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Immobilization of magnetite nanoparticles for the removal of arsenic and antimony from contaminated water

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ABSTRACT: Magnetite (Fe₃O₄) nanoparticles were synthesized and immobilized in a synthetic resin poly-methyl methacrylate (PMMA). The Fe₃O₄ nanoparticle-PMMA composites were studied for their efficiencies of removing dissolved arsenic (As) and antimony (Sb). The effects of major environmental and operating parameters on the removal of As and Sb were investigated in batch experiments. Singular and competitive adsorption of As and Sb onto the composites were studied. The results demonstrated the capability of the Fe₃O₄-PMMA composites for removing dissolved metalloids.

1 INTRODUCTION

Dissolved metalloids, including As and Sb, can be present in contaminated surface waters or groundwater. The health risk posed by arsenic, in particular when it is in inorganic dissolved form, has been widely recognized. Antimony is considered an emerging contaminant that is linked to skin, lung, and eye diseases (Cooper & Harrison, 2009). The removal of these metalloids from contaminated waters is essential for protecting human health and the environment.

A variety of methods have been studied for the removal of the two metalloids, including adsorption, membrane, coagulation/flocculation, oxidation/precipitation, etc. Among these methods, adsorption is considered a low cost conventional technique. Various high-tech or low-tech adsorbents, such as synthetic resins, activated carbon, agro-wastes, and mineral clays, etc., have been studied in lab experiments (Ungureanu *et al.*, 2015). Currently, many researchers are focusing their efforts on discovering innovative adsorbents, to further enhance adsorption efficiency and reduce cost.

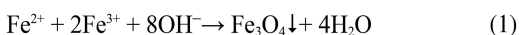
Magnetite (Fe₃O₄) nanoparticles have the potential to be used as an adsorbent or electron donor for the removal of water contaminants, due to their high surface areas, reactivity and well-established synthesis methods. Another advantage of the magnetite-based adsorbents is that they can be recovered through magnetic field. Their disadvantages include mobility in water, agglomeration, and oxidation by non-target compounds in water; these have limited the application of iron-based nanoparticles to treat contaminated waters. Appropriate methods of applying the particles, such as entrapment in porous media or carriers, need to be investigated. In this study, comparative experiments

were carried out to study the efficiency of magnetite nanoparticles immobilized in a synthetic PMMA resin for the adsorption of As and Sb.

2 MATERIALS AND METHODS

2.1 *Synthesis of magnetite nanoparticles*

AR grade ferric chloride hexahydrate (FeCl₃·6H₂O), ferrous chloride tetrahydrate (FeCl₂·4H₂O), 2-(4-chlorosulfonylphenyl) ethyl-trichlorosilane (CTCS), methyl methacrylate (MMA), and 30% ammonium hydroxide (NH₄OH) were purchased from Sigma Aldrich. Milli-q water was produced by a Merck Millipore instrument. Magnetic iron oxide nanoparticles were synthesized by co-precipitation of Fe²⁺ and Fe³⁺ ions to form Fe₃O₄ precipitates via the following reaction.



4.31 g of ferrous chloride tetrahydrate and 11.68 g of ferric chloride hexahydrate were added into a reaction chamber (500 mL triple neck round bottomed flask) pre-filled with 200 ml water (deoxygenated by purging with N₂ gas). The chamber was connected to a condenser and a mechanical mixer and placed in an oil bath. Ammonium hydroxide solution was gradually (dropwise) added to the solution. The color of the solution gradually changed from orange to dark brown and finally black. This was continued until the pH reached 8.0. A magnet was used to separate solid precipitates of Fe₃O₄ from the solution. The Fe₃O₄ precipitates were rinsed repeatedly with milli-q water and saline (0.1 M NaCl) water, vacuum-dried and stored.

