Arsenic Remediation in Contaminated Groundwater By Combination of Iron and Manganese Oxide Coated Materials

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1. INTRODUCTION

Recently As contamination becomes a worldwide issue because of their high toxicity and the consequent risks for human health, and the As contamination of groundwater is serious problem in some regions; Bangladesh, Taiwan, Chile and West Bengal in India. Especially, the geochemical nature of arsenic contamination of the aquifers in soil of Bangladesh region is of serious concern because groundwater is the primary source of drinking water; more than 95% of Bangladesh population drinks well water and total As measurement shows that more than 60% of the tube well water is above 10ppb guideline value (Khan et al., 2000); thus, the device for simple and effective treatment of the groundwater are demanded. Arsenic remediation methods are intensified recently and researched through the accesses of diverse scientific fields. In spite of many types of methods, adsorption methods by iron oxide coated materials, especially using sand, are the most used because of the Fe affinity toward inorganic As species and consequent selectivity of the adsorption process (José et al., 2002).

However, this methods have been restricted to removal of As(III), which is more toxic than As(V) species, as well as tend to lower the pH of solution. Therefore, the main objective of this study was to overcome the limitation of conventional technologies for As(III) contaminated water, with use of oxidation property of manganese oxide, maintaining the circum-neutral pH for use as drinking water. In addition, It is to devise the simple and easily applicable techniques for small scale treatment system.

2. MATERIALS AND MATHODS

Quartz sand and polypropylene textile. The sand, supplied by silicas and co., Joomunjin standard sand, which more sort than 90% was quartz. The sand was pretreated by weak acid for prevention of the interference effect by other metal ions and then prepared as fine sand (0.3 - 0.6mm) through sieving process before use.

The polypropylene textile (PP), supplied by Jeonsan co. ltd., is a low-priced and porous fabric, such as a wad of cotton. The PP was pretreated by methanol solution (24 hr) to prevent the effect of other impurities, rinsed with de-ionized water three times and dried at room temperature (20 hr).

Arsenic stock solution. Arsenic (III) stock solution was prepared by dissolution of sodium arsenite NaAsO₂ (Sigma) in distilled water and As(V) was spiked with sodium heptahydrate salt Na₂HAsO₄·7H₂O (Sigma), adjusted pH.

Synthesis of Iron oxide coated materials (IOCS and IOCP). Iron oxide coated sand or PP was synthesized by coating mechanism of sand or PP surface through mixing of Fe(NO₃)₃ 9H₂O and NaOH solutions. This process was repeated once again for ideal coating (Thirunavukkarasu et al., 2003).

Synthesis of Manganese oxide coated materials (MOCS and MOCP). Manganese oxide coated sand or PP was produced onto materials surface by mixing process with heated KMnO₄ and concentrated HCl solutions (Manning et al., 2002). This coating procedure was also performed twice.

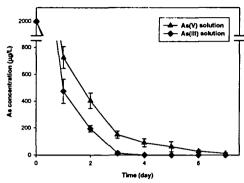
Analytical determinations. The forms of arsenic were separated to As(III) and As(V) using anionic exchange cartridge (LC-SAX) (Yalcin and Le, 2001). The determination of the dissolved As was performed by the hydride generation atomic absorption spectrophotometry (HG-AAS, Perkin-Elmer ZL 5100) method. The reduction of As(V) was performed by KI addition, and the pre-treatment of the samples involved also addition of HCl. For the analysis, sodium borohydride solutions was supplemented with 0.1 M sodium hydroxide and 10-wt% HCl solution (Merck) was used for carrier. (Ko, 2004).

3. RESULTS AND DISCCUSION

Restriction of As(III) adsorption by iron oxide and As(III) Oxidation by Manganese Oxide Arsenic batch experiments for adsorption by IOCS and for Oxidation by MOCS were set at $2000 \, \Box / l$ initial concentration adjusted pH 6.5 and shaken at $100 \, \text{rpm}$ and $25 \, \text{°C}$ for 1 week (ratio of sand : soln. = 1 : 6). On As adsorption experiment for As(III) and As(V), the adsorption degree was presented differently depending on the As species; it is appeared that As(III) adsorption by Iron oxide has some restriction at removal, comparing with As(V) (Fig. 1.).

Accordingly, when As exist as As(V) species rather than As(III) in solution, more effective remediation could be expected. Thus, artificial oxidants need for further effect of the As adsorption process, in this study, manganese oxide was applied for oxidation of As.

