

A REVIEW PAPER ON ARSENIC REMOVAL FROM DRINKING WATER

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ABSTRACT

Arsenic contamination of land and natural water has become a global problem. Arsenic is one of the most toxic trace elements present in drinking water, has a significant impact on human health. In this review paper, sources and serious health effects of arsenic are mentioned and also arsenic effects in various level of global, national and domestic are discussed. Then few effective methods adopted by various researchers in different parts of world in arsenic treatment are explained with their drawbacks. Thus, finally by overcoming those drawbacks, our methodology of treating arsenic using fly ash is clearly explained.

Keyword: - Arsenic, sources, effects, different methods, fly ash

1. INTRODUCTION

Fly ash is a byproduct from burning pulverized coal in electric power generating plants. During combustion, mineral impurities in the coal (clay, feldspar, quartz, and shale) fuse in suspension and float out of the combustion chamber with the exhaust gases. As the fused material rises, it cools and solidifies into spherical glassy particles called fly ash. Fly ash is collected from the exhaust gases by electrostatic precipitators or bag filters.

Arsenic is an element and is a naturally occurring mineral found widely in the environment. Arsenic exists in four common valence states. Arsenic is widely used commercially, a fact that increases the risk of overexposure. Workers may be overexposed occupationally to arsenic. Inorganic arsenic is generally more toxic than organic arsenic.

2. SOURCES OF ARSENIC

Arsenic is widely distributed throughout the earth's crust. It is introduced into water through the dissolution of minerals and ores, and concentrations in groundwater in some areas are elevated as a result of erosion from local rocks. Industrial effluents also contribute arsenic to water in some areas. Arsenic is also used commercially e.g. in alloying agents and wood preservatives. Also, combustion of fossil fuels is a source of arsenic in the environment through disperses atmospheric deposition. Inorganic arsenic can occur in the environment in several forms but in natural waters, and thus in drinking-water, it is mostly found as trivalent arsenite (As(III)) or pentavalent arsenate (As (V)). Organic arsenic species, abundant in seafood, are very much less harmful to health, and are readily eliminated by the body. Drinking-water poses the greatest threat to public health from arsenic. Exposure at work and mining and industrial emissions may also be significant locally.

3. HEALTH EFFECTS

1. Chronic arsenic poisoning, as occurs after long-term exposure through drinking water is very different to acute poisoning. Immediate symptoms on an acute poisoning typically include vomiting, oesophageal and abdominal pain, and bloody "rice water" diarrhoea. Chelation therapy may be effective in acute poisoning but should not be used against long-term poisoning.

2. The symptoms and signs that arsenic causes appear to differ between individuals, population groups and geographic areas. Thus, there is no universal definition of the disease caused by arsenic. This complicates the assessment of the burden on health of arsenic. Similarly, there is no method to identify those cases of internal cancer that were caused by arsenic from cancers induced by other factors.

3. Long-term exposure to arsenic via drinking-water causes cancer of the skin, lungs, urinary bladder, and kidney, as well as other skin changes such as pigmentation changes and thickening (hyperkeratosis).

4. Increased risks of lung and bladder cancer and of arsenic-associated skin lesions have been observed at drinking-water arsenic concentrations of less than 0.05 mg/L.

5. Absorption of arsenic through the skin is minimal and thus hand-washing, bathing, laundry, etc. with water containing arsenic do not pose human health risk.

6. Following long-term exposure, the first changes are usually observed in the skin: pigmentation changes, and then hyperkeratosis. Cancer is a late phenomenon, and usually takes more than 10 years to develop.

7. The relationship between arsenic exposure and other health effects is not clear cut. For example, some studies have reported hypertensive and cardiovascular disease, diabetes and reproductive effects.

8. Exposure to arsenic via drinking-water has been shown to cause a severe disease of blood vessels leading to gangrene in China (Province of Taiwan), known as 'black foot disease'. This disease has not been observed in other parts of the world, and it is possible that malnutrition contributes to its development. However, studies in several countries have demonstrated that arsenic causes other, less severe forms of peripheral vascular disease.

