

Removal of arsenic from groundwater using iron-coated jute-mesh structure

Samia Syeoti Ramim, Habiba Sultana, Tanjinur Akter, Muhammad Ashraf Ali*

Department of Civil Engineering, Bangladesh University of Engineering and Technology, Dhaka, Bangladesh,
email: syeoti@yahoo.com ((S.S. Ramim)), hrupa53@gmail.com (H. Sultana), Tel. +8801713043325, +8801836188498,
+8801627004193, email: ashraf@ce.buet.ac.bd (M.A. Ali)

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ABSTRACT

In Bangladesh water extracted from arsenic (As) contaminated shallow aquifer is extensively used for irrigating dry season (boro) rice fields. This practice is increasing As concentration in top-soils, resulting in elevated concentration of As in rice plants, decreased rice yield and increased human exposure to As. Efforts have been made to reduce As loading to rice fields by amending irrigation channels with jute-mesh structure, which however achieved limited success. The objective of the present study was to assess removal of Arsenite and Arsenate by “iron-oxide-coated jute-mesh structure” through laboratory batch experiments, which could be used in amending irrigation channels for more efficient removal of As. Two different types of locally available jute fabrics have been coated with iron oxide following two different coating methods and two drying methods. The iron-coated jute fabrics were subsequently used to assess their effectiveness in removing arsenic from groundwater in laboratory batch experiments. Results suggest that iron-coated jute fabrics have significant capacity to remove both As(III) and As(V) from groundwater and could potentially be used for reducing As loading to rice fields.

Keywords: Arsenic; Irrigation; Rice; Jute-mesh; Iron-coating

1. Introduction

In Bangladesh, groundwater from shallow aquifer is the principal source of both drinking water and irrigation water. Dry season *boro* rice provides higher yields than traditional varieties of wet season rice and accounts for more than half of the total rice production in Bangladesh [1]. However, dry season *boro* rice requires groundwater irrigation. Shallow groundwater is extensively used for irrigating *boro* rice fields [1]. But presence of high concentration of arsenic (As) in shallow groundwater of many regions of Bangladesh is becoming a major concern [2]. Various studies have reported significant arsenic accumulation in rice field soils [3,4] and in rice plants particularly in roots and stems [5,6], the extent of which primarily depends on the As concentration in the applied irrigation water [7,8]. A number of studies have reported that following years of irrigation with As-contaminated groundwater, concentration of As in soil

has risen, which in turn is decreasing yields and increasing arsenic levels in rice grains [4,6,9–12]. Rice being the staple food in Bangladesh, this could adversely affect both food security and public health in the long run.

Arsenic contamination of the irrigation water-soil-crop system, especially in the rice systems, is a matter of great concern which calls for substantial improvement of understanding of the As problem and protect millions of human beings from potential exposure to high levels of As through food chain. In the absence of alternative sources for irrigation, reduction of arsenic concentration of irrigation water within irrigation channel, before the water reaches the rice field, could be an effective way to reduce arsenic loading in rice fields. But such initiatives are still absent due to lack of practical methods and high cost of treatment for large-scale removal of As from irrigation water [9,13].

To develop a land based arsenic removal scheme, studies have been made to understand arsenic behavior and spatiotemporal distributions of As in flowing

*Corresponding author.

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irrigation water [14]. Studies have revealed that arsenic removal from irrigation water could be improved within distribution channels by amending them with physical structures like jute mesh; but the increased elemental removal in amended channels was found to be largely due to increases in residence time and particle-trapping efficiency, rather than via oxidative particle formation [15,16].

Iron oxide coated sand has been proved to be an effective medium for removal of As from groundwater, drinking water and wastewater [17,18]. Different fiberglass types (cloth, mat and fibers) were coated with iron oxide using different combinations of temperature (25°C and 110°C), pH (1.3, 7 and 8.5) and initial iron concentrations (0.25 M and 2.5 M) for treating drinking water [18]. Fibrous materials offer a high specific surface area alternative to sand as the substrate for the iron oxide coating. Studies revealed that arsenate adsorption densities were highest for iron-oxide-coated fiberglass and cellulose (compared to a number of other iron-oxide-coated fibers and iron-oxide-coated sand), suggesting that these fibrous materials may offer advantages over iron oxide-coated sand [19].

