An overview of main arsenic removal technologies

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Abstract: Arsenic (As) is metalloid, naturally present in the environment but also introduced by human activities. It is toxic and carcinogenic and its exposure to low or high concentrations can be fatal to human health. Arsenic contamination in drinking water threatens more than 150 million peoples all over the world. Therefore, treatment of As contaminated water is of unquestionable importance. The present review begins with an overview of As chemistry, distribution and toxicity, which are relevant aspects to understand and develop remediation techniques. The most common As removal processes (chemical precipitation, adsorption, ion exchange, membrane filtration, phytoremediation and electrocoagulation) are presented with discussion of their advantages, drawbacks and the main recent achievements.

Keywords: arsenic contamination, health hazards, toxicity, removal techniques

Introduction

Arsenic with atomic number 33 (located in group VA) is a metalloid, which naturally occurs in an environment, but is proven to have negative influence on a human health. It is a silver-grey brittle crystalline solid with atomic weight 74.9 $g \cdot mol^{-1}$; specific gravity 5.73 $g \cdot cm^{-3}$; melting point 817 °C and boiling point 614 °C. It ranks as the 20th most occurring trace element in the earth's crust, 14th in seawater, and the 12th in the human body (Mohan and Pittman 2007; Singh et al., 2015).

Nowadays the arsenic occurrence has been proven to be responsible for many diseases, where it occurs in drinking water. Therefore the arsenic problem has been considered as one of the main issues within researches and authorities. And for the past 20 years interest in this problem solution has increased significantly as a consequence of new findings about arsenic effects on human health resulting in implementation of stricter drinking water quality guidelines (van Halem et al., 2009).

Due to the very insidious nature of the problem and the absence of effective monitoring in many cases, it is difficult to assess exactly how many people daily are joining the long list of arsenic victims. To ensure sustainable supplies of safe drinking water to the arsenic-affected areas, certain requirements must be fulfilled: an adequate supply source has to be identified and the best source for a specific area has to be selected; efficient treatment systems should be developed for treating such water to at least the WHO-prescribed level of 10 $\mu g \cdot L^{-1}$; treatment cost of systems should be affordable to the people of the affected regions and long-term trouble-free operation of treatment systems should be ensured through effective monitoring and regular maintenance. Such a system should be able to take care of the total environment without transferring the problem of pollution from one area to another (Pal, 2015).

However, to understand the removal processes and before providing a detailed comparison of the available systems it is needed to know about better arsenic properties, especially the derivates that occur in the environment, because the technologies are based on shifting from one arsenic compound to another.

Arsenic occurrence in water

Arsenic is mobilized by natural weathering reactions, biological activity, geochemical reactions, volcanic emissions and other anthropogenic activities. Most environmental arsenic problems are the result of mobilization under natural conditions. However, mining activities, combustion of fossil fuels, use of arsenic pesticides, herbicides, and crop desiccants and use of arsenic additives to livestock feed create additional impacts. The general pathways of arsenic release are presented in Figure 1 (Mackenzie et al., 1979; Wang and Mulligan, 2006).

