

Comparative Studies for Selection of Technologies for Arsenic Removal from Drinking Water

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Abstract

This paper present the possible alternative removal options for the development of safe drinking water supply in the arsenic-affected areas. Each alternative option was studied in the Environmental Engineering Laboratory at Indian Institute of Technology (IIT) Kharagpur, India. In this paper conventional precipitation (Alum and Iron salt) and adsorption (With various types of adsorbent) method were used for arsenic removal. Various parameters such as, coagulant dose, pH, anions concentration and reaction time were studied to establish optimum conditions. Iron salt as a coagulant and Hydrous Granular Ferric Oxide as a fixed bed adsorbent were found to be effective options for arsenic removal.

INTRODUCTION

The occurrence of geogenic As(III) and As(V) in ground water is a major problem in Bangladesh. A major part of village people are suffering from arsenicosis due to consuming arsenic contaminated water. About 80 million people in 59 districts out of 64 districts poses a serious health threat (BGS, 1999). Numerous recent investigations have indicated that arsenic constitutes a serious health risk at different places and it has been confirmed by the medical studies (Saha, 1999). Ingestion via food or water is

the main pathway of this metalloid in the organism where its absorption takes place in the stomach and intestines, followed by its release into the blood stream. Arsenic is then converted by the liver to a less toxic form, which is eventually largely excreted in the urine (Caroli et al., 1996). Due to the illness of people the nation is loosing million and million of manpower-hour as well as impoverish strength, knowledge, economy, development and finally it begins to kill slowly and painfully.

The removal technology of arsenic and disposal of arsenic bearing waste presents a challenging task to the environmental engineers. Unlike organic waste, inorganic arsenic cannot be degraded biologically to harmless products. The chemistry of arsenic is quite complex and interesting, as it can be stable in four oxidation states, continue changing its states and its removal is dependent on pH of the medium, oxidation state and redox potential.

Recent publications discussing methods of removing arsenic from water are too numerous to mention. A comprehensive search of literature by reference to abstracts and articles, followed by computer search, reveals that aqueous arsenic removal techniques fall into four major categories – chemical precipitation, ion exchange, membrane processes and adsorption.

Since contamination of drinking water due to arsenic compound is a severe problem with regards to health hazards and in this connection a lot of research has been undertaken in this field to remove arsenic from contaminated water using different technique. But most of them were found to be less effective for removing arsenic and many drawbacks when applied in the field. It is very difficult to select a unique method for arsenic removal. Some are effective but economically not feasible, some are economically feasible but are not effective. Some are not user friendly, technologically not sound, energy dependent, post treatment required, skill manpower required, quality of treated water in respect to other parameter in water not maintaining with standard. It is difficult to the meet stringent arsenic standard as well as drinking water quality standard for different parameter and other drawbacks is still a great problem. Even specific information on the major factors affecting arsenic removal is also still incomplete.

Complexities of Arsenic Removal

The following are the various difficulties arise at the removal of arsenic when applied in the field:

- Wide range of initial concentration of arsenic in water, other elements and its variation of concentration in water, optimization of right dose for generalization, filtration of treated water, adjustment of pH in water, analysis of arsenic specially by field test kit method, post treatment difficulties, handling of waste and proper operation and maintenance.

- It is apparent that selection of an arsenic removal method is a really complex decision and the method of choice changes depending on the oxidation state

of arsenic and a number of other factors. There are many technologies, which are successful in the laboratory but in the field condition they do not working properly.

Best Available Technology for Removal of Arsenic

Selection of arsenic removal technology can be done by best available technology (BAT) method, which the administrator finds after examination for efficacy under field conditions and not solely under laboratory conditions is available (taking cost into consideration). BAT can be determined by identifying available technologies, which reduce contaminant concentration levels and by evaluating the costs and commercial availability of the technology. Environmental Protection Agency (EPA) uses required and large metropolitan water systems to serve the basis for making the cost determination for BAT.

In this system, the cost can be measured by the increase in the annual household water bill due to the installation of a technology under consideration.

The arsenic removal performance data for all the treatment technologies could be evaluated to determine the best technologies that should be designated as BAT.