Based on these previous studies, it can be inferred that low cost local materials like jute-mesh could be coated with iron oxide and be used as a lining in the irrigation channel to uptake As and thus reduce As loadings to rice fields. With this goal in mind, the specific objectives of this study were: (1) to develop suitable methods of providing iron oxide coating on jute mesh that could act as sorbents for As removal in irrigation channel; and (2) to assess the effectiveness of iron oxide coated jute mesh in removing arsenite and arsenate from groundwater. It should be noted that arsenic contaminated groundwater contains both arsenite and arsenate, although arsenite is typically the predominant species [20].

2. Methodology

2.1. Materials

Reagent grade chemicals were used for all experiments and deionized water was used to prepare stock solutions. Prior to use, all glassware were properly washed and rinsed with deionized water. Arsenic trioxide (As_2O_3) was used to prepare arsenite [As (III)] stock solution and sodium hydrogen arsenate heptahydrate ($\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$) was used to prepare arsenate [As (V)] stock solution; arsenic stock solutions were prepared freshly prior to each set of experiment. Ferric nitrate [$\text{Fe}(\text{NO}_3)_3$] solution and sodium hydroxide (NaOH) solution were used for providing iron coating.

Two kinds of locally available jute fabrics were used – one white in color and the other brown (Figs. 1 and 2); the white fabric is made of processed/bleached jute fibers and is costlier than the brown fabric. Some properties of the two types of jute fabrics are given in Table 1. ASTM (American Society for Testing and Materials) D5261-92 was followed for conducting mass per unit area test, ASTM D5199-01 was followed to determine the nominal thickness, and ASTM D4751 was followed to determine the apparent opening size of the jute fabrics.



Fig. 1. White jute fabric.

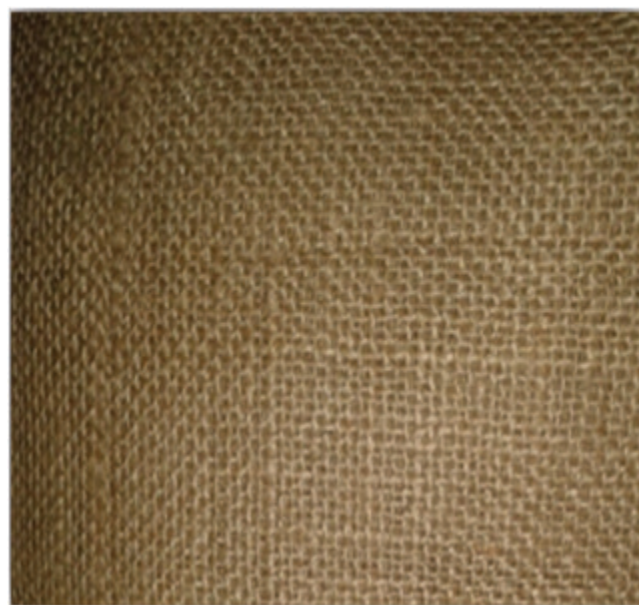


Fig. 2. Brown jute fabric.

2.2. Preparation of iron coated jute fabric

Jute fabrics were first cut into pieces of approximately 5" × 10". Before providing coating on the jute fabrics, they were thoroughly washed with deionized water and air dried so as to get rid of any foreign material adsorbed on the surface. Several methods are available for coating a media with iron oxide/hydroxide. In the present study, methods similar to those reported by Joshi and Chaudhuri [18] and Kumar et al. [19] have been used. Following the methods followed by Kumar et al. [19], two different procedures were used for precipitating iron onto jute fabrics, and two different procedures were used for drying the iron coated fabrics.

Table 1
Properties of jute fabrics

| Properties | White jute fabric | Brown jute fabric |
|-----------------------------------------|-------------------|-------------------|
| Color | White | Brown |
| Mass per unit area (g/m ²) | 269.5 | 310.8 |
| Nominal thickness (mm) | 1.28 | 0.923 |
| Apparent opening size (O95 value in mm) | 1.18 | 0.6 |
| Retail price (USD/m ²) | 1.0 | 0.34 |

In the first method (referred to as Method A), 0.25 M ferric nitrate solution was adjusted to pH 10.0 with 10.0 M NaOH solution and constantly stirred to prevent precipitation of iron. Then the jute fabrics were soaked into the solution. In the second method (referred to as Method B), jute fabric was first soaked into 0.25 M ferric nitrate solution and then iron was precipitated through the addition of 10.0 M NaOH solution, adjusting the pH to 10.0. These two coating methods were used in order to identify the more efficient method of coating that would result in higher removal of arsenic by the fabrics under similar conditions.