Water is one of the major means of transport of arsenic in the environment. Arsenic contamination of surface and subsurface waters is a worldwide problem, with reported studies in a large number of countries including Italy, Hungary, Serbia, Croatia, New Zealand, Bengal, Chile, Taiwan, Vietnam,
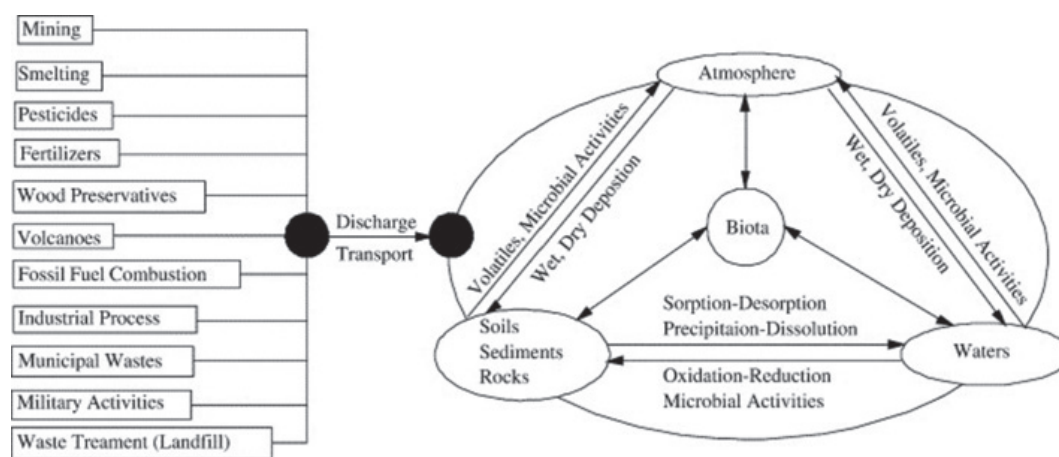
Fig. 1. A simplified diagram of the arsenic cycle (Wang and Mulligan, 2006).

Bangladesh, United States and Canada (Welch et al., 2000; Smedlev and Kinniburgh, 2001). Arsenic occurs naturally in water in many parts of the world usually in the forms of the soluble arsenic species As (III) (arsenite) and As (V) (arsenate). The distribution of arsenic species in natural water is mainly dependent on redox potential (Eh) and pH of the water. Under oxidizing conditions the predominant species is pentavalent arsenic, which is present in arsenic acid and oxyanionic forms (H_3AsO_4, $H_2AsO_4^-$, $HAsO_4^{2-}$ and AsO_4^{3-}). The trivalent arsenic is a thermodynamically stable form (H_3AsO_3, $H_2AsO_3^-$ and $HAsO_3^{2-}$ and AsO_3^{3-}), under anoxic conditions. The pentavalent arsenic species are predominant and stable in oxygen-rich aerobic environments, whereas the trivalent arsenite species are predominant in moderately reducing anaerobic environment such as groundwater. The pH value determines the predominant species present in water and is therefore expected to be a very influential parameter during arsenic removal treatments. At a typical pH of 5.0 to 8.0 of natural water, the predominant pentavalent arsenate species are $H_2AsO_4^-$ and $HAsO_4^{2-}$ and the trivalent arsenite species is H_3AsO_3. The ratio of As (V) to As (III) in natural water is about 4:1. Figure 2 describes the relationships between Eh, pH and aqueous arsenic species. Under oxidizing conditions, $HAsO_4^{2-}$ dominates at a high pH regime whereas H_3AsO_4 and AsO_4^{2-} predominate in extremely acidic and alkaline conditions, respectively. $H_2AsO_4^-$ predominates at low pH (< 6.9). Under reducing conditions at a pH of less than 9.2, the uncharged species H_3AsO_3 will predominate. This means that As (III) remains as a neutral molecule in natural water (Viraraghavan et al., 1999; Smedlev and Kinniburgh, 2001; Katsoyiannis and Zouboulis, 2004).

In natural watercourses, organic can also be found, such as monomethyl arseneous acid (MMA (III)), monomethyl arsenic acid (MMA (V)), dimethyl arseneous acid (DMA (III)) and dimethyl arsenic acid (DMA (V)). This organic arsenic occurs at a concentrations less than 1 $\mu g \cdot L^{-1}$ and is not of major significance in drinking water treatment (Edwards, 1994; Hung et al., 2004). Information on the solubility data of arsenic compounds can be found in Table 1 (IARC, 2012; Pubchem, 2018).
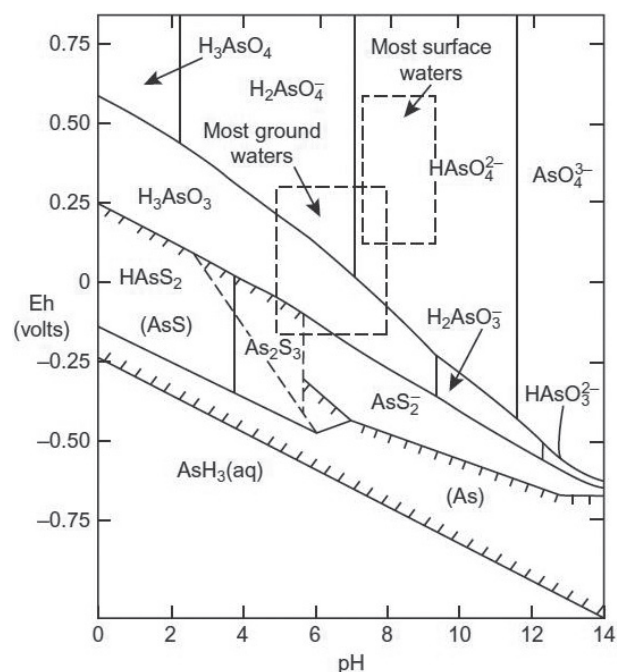

