ARSENIC REMOVAL USING ADSORPTIVE MEDIA TREATMENT PROCESS

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ABSTRACT

Arsenic contamination in ground water has been envisaged as a problem of global concern. However, arsenic contamination of ground water in parts of South East Asia is assuming greater proportions and posing a serious threat to the health of millions of people. A variety of treatment technologies based on oxidation, co-precipitation, adsorption, ion-exchange and membrane process are available for the removal of arsenic from ground water. However, question remains regarding the efficiency and applicability/appropriateness of the technologies, particularly because of low influent arsenic concentration and differences in source water composition. Some of these methods are quite simple, but the disadvantage associated with them is that they produce large amounts of toxic sludge, which needs further treatment before disposal into the environment. Besides, the system must be economically viable and socially acceptable. In this paper an attempt has been made to review and update the recent advances made in the technological development in arsenic removal technologies using adsorptive media process to explore the potential of those advances to address the problem of arsenic contamination in South East Asia.

1. INTRODUCTION

Arsenic is a common, naturally occurring drinking water contaminant that originates from arsenic-containing rocks and soil and is transported to natural waters through erosion and dissolution. Arsenic occurs in natural waters in both organic and inorganic forms. However, inorganic arsenic is predominant in natural waters and is the most likely form of arsenic to exist at concentrations that cause regulatory concern.

The valence and species of inorganic arsenic are dependent on the oxidation-reduction conditions and the pH of the water. As a general rule of thumb, arsenite, the reduced, trivalent form [As(III)], normally is found in ground water (assuming anaerobic conditions); and arsenate, the oxidized pentavalent form [As(V)], is found in surface water (assuming aerobic conditions). This rule, however, does not always hold true for ground water. Some ground waters have been found to have only As(III), others with only As(V), and still others with the combination of both As(III) and As(V).

Various technologies available for removal of arsenic from contaminated water are based mainly on six principles:

i) Oxidation and filtration
ii) Biological oxidation: Oxidation of As(III) to As(V) by microorganisms and then removal of As(V) by iron and manganese oxides.
iii) Co-precipitation: Oxidation of As(III) to As(V) by adding suitable oxidizing agent followed by coagulation, sedimentation and filtration.
iv) Adsorption: Activated alumina, activated carbon, iron based sorbents, zero valent iron and hydrated iron oxide, etc.
v) Ion exchange through suitable cation and anion exchange resins.
vi) Membrane technology: Reverse osmosis, nanofiltration and electrodialysis.

In this paper an attempt has been made to review and update the recent advances made in the technological development in arsenic removal technologies using adsorptive media process to explore the potential of those advances to address the problem of arsenic contamination in South East Asia.

2. ADSORPTION TECHNOLOGY

Adsorption technology has been widely used to treat ground water containing arsenic. The technology typically can reduce arsenic concentrations to less 10 µg L^{-1}. Its effectiveness is sensitive to a variety of untreated water contaminants and characteristics. The adsorption media is usually packed into a column. As the contaminated water is passed through the column, contaminants are adsorbed. When adsorption sites become filled, the column must be regenerated or disposed of and replaced with new media.

The following media are commonly used for removal of arsenic through adsorption technique.

i) Activated alumina (AA),
ii) Iron Based Sorbents (IBS) - Granular Ferric Hydroxide, Zero Valent Iron, Iron Coated Sand, etc.,
iii) Indigenous Filters and Cartridges, and
iv) Other Miscellaneous Adsorbents

The efficiency of sorptive media depends on the use of oxidizing agent as aids to sorption of arsenic. Saturation of media by different contaminants and components of water takes place at different times of operation depending on the specific sorption affinity of the medium to the given component.

2.1 ACTIVATED ALUMINA (AA)

Activated alumina was the first adsorptive medium to be successfully applied for the removal of arsenic from water supplies (EPA, 2000a,b). It is a porous, granular material having good sorption properties. The media, aluminum trioxide, is prepared through the dehydration of aluminum hydroxide at high temperatures. AA grains have a typical diameter of 0.3 to 0.6 mm and a high surface area for sorption. The removal of As(V) by AA adsorption can be accomplished by continuously passing water under pressure through one or more beds packed with AA media. Fig. 1 shows a typical process flow diagram for the adsorption process. Dashed lines and boxes indicate optional streams and processes.

