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ORIGINAL PAPER

Removal of arsenic from water streams: an overview of available techniques

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Abstract Arsenic poisoning has become one of the major environmental worries worldwide, as millions of people, which have been exposed to high arsenic concentrations (through contaminated drinking water), developed severe health problems. The high toxicity of this element made necessary the enforcement of stringent maximum allowable limits in drinking water. So, the development of novel techniques for its removal from aqueous streams is a very important issue. This paper offers an overview of geochemistry, distribution, sources, toxicity, regulations and applications of selected techniques for arsenic removal. The contribution briefly summarizes adsorption processes and mechanism of arsenic species removal from water streams by means of iron oxide/oxyhydroxide based materials. Sorption capacities of various sorbents (e.g. akaganeite, goethite, hydrous ferric oxide, iron oxide coated sand, Fe(III) loaded resin, granular ferric hydroxide, Ce(IV) doped iron oxide, natural iron ores, iron oxide coated cement, magnetically modified zeolite, Fe-hydroxide coated alumina) have been compared.

Keywords Arsenic · Characterization and distribution · Toxicity · Iron oxides · Iron oxyhydroxide · Akaganeite · Nanoparticles · Adsorption · Wastewater treatment

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Introduction

Water is a very limited natural resource and in many cases there is not enough water supply of appropriate quality for industrial and domestic use. Many pollutants in water streams have been identified as toxic and harmful to environment and human health. Among them arsenic is considered as a high priority one. It occurs naturally in rocks and soils, water, air, plants, and animals. Volcanic activity, the erosion of rocks and minerals, and forest fires are also sources that can release arsenic into the environment. Anthropogenic activities are also responsible for arsenic release into the environment. Wood preservatives, paints, drugs, dyes, metals, and semi-conductors contain arsenic. Agricultural applications (pesticides, fertilizers), fossil fuel combustion, mining, smelting, landfilling and other industrial activities contribute to arsenic releases as well. These natural and anthropogenic sources introduce a certain amount of arsenic to the environment and increase its concentration and distribution. Figure 1 shows the occurrence and flow paths of arsenic in the environment (Wang and Mulligan 2005). Arsenic has been found at higher levels in underground drinking water sources than in surface waters. In some cases, arsenic poisoning is so severe that made worldwide headlines when reported (e.g. Bangladesh, West Bengal, India, etc.). Water containing 300–4,000 μg L^{-1} of arsenic is not so rare (Berg et al. 2001; Hossain 2006). Several studies have linked long-term exposure even to small concentrations of arsenic with cancer and cardiovascular, pulmonary, immunological, neurological and endocrine effects. As a consequence, arsenic is considered highly toxic and there is a tremendous demand for developing efficient methods for arsenic removal from drinking waters (Shih 2005).

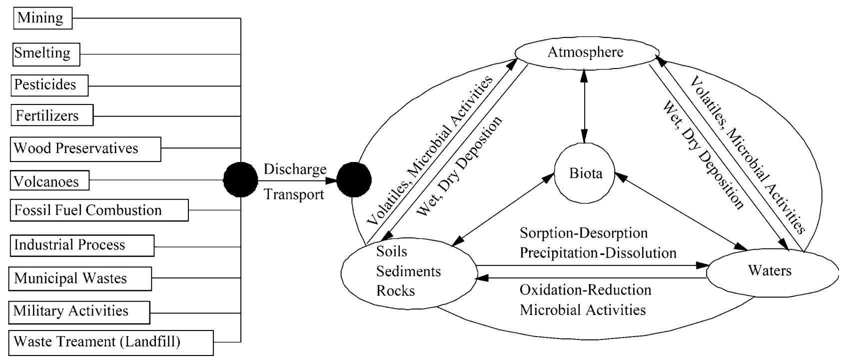
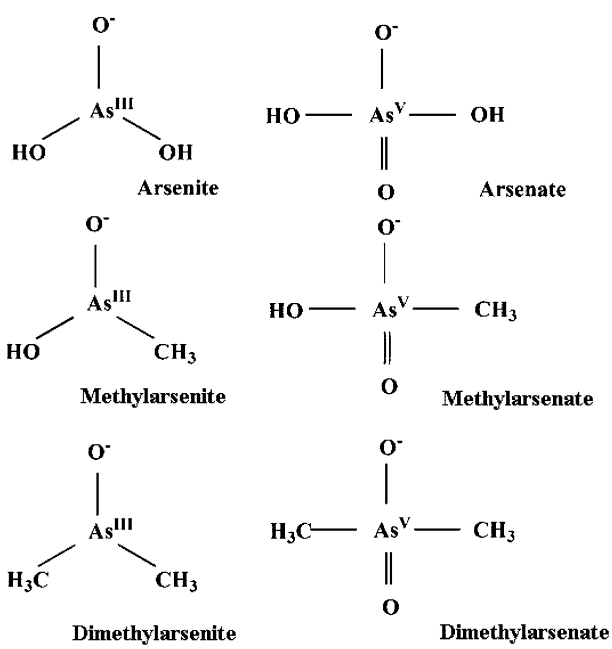