Following the methods followed by Kumar et al. [19], the wet jute fabrics were dried under two conditions: (1) heating at $45 \pm 2^\circ\text{C}$ in an oven for 24 h; and (2) drying at room temperature for one week. After drying, the iron-coated jute fabrics were washed with deionized water in a beaker to remove loose iron oxide particles. After that the samples were kept at room temperature for 1–2 d for drying; adsorption experiments were conducted within one week after drying. The iron-coated jute fabric samples were then cut into $2.5 \pm 0.1'' \times 5 \pm 0.1''$ pieces for carrying out batch experiments. Drying temperature affects formation of iron-oxide solids, and the two drying methods were used to assess the impact of drying temperature on arsenic removal by the fabrics.

2.3. Batch experiments for Arsenic removal

Laboratory batch experiments were carried out using natural groundwater, collected from a deep tube well pump station at Bangladesh University of Engineering and Technology (BUET). The characteristics of the groundwater are presented in Table 2. The arsenic concentration of the groundwater is below 1 $\mu\text{g/l}$. The groundwater also contains low concentration (0.325 mg/l) of phosphate, which is known to compete with arsenic for adsorption onto iron minerals. The other constituents present in the groundwater are unlikely to have significant impact on arsenic adsorption. For laboratory experiments, this groundwater was spiked with arsenic (both arsenite and arsenate) stock solutions to attain desired level of initial arsenic concentration; initial arsenic concentration was varied from 100 to 567 $\mu\text{g/L}$. Batch experiments were carried out with iron-coated jute fabrics (both white and brown types) prepared in the following ways: (1) Method A, dried at room temperature; (2) Method B, dried at room temperature; (3) Method A,

Table 2
Characteristics of groundwater used for preparation of water samples for adsorption experiments

| Sl. No. | Parameter | Concentration |
|---------|------------------------------------------------|---------------|
| 1 | pH | 7.1 ± 0.2 |
| 2 | Carbon-di-oxide, mg/l | 71.0 |
| 3 | Total alkalinity, mg/l as CaCO_3 | 216.0 |
| 4 | Total hardness, mg/l as CaCO_3 | 256.0 |
| 5 | Chloride, mg/l | 55.0 |
| 6 | Total ammonia as $\text{NH}_3\text{-N}$, mg/l | < 0.017 |
| 7 | Nitrate ($\text{NO}_3\text{-N}$), mg/l | 0.30 |
| 8 | Nitrite ($\text{NO}_2\text{-N}$), mg/l | < 0.008 |
| 9 | Orthophosphate, mg/l | 0.325 |
| 10 | Sulfate, mg/l | 14.2 |
| 11 | Iron (Fe), mg/l | 0.01 |
| 12 | Manganese (Mn), mg/l | 0.022 |
| 13 | Arsenic (As), mg/l | < 0.001 |

dried by heating at $45 \pm 2^\circ\text{C}$; (4) Method B, dried by heating at $45 \pm 2^\circ\text{C}$. Batch experiments were carried out to evaluate removal of both As (III) and As (V) by these iron-coated jute fabrics. Arsenic removal capacity of uncoated jute fabrics (both white and brown) was also evaluated.

