Use of magnetic filtration in waste water treatment

Katarína Štefušová¹, Miroslava Václavíková¹, Michal Lovás¹ and Slavomír Hredzák¹

The materials based on iron oxides are widely used for toxic elements removal. Magnetite nanoparticles are good sorbent of arsenic from water, but their practical use is quite limited. The solid/liquid separation of material in the nano-scale range is difficult. In this study, the synthetic magnetite was studied as arsenic sorbent from aqueous solutions with maximum sorption capacity of 40.4 mg/g. Magnetic properties of magnetite allow a relatively simple magnetic separation after arsenic sorption.

Key words: arsenic, sorption, magnetite, magnetic filtration, centrifugation

Introduction

Arsenic contamination of water supplies has become one of the major problems worldwide. Arsenic in drinking water can lead to health problems as cancer, pigmentation changes, hyperkeratosis, neurological disorders, cardiovascular diseases and conjunctivitis (Mandal and Suzuki, 2002). Arsenic can be removed by various treatment methods, such as coagulation and flocculation, precipitation, ion exchange, reverse osmosis, membrane and adsorption technologies. Among them, coagulation/precipitation followed by filtration is the most commonly used technique. Ion exchange is effective in As(V) removal but may not be suitable for As(III) removal. Use of reverse osmosis and membrane technologies is quite expensive compared to other methods (Pokhrel and Viraraghavan, 2007; Vaclavikova et al., 2008). Adsorption processes are commonly applied and show a good efficiency to cost ratio for arsenic removal. However, they fail to remove arsenic concentrations below the maximum contaminant level (10 μg.L⁻¹) (Mohan and Pittman, 2007). Therefore, there is a tremendous demand for developing cheap efficient methods for removal of arsenic species from drinking water. Sorption methods are still considered promising, in regard to the cost/efficiency factor and new sorbents are being developed with the aim of obtaining the target limit (Vaclavikova et al., 2008).

Recently, there has been increased interest in the use of magnetic materials based on iron oxides in the arsenic removal. Generally, their effectiveness increases with a decrease of the particle size. Nanosize materials are found to be very effective on the removal of arsenic from water (Deliyanni et al., 2010; Mayo et al., 2006). However solid/liquid (S/L) separation is more difficult as the particle size decreases. On the other side, in case of magnetic sorbents based on Fe oxides, the magnetic filtration may be applied for S/L separation. Furthermore, the removal of particles from solution with the use of magnetic fields is more selective and efficient (and often much faster) than centrifugation or filtration (Yauvuz et. al., 2006).

High-gradient magnetic separation (HGMS) is commonly used term in magnetic separations. HGMS device comprises of a bed of magnetically susceptible filling (wires or balls) placed inside an electromagnet, as shown in Fig. 1.

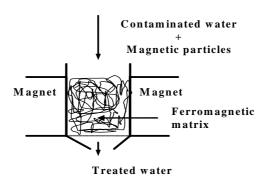


Fig. 1. A scheme of high gradient magnetic separation.

¹ Ing. Katarína Štefušová, PhD., Ing. Miroslava Václavíková, PhD., RNDr. Michal Lovás, PhD., Ing. Slavomír Hredzák, PhD., Institute of Geotechnics of the Slovak Academy of Sciences, Watsonova 45, SK-04353 Kosice, Slovakia, stefusova@saske.sk

When a magnetic field is applied across the column, the magnetic filling dehomogenizes the magnetic field in the column, producing large field gradients around the magnetic matrix that attract magnetic particles to their surfaces and trap them there. The collection of particles strongly depends on the creation of these large magnetic field gradients, as well as on the particle size and magnetic properties. For successful collection of magnetic particles by HGMS, the magnetic force attracting particles toward the magnetic matrix must dominate the fluid drag, gravitational, inertial, and diffusion forces as the particle suspension flows through the separator (Ambashta and Sillanpää, 2010).

Experimental

Materials and chemicals

Analytical grade chemicals were used in all experiments. Model solutions were prepared by dissolving Na₂HAsO₄.7H₂O in deionized water. The pH of the solutions was adjusted with suitable concentrations of NaOH and HNO₃.