Fig. 2. Eh-pH diagram for arsenic at 25 °C and 1 bar total pressure, with total arsenic 10^{-5} mol $\cdot L^{-1}$; symbols for solid species are enclosed in parentheses in crosshatched area, which indicates solubility less than 10^{-5} mol $\cdot L^{-1}$ (Pal, 2015).

Toxicity of arsenic and effect on human health

Many pollutants in water streams have been identified as toxic and harmful to the environment and

Tab. 1. Chemical name, molecular formula and solubility data of arsenic compounds (IARC, 2012; Pubchem, 2018).

Chemical name	Formula	Solubility
Arsanilic acid	$C_6H_8AsNO_3$	Slightly soluble in cold water, alcohol and acetic acid. Soluble in hot water and amyl alcohol.
Arsenic pentoxide	As_2O_5	Soluble in water (658 g·l⁻¹ at 20 °C) and in alcohol.
Arsenic trihydride (Arsine)	AsH_3	Very poor solubility in water (0.28 g·l⁻¹ at 20 °C), in alcohol and alkalies. Soluble in chloroform and benzene.
Arsenic trichloride	$AsCl_3$	Hydrolysis in water. Soluble in alcohol, ether, hydrochloric and hydrobromic acid.
Arsenic trioxide	As_2O_3	Very slightly soluble in water (37 g·l⁻¹ at 20 °C). Soluble in dilute acids and alkalies.
Arsenic trisulfide	As_2S_3	Insoluble in water. Soluble in alkalies, carbonates, alkali sulfides and alcohol.
Calcium arsenate	$Ca_3(AsO_4)_2$	Very poor solubility in water (0.13 g·l⁻¹ at 25 °C). Soluble in acids.
Dimethylarsenic acid	$C_2H_7AsO_2$	Soluble in water (667 g·l⁻¹ at 20 °C), ethanol, acetic acid.
Disodium hydrogen arsenate	Na_2HAsO_4	Soluble in water (610 g·l⁻¹ at 15 °C). Slightly soluble in alcohol.
Lead arsenate	$PbHAsO_4$	Insoluble in water. Soluble in nitric acid and alkalies.
Methylarsonic acid	CH_5AsO_3	Soluble in water (256 g·l⁻¹ at 20 °C) and in ethanol.
Sodium arsenite	$NaAsO_2$	Soluble in water. Slightly soluble in alcohol.

human health. Among them arsenic is considered a high priority, because it has been identified as a human carcinogen (group 1) and can cause chronic diseases. The toxicity and carcinogenicity of arsenic is dependent on their forms and oxidation states (Ungureanu et al., 2015).

Inorganic forms of arsenic dissolved in drinking water are the most significant forms of natural exposure and are more toxic than organic ones. Generally it is stated that the As (III) forms are more toxic than the As (V). The pentavalent arsenic (arsenate) can replace the role and position of phosphate in the human body due to its similar structure and properties with phosphate. The toxicity of arsenic species follows the order (highest to lowest): arsines > inorganic arsenites > organic trivalent compounds (arsenoxide) > inorganic arsenates > organic pentavalent compounds > arsonium compounds > elemental arsenic. Toxicity of As (III) is referred to be nearly 70 times higher than As organic forms and 10 times higher than As (V).