Fig. 1 Arsenic cycle in the environment (Wang and Mulligan 2005)

Chemistry and toxicity of arsenic

Arsenic is the twentieth most abundant element in the earth's crust, fourteenth in the seawater and the twelveth most abundant element in human body (Mandal and Suziki 2002). Arsenic is a metalloid or inorganic semiconductor. It occurs with valence states of –3, 0, + 3 (arsenite, As[III]), and +5 (arsenate, As[V]). Because the valence states –3 and 0 occur rarely, this discussion of arsenic chemistry focuses on As(III) and As(V). Arsenic forms inorganic and organic compounds. Inorganic compounds of arsenic include hydrides (e.g., arsine), halides, oxides, acids, and sulphides (Kirk-Othmer 1992; Smedley and Kinniburgh 2002). The dominant organic forms found in water are methyl and dimethylarsenic compounds (Scheme 1, Hung et al. 2004). Methylated arsenic species, such as monomethylarsonous acid (MMA(III)), monomethylarsonic acid (MMA(V)), dimethylarsonous acid (DMA(III)), dimethylarsonic acid (DMA(V)) can be formed through biomethylation by microorganisms under favourable conditions (Cullen and Reimer 1989; Hasegava et al. 2001). Generally, As(V) is more prevalent in surface water while As(III) is more likely to occur in anaerobic ground waters. However, redox potential (Eh) and pH are the most important factors controlling As speciation. Under oxidising conditions, $H_2AsO_4^-$ is dominant at low pH (less than about pH 6.9), whilst at higher pH, $HAsO_4^{2-}$ becomes dominant ($H_3AsO_4^0$ and AsO_4^{2-} may be present in extremely acidic and alkaline conditions, respectively). Under reducing conditions at pH less than about 9.2, the uncharged arsenite species $H_3AsO_3^0$ will predominate (Fig. 2; Smedley and Kinniburgh 2002; Yong and Mulligan 2004; Kundu and Gupta 2005; Wang and Mulligan 2005). The toxicity and mobility of arsenic are determined by its oxidation state, thus the behaviour of arsenic will change depending on the biotic and abiotic conditions of water (Meng at al. 2003; Thomas et al 2001).



Scheme 1 Arsenic species found in water (Hung et al. 2004)