9. According to some estimates, arsenic in drinking-water will cause 200,000 -- 270,000 deaths from cancer in Bangladesh alone (NRC, 1998; Smith, et al, 2000).

4. WHO'S ACTIVITIES IN ARSENIC

WHO's norms for drinking-water quality go back to 1958. The International Standards for Drinking-Water established 0.20 mg/L as an allowable concentration for arsenic in that year. In 1963 the standard was re-evaluated and reduced to 0.05 mg/L. In 1984, this was maintained as WHO's "Guideline Value"; and many countries have kept this as the national standard or as an interim target. According to the last edition of the WHO Guidelines for Drinking-Water Quality (1993):

1. Inorganic arsenic is a documented human carcinogen.
2. 0.01 mg/L was established as a provisional guideline value for arsenic.
3. Based on health criteria, the guideline value for arsenic in drinking-water would be less than 0.01 mg/L.
4. Because the guideline value is restricted by measurement limitations, and 0.01 mg/L is the realistic limit to measurement, this is termed a provisional guideline value.

Information on arsenic in drinking-water on a country-by-country basis is being collected and will be added to the UN report and made available on the web site. As part of WHO's activities on the global burden of disease, an estimate of the disease burden associated with arsenic in drinking-water is in preparation. A report entitled "Towards an assessment of the socioeconomic impact of arsenic poisoning in Bangladesh" was released in 2000.

A United Nations Foundation grant for 2.5 million approved in July 2000, will enable UNICEF and WHO to support a project to provide clean drinking-water alternatives to 1.1 million people in three of the worst affected sub-districts in Bangladesh. The project utilizes an integrated approach involving communication, capacity building for arsenic mitigation of all stakeholders at sub district level and below, tube-well testing, patient management, and provision of alternative water supply options.

5. GLOBAL EFFECTS

The delayed health effects of exposure to arsenic, the lack of common definitions and of local awareness as well as poor reporting in affected areas are major problems in determining the extent of the arsenic-in-drinking-water problem. Reliable data on exposure and health effects are rarely available, but it is clear that there are many countries in the world where arsenic in drinking-water has been detected at concentration greater than the Guideline Value, 0.01 mg/L or the prevailing national standard. These include Argentina, Australia, Bangladesh, Chile, China, Hungary, India, Mexico, Peru, Thailand, and the United States of America. Countries where adverse health effects have been documented include Bangladesh, China, India (West Bengal), and the United States of America. Examples are:

1. Seven of 16 districts of West Bengal have been reported to have ground water arsenic concentrations above 0.05 mg/L; the total population in these seven districts is over 34 million (Mandal, et al, 1996) and it has been estimated that the population actually using arsenic-rich water is more than 1 million (above 0.05 mg/L) and is 1.3 million (above 0.01 mg/L) (Chowdhury, et al, 1997).

2. According to a British Geological Survey study in 1998 on shallow tube-wells in 61 of the 64 districts in Bangladesh, 46% of the samples were above 0.010 mg/L and 27% were above 0.050 mg/L. When combined with the estimated 1999 population, it was estimated that the number of people exposed to arsenic concentrations above 0.05 mg/l is 28-35 million and the number of those exposed to more than 0.01 mg/l is 46-57 million (BGS, 2000).

3. Environment Protection Agency of The United States of America has estimated that some 13 million of the population of USA, mostly in the western states, are exposed to arsenic in drinking- water at 0.01 mg/L, although concentrations appear to be typically much lower than those encountered in areas such as Bangladesh and West Bengal. (USEPA, 2001)

Table 1. Arsenic exposure concerns worldwide.