Many studies have indicated that arsenic ingestion may result in internal malignancies, including cancers of the kidney, bladder, liver, lung and other organs. It also has noncancer effects that include cardiovascular, pulmonary, immunological, neurological, reproductive and endocrine (e.g. diabetes) disorders. Bedsides its tumorigenic potential, arsenic has been shown to have genotoxicity. The health effects caused by acute arsenic poisoning are called arsenicosis, which has been also responsible for keratosis, skin changes and hyperkeratosis, skin lesions (Squibb and Fowler, 1983; Karim, 2000; Mascher et al., 2002).

Strong epidemiological evidence of arsenic carcinogenicity and genotoxicity has forced the World Health Organization (WHO) to lower the Maximum Contaminant Level (MCL) in drinking water to 10 µg·L⁻¹ from earlier limit of 50 µg·L⁻¹ in 1993, followed by the Unites States Environmental Protection Agency adoption of the same in 2001. However, the prescribed MCL of arsenic in drinking water (Table 2) is found to vary from country to country.

Tab. 2. Maximum contaminant level (MCL) of arsenic set by different countries (Pal, 2015).

Countries/others	MCL (µg·L⁻¹)
WHO/USEPA/European Union	10
Australia	7
France	15
India, Bangladesh, Vietnam, Mexico	50
Malaysia	10—50

Methods of arsenic removal

Different technologies have been used and proposed to remove arsenic from aqueous media. The presently available technologies are chemical precipitation, adsorption, ion exchange, membrane filtration, phytoremediation and electrocoagulation.

Tab. 3. A Comparison of Main Arsenic Removal Technologies (Viraraghavan et al., 1999; Choong et al., 2007; Han et al., 2013; Pal, 2015; Singh et al., 2015).

Technology	Advantages	Disadvantages
Chemical precipitation	Relatively low capital cost, simple in operation, common chemicals are available.	Produces toxic sludge. Pre-oxidation may be required, which can produce harmful disinfection by-products. Mainly removes As (V), medium removal of As (III). Chemicals dosing, oxidation step, sedimentation and filtration is needed.
Sorption techniques	Relatively well know and commercially available. High removal efficiency. Easy operation and handling. More cost effective. Additional chemical and sludge free. No harmful by-products.	Adsorption bed gets exhausted. Periodic replacement of adsorbent material. Yet to be standardized. Produces toxic solid waste.
Ion exchange	Well-defined medium and capacity. The process is less dependent on pH of water. Exclusive ion-specific resin to remove arsenic.	High cost medium. Requires high-tech operation and maintenance. Regeneration creates a sludge disposal problem. As (III) is difficult to remove. Exhausted resin regeneration. Applicable for low-TDS. Life of resin.
Membrane techniques	Well-defined and high removal efficiency. No toxic solid waste is produced. Capable of removal of other contaminants and microorganisms. Chemicals free.	Very high capital cost. Pre-treatment is needed. High water rejection. High-tech operation and maintenance. Toxic wastewater is produced.
Phyto-remediation	Environmental friendly. Chemicals free. Long-term remediation.	Strong developing is needed. No commercially available.
Electro-coagulation	Alternative to chemical precipitation. Chemicals free. Novel and promising strategy. Efficient, low cost and easy to maintain.	Strong developing is needed. No commercially available. Focus on effective design and operation parameters is needed.

Nevertheless, each technology has disadvantages and advantages, especially regarding efficiency and costs, which decide on implemented treatment (Table 3). Other factors influencing the choice of proper arsenic treatment are the following: local guidelines for arsenic level in drinking water; country development stage; authority's requirements and restrictions for water treatment technologies. Therefore, many different technologies can be found around the world. Additionally, the general trend in water treatment is to use as less of chemicals and energy as possible, to reduce the costs. For better understanding the arsenic removal techniques next part will describe them.