2.2 Synthesis and characterization of Fe_3O_4 -PMMA composite

Three grams of the Fe_3O_4 precipitates were mixed in 40 mL of dehydrated toluene. 1.3 mmol CTCS was added to the solution, and kept in room temperature for 24 h, to produce Fe_3O_4 particles immobilized with CTCS. The particles were separated from the solution, washed, dried and weighted. For polymerization, 1.6 g of these particles were added to 80 mL toluene and mixed with 50 g MMA in the reaction chamber in N_2 environment. The reactor was submerged in the oil bath at $80^\circ C$, and the solution was stirred by a mechanical impeller for 4 h. By the end of the polymerization period, magnetic composites in the liquid were separated by the magnet, collected, washed, centrifuged, and dried. Surface characteristics of the Fe_3O_4 nanoparticles and Fe_3O_4 -PMMA composites were analyzed by a scanning electron microscope (JCM-6000, JOEL). An X-ray powder diffraction apparatus (PANAnalytical) was used to analyze the chemical compositions of the Fe_3O_4 nanoparticles.

2.3 Batch testing of As and Sb removal by the nanoparticles

An arsenic stock solution was prepared by diluting 1000 mg L^{-1} As(III) standard solution (Agilent) to 100 mg L^{-1} . An 274 mg L^{-1} antimony (III) stock solution by dissolving measured amount of antimony potassium tartrate $K_2Sb_2(C_4H_2O_6) \cdot 2.3H_2O$ in milli-q water. A series of batch adsorption tests were then carried out to study As and Pb removal by the Fe_3O_4 nanoparticles, in mobile (bare particles) and PMMA immobilized forms. The batch adsorption studies were carried out using 100 mL flasks, each filled with 50 mL of As, Sb, or mixed As and Sb solutions, which were placed on an orbital shaker rotated at 180 rpm for 90 minutes. As and Pb concentrations of the solutions, at the beginning and end each batch test, were analyzed by a MP-AES instrument (Agilent).

3 RESULTS AND DISCUSSION

3.1 Characteristics of the synthesized nanoparticles and Fe_3O_4 -PMMA Composites

Figure 1 shows the surface of the synthesized nanoparticles and Fe_3O_4 -PMMA composites. It appears that the sizes of the particles were indeed in nm range. The Fe_3O_4 impregnated PMMA composites were observed to have more rugged surfaces, compared with the surfaces of blank (without particle impregnation) MMA polymer.

XRD analyses of the synthesized nanoparticles and a commercial Fe_3O_4 sample produced similar profiles. Both synthesized and commercial samples had six significant diffraction peaks observed in both samples (at 2θ of 511, 442, 440, 311 and 220), confirming the presence of hematite in the synthesized nanoparticles.

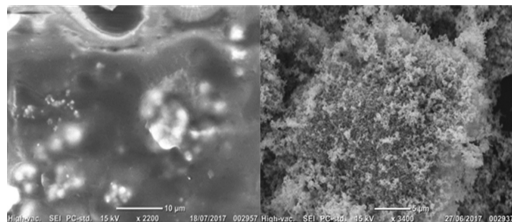


Figure 1. The SEM images of the Fe_3O_4 nanoparticles (left) and Fe_3O_4 -PMMA composites (right).

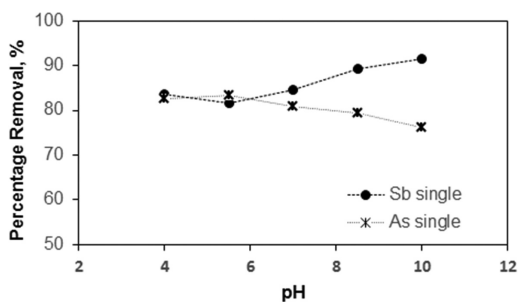


Figure 2. Percentage removal of As and Sb by the Fe_3O_4 -PMMA composites at different pH.

3.2 Batch testing results of As and Sb removal

Figure 2 shows the percentage removal of As and Sb at different pH, when As and Sb were present as single contaminants in separate solutions. Efficiencies of commercial and synthesized nanoparticles, dosage, and co-existence (competitive removal) of these metalloids were also investigated.

4 CONCLUSION

Magnetite nanoparticle-PMMA composites were synthesized and tested for the removal of As and Sb. The results demonstrated their potential as an effective, recoverable adsorbent for *in situ* remediation of metalloid contaminated water.

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