Synthetic magnetite was prepared by co-precipitation of Fe(II) and Fe(III) in the presence of NaOH.

Sorbents were characterized by high-resolution transmission electron microscopy (JEOL 1200 EX) (Fig. 2), surface area measurements (Gemini 2360), zeta potential measurements (ZetaPlus, Brookhaven Instruments) and magnetic susceptibility measurements (KappaBridge KLY-2). The characteristics are given in Table 1.

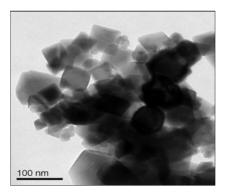


Fig. 2. HRTEM image of synthetic magnetite.

Tab. 1. The characteristics of synthetic magnetite.

Surface area [m².g¹]	125.7
pH _{PZC}	6.5
Magnetic susceptibility [10 ⁻⁶ SI units]	702 778

Methods

The sorption properties of synthetic magnetite were tested under batch conditions. The initial arsenic concentration was 100 mg.L⁻¹. The sorbent concentration was 2 g.L⁻¹. The experiments were performed at constant temperature $23\pm1^{\circ}$ C in a rotary shaker set at 30 rpm and equilibrium time 24 hours. The arsenic quantity in solutions was determined by AAS (Varian 240 RS/240 Z, Australia) before and after the sorption experiments. The amount of the arsenic sorbed (mg) per unit mass of magnetite (g) q_{eq} , was calculated using the Eq. (1)

$$q_{eq} = \frac{C_0 - C_{eq}}{C_s} \tag{1}$$

where C_0 and C_{eq} are initial and equilibrium arsenic concentration (mg.L⁻¹), respectively and C_s is the sorbent concentration in solution (g.L⁻¹).

Magnetic separation was performed using the high gradient magnetic separator (Jones), with the separation column of 30 mm in inner diameter and 320 mm in length. The separation column was packed with magnetically susceptible filling. The magnetic field was applied and then the sample was passed through the column. The magnetite nanoparticles were retained in the column as the solvent passed through and the solution without magnetite nanoparticles was obtained at the outlet of the column. The efficiency of magnetic separation was studied under various conditions. The effect of the magnetically susceptible filling type (stainless-steel wool or stainless-steel balls) and the intensity of magnetic field were investigated.

Results and discussion

The pH of arsenic solution is the most important factor influencing the sorption capacity of iron oxides. The effect of pH on the removal of As(V) within a range of 2-9 is given in Fig. 3. The removal of As(V) from aqueous solution decreased with increasing pH and the greatest arsenic uptake was observed at pH 2. The equilibrium As(V) uptake was 40.4 mg.g⁻¹ at this pH value. However, at pH 7.0 (typical for natural waters), gives also satisfactory results. The equilibrium As(V) uptake was 16.8 mg.g⁻¹ at this pH value. pH dependence of arsenic sorption onto magnetite was confirmed by others authors as well (Chunming and Puls, 2008; Giménez et al., 2007; Yean et al., 2005).

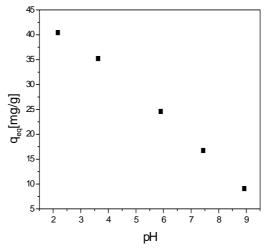


Fig. 3. The effect of pH on arsenic sorption.

The pH of solution plays an important role in arsenic sorption on iron oxides. In acidic environment, where maximum As(V) removal was observed, predominate form of As(V) is the single negatively charged H_2AsO_4 . In this pH range magnetite surface is positively charged. So, the attractive electrostatic forces between the negatively charged As(V) species and positively charged magnetite surface favour the sorption. The point of zero charge of sorbent was found at pH 6.5 (Table 1). Above this pH value magnetite surface is negatively charged and the double negatively charged form of $HAsO_4^{2-}$ predominates. The electrostatic forces are repulsive but sorption is still achieved due to the existence of specific forces.