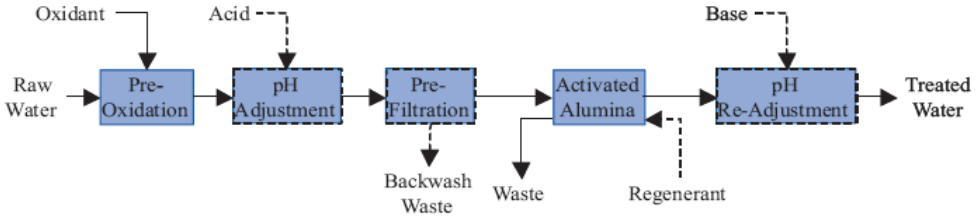

Fig. 1. A Typical Process Flow Diagram for Adsorption Process

The reported adsorption capacity of AA ranges from 0.003 to 0.112 grams of arsenic per gram of AA. It is available in different mesh sizes and its particle size affects contaminant removal efficiency. Up to 23,400 bed volumes of wastewater can be treated before AA requires regeneration or disposal and replacement with new media. The selectivity of AA towards As(III) is poor, owing to the overall neutral molecular charge at pH levels below 9.2. Therefore, pre-oxidation of As(III) to As(V) is critical. Several different studies have established the optimum pH range as 5.5-6.0, and demonstrated greater than 98% arsenic removal under these conditions (Rosenblum and Clifford, 1984; EPA, 2000a,b). AA column runs operated under acidic pH conditions are 5 to 20 times longer than under natural pH conditions (6.0-9.0). The arsenic removal capacity of activated alumina is pH sensitive and therefore requires pre- and post- pH adjustment using caustic soda and sulphuric acid. Various hand pumps attached arsenic removal plants with activated alumina as adsorbent have been installed in West Bengal (India) and Bangladesh through various initiatives.

The technologies and market for alumina-based adsorptive media is continuously expanding. There are several emerging proprietary media, commonly referred to as modified AA, which contain alumina in a mixture with other substances such as iron and sulfur. In some instances, these media have greater overall adsorptive capacities, enhanced selectivity towards arsenic, and/or greater overall operational flexibility than conventional AA, thus making them more cost-effective. Some of the activated alumina based sorptive media include BUET Activated Alumina, Alcan Enhanced Activated Alumina, Arsenic Removal Units (ARUs) of Project Earth Industries Inc., USA, and Apyron Arsenic Treatment Units.

The BUET and Alcan activated alumina have been extensively tested in field condition in different parts of Bangladesh and found very effective in arsenic removal (BAMWSP, DFID, Water Aid, 2001). The Arsenic Removal Units (ARUs) of Project Earth Industries Inc., USA used hybrid aluminas and composite metal oxides as adsorption media and were able to treat 200-500 Bed Volume (BV) of water containing 550 mg L^{-1} of arsenic and 14 mg L^{-1} of iron (Ahmed et al., 2000). The Apyron Technologies Inc. (ATI) also uses inorganic granular metal oxide based media that can selectively remove As(III) and As(V) from water. The Aqua-Bind TM arsenic media used by ATI consist of non-hazardous aluminium oxide and manganese oxide for cost-effective removal of arsenic. The proponents claimed that the units installed in India and Bangladesh consistently reduced arsenic to less than 10 µg L^{-1}.

Recently, Ike et al. (2008) have examined the efficacy of activated alumina in removing arsenic through bacterial oxidation of As(III) with a chemical adsorption process. By acclimation to As(III) of high concentrations, a mixed culture of heterotrophic bacteria with high As(III)-oxidizing activity was obtained from a soil sample that was free from contamination. The mixed culture contained several genera of heterotrophic As(III)-oxidizing and arsenic-tolerant bacteria: *Haemophilus*, *Micrococcus*, and *Bacillus*. Arsenic removal by activated alumina was greatly enhanced by bacterial oxidation of As(III) to As(V). The isotherms of As(III) and As(V) onto activated alumina verified that bacterial As(III) oxidation is a helpful pretreatment process for the conventional adsorption process for arsenic removal.

