Iron-Coated Sponge as Effective Media to Remove Arsenic from Drinking Water

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Arsenic (As) contamination in drinking water is a serious problem in a number of countries in the world, especially in small communities and developing countries. Arsenic is related to many health diseases. Several technologies such as coagulation, adsorption, ion exchange and membrane processes, etc., are used in removing arsenic from water. In this study, a new material, namely iron-coated sponge (IOCSp), was developed and used to remove As, and it was found that IOCSp has a high capacity for removing both As(V) and As(III). Each gram of IOCSp adsorbed about 160 µg of As upon a 9-h contact of IOCSp with As solution. A dynamic filter column conducted showed that even a small quantity of IOCSp (8 g) could reduce As from 156 µg/L to a concentration of less than 50 µg/L while treating 75 L of groundwater contaminated with arsenic. Both the filtration rate and the size of the sponge had an effect on effluent quality, and the amount of water treated to the acceptable quality.

Key words: arsenic, drinking water, iron-coated sponge (IOCSp), water treatment technology

Introduction

A number of natural and human activities are responsible for the arsenic (As) contamination of groundwater sources. Arsenic contamination of the subsurface water is believed to be geological and it may result from dissolution of, or desorption from, iron oxide and oxidation of arsenic pyrites (Thirunavukkarasu et al. 2001). Arsenic is the major constituent of a number of minerals. These minerals are mostly ores containing sulfide, along with copper, nickel, lead, cobalt and other minerals. The possible As contamination could be from mine tailings (wastewater from exploring gold, copper and zinc ore, etc.), industrial waste discharges, fertilizers, pesticides/insecticides, smelting of metals and burning of fossil fuels (Altundogan et al. 2000; Thirunavukkarasu et al. 2001).

Arsenic can exist in several oxidation states (-III, 0, +III and +V) in the environment. However, in natural water As is mostly found in inorganic form as oxanionns of trivalent arsenite [As(III)] or pentavalent arsenate [As(V)]. pH is the major factor controlling the differences in aqueous As speciation in freshwater (Cullen et al. 1989).

The toxicity of As highly depends on its speciation. The toxicity of arsenite is 25 to 60 percent higher than that of arsenate and toxicity decreases in the order of inorganic As(III) > organic As(III) > inorganic As(V) > organic As(V) > arsonium compounds and elemental As (Thirunavukkarasu et al. 2001). Arsenite is more prevalent in groundwater than arsenate.

Arsenic is a toxic and carcinogenic substance. Ingestion of inorganic As can result in both cancer (skin, lung and bladder) and non-cancerous effects (Smith et al. 1992; Chen et al. 1994). In addition, inorganic As in drinking water may affect many organs such as the central and peripheral nervous systems, dermal, cardiovascular, gastrointestinal and respiratory systems. A health effects study conducted by the National Academy of Sciences (National Academy of Sciences 1977) concluded that the cancer risk associated with As exposure has been greatly underestimated by the United States Environmental Protection Agency (U.S. EPA). The World Health Organization (WHO) estimated that a lifetime exposure to As in drinking water at a concentration of 200 µg/L might give a 5% risk of getting skin cancer (Chen et al. 1994). The U.S. Environmental Protection Agency (U.S. EPA 2000) estimated the lifetime risk of developing skin cancer was in 1 or 2% per 1000 people/µg of inorganic As/L of drinking water.

In Bangladesh, it is estimated that 30 to 40 million people are at risk from arsenic in drinking water (Ahmad et al. 2005). Thousands more people in India, Taiwan, Mongolia, Chile and Argentina, etc., suffer from the same health problems. Acid mine water containing As and heavy metals is considered a major environmental hazard in the western United States. A report indicated that 2.5 million people in the United States may be sup-
plied with water with more than 25 µg/L As and 330,000 people are supplied with water of more than 50 µg/L As, the current U.S. limit (Altundogan et al. 2000).

The As standard concentration limit set by different health and quality controlling organizations varies from 7 to 50 µg/L. Different treatment technologies are reported in literature for As removal. The U.S. EPA suggested ion exchange, activated alumina, reverse osmosis, modified coagulation/filtration and modified softening as best available technologies (BAT) and especially on iron-based coagulation assisted by microfiltration, iron oxide-coated sand (IOCS), manganese greensand filtration and granular ferric hydroxide (GFH) (U.S. EPA 2000). Appropriate treatment depends on many factors, for example, concentration of As, water composition, pH and cost effectiveness.