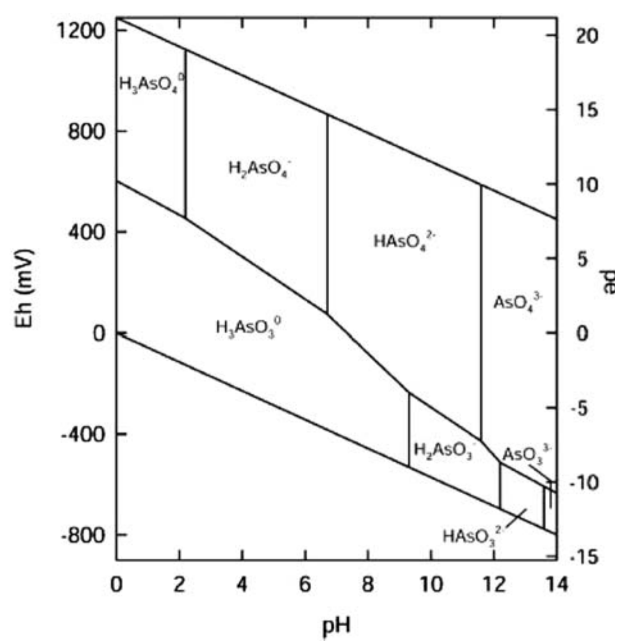


Fig. 2 Eh–pH diagram for aqueous As species in the system As–O_2–H_2O at 25°C and 1 bar total pressure (Smedley and Kinniburgh 2002)

Generally, inorganic forms are more toxic and mobile than organoarsenic species, while arsenite is considered to be more toxic than arsenate. It has been reported that As(III) is 4 to 10 times more soluble in water than As(V) (Squibb and Fowler 1983; Xu et al. 1988; Lambe and Hill 1996; US EPA-542–S-02-002, 2002a). Moreover, it has been found that As(III) is 10 times more toxic than As(V) and 70 times more toxic then MMA(V) and DMA(V). However, the trivalent methylated arsenic species, i.e., MMA(III) and DMA(III) have been found to be more toxic than inorganic arsenic because they are more efficient at causing DNA breakdown (Styblo et al. 2000; Dopp et al. 2004). The toxicity of different arsenic species varies in the order: arsenite > arsenate > mono-methylarsonate (MMA) > dimethylarsinate (DMA) (Jain and Ali 2000).

Methods of arsenic removal

As the presence of arsenic species in drinking water, even in low concentrations, is a threat to human health, the World Health Organization (WHO) recommended the new maximum contaminant level (MCL) of arsenic in drinking water to 10 μg L^{-1} from an earlier value 50 μg L^{-1} (WHO 2004). This greatly reduced limit was recently accepted by authorities of European Union (EU) as well as by United States Environmental Protection Agency (US-EPA) (Directive 98/83/EC 1998; US EPA 2002b). With the new standard of 10 μg L^{-1} from the 74,000 total community water systems in USA around 5,000 will contain arsenic above the limit and should take corrective actions. As 94% of these water systems serve fewer than 10,000 people each cost effective procedures should be applied.

Various treatment methods have been developed for the removal of arsenic from water streams, such as sorption and ion-exchange (Jekel 1994; Rau et al. 2003; Shevade and Ford 2004; Singh and Pant 2004), precipitation (Hering et al. 1997; Wickramasinghe et al. 2004; Kartinen and Martin 1995), coagulation and floculation (Hering et al. 1997; Han et al. 2002; Kumar et al. 2004; Song et al. 2006) reverse osmosis (Ning 2002; Kang et al. 2000), membrane technologies (Han et al. 2002; Sato et al. 2002; Shih 2005), electrodialysis (Kundu and Gupta 2005), biological processes (Katsoyiannis and Zouboulis 2004; Pokhrel and Viraraghavan 2006) as well as lime softening (Dutta and Chaudhuri 1991), etc. Among them, coagulation-precipitation followed by filtration has been recognized as a popular one. Reverse osmosis and electrodialysis have also been found effective, but they were costly and water recovery was not optimised (US-EPA 2003).