Fig. 4 shows the effect of the magnetic matrix type and the intensity of magnetic field on S/L separation after arsenic adsorption. The results have shown that magnetite can be easily separated from the solution after arsenic sorption by magnetic filtration in high-gradient magnetic field. The retention of magnetite nanoparticles was around 99.9 %. There is a negligible effect of studied parameter on magnetic filtration of synthetic magnetite.

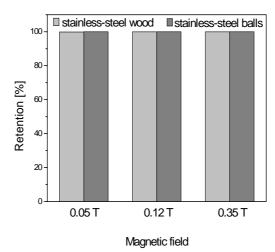


Fig. 4. The effect of magnetic matrix type and magnetic field on the magnetite retention.

Using of magnetic separation after arsenic sorption onto materials based on iron oxides was studied by several authors as well. Mayo et al. (2007) studied magnetic separation of the nanocrystalline magnetite in high-gradient magnetic column separator and they found that increasing the magnetic field the magnetic separation was more effective with increasing of. The size dependence of nanocrystalline magnetite particle in magnetic separation using high-gradient magnetic separator was studied by Yavuz et al. (2006). It was found that as the particle size became smaller, more magnetic field was required to ensure their complete separation.

Conclusion

Results of this study indicates that synthetic magnetite is a suitable sorbent for arsenic removal from water, especially at acidic environment. The maximum capacity of sorbent was 40.4 mg/g. It was found that magnetite nanoparticles can be easily removed from solution after arsenic sorption by high-gradient magnetic separation.

Acknowledgements: This work has been supported by Science and Technology Assistance Agency, project No. APVT-51-017104 and Scientific Grant Agency VEGA, project No. 2/0087/08.

References

- Ambashta, R. D., Sillanpää, M.: Water purification using magnetic assistance: A review. *J. Hazard. Mater.*, 2010, Vol. 180, 38-49.
- Deliyanni, E. A., Peleka, E. N., Gallios, G. P., Matis, K. A.: A critical review of the separation of arsenic oxyanions from dilute aqueous solution (the contribution of LGICT). *Int. J. Environment and Pollution*, accepted for publication (2010).
- Chunming, S., Puls, R. W.: Arsenate and Arsenite Sorption on Magnetite: Relations to Groundwater Arsenic Treatment Using Zerovalent Iron and Natural Attenuation. *Water Air Soil Pollut.*, 2008, Vol. 193, 65-78.
- Giménez, J., Martínez, M., Pablo, J., Rovira, M., Duro, L.: Arsenic sorption onto natural hematite, magnetite, and goethite. *J. Hazard Mater.*, 2007, Vol. 141, 575-580.
- Mandal, B. K., Suzuki, K. T.: Arsenic round the world: a review. Talanta, 2002, Vol. 58, 201-235.
- Mayo, J. T., Yavuz, C., Yean, S., Cong, L., Shipley, H., Yu, W., Falkner, J., Kan, A., Tomson, M., Colvin, V. L.: The effect of nanocrystalline magnetite size on arsenic removal. *Sci. Technol. Adv. Mater.*, 2007, *Vol.* 8, 71-75.
- Pokhrel, D., Viraraghavan, T.: Arsenic removal from an aqueous solution by modified A. niger biomass: Batch kinetic and isotherm studies. *J. Hazard. Mater.*, 2008, Vol. 150, 818-825.
- Vaclavikova, M., Gallios, G. P., Hredzak, S., Jakabsky, S.: Removal of arsenic from water streams: an overview of available techniques. *Clean Techn. Environ. Policy*, 2008, Vol. 10, 89-95.
- Yavuz, C. T., Mayo, J. T., Yu, W. W., Prakash, A., Falkner, J. C., Yean, S., Cong, L., Shipley, H. J., Kan, A., Tomson, M., Natelson, D., Colvin, V. L.: Low-field magnetic separation of monodisperse Fe₃O₄ nanocrystals. *Science*, 2006, Vol. 314, No. 5801, 964-967.
- Yean, S., Cong, L., Yavuz, C. T., Mayo, J. T., Yu, W. W., Kan, A. T., Colvin, V. L., Tomson, M. B.: Effect of magnetite particle size on adsorption and desorption of arsenite and arsenate. *J. Mater. Res.*, 2005, Vol. 20, 3255-3264.