Batch experiments were carried out in a series of 1 L glass beakers. In each beaker, 500 ml groundwater sample was taken; it was then spiked with freshly prepared arsenite or arsenate stock solution to attain the desired level of initial As concentration. It should be noted that oxidation of arsenite to arsenate in oxygenated water, though thermodynamically favored, usually takes days, weeks or even months depending on specific conditions [21]. Roberts et al. [20] reported only limited oxidation of arsenite to arsenate in irrigation channel of rice fields, and this oxidation was facilitated by oxidation of Fe(II) (present at an initial concentration of 10.9 ± 0.2 mg/l). Therefore, arsenite present in the experimental water samples (containing negligible iron concentration) is unlikely to be oxidized to a significant extent to arsenate during the course of the experiments. No effort was made to adjust pH of the water samples during the batch experiments; initial pH of the water samples was 6.9 ± 0.1 . One piece of iron-coated jute fabric ($2.5'' \times 5''$) prepared by different methods was then added to each beaker; similar uncoated pieces of white and brown jute fabrics were also added to As-bearing water in separate beakers. Each set of experiment was carried out in duplicates and the results averaged. The contents of the beakers were stirred with a glass rod for 15 min, and then the beakers were kept at rest for 30 min. After that, water samples were taken from the beakers for analysis of arsenic concentration, and final pH of the water samples was measured. pH was measured with a pH meter (HACH Co., USA), measurement of arsenic concentration was carried out with an Atomic Absorption Spectrophotometer (Shimadzu, AA 6800); detection limit for arsenic was 1 $\mu\text{g/l}$. Arsenic uptake/ removal by the jute fabrics was estimated by subtracting final As concentration from ini-

tial As concentration, and then expressing the removal/uptake as mg As/m² of jute fabric.

In order to evaluate arsenic uptake capacity, similar batch experiments were carried out with iron oxide coated brown jute fabrics (dried at 45 ± 2°C and at room temperature), where initial As (III) and As (V) concentration in water was gradually increased to around 1000 mg/l, and corresponding arsenic uptake was determined.

3. Results and discussions

3.1. Arsenic uptake by coated white and brown jute fabrics

Figs. 3 and 4 show average uptake of As (III) and As (V), respectively, by different types of jute fabrics, where arsenic uptake has been expressed as mg As/m² of jute fabric. It is clear from these figures that iron-coated jute fabrics could uptake/remove significant amount of As from groundwater compared to uncoated fabrics. The figures also show that uptake of As varies significantly with the type of fabric (i.e., whether white or brown fabric) and coating method used to prepare the fabrics. Fig. 3 shows that As (III) uptake by iron-

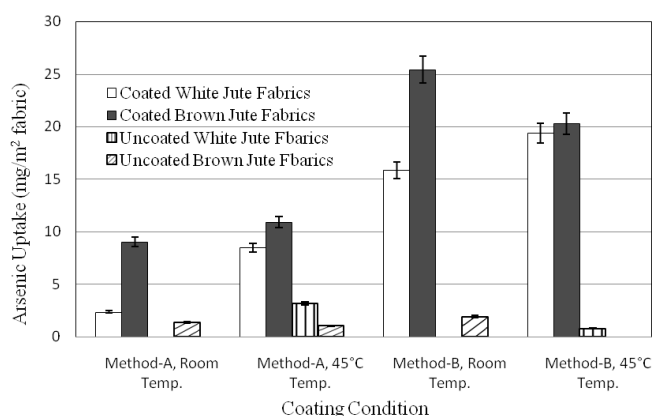


Fig. 3. As (III) uptake by different types of iron-oxide coated white and brown jute fabrics; the error bars represent standard deviation of duplicate test results.

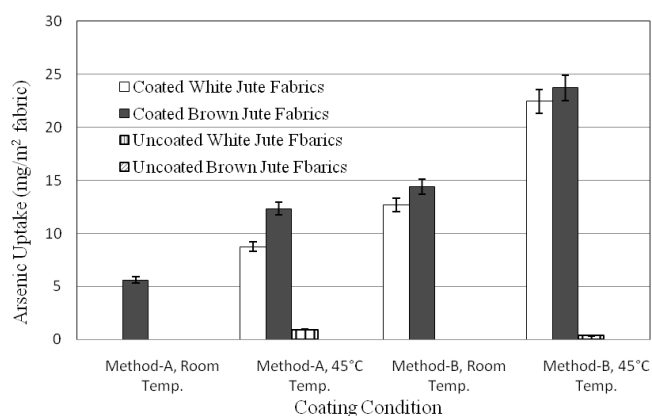


Fig. 4. As (V) uptake by different types of iron-oxide coated white and brown jute fabrics; the error bars represent standard deviation of duplicate test results.

coated jute fabrics prepared by Method B (at both room temperature and at 45 ± 2°C) is significantly higher compared to that by samples prepared by Method A. This implies that Method B, which involved soaking of fabric in ferric nitrate solution followed by addition of sodium hydroxide, was more efficient in imparting iron-oxide coating on jute fabrics. As (III) uptake by iron-coated “brown” fabrics has been found to be slightly higher than that by “white” fabrics in all cases. Highest As (III) uptake of 25.1 mg/m² was recorded for iron-coated brown fabric prepared by Method B and dried at room temperature. It should be noted that it was not possible to quantify iron-oxide coating on the jute fabrics (e.g., in terms of mass Fe per unit area of fabric). Therefore, arsenic removal efficiency was used to indirectly assess effectiveness of iron-oxide coating.