BAT can be designated based upon the following criteria:

- High removal efficiency
- Affordability (using large system as the basis)
- General geographic applicability
- Compatibility with other water treatment processes, and
- Process reliability

MATERIALS

Glassware and apparatus

All glassware used in the present study were manufactured by M/S Bhattacharya Co. Ltd. (Calcutta, India) and marked under brand name 'Borosil'. 150 ml capacity special types of arsine generator apparatus were used throughout the experiment for determination of total arsenic. All glassware except arsine generator apparatus were cleaned by soaking in 10% HCl for 24 hours followed by washing with dilute soap solution (Rankleen, Ranbaxy Laboratories Ltd., Punjab, India), tap water and then distilled water. Arsine generation apparatus were cleaned by soaking 24 hours in 8M nitric acid followed by other method as mentioned.

Water

Distilled water was obtained by distilling tap water in glass distillation apparatus. All the working arsenic solutions were prepared in freshly prepared double distilled water unless otherwise mentioned. The pH of the distilled water was around 6.9.

Chemicals

All chemicals were of analytical reagent grade and were used without further purification. All solutions were prepared using double distilled water (Standard method, 1989). The arsenite As(III) stock solution was prepared by NaAsO₂ (E Merck, Germany) in double distilled water from 1000 mg/l to 100 mg/l. The arsenate As(V) stock solution was prepared from sodium salt heptahydrate, Na₂HAsO₄·7 H₂O E-Merck, (Germany) dissolved in double distilled water. Prior to each analysis, intermediate standard As(III) and As(V) solutions were diluted with double distilled water to 10 mg/l from the arsenic stock solutions. Secondary standard solutions 1 mg/l were also freshly prepared for each experiment from the 10 mg/l intermediate stock solutions to 1 mg/l. Stock solutions of different element were prepared from metal nitrate salts. These solutions were diluted to prepared working solutions.

Analysis of water samples

Total arsenic content in water sample was measured spectrophotometrically using Silverdiethyldithiocarbamate (SDDC) method. (Model Shimadzu Spectrophotometer UV-160A). Graphite furnace AAS method was used for cross-check at lower concentration of arsenic and other metal.

EXPERIMENTAL METHOD

Precipitation Method

Among them arsenic removal by chemical precipitation with aluminium or iron hydroxides is the best-known and most frequently applied technique. At present many countries successfully using the conventional method of arsenic removal. The treatment process is consists of coagulation followed by flocculation, sedimentation and filtration.

Removal of arsenic by alum (aluminium sulphate) and iron salt (ferric sulphate) from drinking water were studied separately in the laboratory as well as in the field. To avoid chlorine natural sunlight was used as an oxidizing agent to convert arsenite to arsenate. Alum and iron salt were added in the arsenic contaminated water in the range of 30-75mg/l and 20-50 mg/l respectively. Concentration of arsenic in water was maintained in the range 0.1 to 1.0 mg/l.

The findings of different studies are briefly presented below:

- Oxidation of arsenite to arsenate by sunlight is a very slow process. An investigation of precipitation methods indicates that the removal of arsenic by coagulation is a function of the oxidation state of the arsenic, the type and dose of coagulant, the pH of the water and the initial arsenic concentration.
- Iron salt is more preferable than alum. Because iron is more soluble than aluminum in water. When ferric coagulant are added, all the iron forms particulate $\text{Fe}(\text{OH})_3$. However, not all aluminum added as alum coagulant precipitates as $\text{Al}(\text{OH})_3$. Because only particulate metal hydroxides can mediate arsenic removal, alum plants must carefully considered all solubility when improved arsenic removal is desired. 92% removal was achieved using 20 mg/l of alum in 0.1 mg/l of arsenic in water at pH 6.6 and 96% removal was achieved using 10 mg/l ferric sulphate in 0.1 mg/l of arsenic in water at neutral pH range with 6 hour retention time. Doses of coagulant increase with increasing concentration of arsenic in water.