2.2 IRON BASED SORBENTS (IBS)

Adsorption on IBS is an emerging treatment technique for arsenic removal. Examples of IBS products currently available in the market include granular ferric hydroxide, zero valent iron, iron coated sand, modified iron and iron oxide based adsorbents. The sorption process has been described as chemisorption (Selvin et al., 2000), which is typically considered to be irreversible. It can be applied in fixed bed pressure columns, similar to those for AA. The development of iron based sorbents for arsenic removal has been considered as one of the most promising solution, leading to the development of adsorbents with better performance than activated alumina (Jekel, 1994; Driehaus et al., 1998; Lackovic et al., 2000; Farrell et al., 2001; Su and Puls, 2001; Manning et al., 2002; Melitas et al., 2002; Nikolaidis et al., 2003; Su and Puls, 2003; Hussam and Munir, 2007).

The studies conducted with IBS media have revealed that the affinity of this media for arsenic is strong under natural pH conditions, relative to AA. This feature allows IBS to treat much higher bed volumes without the need for pH adjustment. However, similar to AA, optimal IBS performance is obtained at lower pH values. Phosphate has been shown to compete aggressively with As(V) for adsorption sites. Each 0.5 mg L^{-1} increase in phosphate above 0.2 mg L^{-1} reduce adsorption capacity by roughly 30%. The exhausted IBS media can be disposed of in a municipal solid waste landfill (MacPhee et al., 2001).

The Technical University, Berlin, Germany has developed an effective Granular Ferric Hydroxide (GFH) adsorbent for arsenic removal from natural water (Driehaus et al., 1998). The adsorbent has a specific surface area of 250-300 $m^2 g^{-1}$ and a porosity of 75-80%. The bulk density of GFH saturated with water is 1.32 g cm^{-3}. The grain size ranges from 0.2 to 2.0 mm with all the pores filled with water leading to high density of the available adsorption sites and a high adsorption capacity. The adsorbent has been successfully applied in simple fixed bed reactors, similar to those of activated alumina or activated carbon. The specific capacity of fixed bed adsorbers with GFH depends on pH, phosphate content and the raw water concentration of As(V). It has been shown that in technical scale adsorbers more than 50,000 bed volumes can be treated and filter rates of 15 m h^{-1} are possible. The residue of the technique is a solid waste with residual mass in the range 5-25 g m^{-3} treated water (Driehaus et al., 1998). The adsorbent has been successfully marketed under the trade name GEH®. The technology requires only a small contact time between 3 to 10 minutes, whereas the treatment capacities are up to 150,000 bed volumes. The average treatment costs,

including media exchange service and disposal is 0.04 EURO per m^3 treated water (Driehaus, 2002). M/s Pal Trockner (P) Ltd, India and M/s Sidko Limited, Bangladesh installed several GFH based arsenic removal units in India and Bangladesh. The unit requires iron removal as pre-treatment to avoid clogging of filter bed. The proponents of the unit claim to have high treatment capacity between 40,000 to 60,000 bed volumes, until the permissible limit of 0.01 mg L^{-1} is exceeded and the unit produces non-toxic spent granular ferric hydroxide. As iron content in ground water is generally high, all hand pump attached arsenic removal plants working under adsorption principles also require regular backwashing/cleaning for removal of arrested iron particles. Such backwashing need to be done periodically for optimal operation of the units.

The removal of arsenic by zero valent iron (ZVI) has received increased attention during recent years due to high arsenic removal capacity of ZVI. Several research groups have used ZVI for the removal of arsenic from water (Lackovic et al., 2000; Farrell et al., 2001; Su and Puls, 2001; Manning et al., 2002; Melitas et al., 2002; Nikolaidis et al., 2003; Su and Puls, 2003) and reported that arsenic species are removed by zero valent iron through adsorption on iron oxides. Column experiments have been conducted to evaluate the effectiveness of ZVI for arsenic removal (Melitas et al., 2002; Lackovic et al., 2000; Nikolaidis et al., 2003; Su and Puls, 2003). Melitas et al. (2002) reported that the removal rates of arsenic were up to 10 times faster near the inlet end of the iron column than near the effluent end and attributed this to rapid oxidation of ZVI by small amounts of DO in the influent. On the other hand, Ramaswami et al. (2001) reported that the presence of air in batch reactors hindered arsenic removal by ZVI. All the column experiments were conducted under anoxic conditions and at relatively long hydraulic contact time. The results indicate that a batch-mixed iron decantation system is effective in the treatment of water containing arsenic in the range of 200–2000 µg L^{-1} at contact times of 30 min–3 h, with iron dose ranging from 2500 to 625 mg L^{-1}.