Co-precipitation/adsorption processes are commonly applied today to meet the current drinking water standards and show a good efficiency to cost ratio for higher arsenic concentrations. However, they fail to remove arsenic to concentrations below the new decreased limits (10 μg L^{-1}) (Deschamps et al. 2005). Arsenic is more difficult to be removed when it occurs in As(III) form. Sorption methods are based on the development of attractive forces between the arsenic soluble species and the sorbent surface. Electrostatic forces contribute quite a lot in the sorption mechanisms. In order to understand the different sorption behaviour of As(III) and As(V) the thermodynamic equilibrium diagrams have been constructed with the computer code Mineql Plus (1998) using the Mintequa II databases and are presented in Figs. 3 and 4, respectively. It is observed that, in As(III), the neutral H_3AsO_3 species predominate at pH values 2–8, while in As(V) the single negatively charged $H_2AsO_4^-$ predominates at pH values 3–6 and then the double negatively charged $HAsO_4^{-2}$ at pH values up to 11. So, the electrostatic forces between the negatively charged As(V) species and the usually positively charged iron oxide surface, at the pH ranges of natural waters, are much stronger. This might be a reason for the better As(V) sorption removal. However, with decreased arsenic concentrations the contribution of electrostatic forces to sorption is not enough to remove arsenic below the new stringent limits. Therefore, there is a tremendous demand for developing cheap efficient methods for removal of arsenic species from drinking water. Sorption methods are still considered promising, in regard to the cost/efficiency factor and new sorbents are being developed with the aim of obtaining the new target limit. The most commonly used sorbents can be classified in two main groups: (1) those based on iron compounds, which are the most frequently used (Xu et al. 2002; Rau et al. 2003; Singh and Pant 2004) and (2) those based on aluminium compounds, e.g., activated alumina γ-Al_2O_3 or gibbsite $Al(OH)_3$. Other sorbents that have been studied comprise coconut husk carbon (Manju et al. 1998), carbon from fly

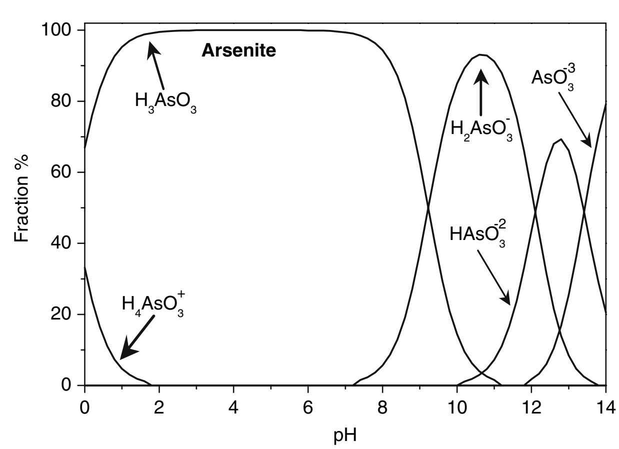


Fig. 3 Arsenite speciation as a function of pH for total As(III) concentration 50 mg L^{-1} (constructed by Mineql Plus)

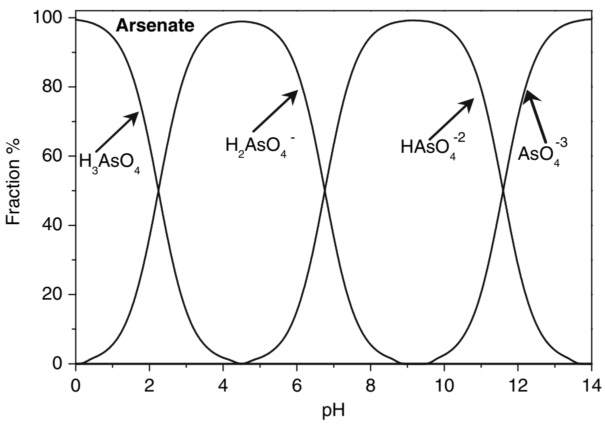


Fig. 4 Arsenate speciation as a function of pH for total As(V) concentration 50 mg L^{-1} (constructed by Mineql Plus)

ash (Patanayak et al. 2000, Bertocchi et al. 2006), hybrid polymeric materials (DeMarco et al. 2003), red mud (Genc-Fuhrman et al. 2005, Altundogan et al. 2002), titanium dioxide (Dutta et al. 2004; Jing et al. 2005), manganese dioxide (Lenoble et al. 2004; Deschamps et al. 2005), orange waste (Ghimire et al. 2003) and also biosorbents, e.g., fungal biomass (Loukidou et al. 2003; Pokhrel and Viraraghavan 2006).