Chemical precipitation — coagulation/flocculation

In this process, anions combine with cations resulting in precipitation. It is one of the mostly employed treatments to produce water for drinking-purposes. The addition of a coagulant followed by the formation of a floc is a potential way for the removal of As from groundwater. Coagulants change the surface charge properties of solids to allow the agglomeration or enmeshment of particles into a flocculated precipitate. The final products are larger particles or floc, which settle under the influence of gravity or filtered more readily. Commonly used chemicals are aluminium salts such as aluminium sulfate $[Al_2(SO_4)_3 \cdot 18H_2O]$ and ferric salts such as ferric chloride $[FeCl_3]$ or ferric sulfate $[Fe_2(SO_4)_3 \cdot 7H_2O]$. Both aluminum and ferric salts have proven to be efficient coagulants for arsenic removal from $mg \cdot L^{-1}$ and $\mu g \cdot L^{-1}$ levels. In this process, chemicals transform dissolved As into solid (insoluble) which is precipitated later. The alum or iron sludge generated in the clarifier contains arsenic removed from the water. The particles of alum/iron/arsenic that are not settled out in the clarifier are removed by employing a filter, followed by a clarifier.

Dissolved As may also be adsorbed on the solid hydroxide surface site and be coprecipitated with other precipitating species. The principle of co-precipitation is oxidizing the iron and/or manganese from their soluble state (oxidation state 2+) to a higher oxidation state to form iron and/or manganese precipitates. The arsenic is apparently removed as iron/arsenic or manganese/arsenic precipitates, which can be filtered and then are backwashed off of the filter media (Smedley and

Kinniburgh, 2002; Mondal et al., 2006; Choong et al., 2007; Ungureanu et al., 2015).

Other chemical required for this technique are oxidizing agent (chlorine, chlorine dioxide, ozone, hydrogen peroxide, chloroamine, permanganate, air and pure oxygen), acid and caustic soda. The main purpose of oxidation is to convert the soluble As (III) to As (V), which is then followed by precipitation of As (V). Acid is required to maintain pH at the desired level. Caustic soda would be added to increase pH to an acceptable level in the posttreatment of clarified water. Chlorine is an oxidizing agent, although it is not recommended due to adverse effects related to the formation of disinfection by-products and release of flavor and odor. The most common and easiest used oxidative agent is molecular oxygen that can be delivered to solution in contact with atmospheric air. Although oxygen is not as efficient as other chemical oxidants, but it has some advantages — low cost and easy access, oxygen can be delivered in an aeration process (Gregor, 2001; Smedley and Kinniburgh, 2002; Sharma et al., 2007).

The use of advanced oxidative processes has also been proposed, as an involvement of highly reactive radicals seemed to be a good solution. UV-light assisted oxidation of As (III) is the most widely tested chemical oxidant in presence of naturally occurring iron. The oxidation rate of As (III) in the water can be increased by UV irradiation in the presence of oxygen. UV/solar light helps to generate hydroxyl radicals through the photolysis of Fe (III) species: ($FeOH^{2+}$) and in the presence of both hydroxyl radicals and oxygen, the oxidation rate becomes faster. Several studies have investigated the photochemical oxidation of As (III) using UV light irradiation. This system was also found to be useful under natural water conditions. Instead of UV-light, solar-light can also remove As from natural water upon addition of iron and citrate (Sharma et al., 2007; Ryu et al., 2013).

Generally, with increasing coagulant dosages higher As removal efficiencies can be achieved. The chemical precipitation has: relatively low capital costs, simplicity in operation and chemicals are available. On the other side, it produces toxic sludge, pre-oxidation may be required, sedimentation and filtration is needed (Han et al., 2013).