Hussam and Munir (2007) have developed a simple and effective arsenic filter based on composite iron matrix (zero valent iron) and won the prestigious Grainger gold medal award for their work. The development of this innovative filter (SONO filter) has been considered as one of the best treatment options for over 30 million people in Bangladesh who are still exposed to high arsenic concentrations. The filter has been approved by the Bangladesh Government and about 30,000 SONO filters were deployed all over Bangladesh and continue to provide more than a billion liters of safe drinking water. The filter removes arsenic species primarily by surface complexation reactions: $\equiv FeOH + H_2AsO_4^- \rightarrow \equiv FeHAsO_4^- + H_2O$ ($K = 10^{24}$) and $\equiv FeOH + HAsO_4^{2-} \rightarrow \equiv FeAsO_4^{2-} + H_2O$ ($K = 10^{29}$) on a specially manufactured composite iron matrix. The filter water meets WHO and Bangladesh standards, has no breakthrough, work without any chemical treatment (pre- or post-), without regeneration and without producing toxic wastes. It costs about $40/5 years and produce 20-30 L/hour for daily drinking and cooking need of 1-2 families. The spent material is completely non toxic-solid self contained iron-arsenate cement that does not leach in rainwater (Hussam and Munir, 2007). Recently, Mohan and Pittman (2007) reviewed various adsorption based technologies for the removal of arsenic from water and waste water while Cundy et al. (2008) have reviewed iron based technologies for the removal of arsenic from ground water.

2.3 INDIGENOUS FILTERS AND CARTRIDGES

There are several filters available that use indigenous material as arsenic adsorbent. Red soil rich in oxidized iron, clay minerals, iron ore, iron scrap or fillings and processed cellulose materials are known to have capacity for arsenic adsorption. Some of the filters manufactured using these materials include Sono 3-Kolshi Filter, Granet Home-made Filter, Chari Filter, Adarsha Filter, Shafi Filter, and Bijoypur Clay/Processed Cellulose filters.

Milton et al. (2007) have examined the effectiveness of three-pitcher filters used in Bangladesh. Data were collected at baseline and at 1, 6 and 12 months after the intervention. The study demonstrates that arsenic removal technologies such as three-pitcher filters are an effective option as a short-term measure. However, the three-pitcher filters that are not adequately maintained are not an effective option for long duration. The filter may be even harmful in the long term if the resultant water quality is not properly monitored.

The Garnet home-made filter contains relatively inert materials like brick chips and sand as filtering media. No chemical is added to the system. Air oxidation and adsorption on iron-rich brick chips and flocs of naturally present iron in ground water could be the reason for arsenic removal from ground water. The unit produced inadequate quantity of water and did not show reliable results in different areas of Bangladesh and under different operating

conditions. The Chari filter also uses brick chips and inert aggregates in different Charis as filter media. The effectiveness of this filter in arsenic removal is not known.

The Shafi and Adarshs filters use clay material as filter media in the form of candle. The Shafi filter was reported to have good arsenic removal capacity but suffered from clogging of filter media. The Adarsha filter participated in the rapid assessment program but failed to meet the technical criterion of reducing arsenic to acceptable level (BAMWSP, DFID and Water Aid, 2000). Bijoypur clay and treated cellulose were also found to adsorb arsenic from water (Khair, 2000).

Filter units with cartridges filled with sorptive media or ion-exchange resins are readily available in the market. These units remove arsenic like any other dissolved ions present in water but are not suitable for water having high impurities and iron content in water. Presence of ions having higher affinity than arsenic can quickly saturate the media requiring regeneration or replacement. Two household filters tested at BUET laboratories include Chiyoda Arsenic Removal Unit, Japan, and Coolmart Water Purifier, Korea.

The Chiyoda Arsenic Removal Unit could treat 800 BV meeting the WHO guideline value of 10µg L^{-1} and 1300 BV meeting the Bangladesh Standard of 50µg/L when the feed water arsenic concentration was 300 µg L^{-1}. The Coolmart Water Purifier could treat only 20 L of water with a effluent arsenic content of 25 µg L^{-1} (Ahmed et al., 2000). The initial and operation costs of these units are high and beyond the reach of the rural people.