Sorbents based on iron oxides/oxyhydroxides

Iron compounds have been reported to be effective for the removal of metal ions. Several iron(III) oxides/oxyhydroxides, such as amorphous hydrous ferric oxide (FeO-OH), poorly crystalline hydrous ferric oxide–ferrihydride (Wilkie and Hering 1996), goethite (α-FeOOH) and akaganeite (*β*-FeOOH) are promising sorbents for removing both As(V) and As(III) from aqueous solutions (Lakshmipathiraj et al. 2006; Vaclavikova et al. 2005b; Deliyanni et al. 2003; Matis et al. 1999). Other type of sorbents based on iron oxides/oxyhydroxides such as iron oxide-coated polymeric minerals (Katsoyiannis and Zouboulis 2002), iron oxide-coated sand (Thirunavukkaresu et al. 2003), granular ferric hydroxide (Badruzzaman et al. 2004; Sperlich et al. 2005), iron oxide-coated cement (Kundu and Gupta 2005), iron-hydroxide coated alumina (Hlavay and Polyak 2005), Ce(IV)-doped iron oxide adsorbent (Zhang et al. 2003), silica-containing iron(III) oxide (Zeng 2003), magnetically modified zeolite (Vaclavikova et al. 2005a), natural iron ores (Zhang et al. 2004) and waste materials containing iron particles e.g. fly ash and red mud (Bertocchi et al. 2006) have been investigated as well. A comparison of the removal capacities of selected sorbent materials towards As is given in Table 1.

Recent data indicated that hydrous ferric oxide (HFO) such as granular ferric hydroxide (GFH), ferrihydride, goethite as well as akaganeite sorb strongly arsenic species. Sperlich et al. (2005) studied the breakthrough behaviour of GFH fixed bed adsorption filter for removal of arsenic. Equilibrium adsorption isotherms were developed for initial concentrations (1–8 mg L^{-1}) of arsenate spiked in deionized water. The maximum uptake values for low (0–1 mg L^{-1}) and high initial concentrations (1–8 mg L^{-1})

Table 1 Arsenic removal capacities of selected sorbent materials

Adsorbent	Q_{max} (mmol g^{-1})		pH	Reference
	As(III)	As(V)		
Akaganeite	N/A	1.79	7.5	Deliyanni et al. (2003)
Akaganeite	N/A	0.93	3.5	Vaclavikova et al. (2005b)
Goethite	N/A	0.33	5.0	Matis et al. (1999)
Hydrous ferric oxide	N/A	1.34	4.0	Wilkie and Hering (1996)
Iron oxide coated sand	0.0057	0.0055	7.6	Thirunavukkarasu et al. (2003)
Fe(III) loaded resin	0.15	0.80	1.7	Rau et al. (2003)
Granular ferric hydroxide				
low initial conc.	N/A	0.39	7.0	Sperlich et al. (2005)
higher initial conc.	N/A	0.57	7.0	
Ce(IV) doped iron oxide	N/A	0.93	5.0	Zhang et al. (2003)
Natural iron ores	N/A	0.0053	4.5–6.5	Zhang et al. (2004)
Iron oxide coated cement	0.0093	N/A	7.0	Kundu and Gupta 2005
Magnet. modified zeolite	N/A	0.93	3.5	Vaclavikova et al. (2005a)
Fe-hydroxide coated alumina	0.102	0.212	6.6–7.2	Hlavay and Polyak (2005)
Sand-red mud columns	0.012	N/A	5.8–7.5	Altundogan et al. (2002)
	N/A	0.013	1.8–3.5	
Fly ash	N/A	0.0026	6.9	Bertocchi et al. (2006)