Country	Estimated exposed population (millions) ^a	Arsenic concentration in drinking water (µg/L)	References
Argentina	2.0	< 1 to 7,550	Bates et al. 2004; Moore et al. 2004; Stein Maus et al. 2010
Bangladesh	35–77	< 10 to > 2,500	Keansburg and Smedley 2001
Chile	0.4	600 to 800	Faurecia et al. 2000; Smith et al. 1998, 2000a
China	0.5–2.0	< 50 to 4,400	Yu et al. 2007
Ghana	< 0.1	< 2 to 175	Asante et al. 2007; Smedley 1996
India	> 1.0	< 10 to > 800	Acharya et al. 1999
Mexico	0.4	5 to 43	Calderón et al. 2001; Camacho et al. 2011; Meza et al. 2004, 2005
Taiwan	NA	< 1 to > 3,000	Chen et al. 2010a, 2010b
United States	> 3.0	< 1 to > 3,100	Anning et al. 2012; Ayotte et al. 2003; Burgess et al. 2007; Nielsen et al. 2010; NRDC 2000; Peters 2008; Sanders et al. 2012; Thandie et al. 2007; Xu et al. 2010
Vietnam	> 3.0	< 0.1 to 810	Winkle et al. 2011

6. NATIONAL LEVEL EFFECTS

Arsenic was a very rarely heard term in the water sector till a few decades back. However, in recent years, the number of areas reporting arsenic contamination have gone up drastically with over 20 countries from different parts of the world reporting arsenic contamination of groundwater (Bordello, 2012). With the constantly increasing number of occurrences, especially in the South Asian region, it is now recognized as a major public health concern affecting a large number of people around the world.

In South Asia, arsenic contamination in groundwater in the Ganga- Brahmaputra fluvial plains in India and Padma-Meghna fluvial plains in Bangladesh has been found to have a huge impact on human health and its consequences have been reported as the world's biggest natural groundwater calamities. In India, West Bengal, Jharkhand, Bihar, Uttar Pradesh in the flood plains of the Ganga, Assam and Manipur in the flood plains of the Brahmaputra and Imp Hal rivers and Rajnandgaon village in Chhattisgarh state have been reported to be affected by arsenic contamination in groundwater.

Table 2. State-wise Districts with Arsenic Contaminated Groundwater (2013)

States	Arsenic contaminated districts		Total no of districts in a state
	Number	Proportion %	
Andhra Pradesh	-	-	23
Arunachal Pradesh	-	-	16
Assam	1	4	27
Bihar	15	39	38

Chhattisgarh	1	6	18
Delhi	-	-	9
Gujarat	-	-	26
Haryana	-	-	21
Jammu & Kashmir	-	-	22
Jharkhand	-	-	24
Karnataka	-	-	30
Kerala	-	-	14
Madhya Pradesh	-	-	50
Maharastra	-	-	35
Odisha	-	-	30
Punjab	-	-	20
Rajasthan	-	-	33
Tamil Nadu	-	-	32
Uttar Pradesh	9	13	71
West Bengal	8	42	19
Total	34	6	558

7. EFFECTS OF ARSENIC IN TAMILNADU

Now-a-days heavy metal arsenic poses a health risk problem throughout the world. Arsenic may be found in water which has flowed through arsenic-rich rocks. Severe health effects have been observed in populations drinking arsenic-rich water over long periods in countries world-wide.

A study of this kind involves the accurate measurement of arsenic in drinking-water at levels relevant to health requires laboratory analysis, using sophisticated and expensive techniques and facilities. Analytical quality control and external validation remains problematic in the absence of well trained staff in many parts of the world. Furthermore, field test kits can detect high levels of arsenic but are typically unreliable at lower concentrations of concern for human health. The reliability of field methods is yet to be fully evaluated.

As a part of the study about 412 groundwater samples were collected in the entire state of Tamil Nadu. These samples were analysed using sophisticated Atomic Absorption Spectrophotometer (Perkin Elmer) using expensive techniques of Mercury Hydride System (MHS) with a well-trained and skilled staff, since arsenic was present in the groundwater sample in very low quantity.