2.4 OTHER MISCELLANEOUS ADSORBENTS

During recent years, a wide variety of adsorbent systems have been developed for removal of arsenic. Hlavay and Polya´k (2005) prepared and characterized iron hydroxide-coated alumina for the removal of arsenic from drinking water. Andreas et al. (1998) studied the removal of arsenic by adsorption on granulated $Fe(OH)_3$ on a pilot-plant scale and reported a specific load of 1.4 mg g^{-1}. Manganese greensand (Subramanian et al., 1997) and iron oxide-coated sand and ferrihydrite (Thirunavukkarasu et al., 2001) were used for the removal of arsenic from drinking water. Activated carbon (Huang and Fu, 1984), fly ash (Diamadopoulos et al., 1993), modified fly ash (Goswami and Das, 2000), aluminum-loaded coral limestone (Ohki et al., 1996), nanoparticles of hydrous iron oxide (Sylvester et al., 2007), chitosan (Chen and Chung, 2006), chitosan derivatives (Laurent et al., 2002), modified fungal biomass (Pokhrel and Viraraghavan, 2006), iron oxide minerals (Suvasis and Janet, 2003), activated neutralized red mud (Hulya et al., 2004), iron containing mesoporous carbon (Zhimang and Baolin, 2007), natural hematite, magnetite, and goethite (Javier et al., 2007), and iron oxide-impregnated activated carbon (Ronald et al., 2007) were used as adsorbents for the removal of arsenic from aqueous environments.

3. DISCUSSION AND CONCLUSION

Activated alumina has been used for more than two decades to adsorb As(V)-containing waters. The adsorbent does not require regeneration due to its limited life cycle. The optimum pH for arsenic removal with activated alumina is around 6.0; thus raw water usually needs to be pH adjusted by adding mineral acids or CO_2. Activated alumina is ineffective in As(III) adsorption. Dilute caustic soda can be used to rinse and regenerate activated alumina. This regeneration technique may cause problem because it significantly lessens capacity and, more importantly, produces an arsenic rich liquid waste stream.

A new removal technique, which has been developed at Technical University, Berlin (Germany), is adsorption on granular ferric hydroxide in fixed bed reactors. An activated ferric oxide or ferric hydroxide should demonstrate a larger capacity for arsenic removal than activated carbon, as within the coagulation-filtration ferric salts show better removal compared to alum at equal dosage. Furthermore, a granulated ferric hydroxide should be easy to apply as is activated alumina in fixed bed reactors. It is the aim of this technique to combine the advantages of the coagulation-filtration technique, efficiency and small residual mass, with the fixed bed adsorption on activated alumina, simple processing. The high adsorption capacity coupled with the fact that it does not produce contaminated sludge or require chemical dosing, gives it an edge over other arsenic removal technologies. The adsorption technique rely on a simple filtration process over granular adsorbents like activated alumina or granular iron oxides and ferric hydroxides. Chemical dosing is usually not necessary. These techniques do not produce backwash sludge, the actual residual is the arsenic loaded adsorbent.

As a simple filtration process over granular adsorbent media, the technique usually does not require chemical dosing or pH adjustment. The media lasts five to 20 times longer than activated aluminas, and is applied as a non-regenerative media. GEH Wasserchemie GmbH & Co. KG of Germany manufactures and distributes granular ferric hydroxide (GEH^{R}) media. The Siemens Company USFilters is the exclusive North American distributor of the adsorbent, marketing it under the trade name GFH^{TM} media. A number of arsenic treatment plants are in operation in Germany, Japan, the United States and elsewhere within Europe. A large number of hand-pump plants are also operating in West Bengal (India).

Driehaus et al. (1998) reported that approximately 35,000 bed volumes of water containing 16-17 µg L^{-1} of As(V) were filtered by granular ferric hydroxide (GFH) before the effluent arsenic increased to 10 µg L^{-1}. Fe(0) filings, AA, and GFH costs were about $0.20/lb, $0.41/lb, and $4.60/lb, respectively. The results indicate that a cost-effective filtration system can be developed for the removal of arsenic using ZVI. Aeration or/and pH adjustment may be required for treatment of ground water with low DO content and high pH.

Filters with ZVI that are currently in use in Bangladesh (SONO filters) contain a large excess of iron so that sufficient Fe(0) surfaces probably remain after the formation of scales to sustain long-term corrosion. Furthermore, columns with sand and iron operating for weeks to years will most likely be populated with Fe(II)-, Mn(II)-, and As(III)-oxidizing microorganisms that support As(III) oxidation (Katsoyiannis et al., 2004; 2008). The complex reactions in ZVI-filters that are used for months to years are a subject of ongoing studies. Further research should be directed at improving the efficiency and extending the reactive life of ZVI-filters.

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