Adsorption Method

Screening of Existing Adsorbents

Arsenic can be removed by adsorption onto many adsorbent materials. Some of adsorbent materials are very costly and some are less effective. The criteria for selection of suitable adsorbent include: the cost of the medium, the ease of operation or handling, the cost of operation, the useful service life per cycle / the adsorption capacity of the adsorbent, the potential of reuse, the number of useful cycles and the possibilities of regeneration of adsorbent.

For selecting an appropriate adsorbent for removal of arsenic, a number of available adsorbents such as Kimberlite tailing, wood charcoal, banana pith, coal fly ash, spent tea leaf, mushroom, saw dust, rice husk, sand, water hyacinth, activated carbon, bauxite, hematite, laterite, iron-oxide coated sand, activated alumina, CaSiCo and Hydrous Granular Ferric Oxide were evaluated for arsenic removal. The adsorbents were compared on the basis of percentage removal of As(III) and As(V) with activated alumina being the reference. Preliminary screening of the adsorbents was made through the batch sorption screening test with 6 hr contact time. The results are presented in Table 1.

Table 1: Arsenic removal efficiency of different adsorbent materials.

Adsorbent	Dose (g/l)	% Removal	
		As(III)	As(V)
1. Kimberlite tailing	10	25	40
2. Water hyacinth	10	45	70
3. Wood charcoal	10	19	37
4. Banana pith	10	12	18
5. Coal fly ash	10	20	28
6. Spent tea leaf	10	25	42
7. Mushroom	10	22	35
8. Saw dust	10	28	36
9. Rice husk ash	10	5	12
10. Sand	10	15	22
11. Activated carbon	10	50	65
12. Bauxite	10	58	80
13. Hematite	10	40	60
14. Laterite	10	45	70
15. Iron-oxide coated sand	10	72	90
16. Activated alumina	10	90	96
17. CalSiCo	5	90	98
18. Hydrous granular ferric oxide	2	92	99

Most of the adsorbent were found poor arsenic removal but iron-oxide coated sand, activated alumina, CalSiCo and Hydrous Granular Ferric Oxide showed a considerable amount of arsenic removal.

Three adsorbent named iron oxide coated, CalSiCo and Hydrous granular ferric oxide were prepared in the laboratory and activated alumina was collected from the market. All the four adsorbent were evaluated its capacity to adsorb arsenic under different conditions using synthetic As(III) and As(V) solutions. The experiments were conducted in three phases: (i) batch studies, (ii) column studies, (iii) desorption and regeneration studies.

In batch (static) studies, apart from kinetic and isotherm studies, the effects of different parameters such as pH, adsorbent dose, adsorbent size, presence of chloride, sulphate, nitrate, chromate, calcium, magnesium, and iron were studied. All the above parameters were varied one at a time to study their effect on the adsorption process. Interruption test was conducted to determine the rate-limiting step by kinetic data; by

the effect of initial adsorbate concentration and adsorbent size on removal rate; and by multiple interruption test. Effect of anionic ligands such as EDTA, fluoride and phosphate on the removal of As(III) and As(V) was also studied. All the above parameters taken together were varied to study their combined effect on removal efficiencies.

Column (dynamic) studies were conducted to ascertain the engineering aspect of arsenic removal by CalSiCo. The experiments were carried out with distilled and tap water spiked with 1 mg/l of arsenite and 1 mg/l of arsenate separately. A glass column of 15 mm diameter having 220 mm bed depth of all the adsorbent about 0.2 mm geometric mean size was used. The flow of influent was maintained at 10 ml/min for all the adsorbents.

Desorption and regeneration studies were conducted to investigate the possibility of reuse of spent adsorbent. Various eluents such as distilled water, NaOH, NaHCO₃, Na₂CO₃, and H₂O₂ in HNO₃ were used in batch experiment. The regeneration study was conducted for 3 cycles. H₂O₂ in HNO₃ for CalSiCo and NaOH for rest three were used as an eluent in the dynamic desorption studies carried out for three cycles.

