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Arsenic removal from aqueous solution using pyrite

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ABSTRACT

Arsenic is one of the major pollutants and a worldwide concern because of its toxicity and chronic effects on human health. This investigation presents the results of arsenic adsorption onto four adsorbents such as pure pyrite, waste pyrite, dried waste pyrite and ground dried waste pyrite. The findings show that the addition of 5 g/L ground dried waste pyrite into the solution proved a high arsenic removal efficiency, with the ability to reduce the concentration of As(V) in the aqueous solution from an initial value of 10 mg/L to 10 µg/L at pH 5. The pH range of 4–6 for ground dried waste pyrite is effective to adsorb arsenic from solution. It is found that oxidation and the particle size of the pyrite has a big influence on arsenic removal. Following the adsorption, the separation of the adsorbent from the liquid phase was achieved using flotation and flocculation methods. Both methods were found to be effective for removing arsenic loaded pyrite from the solution. Test results were confirmed using arsenic containing process water sample to remove arsenic. 15 g/L of pyrite addition provided 99% arsenic removal at pH 5. The arsenic concentration was decreased from the as received 38.5 mg/L to 0.03 mg/L. High arsenic uptake capability and cost-effectiveness of waste pyrite make it potentially attractive for arsenic removal.

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1. Introduction

Arsenic is toxic and it is long term exposure can cause stomach-intestine, liver, kidney and heart disorders as well as neurological, dermal haematopoietic, reproductive, and carcinogenic diseases (Gehle, 2009; Nguyen et al., 2009). Problems concerning arsenic have been reported recently in the USA, China, Chile, Bangladesh, Taiwan, Mexico, Romania, the United Kingdom, Argentina, Poland, Canada, Hungary, New Zealand, Vietnam, Japan and India (Smedley and Kinniburgh, 2002). Turkey is one of the countries under the threat of arsenic pollution. Arsenic concentrations in aqueous systems range from 10 µg/L to 7754 µg/L in the west regions of Turkey (Başkan and Pala, 2009a; 2009b).

Depending on the redox conditions arsenic exists in the –3, 0, +3, +5 oxidation states. Arsenite (+3) and arsenate (+5) exists as oxyanions. While the monovalent anionic species of arsenate (H_2AsO_4^-) is predominant at pH 2–6, arsenite H_2AsO_3^- is significant at the pH range 9–12. In surface waters the oxidized forms as arsenite and arsenate are the most widespread species (Fig. 1). In soils arsenate is the predominant form under oxidizing conditions whereas arsenite is the major form of arsenic that occurs under reducing conditions (Turpeinen et al., 1999).

Arsenic contamination in aqueous systems required development of various arsenic removal methods. Iron or aluminium salts are commonly used in water purification (Lakshminathiraj et al., 2006; Aydoğan et al., 2012; Yenial et al., 2012). Adsorption is an economical and reliable technique (Kanel et al., 2006) and much more efficient alternative for removal of inorganic compounds than the conventional treatment (Vaishya and Gupta, 2006). The removal of arsenic accomplished by adsorption techniques wherein the use of adsorbents such as natural iron containing minerals (Aredes et al., 2012), titanium dioxide (Guan et al., 2012), zero-valent iron (Mamindy-Pajany et al., 2009), red mud (Li et al., 2010; Lopes et al., 2012), magnetic nanoparticles (Kim et al., 2012; Jin et al., 2012), have been investigated. Arsenic sorption onto natural and synthetic pyrite mineral has been examined by some researchers (Zouboulis et al., 1993; Jingtai and Fyfe, 2000; Farquhar et al., 2002; Han et al., 2013).

Pyrite is the most abundant sulphide mineral in the earth's crust and thermodynamically stable in many reducing conditions (Sun, 2010). Pyrite has a very low economic significance and it is mainly produced as a waste material in mineral processing plants. Pyrite is used for only sulphuric acid generation and usually stored in waste dumps or dry storage locations. The affinity of pyrite to arsenic makes it a favourable adsorption material. Furthermore, waste pyrite being a by-product of mineral processing plants is a cheap adsorbent source (Zouboulis et al., 1993).

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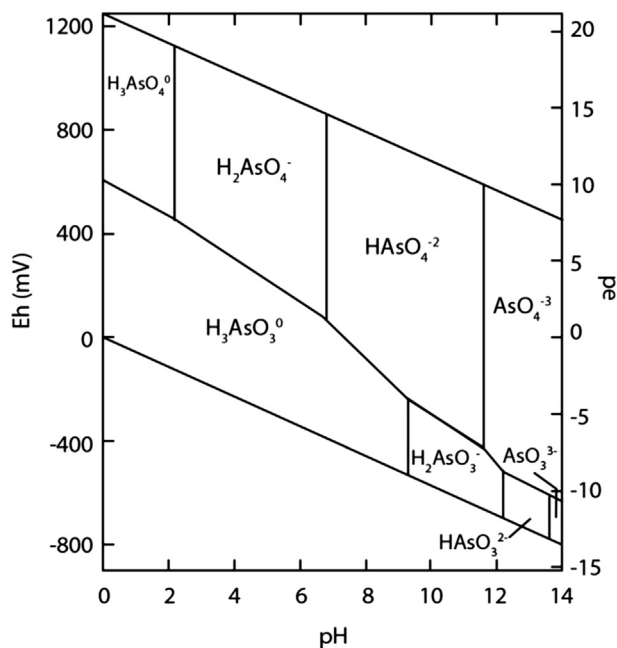


Fig. 1. Eh-pH diagram for As–O₂–H₂O (Smedley and Kinniburgh, 2002).