Arsenic reaction with MOCS appeared remarkable As oxidation effect. As(III) species in As(III) solution was transformed drastically to As(V) species (Fig. 2.). Accordingly, if As oxidation by manganese oxide is embarked with As remediation technique by iron oxide, it could be faster and more eminent remediation techniques.



2000

As tool conc.

As(III) conc.

As(V) conc.

1500

2 4 6

Time (day)

Figure 1. Difference of As removal by IOCS on As(III) and As(V) species.

Figure 2. As(III) oxidation effect by MOCS.

Combination application of IOCS and MOCS for As effective removal For efficient removal of As in water, the combined media of MOCS with a little IOCS was applied at batch experiments.

The experiment was performed at 50ml As(III) solution of 500 \square /l and it was contacted with mixture of MOCS 1g and IOCS 0.2g for 6 hrs. This mass ratio conditions were drawn by base experiments of several times before the principal removal experiments. At the experiment of mixture, As concentration after reaction was appeared that about 86 % of initial As was eliminated, and its removal efficiency was much higher than that of IOCS or MOCS reacted independently; IOCS was for 78 % and MOCS was for 37 % (Fig. 3.).

Although the adsorption by mixture, comparing with removal by IOCS, was not appeared significantly, the additional advantages of this combined media was the ability of maintaining the circum-neutral pH of water and the ability to intend the remained As to remain as As(V). Iron oxide tends generally to lower the pH level in As solution. However, oxidation of manganese oxide complemented the pH-dropping phenomenon and preserved the solution to neutral state, while the solution with only iron oxide changed to acid state (Fig. 4). It shows the possibility for application of drinking water, if the treatment with the combined media of IOCS and MOCS is accomplished on As conta-minated water.

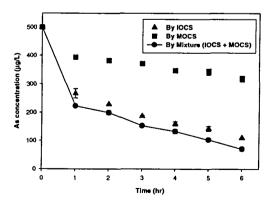


Figure 3. As(III) remediation on As remediation by IOCS, MOCS and their combination.

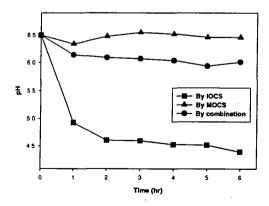
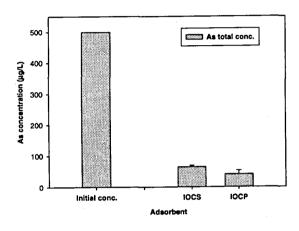


Figure 4. pH variation on As remediation by IOCS, MOCS and their combination.

Application of coated PP on As remediation IOCP and MOCP were contacted separately with solution for As adsorption and oxidation efficiency tests for 6 hrs reaction in 50 mL As(III) solution for each 1 g adsorbent.

On reaction with IOCP, As removal by IOCP appeared somewhat higher efficiency than that by IOCS (Fig. 5.). On reaction with MOCP, the efficiency was very superior relatively, comparing with As oxidation by MOCS. Arsenic removal efficiency and oxidation efficiency of MOCP was very superior relatively, comparing with As oxidation by MOCS. (Fig. 6.).



As total conc.

As(III) conc.

As(IV) conc.

As(V) conc.

Initial conc.

MOCS MOCP

Adsorbent

Figure 5. Arsenic adsorption effect of IOCP.

Figure 6. Arsenic oxidation effect of MOCP.

Removal Reaction by combination of IOCP and MOCP was performed in As(III) solution and their remediation efficiency was compared with removal by coated sand. All experimental parameters were controlled identically with the condition at experiments by sand combination. Arsenic amount at reaction

by coated sand was removed into about 86 % after 6 hrs, however, at removal by coated PP, As was eliminated very drastically, so over 99 % was reduced (Fig. 7.).

It was shown much more prominent remediation effect of rapid and high efficiency, as well as it has practical advantages at replacement or disposal of adsorbent for practical use. Consequently, this remediation technology applying the coated PP was ascertained to be superior for use as drinking water of As contaminated groundwater.

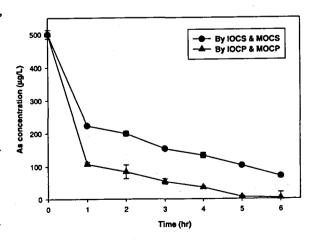


Figure 7. Arsenic remediation by IOCP and MOCP.

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Key Words; arsenic remediation, adsorption, oxidation, iron oxide, manganese oxide, sand, Polypropylene

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