Similarly, Fig. 4 shows that As (V) uptake by iron-coated jute fabrics prepared by Method B is significantly higher compared to that by samples prepared by Method A; As (V) uptake was highest for samples prepared by Method B and dried at 45 ± 2°C. Similar to the case of As (III), As (V) uptake by iron-coated “brown” fabrics has been found to be slightly higher than that by “white” fabrics in all cases. Highest As (V) uptake of 23.7 mg/m² was recorded for iron-coated brown fabric prepared by Method B and dried at 45 ± 2°C.

Tables 3 and 4 show detail results from two sets of batch experiments carried out for evaluating As (III) and As (V) removal/uptake by iron-coated jute fabrics prepared by Method B and dried at 45 ± 2°C. These two sets of experiments were carried out with similar initial As concentrations, and comparison of results of these two sets of experiments provide useful insights into the As uptake by brown and white jute fabrics. Both Tables 3 and 4 show that under similar experimental conditions, uptake of both As (III) and As (V) is slightly higher for iron-coated brown fabrics, compared to the iron-coated white fabrics; but the differences are not very significant. For example, average As (III) uptake by iron-coated white and brown fabrics were 19.4 mg/m² and 20.3 mg/m², respectively. Tables 3 and 4 show that the adsorption of both arsenite and arsenate is accompanied by a rise in pH; the rise in pH is more prominent for arsenate, as expected [22], which exists primarily as anionic species in the pH range of the experiments.

3.2. Effectiveness of iron coated jute fabric in Arsenite and Arsenate uptake

Most adsorptive media, including iron-coated sand, have been found to remove As (V) more efficiently than As (III). Since majority of arsenic in natural groundwater exists as As (III) [20], this often necessitates oxidation of As (III) to As (V) prior to its removal by adsorption. Therefore, ability of iron-coated jute fabrics in removing As (III) and As (V) was carefully evaluated. Figs. 5 and 6 show uptake of As (III) and As (V) by different types of brown and white jute fabrics, respectively. It is clear from these two figures (as well as Figs. 3 and 4) that fabrics prepared by Method B are more efficient in removing As than those prepared by Method A. Therefore, subsequent discussion is focused on As uptake by fabrics prepared by Method B. The figures show that the iron-coated jute fabrics (both brown and white, prepared by Method B) are efficient in removing both As (III) and As

Table 3
As (III) uptake by brown and white jute fabrics coated with iron by Method B and dried by heating at $45 \pm 2^\circ\text{C}$ in an oven

| Initial As (III) concentration = 551 $\mu\text{g/L}$ | | | | | | |
|------------------------------------------------------|----------------|------------|----------|---------------------------------|----------------------------------|------------------------------------------|
| Serial | Description | Initial pH | Final pH | Final As (III), $\mu\text{g/L}$ | As (III) uptake, mg/m^2 | Average As (III) uptake, mg/m^2 |
| 1 | Uncoated White | 6.98 | 7.31 | 538 | 0.81 | 0.81 |
| 2 | Uncoated Brown | 6.98 | 7.19 | 577 | 0.00 | 0.00 |
| 3 | Coated White-1 | 6.98 | 7.38 | 237 | 19.5 | 19.4 |
| 4 | Coated White-2 | 6.98 | 7.40 | 240 | 19.3 | |
| 5 | Coated Brown-1 | 6.98 | 7.32 | 233 | 19.7 | 20.3 |
| 6 | Coated Brown-2 | 6.98 | 7.31 | 215 | 20.8 | |