The data were compiled district-wise and the range of arsenic present in the groundwater samples (microgram/litre) are given in the statement. As per Bureau of Indian Standards (BIS), the maximum permissible limit of arsenic in the drinking water is 0.05 mg/L (50 µg/L).

As per the study, from the data it is seen that arsenic present in the groundwater sample of Tamil Nadu within the safe limit of 0.05 mg/L (50 µg/L) as fixed by the Bureau of Indian Standards for the drinking water. However the highest amount of arsenic 8.62, 16.67 and 11.5 µg/L have been noticed in the districts of Kancheepuram, Preamble and Virudhunagar respectively. In Theni and Erode district the lowest amount of Arsenic 1.00 µg/L have been noticed.

8. VARIOUS METHODOLOGIES ADOPTED IN ARSENIC TREATMENT

8.1. Arsenic removal from drinking water using iron oxide coated sand by O.S.Thirunavukkarasu, T.Viraraghavan, and K.S.Subramanian.

This article describes experiments in which iron oxide-coated sand (IOCS) was used to study the removal of both As(V) and As(III) to a level less than 5 µg L⁻¹ in drinking water. Iron oxide-coated sand 2 (IOCS-2) prepared through high temperature coating process was used in batch and column studies to assess the effectiveness and suitability. The isotherm study results showed that the observed data fitted well with the Langmuir model, and the adsorption maximum for IOCS-2 at pH 7.6 was estimated to be 42.6 and 41.1 µg As g⁻¹ IOCS-2 for As(V) and As(III), respectively. In the fixed bed column tests to study arsenic removal from the tap water, good performance of IOCS-2 was observed in respect of bed volumes achieved and arsenic removal capacity. Five cycles of column tests were conducted to evaluate the performance of IOCS-2, and arsenic was

successfully recovered from the media through regeneration and backwash operations. High bed volumes (860 to 1403) up to a breakthrough concentration of $5 \mu\text{g L}^{-1}$ were achieved in the column studies with tap water, and the bed volumes achieved in the studies with natural water (containing arsenic) were 1520. The results of both the batch and column studies showed that iron oxide-coated sand filtration could be effectively used to achieve less than $5 \mu\text{g L}^{-1}$ As in drinking water.

8.1.1. Drawbacks

This method involves high temperature for making iron coated sand bed and also requires large investment of money. The adsorption of arsenic is not fully done and also there are chances of seeing some traces of chemicals in final obtained water.

8.2. Arsenic removal from drinking water by flocculation and Microfiltration by Binbing Hana, Timothy Runnellsb, Julio Zimbronb, Ranil Wickramasinghea

Arsenic removal from drinking water is a major problem in many parts of the world. We have investigated arsenic removal by flocculation and microfiltration. Ferric chloride and ferric sulphate have been used as flocculants. The use of small amounts of cationic polymeric flocculants, as flocculation aids in the presence of ferric ions, has also been investigated. The results obtained here show that flocculation prior to microfiltration leads to significant arsenic removal in the permeate. Further, the addition of small amounts of cationic polymeric flocculants lead to significantly improved permeate fluxes during microfiltration. The residual turbidity, after flocculation and microfiltration, may be used as a guide to the level of arsenic removal. Since energy requirements for microfiltration are low and fluxes high, compared to other membrane processes such as reverse osmosis and nanofiltration, flocculation and microfiltration may be a cost-effective method for arsenic removal from drinking water.

8.2.1. Drawbacks

Removal of arsenic by flocculation and microfiltration depends on the effectiveness of arsenic adsorption onto the ferric complexes present and on the rejection of the arsenic containing flocs formed by the membrane. The larger the membrane pore size, the lower the resistance to filtrate flow and hence the lower the pumping costs for a given filtrate flow rate. However, the larger the pore size the larger the number of smaller flocs that are able to pass through the membrane into the filtrate.

