major ionic components by removing together with the contaminants the essential ions from water.
- Coagulation with ferric or aluminum salts is an efficient method but the efficiency greatly depends on the pH value of water. Removal efficiencies of more than 80% can be achieved only at pH values near 6 and 10. Lime softening can remove uranium at rates of more than 90%, though only at pH values above 9.5. This means that at pH values typical of natural waters coagulation and lime softening is not effective and thus not recommended.
- The removal of uranium by adsorption media such as iron oxides and titanium oxide has also been investigated. Removal efficiencies are reported to be more than 90%; however, its efficiency is limited by the concentration of carbonates in water.
- The use of PRBs, with appropriate reactive materials such as zero valent iron or hydroxyapatite, has been widely investigated. The PRBs are placed into the groundwater aquifer and remove uranium mainly through reduction of the hexavalent to the tetravalent form of uranium, which easily forms insoluble products.

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