Adsorption

Adsorption is a process that uses solids for removing substances from either gaseous or liquid solutions. Adsorption process has been used most widely because of its high removal efficiency, easy operation and handling, low cost and sludge-free. The adsorptive behavior of an adsorbent is strongly dependent on the chemical form of the adsorbate. Conventional sorbents used in water treatment include commercial activated carbons, activated alumina, iron based sorbents, zeolites, etc. Many other materials, such as synthetic and modified activated carbons, clay minerals, other natural and synthetic oxides, sand and biomaterials have been proposed as potential low-cost adsorbents for arsenic removal. The aim is to find a cost-effective treatment for contaminated water treatment, especially useful for developing countries. Several materials have been used in their naturals forms, without significant further treatment (Zhu et al., 2013; Singh et al., 2015).

Generally, the remove of As by adsorption techniques depends on pH and the speciation of As thus, at pH lower than 7 showing better As (V) removals compared to the As (III). The capacity and adsorption rate further depends on the presence of other ions like phosphate, silica, alkalinity and Ca^{2+} competing for the adsorption sites (Sen and Pal, 2009).

Adsorption has attracted much attention due to the following advantages: it usually does not need a large volume and additional chemicals; it is easier to set up as a As removal process and it doesn't produce harmful by-products and can be more cost effective. But on the other side the sorbents need replacement after four to five regeneration or with time, as the adsorption bed gets more and more saturated and exhausted, it loses its capacity of separation and eventually no further separation is done. From a quality point of view, because the adsorption-based process is not self-monitoring, it continues to produce water even after the adsorption bed gets exhausted. Periodic replacement of adsorbent material is a must for such units (Dambies, 2004; Höll, 2010; Zhu et al., 2013).

Ion Exchange

Ion exchange is a physical/chemical process by which an anion on the solid resin phase is exchanged for an ion in the feed water. The solid resin is typically an elastic three-dimensional hydrocarbon network containing a large number of ionizable groups electrostatically bound to the resin. These groups are exchanged for ions of similar charge in solution that have a stronger exchange affinity (i.e. selectivity) for the resin. Typically, strong base anion exchange resins are commonly used for the removal of As where the oxy-anionic species of As (V) (such as $H_2AsO_4^-$, $HAsO_4^-$ and AsO_4^{3-}) are effectively exchanged with the anionic charged functional group of the resin, thus produces effluents with low concentration of As (V) (Choong et al., 2007).

The As (V) can be easily removed through the use of strong-base anion exchange resin either in the form of chloride or hydroxide. An ion exchange resin, attached with chloride ions at the exchange sites, is placed in a vessel. The arsenic-containing water is passed through the resin bed and the chloride ion is exchanged by arsenic anions. The water coming out from the resin bed is lower in arsenic but higher in chloride than the water entering the vessel. When all or most of the exchange sites are occupied by arsenic or other anions by replacing chloride ions, the resin gets exhausted. The exhausted resin is regenerated with salt (sodium chloride) (Sarkar et al., 2007; Pal, 2015).

The efficiency of ion exchange process is improved by pre-oxidation of As (III) to As (V) but before the ion exchange, the excess of oxidant often needs to be removed in order to avoid the damage of sensitive resins. Therefore, the efficiency of the ion exchange process for As (V) removal strongly depends on the solution pH and the concentration of competing ions most notably sulfates and nitrates, resin type, alkalinity and influent. The resin prefers sulfate ions to arsenic anions, so the sulfate ions are exchanged for chloride ions before the arsenic ions. The performance of an ion exchange system can be adversely affected by high levels of total dissolved solids (TDS). In general, ion exchange for As removal is only applicable for low-TDS, low-sulfate source waters. Metal-loaded polymers (chelating or ion-exchange metal-loaded resins) have been proposed as advantageous comparing to strong-base ion-exchange resins, since these materials can overcome interferences from anions and present the possibility to remove both As (III, V). The ion exchange process has the disadvantage of releasing noxious chemical reagents used in the resin regeneration into the environment (Donia et al., 2011; Pal, 2015).

Membrane filtration

Membranes are typically synthetic materials with billions of pores or microscopic holes that act as a selective barrier; the structure of the membrane allows some constituents to pass through, while others are excluded or rejected. The movement of molecules across the membrane needs a driving force, such as pressure difference between the two sides of the membrane. It produces large residual volumes and is more expensive than other As treatment technologies. Membrane filtration processes are classified into four categories: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). The separation by these processes depends on the pore size of the membrane; for MF and UF membranes, mechanical sieving is responsible for separation while for NF and RO membranes, separation is achieved via capillary flow or solution diffusion (Sen et al., 2010; Pal et al., 2012).