8.3. Effects of water chemistry on arsenic removal from drinking water by electrocoagulation by Wei Wan, Troy J. Pepping, Tuhin Banerji, Sanjeev Chaudhari, Daniel E. Giammar

Exposure to arsenic through drinking water poses a threat to human health. Electrocoagulation is a water treatment technology that involves electrolytic oxidation of anode materials and in-situ generation of coagulant. The electrochemical generation of coagulant is an alternative to using chemical coagulants, and the process can also oxidize As(III) to As(V). Batch electrocoagulation experiments were performed in the laboratory using iron electrodes. The experiments quantified the effects of pH, initial arsenic concentration and oxidation state, and concentrations of dissolved phosphate, silica and sulphate on the rate and extent of arsenic removal. The iron generated during electrocoagulation precipitated as lepidocrocite (g-FeOOH), except when dissolved silica was present, and arsenic was removed by adsorption to the lepidocrocite. Arsenic removal was slower at higher pH. When solutions initially contained As(III), a portion of the As(III) was oxidized to As(V) during electrocoagulation. As(V) removal was faster than As(III) removal. The presence of 1 and 4 mg/L phosphate inhibited arsenic removal, while the presence of 5 and 20 mg/L silica or 10 and 50 mg/L sulphate had no significant effect on arsenic removal. For most conditions examined in this study, over 99.9% arsenic removal efficiency was achieved. Electrocoagulation was also highly effective at removing arsenic from drinking water in field trials conducted in a village in Eastern India. By using operation times long enough to produce sufficient iron oxide for removal of both phosphate and arsenate, the performance of the systems in field trials was not inhibited by high phosphate concentrations.

8.3.1. Drawbacks

Arsenic removal by electrocoagulation involved lepidocrocite formation followed by arsenic adsorption. As removal was slower at higher pH and higher initial arsenic concentrations. As(III) was partially Oxidized to As(V) during electrocoagulation. As(V) removal was faster than As(III) removal.

8.4. A Hybrid Sorbent Utilizing Nanoparticles of Hydrous Iron Oxide for Arsenic Removal from Drinking Water by P. Sylvester, Westerhoff, T.Möller, M. Badruzzaman and O. Boyd

Arsenic in drinking water, present primarily as inorganic As(III) (arsenite) or As(V) (arsenate), has been linked to a number of human ailments. This has prompted the United States Environmental Protection Agency (U.S. EPA) to lower the maximum contaminant level for arsenic in drinking water from 50 to 10 $\mu\text{g/L}$. ArsenXnp was developed to satisfy the need for an efficient, reliable, and reusable material to meet this new arsenic standard. ArsenXnp is a hybrid sorbent consisting of nanoparticles of hydrous iron oxide distributed throughout a porous polymeric bead. This combines the excellent handling, flow characteristics and attrition resistance of conventional ion exchange resins with the well-documented arsenic affinity of hydrous iron oxides. In field tests, the resin maintained an effluent arsenic concentration below 10 $\mu\text{g/L}$ for 4 months of continuous operation with no mechanical problems and no pressure build up across the resin bed. Arsenic is removed from solution due to the interaction with the nanoscale hydrous iron oxide surfaces rather than the anion exchange groups associated with the polymeric substrate, and the arsenic capacity is unaffected by common anions such as sulfate, chloride, or bicarbonate.

8.4.1. Drawbacks

Accurate measurement of the surface area of ArsenXnp is not possible, since dewatering of the resin (a necessary step prior to surface area and porosity measurements) results in shrinkage of the polymeric resin bead and distortion of the porous network. The chemical form of the embedded hydrous iron nanoparticles will also be impacted due to loss of surface water and hydroxyl groups.