Sponge is seen to be an ideal material for adsorption because of its characteristics. Sponge has high internal porosity and specific surface which facilitate As absorption. In addition, sponge can be easily compacted to a very small volume to help with disposal. Importantly, sponge is cost effective so it can be applied in small communities as well as in developing countries. To achieve high As adsorption, sponge should be coated with metal oxide. After adsorption of As onto a sponge matrix, the sponge can be directly disposed or regenerated with chemical solutions.

The objective of this study is to evaluate the capacity and effectiveness of a new material (metal oxide-coated sponge [IOCSp]) in removing arsenic from synthetic and real groundwater.

Materials and Methods

The experiments were conducted with synthetic water and natural groundwater containing arsenic (Kelliher water treatment plant, Kelliher, Saskatchewan, Canada, and wells in Van phuc village, Thanh Tri, Hanoi, Vietnam). Synthetic water was prepared by adding As in tap water to obtain an arsenic concentration from 260 to 530 µg/L. The pH of synthetic water was adjusted to a designed value (6, 7 and 8) with 1 M HNO₃ and 1 M NaOH. Groundwater in wells in Kelliher water treatment plant, Canada, and Vanphuc village, Vietnam, was collected using 20-L low-density polyethylene containers and used in the studies immediately upon arrival to the laboratory.

The characteristics of the synthetic water and the groundwater used are presented in Table 1.

### Experiment with Uncoated Sponge

Typical properties of the polyurethane sponge are summarized in Table 2. It has high internal porosity and specific surface. The pore size of the sponge may be large or small and a multitude of additives may be incorporated into the main material.

Initially, experiments were conducted with uncoated sponge (half-cylinder size with 3-cm diameter and 1.5-cm length). These sponges were put into 100 mL As solution (total arsenic = 530 µg/L) and shaken for one to four days.

### Optimization of Iron Oxide Coating on Sponge

The principal method for preparation of IOCSp is presented in Fig. 1. The experiments were conducted at various operating conditions to find the optimum condition for coating on sponge with iron oxide. The conditions studied were: (i) pH value of coating solution (pH from 1, 2… to 12); (ii) time of contact between iron oxide and the sponge, t₁ (5, 10, 15, 20, 30 min and up to 10 h);

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Synthetic water</th>
<th>Kelliher groundwater</th>
<th>Hanoi groundwater</th>
</tr>
</thead>
<tbody>
<tr>
<td>Arsenic</td>
<td>0.26–0.53</td>
<td>0.056</td>
<td>0.156</td>
</tr>
<tr>
<td>Iron</td>
<td>0.07–0.09</td>
<td>2.1</td>
<td>6.2</td>
</tr>
<tr>
<td>Manganese</td>
<td>0.02</td>
<td>1.2</td>
<td>1.5</td>
</tr>
<tr>
<td>Nitrate</td>
<td>1.7–2</td>
<td>2.9</td>
<td>N/A b</td>
</tr>
<tr>
<td>Turbidity (NTU)</td>
<td>0.22–0.28</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>Chloride</td>
<td>10–12</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>Copper</td>
<td>0.001</td>
<td>0.04</td>
<td>0.08</td>
</tr>
<tr>
<td>Zinc</td>
<td>&lt;0.005</td>
<td>0.01</td>
<td>0.01</td>
</tr>
<tr>
<td>Lead</td>
<td>&lt;0.002</td>
<td>0.002</td>
<td>N/A</td>
</tr>
<tr>
<td>Cadmium</td>
<td>&lt;0.001</td>
<td>&lt;0.001</td>
<td>N/A</td>
</tr>
<tr>
<td>Chromium</td>
<td>&lt;0.001</td>
<td>0.001</td>
<td>N/A</td>
</tr>
<tr>
<td>Chlorine (residual)</td>
<td>0.3–0.4</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>Phosphate</td>
<td>0.004</td>
<td>N/A</td>
<td>1.5</td>
</tr>
<tr>
<td>Silicate</td>
<td>0.8</td>
<td>N/A</td>
<td>15</td>
</tr>
</tbody>
</table>

*All parameters except pH and turbidity are in mg/L.

bN/A; Not available.
(iii) coating temperature, T°C (110°C, 220°C); and (iv) time of drying of sponge after the coating, t 2 (8, 12, 14, 18, 20 h and up to 34 h). After determining the optimum coating conditions for preparing IOCSp, the IOCSp was prepared at optimum conditions for all the experiments in the subsequent batch and column studies.

The weight of iron oxide coated on sponge (as dry weight) was calculated on a regular basis. Both coated and uncoated sponge were dried in an oven at 100°C for 24 h and desiccated prior to the measurement of the total amount of coated iron oxide.

**Batch Kinetic Studies**

Batch adsorption kinetic studies were conducted with the IOCSp prepared at the optimum coating conditions to investigate the removal of both As(III) and As(V) from the synthetic water.