All these membrane processes are effective to remove arsenic in order to respect maximum admissible concentrations, especially the high pressure processes, NF and RO, which represent excellent removal efficiencies. As (V) rejections observed in NF and RO ranged from 85 to 99 % and As (III) rejections between 61 and 87 %. A previous oxidation to convert As (III) to As (V) is not advisable due to the possible damage of the membrane. These processes are essentially disadvantageous due to the high costs (installation and energy consumption) and the high rejection of water. The advantages of using membrane in arsenic removal are: the membrane technologies can effectively remove portions of all dissolved solids including arsenic from feed water and even prevent the microorganisms passing through the membrane to diminish the harmful diseases and the membrane itself does not accumulate arsenic, so disposal of used membranes would be simple, maintenance and operation requirements are minimal, and no chemicals need to be added (Shih, 2005; Choong et al., 2007; Ungureanu et al., 2015).

Phytoremediation

Phytoremediation is the plant based environmental-friendly technology, for the remediation of As contaminated sites, using plants and microbes to clean up contaminated water. The Pteris vittata (Chinese brake fern) was found to be resistant to As, having the capability of hyperaccumulating large amounts of As in its fronds by area contaminants are picked up by the roots of plants and transported to their over ground parts, and then removed together with the crops (phytostabilization, phytoextraction and phyto-volatilization). The As hyperaccumulation capacity has also been demonstrated in other plants. Besides phytoremediation, phytostabilization methods using plants can also be applied for long-term remediation of As. This method limits uptake and excludes mobilization of As. The major benefit of phytostabilization is that the vegetative biomass above ground is not contaminated with As, thus reduces the risk of As transfer through food chains. Furthemore, the bioremediation techniques, including a variety of sulfate reducing bacteria and other species such as Paenibacillus, Pseudomonas, Haemophilus, Micrococcus and Bacillus may be involved to remediate As from contaminated environments. However, there is still a strong challenge in developing economical and commonly available biosorbents for the As removal (Ma et al., 2001; Madejón et al., 2002; Yamamura et al., 2003).

Electrocoagulation (EC)

It is an alternative process to chemical precipitation (CP) using electrodes made from iron scrap. Instead of adding a chemical reagent as ferric chloride, metallic cations are directly generated in the effluent to be treated by applying a current between iron electrodes to dissolve soluble anodes. In EC, electrolytic oxidation of a sacrificial iron Fe (0) anode produces Fe (III) oxyhydroxides/precipitates in As contaminated water. With Fe (III) precipitates As forms binuclear, inner-sphere complexes, which aggregate to form a floc. It is a novel and promising As removal strategy for arsenic for drinking water as, it is efficient, low cost and easy to maintain and operate with locally available materials; EC introduces Fe (II)/Fe (III) without introducing undesirable anions into the solution; the release of H_2 (g) from the cathode neutralizes the consumption of hydroxide by the Fe (III) hydrolysis and therefore likely to buffer the system better than chemical coagulation; and the gradual release of Fe (II)/Fe (III) in EC may produce intermediate oxidants that enhance the efficiency of As (III) oxidation as compared to CP. Previous EC researchers have mostly focused on the effect of design and operation parameters or water matrix on As removal and propose some qualitative conclusions (van Genuchten et al., 2012; Li et al., 2014).

Conclusion

Present overview shows that the removal of arsenic from contaminated water could be the only effective option to minimize health hazard. To achieve this, various processes are being used. It is difficult to select a best technique, since each one has some advantages and drawbacks and their by-products can be a further potential source for secondary As pollution. Therefore, new technologies with the options of new hybrid techniques are needed to challenge the menace of As. Ease of maintenance, simplicity and flexibility of the system are other important parameters that should be considered when identifying a sustainable technology.

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