8.5. A method for preparing ferric activated carbon composites adsorbents to remove arsenic from drinking water by Qiao Li Zhang, Y.C. Lin, X. Chen, Nai Yun Gao

Iron oxide/activated carbon (FeO/AC) composite adsorbent material, which was used to modify the coal-based activated carbon (AC) 12 \times 40, was prepared by the special ferric oxide microcrystal in this study. This composite can be used as the adsorbent to remove arsenic from drinking water, and Langmuir isotherm adsorption equation well describes the experimental adsorption isotherms. Then, the arsenic desorption can subsequently be separated from the medium by using a 1% aqueous NaOH solution. The apparent characters and physical chemistry performances of FeO/AC composite were investigated by X-ray diffraction (XRD), nitrogen adsorption, scanning electron microscopy (SEM) and X-ray photoelectron spectroscopy (XPS). Batch and column adsorption experiments were carried out to investigate and compare the arsenic removal capability of the prepared FeO/AC composite material and virgin activated carbon. It can be concluded that: (1) the main phase present in this composite are magnetite (Fe_3O_4), maghemite ($\gamma\text{-Fe}_2\text{O}_3$), hematite ($\alpha\text{-Fe}_2\text{O}_3$) and goethite ($\gamma\text{-FeO(OH)}$); (2) the presence of iron oxides did not significantly affect the surface area or the pore structure of the activated carbon; (3) the comparisons between the adsorption isotherms of arsenic from aqueous solution onto the composite and virgin activated carbon showed that the FeO/AC composite behave an excellent capacity of adsorption arsenic than the virgin activated carbon; (4) column adsorption experiments with FeO/AC composite adsorbent showed that the arsenic could be removed to below 0.01 mg/L within 1250mL empty bed volume when influent concentration was 0.5 mg/L.

8.5.1. Drawbacks

The efficiency of arsenic removal is presented by breakthrough curves, which show the concentration ratios (C/C_0) is a function of throughput volumes (C is the instantaneous concentration of effluents and C_0 is the initial concentration of influent). Which is not constant throughout the process and the results may vary at any time.

9. OUR METHODOLOGY OF REMOVING ARSENIC(III) FROM DRINKING WATER USING FLY ASH AGGLOMERATES

Batch experiments of arsenic(III) adsorption from drinking water will be carried out on the fly ash agglomerates of the various sizes. The fly ash from the Mettur Thermal Power Plant will be brought to the laboratory and stored. The fly ash chemical composition and the presence of various minerals will be studied using the X-ray diffraction. The particle size analysis will be undertaken using a laser diffractometer. The

density of fly ash will be determined using a pycnometer. The surface area will be measured by Brunauer-Emmett-Teller (BET) method for helium/nitrogen mixture using Flow Sorb II apparatus for fly ash powder. Tumble agglomeration experiments will be conducted by taking 200g- 250g of fly ash samples and the agglomeration process will be carried out for 1 hour. The wet agglomerates will be cured in a curing chamber at room temperature for one week to fulfil the hydration of cementitious components. For different fractions of agglomerates the mechanical strength will be conducted on the Universal Testing Machine(UTM). Adsorption experiments to know the effect of pH will be studied with initial arsenic(III) concentration of 500 mg/l and with lesser dose of adsorbent. The contact time will be 24 hours and the experiments will be executed at room temperature (25°C). The initial solution pH will be adjusted by using 0.1 M HCL and 0.1 M NaOH. The uptake of arsenic(III) by fly ash agglomerates will be determined for the known adsorbent/arsenic ratios. And finally, to optimize the adsorption conditions for arsenic(III) removal from drinking water by fly ash agglomerates, isotherm and kinetic studies will be performed. In order to validate the lab scale batch experiments, result using the lab scale optimised parameters, the batch experiments will be conducted for scaled up process and the results will be documented. Finally, upon obtaining the concurrent results of adsorption studies from the lab scale and scaled up process, a water purification column will be fabricated for the optimised adsorption parameters.

10. CONCLUSION

Thus, understanding the various ill effects of Arsenic (III) in drinking water, various treatment methods have been carried out by various researchers and better results have been found out. Apart from those methods, our method of removing arsenic(III) by using fly ash agglomerates involves cheap investment, better results, and mainly involving the waste by product from thermal power plants.

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