were 28.9 and 42.7 mg g^{-1}, respectively, at pH 7. Badruzzaman et al. (2004) presented an arsenate adsorption density of about 8 mg As per g of dry GFH at pH 7. Goethite and akaganeite have also been studied. Goethite showed a satisfactory sorption capacity of about 25 mg g^{-1} (Matis et al. 1999). Synthetic akaganeite (β-FeOOH) has shown an uptake of 65 mg of As per g of akaganeite at pH 3.5 and temperature 22°C (Vaclavikova et al. 2005b) and of 120 mg of As per g of sorbent at pH range 4.5–7 and temperature 25°C (Deliyanni et al. 2003). The higher removal capacity of akaganeite could be attributed to its higher surface area (i.e., 330 m^2 g^{-1} for akaganeite and 130 m^2 g^{-1} for goethite), which is an important factor for an effective sorption process. The particle size of the above mentioned iron oxyhydroxides is in the range of nanomaterials, and the solid/liquid separation as well as the use of the materials in sorption columns is problematic. Here, a good challenge is the development of novel advanced sorbents that will include nano-sized guests (iron oxide/oxyhydroxide) into a well organized host matrix. Zeolites are ideal hosts for the accommodation of organic and inorganic molecules, polymer chains, etc. because of their uniform pore size and their ability to adsorb molecular species. They are known to be good sorbents/ion-exchangers for cations and their surface modification can create localized functional groups with a good affinity to anions too. Recently, Vaclavikova et al. (2005a) presented the synthesis of magnetic iron oxide based nano-partiles inside porous natural zeolite. Initial sorption experiments, have shown a sorption capacity of around 70 mg of As per g of sorbent. This novel synthesised sorbent had good magnetic properties and the solid/liquid (S/L) separation is expected to be easy in a high gradient magnetic field. Moreover, the material has a porous microstructute that might make it suitable for sorption columns; fixed-bed sorption studies are planned.

Recently, Katsoyiannis and Zouboulis (2004) reported a promising technology for the effective removal of arsenic from groundwaters. They used the naturally occurring microorganisms Gallionella ferruginea and Leptothrix ochracea to oxidize the iron which exist (or it is added) to natural waters and form new iron oxide precipitates on filter medium which adsorb the arsenic and remove it from the aqueous streams. Under optimized experimental conditions, trivalent arsenic was found to be oxidized by these microorganisms, contributing to increased overall arsenic removal (up to 95%) even when initial arsenic concentrations were 200 µg L^{-1}. In addition, the pentavalent arsenic content, under the same experimental conditions was removed effectively, leading to residual concentrations below the newly enforced limit of 10 µg L^{-1}.

Conclusions

The presence of arsenic in groundwater has been recognized as a major problem for many parts of the world. Due to its high toxicity, even in low concentrations, it is a threat to human health. Hence, the new maximum contaminant level (MCL) of arsenic in drinking water has been set by authorities worldwide to 10 µg L^{-1} from an earlier value of 50 µg L^{-1}. There are many techniques, which can be effectively applied to remove arsenic from water streams with high arsenic concentrations. However, the same techniques are not very effective at lower initial arsenic concentrations and very often it is not possible to reach the regulatory limit of 10 µg L^{-1}. Moreover, some of the existing techniques, are costly and they are not economically applicable in small community systems. Therefore, there is a need for developing cheap efficient methods for the removal of arsenic species from drinking water. So, from our point of view, it is important to develop efficient sorbent materials suitable for easy S/L separation and/or for column operations. A novel developed sorbent (by incorporation of iron oxide based magnetic nano-particles into a zeolite structure) seems to be a promising one for economic application in small systems. Another promising technique, presented by Katsoyiannis and Zouboulis (2004), capable to remove both As(III) and AS(V) from ground waters to concentrations below the limit of 10 µg L^{-1}, involves the biological oxidation of Fe(II) to Fe(III) (by microorganisms naturally found in the ground waters) and the subsequent sorption of arsenic in the newly formed iron hydroxide.

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