Table 4
As (V) uptake by brown and white jute fabrics coated with iron by Method B and dried by heating at $45 \pm 2^\circ\text{C}$ in an oven

| Initial As (V) concentration = 567 $\mu\text{g/L}$ | | | | | | |
|----------------------------------------------------|----------------|------------|----------|-------------------------------|--------------------------------|----------------------------------------|
| Serial | Description | Initial pH | Final pH | Final As (V), $\mu\text{g/L}$ | As (V) uptake, mg/m^2 | Average As (V) uptake, mg/m^2 |
| 1 | Uncoated White | 6.91 | 7.33 | 561 | 0.37 | 0.37 |
| 2 | Uncoated Brown | 6.91 | 7.20 | 585 | 0.00 | 0.00 |
| 3 | Coated White-1 | 6.91 | 7.46 | 196 | 23.0 | 22.4 |
| 4 | Coated White-2 | 6.91 | 7.45 | 214 | 21.9 | |
| 5 | Coated Brown-1 | 6.91 | 7.39 | 196 | 23.0 | 23.7 |
| 6 | Coated Brown-2 | 6.91 | 7.40 | 173 | 24.4 | |

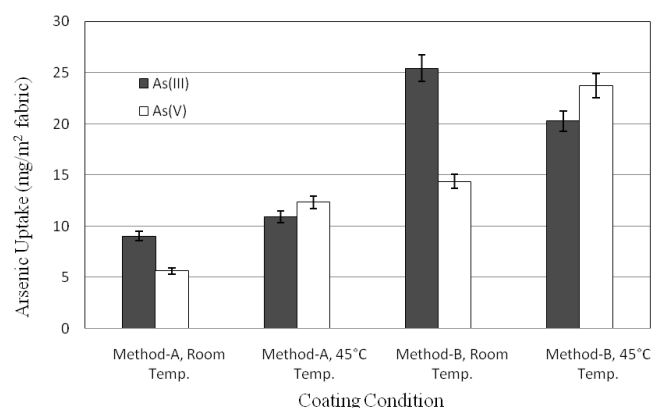


Fig. 5. Comparison between As (III) and As (V) uptake by different types of iron-coated brown jute fabrics; the error bars represent standard deviation of duplicate test results.

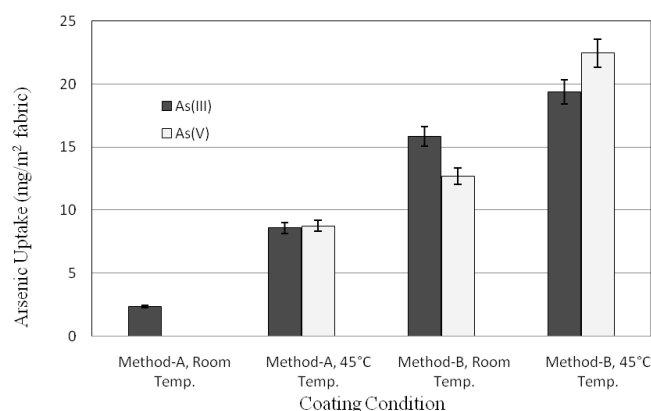


Fig. 6. Comparison between As (III) and As (V) uptake by different types of iron-coated white jute fabrics; the error bars represent standard deviation of duplicate test results.

(V). These figures also clearly show that the drying method has a significant influence on the As uptake characteristics of the jute fabrics. The jute fabrics prepared by drying at room temperature are more efficient in removing As(III), while the jute fabrics prepared by heating the fabrics at $45 \pm 2^\circ\text{C}$ are more efficient in removing As (V). Adsorption of arsenic has been reported to vary with the type of iron oxide mineral and pH [23]. Dixit and Hering [23] studied sorption of As (V) and As (III) onto amorphous iron oxide (HFO),

goethite, and magnetite, and reported that sorption of As (V) onto HFO and goethite was more favorable than that of As (III) below pH 5–6, while at higher pH values, As (III) has higher affinity for the solids. The variation of As (III) adsorption onto jute fabrics prepared under different drying conditions is most likely due to differences in the precipitated forms of iron under different heating conditions and relatively high pH conditions under which the experiments were performed.