The experiments were conducted at three different pH values of 6, 7 and 8. The IOCSp was first cut into half-cylinder pieces each with 1.5 cm in length and 3 cm in diameter. They weighed 0.15 g each. The IOCSp was then placed into 250-mL Erlenmeyer flasks containing the synthetic water. The flasks were sealed with laboratory film. The samples were placed on a mechanical shaker and shaken at 175 rpm. Samples from the flask were collected at regular time intervals and analyzed for residual As.

**Column Studies**

Column experiments were conducted to study the removal of As in both synthetic water and groundwater. A glass column of 20-mm diameter and 700-mm height was used in this study. The water was run through the packed column at a filtration velocity from 1.5 to 3 m/h. The column was packed with different weights of IOCSp (6, 8 and 10 g). The column tests were conducted in the down-flow mode and at the normal pH (6.7–7.6) of the water.

**Results and Discussion**

**Uncoated Sponge in the Removal of Arsenic**

The experimental results showed that uncoated sponge did not have sufficient capacity for removing arsenic, resulting in only 10% removal with arsenic solution of 530 µg/L (Fig. 2) or only 23.3 and 29.3 µg/g sponge after 24 and 96 h contact, respectively.

**Preparation of IOCSp**

Experimental results showed that sponge could not be coated in an adequate manner when the soaking time (t1) of the sponge in the FeNO₃ solution was less than 1 h. The sponge was damaged when it was coated at a temperature (T) of 220°C and above. Experimental results also indicated that sponge could not be coated

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**TABLE 2.** Characteristics of the polyurethane sponge (Moe and Irvine 2000)

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dry density (kg/m³)</td>
<td>84</td>
</tr>
<tr>
<td>Density at 65% moisture content (kg/m³)</td>
<td>113</td>
</tr>
<tr>
<td>Swelling in vertical direction when at 65% moisture content (%)</td>
<td>24</td>
</tr>
<tr>
<td>Swelling in horizontal direction when at 65% moisture content (%)</td>
<td>31</td>
</tr>
<tr>
<td>Swelling in volume when at 65% moisture content (%)</td>
<td>113</td>
</tr>
<tr>
<td>Porosity at 65% moisture content</td>
<td>0.85</td>
</tr>
</tbody>
</table>

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**Fig. 1.** Procedure for coating the sponge with iron oxide.

**Fig. 2.** Arsenic removal by uncoated sponge.
well at a pH above 5 and was damaged when it was allowed to dry more than 34 h.

Thus, the optimum conditions for coating of the sponge with iron oxide were found to be: pH 4, $t_1$ of 10 h, $T$ of 110°C and $t_2$ of 20 h. These optimum conditions were used for the media preparation in the subsequent batch and dynamic experiments.

It was difficult to assess the homogeneity of the iron oxide coating on the sponge. Even with the determination of the specific surface area (S_BET) of IOCSp, the results could not be interpreted due to the sample configuration. However, detailed experiments conducted with different sizes of IOCSp showed uniform amounts of iron oxide per volume of sponge. As such one can safely assume the coating of iron oxide was uniform. The weight measurement showed that 12% weight of IOCSp was iron oxide.

**Adsorption Kinetics**

Figures 3 and 4 present the removal of As(III) and As(V) by IOCSp, respectively. The results showed that IOCSp could remove 47.3 to 65% of As(III) and As(V) after a 1-h contact with IOCSp. A 9-h adsorption led to a very high amount of As adsorption (less than 18 µg/L of As remaining in the water). In these experiments, 0.15 g of IOCSp was placed in a flask containing 100 mL of As solution. Each gram of IOCSp adsorbed about 160 µg of As(III) and As(V) upon a 9-h contact of IOCSp with the As solution (Fig. 3B and 4B) or each gram of iron oxide adsorbed about 1330 µg of As. The results showed that the pH (in the range used in this study) did not have any significant effect on the removal of As(III) and As(V). Further, the removal of As(III) and As(V) was the same magnitude. The difference is hardly 5 to 6% which cannot be taken as a significant value considering the accuracy limit of the arsenic analysis.

**Comparison of Removal Efficiency between IOCSp and Iron-Coated Sand (IOCS)**

Table 3 compares the As removal efficiency of IOCSp and IOCS (as a function of weight of media). IOCS is the iron oxide-coated sand and the coating methods adopted are discussed in detail elsewhere (Thirunavukkarasu et al. 2001).

The arsenic removal capacity was observed to be much higher for IOCSp than IOCS when the comparison was made in terms of weight of media. The density of IOCS is much higher than that of IOCSp and one cannot assume that IOCSp is better than IOCS from the results obtained.