Table 1
Physical properties of pyrite samples used in the tests.

Sample name	Surface area, (m ² /g)	d80, mm	d50, mm
Pure pyrite (PP)	4.036	0.037	0.017
Waste pyrite (WP)	2.751	0.090	0.052
Dried waste pyrite (DP)	2.751	0.090	0.052
Ground dried waste pyrite (GP)	4.240	0.015	0.003

Siirt Madenköy Copper Plant in Turkey as the tailings of chalcopyrite flotation circuit where pyrite is depressed at pH 11.5 and chalcopyrite is floated using Aerophine 3418A as a collector. After floating chalcopyrite in the process, pyrite together with other re-jects are sent to the tailing dam. The pyrite waste contained 15% moisture and 66% pyrite. The samples used in the experiments were called pure pyrite (PP), waste pyrite (WP), dried waste pyrite (DP), and ground dried waste pyrite (GP). The sample tagged as “waste pyrite” is the original sample from tailings dam. “Dried waste pyrite” was the same sample that was dried for 2 days in an oven at 105 °C. “Ground dried pyrite” was obtained by dry grinding the waste pyrite in a vibrating ring mill. The physical and chemical properties of pyrite samples are shown in Tables 1 and 2, respectively. Surface areas of pyrite samples were determined by N₂ BET and the particle sizes were measured with Malvern (Mastersizer, 2000) particle sizer. Electrophoretic mobility measurements were carried out employing Zeta Metre 3.0 equipment using suspensions with 0.01 wt% solids content where HCl and NaOH were the pH regulators. All FTIR experiments were conducted using a Perkin Elmer Spectrum 65 FT-IR Spectrometer fitted with a horizontal ATR accessory as well as a Perkin–Elmer diffuse reflectance (DRIFT) accessory were used for sampling. Spectra of the samples were the result of 1000 co-added scans at a resolution of 4 cm⁻¹ unless otherwise noted.

2.2. Adsorption experiments

Adsorption experiments were designed to search for the optimal conditions under which pyrite samples could remove arsenic from the solutions. Tests were conducted using 100 mL solution containing 10 mg/L As(V), otherwise stated. The amount of pyrite in the solution was kept constant as 5 g/L. The beaker containing 100 mL solution and pyrite sample was agitated for 30 min using a magnetic stirrer while a pH metre monitored the pH of the solution. At the end of the duration, 10 mL of solution was drawn and clarified by passing through a disk filter with 0.20 μm pore size for arsenic analysis.

2.3. Solid-liquid separation experiments

After the adsorption of As(V) onto pyrite test samples settling and flotation methods were carried out to remove solids (pyrite)

Table 2
Chemical analysis of pure pyrite and waste pyrite.

Pure pyrite		Waste pyrite	
Component	Weight, %	Component	Weight, %
S	48.62	S	35.71
Fe	43.70	Fe	35.52
Al ₂ O ₃	0.68	Al ₂ O ₃	5.08
Cu	–	Cu	0.24
Zn	–	Zn	0.35
MgO	0.27	MgO	2.26
SiO ₂	0.97	SiO ₂	17.3
CaO	0.57	CaO	–

Arsenic is frequently associated with sulphide minerals and typically forms its own minerals like arsenopyrite, realgar and orpiment (Darnley et al., 1995). Iron-bearing sulphide minerals are largely responsible for the generation of acid mine drainage and consequently the release of toxic metals such as arsenic into the environment. Pyrite (FeS₂), the most common iron sulphide mineral, can incorporate large amounts of arsenic (Abraitis et al., 2004). In another words, pyrite can be both the source and the collector of arsenic (Han et al., 2013).

In this study, the uptake of As (V) species from aqueous solutions by adsorbing onto different pyrite samples was investigated. Flotation and flocculation methods were investigated for solid liquid separation. The test results were confirmed using process water sample collected from an operating colemanite concentrator.

2. Materials and methods

2.1. Materials

All chemicals used in this study were analytical grade. All experiments were carried out with distilled water. The pH was adjusted using NaOH and HCl when necessary. Synthetic water samples were prepared from 1000 ppm As (V) standard solution and diluted to desired concentrations for experiments. Process water sample was taken from the effluents of Kütahya Emet Colemanite Plant. Potassium ethyl ksantate (KEX), Methyl isobutyl carbinol (MIBC) were used as flotation chemicals, Superfloc A150 (Cyanamid), Superfloc N-300 (Cyanamid) and Zetag 7635 were used as flocculants.

Pure Pyrite samples used in the experiments originated from Mexico. Although the pyrite was of high purity, some further purification was needed. Therefore, after crushing the sample below 10 mm, coarse gangue minerals were removed by hand picking. Eventually the sample size reduced below 2 mm and the –0.3 mm portion was discarded. In order to achieve satisfactory purification –2+0.3 mm size fraction was successively fed to a high tension separator. The final product was washed with ethyl alcohol and stored in a glass bottle. The waste pyrite was obtained from

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