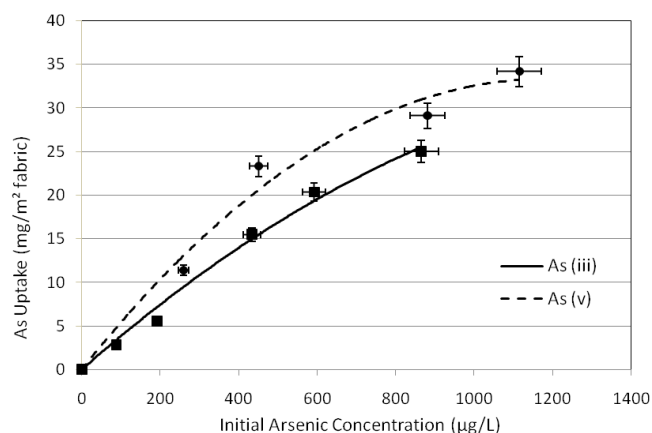


Fig. 7. Comparison between As (III) and As (V) uptake by iron-coated brown jute fabrics, dried at $45 \pm 2^\circ\text{C}$, as a function of initial As concentration; the error bars represent standard deviation of duplicate test results.

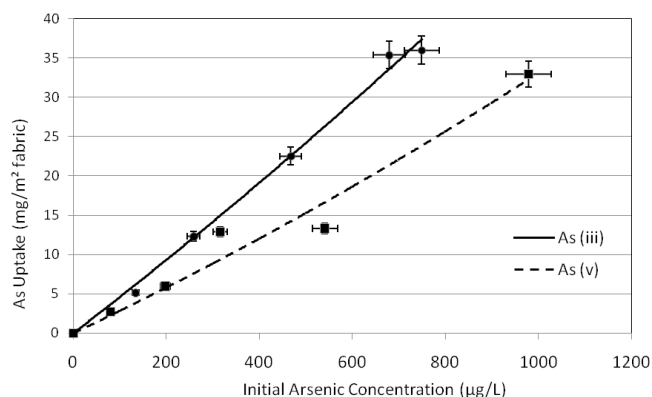


Fig. 8. Comparison between As (III) and As (V) uptake by iron-coated brown jute fabrics, dried at room temperature, as a function of initial As concentration; the error bars represent standard deviation of duplicate test results.

3.3. Arsenic uptake capacity of iron coated brown jute fabric

Figs. 7 and 8 illustrate As uptake (expressed as mg/m^2) vs. initial As concentration (expressed as $\mu\text{g}/\text{L}$) for iron coated brown jute fabrics prepared by Method B and dried at 45°C and at room temperature, respectively. Fig. 7 shows that with increasing initial arsenic concentration in water, uptake of both Arsenite and Arsenate by brown jute fabrics (dried at $45 \pm 2^\circ\text{C}$) increases; the uptake of As (V) is higher than As (III). As expected, initially the slope of the As uptake vs. As concentration graph is steeper and gradually it becomes flatter, at the fabrics approach their capacity. As shown earlier (see Fig. 5), brown fabrics dried at 45°C have been found to be more efficient in removing As(V) than As(III).

On the other hand, Fig. 8 shows that higher uptake of As (III) than As (V), by iron oxide coated jute fabrics dried at room temperature, which is consistent with data presented in Fig. 5. While arsenic uptake increased with increase in initial arsenic concentration, no plateauing effect of arsenic uptake was observed under the experimental condition, possibly indicating very high adsorption capacities of these fabrics.

4. Conclusions

A number of methods were employed to coat two different types of jute fabrics with iron oxide/hydroxides. Uptake/removal of As (III) and As (V) by iron-coated jute fabrics were then evaluated in batch experiments. The method of coating has been found to have a significant influence on arsenic removal efficiency of the iron-coated jute fabrics. The fabric drying method/temperature appear to influence its ability in removing As (III) and As (V). The iron coated jute fabrics have been found to be very efficient in removing both As (III) and As (V) from groundwater, compared to uncoated fabrics. The results of this study, particularly the ability of the iron-coated jute fabrics in removing As (III), suggest that these coated fabrics could potentially be used for reducing As loading to agricultural soil from As-contaminated irrigation water. The white jute fabrics used in this study are made of washed/bleached jute fibers and are more expensive (see Table 1); the less expensive brown jute fabric with comparable or slightly higher arsenic adsorption capacity therefore appears to be the better choice for removing arsenic from groundwater. More work is needed to standardize the iron coating methods, assess kinetics of arsenic adsorption and to evaluate effectiveness of iron coated jute fabrics in removing arsenic from water flowing through a channel at bench and pilot scales.

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