As a function of weight of media, the amount of arsenic adsorbed onto IOCSp is much higher than other materials such as activated carbon (20 µg As/g), copper-impregnated carbon (48 µg As/g), activated alumina (112 µg As/g) (U.S. EPA 2002) and granular ferric hydroxide (112–159 µg As/g) (Thirunavukkarasu et al. 2003).
In this study, the adsorption column experiments were conducted with throughput (filtration) rates of 1.5 and 3 m/h. The filtration rate was kept constant during the experimental run using a constant head tank. The IOCSp medium of 0.3 and 0.5 cm (length, width and height) in size was used.

The results of the removal of As from synthetic water using the IOCSp column system are presented in Fig. 5. For both filtration rates (1.5 and 3 m/h) employed in this study, the columns could be operated for a long time with an effluent As concentration of less than 50 µg/L of As(V). The bed volume achieved was about 260 (which corresponds to a throughput volume of more than 50 L).

The results also showed that the effluent quality became inferior with the increase in filtration rates. However, the difference in removal efficiency was not significant in the initial stage. The drop in the arsenic removal efficiency with the increase in the filtration rate was due to the decrease in the empty bed contact time (Fig. 6).

Kelliher groundwater, Canada. After the operation of the IOCSp column with synthetic wastewater, the IOCSp column was tested with Kelliher groundwater in Canada. The arsenic removal efficiency by the IOCSp column of the Kelliher groundwater is shown in Fig. 7. The arsenic concentration in the effluent increased gradually but was still less than 18 µg/L even after a continuous input of throughput volume of more than 60 L (which corresponds to a bed volume of 310). Two different amounts of adsorption medium (IOCSp) of 6 and 8 g were used in order to study the effect of filter medium amount (Fig. 7). The results indicated that there was not much difference in removal efficiency when the weight of IOCSp was decreased from 8 to 6 g during the experimental period.

![Fig. 5. Removal of As(V) in synthetic water by IOCSp column [initial As(V) concentration = 260 µg/L; weight of IOCSp = 10 g]. (A) Q = 1.5 m/h; (B) Q = 3 m/h.](https://iwaponline.com/wqrj/article-pdf/41/2/164/230070/wqrj0410164.pdf)

![Fig. 6. Effect of filtration rate on As(V) removal by IOCSp column [initial As(V) concentration = 260 µg/L; weight of IOCSp = 10 g].](https://iwaponline.com/wqrj/article-pdf/41/2/164/230070/wqrj0410164.pdf)
The IOCSp column study was also carried out with groundwater in Hanoi to evaluate the arsenic removal capacity of IOCSp. The columns were run with a filtration rate of 3 m/h and IOCSp media sizes of 0.3 and 0.5 cm.

During the initial stage, when the throughput volume was less than 22 L, the effluent arsenic concentration was less than 10 µg/L for both media sizes (Fig. 8). After this stage, the arsenic in the effluent increased slowly with the IOCSp media size of 0.3 cm but the effluent concentration was lower than 50 µg/L even when the bed volume was about 390 (which corresponds to a throughput volume of about 75 L). The results also showed that with the increase in size of IOCSp, the arsenic removal became inferior, especially at the later stages of the experiment. The improvement of the arsenic removal efficiency with the decrease in the IOCSp size was due to the increase in the surface area. This shows the importance in optimizing the size of IOCSp medium.

During the experiment with natural groundwater, As was removed both by iron oxides incorporated in IOCSp and iron present in the natural groundwater. The amount of iron present in the total volume of raw water treated represents about 30% of the total amount of iron available in the system. Even if it assumed that all iron present in the water contributed to the arsenic removal, the major removal was due to adsorption onto the IOCSp.

Conclusions

The results of this study showed that sponge coated with iron oxide under appropriate conditions (soaked in the solution of ferric nitrate with pH = 4 for 10 h and heated at 110°C for 20 h) could remove effectively both trivalent arsenite and pentavalent arsenate from water. In terms of weight of adsorption media, arsenic adsorption capacity on the iron-coated sponge was much higher than that on the other media. A filter column system packed with IOCSp led to high arsenic removal efficiency. The removal efficiency depends on several factors such as the characteristics of water, and the size of media, etc. A column of 10 g of IOCSp with a size of 0.5 cm (in length, width and height) could treat more than 50 L of synthetic water (initial As concentration 260 µg/L) to a value less than 50 µg/L, whereas a smaller amount of IOCSp of 8 g could treat up to 63 L of Kelliher groundwater (Canada) and 75 L of Hanoi groundwater (Vietnam) to a value less than 18 and 50 µg/L, respectively. The initial As concentration in Kelliher and Hanoi groundwater was 56 and 156 µg/L, respectively.

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References