EXPERIMENTAL RESULTS AND DISCUSSION

It was found that all the four iron-oxide coated sand, activated alumina, CalSiCo and Hydrous Granular Ferric Oxide adsorbent could be effectively used as media for the removal of arsenic from ground water. Removal was found to be 90, 96, 98 and 99% respectively for As(V) with initial concentration of 1 mg/l and around 85% for As(III) with initial concentration of 1 mg/l in neutral pH range. Activated alumina, CalSiCo and Hydrous Granular Ferric Oxide could be used for the selective removal of arsenic from water environment upto certain extent over other ions, such as Cl⁻, SO₄²⁻, PO₄³⁻, NO₃⁻, F⁻, CrO₄²⁻, Ca²⁺, Mg²⁺ and Fe²⁺. The overall reaction kinetics could be represented by first order reaction equation. Film diffusion was the rate-limiting step for arsenic removal as indicated by kinetic data; by the effect of initial adsorbate concentration and adsorbent size on removal rate; and by multiple interruption test. The equilibrium adsorption data fitted to Langmuir and Freundlich isotherm models. However, the Langmuir equation gave better fit. Arsenic removal by these adsorbent was found pH dependent. In drinking water neutral pH range, it exhibited excellent arsenic removal. The best arsenic removal was achieved at pH 7.2 for As(V) and above 9 for As(III) for CalSiCo and 7.1-7.5 for others. Removal achieved was typically from 1 mg/l to 0.01-0.05 mg/l. The removal of As(V) was easier than that of As(III), so As(III) could be converted to As(V) by adding oxidising agent for better removal. Removal efficiency decreased with the presence of phosphate (more than 10 mg/l), fluoride (more than 2 mg/l) and EDTA (more than 0.01 moles/l) but the presence of nitrate, sulphate, chloride, chromate, calcium, magnesium and iron did not affect removal significantly. Spent adsorbent could be regenerated with 15% H₂O₂ in

0.2 M HNO₃ for CalSiCo and 1N NaOH for iron-oxide coated sand, activated alumina and Hydrous Granular Ferric Oxide.

The adsorbent coefficients determined by Logit method could be used successfully to predict the breakthrough of another column.

Table 2 : Results of mini-column study in the field at the 20th day.

Parameter	Influent Concentration, (mg/l)	Effluent concentration for CalSiCo, (mg/l)	Effluent concentration for Hydrous Granular Ferric Oxide , (mg/l)
Hardness	145	135	140
Cl ⁻	8	8	8
SO ₄ ²⁻	5	4	3
NO ₃ ⁻	0	0	0
F ⁻	0	0	0
PO ₄ ³⁻	0	0	0
Ca	26	25	24
Mg	5	4	4
Na	10	10	8
Fe	4.61	0.3	0.2
As	0.32	0.015	0.01

Table 2 shows the performance of CalSiCo and Hydrous Granular ferric Oxide were evaluated for its efficiency in removing arsenic in the actual field situation by means of a pilot plant study. Two 30 cm long mini-column of 40 mm diameter each containing 250 gm of CalSiCo and Hydrous Granular ferric Oxide (15 and 18 cm bed depth respectively) was installed at Deganga (North 24-Parganas District) and was run continuously for 20 days at a flow rate of 20 ml/min containing 0.32 mg/l of arsenic. It was found that the columns were able to supply 28 l/day for CalSiCo and 45 l/day for Hydrous Granular ferric Oxide of treated water containing arsenic less than 0.015 mg/l for CalSiCo and less than 0.01 Hydrous Granular ferric Oxide for 20 days. It clearly demonstrates that these adsorbents can be used as a filter media for removing arsenic from ground water in arsenic-affected areas of Bangladesh and West Bengal, India. A 20-liter capacity home water purifier for arsenic was developed for community as well as individual family, which should be subjected to a field trial to assess the long-term performance of the medium.

CONCLUSIONS

Bangladesh and West Bengal (India) are now facing tremendous difficulties in

removing arsenic from the ground water. It is apparent that selection of an arsenic removal method is a really complex which continuously changes its form of oxidation states of arsenic and it is dependent on many factors. Conventional coagulation method of arsenic removal using iron salt is a prefer option in many respect. Hydrous Granular ferric Oxide was found best for arsenic removal from contaminated drinking water as a fixed bed adsorbent. Iron salt and Hydrous Granular ferric Oxide will be able to meet national drinking water standards.

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