



Modified natural magnetite with Al and La ions for the adsorption of fluoride ions from aqueous solutions



J.J. García-Sánchez^{a,*}, M. Solache-Ríos^b, J.M. Martínez-Gutiérrez^c, N.V. Arteaga-Larios^c, M.C. Ojeda-Escamilla^c, I. Rodríguez-Torres^a

^a Instituto de Metalurgia–Facultad de Ingeniería, Universidad Autónoma de San Luis Potosí, Av. Sierra Leona 550, Lomas 2^a sección, 78210 San Luis Potosí, SLP, Mexico

^b Departamento de Química, Instituto Nacional de Investigaciones Nucleares, Apdo. Postal 18-1027, 11801 Mexico, D.F., Mexico

^c Instituto de Metalurgia, Universidad Autónoma de San Luis Potosí, Av. Sierra Leona 550, Lomas 2^a sección, 78210 San Luis Potosí, SLP, Mexico

ARTICLE INFO

Article history:

Received 22 January 2016

Received in revised form 25 April 2016

Accepted 2 May 2016

Available online 3 May 2016

Keywords:

Fluoride

Sorption

Magnetite

Aluminum

Lanthanum

ABSTRACT

Natural magnetite ore (Fe_3O_4) was modified with aluminum and lanthanum ions to increase the fluoride removal efficiency from aqueous solutions. The modified and unmodified magnetite samples were characterized by X-ray diffraction (XRD), scanning electron microscopy and energy dispersive spectroscopy (SEM/EDS), infrared spectroscopy (IR), surface area (BET) and zeta potential (ζ). The adsorption and stability of fluoride on magnetite were investigated using modified magnetite in batch systems. The SEM results indicate the formation of a lanthanum oxide layer around the magnetite and that aluminum oxide is dispersed between the particles. Al and La were deposited as hydroxides (IR), which increased the surface area by 7.5 and 9.5 times higher than natural magnetite and shifted the surface potential toward positive values. The IR results indicated that fluoride replaces a hydroxyl group from the aluminum hydroxides whereas no such replacement was detected for materials with La. Batch tests showed that adsorption increased from 30 to 90% and 96% for the aluminum and lanthanum modified materials, respectively.

© 2016 Elsevier B.V. All rights reserved.

1. Introduction

Groundwater and drinking water can become contaminated through naturally occurring chemicals and minerals (e.g., arsenic, nitrate, fluoride), and the presence of such contaminants can lead to adverse health effects. Excessive fluoride intake can result in the serious health issue called fluorosis, which is characterized by dental mottling and skeletal manifestations such as crippling deformities, osteoporosis, and osteosclerosis [1].

Fluoride occurs naturally in underground water, which may contain high levels of fluoride as the water table may be exposed to many inorganic fluoride-containing minerals. In Mexico, ~75% of the total population relies on groundwater for drinking. The co-occurrence of fluoride is especially found in arid and semi-arid regions, areas of geothermal activity, and in the mining areas in

north-central Mexico [2]. Generally, ingestion of water having a fluoride concentration above 1.5–2.0 mg/L may lead to dental mottling (fluorosis), which is characterized initially by opaque white patches on the teeth. In the advanced stages of dental fluorosis, teeth display a brown to black staining, followed by pitting of their surfaces [3]. Severe chronic and cumulative over exposure of fluoride can cause the incurable and crippling disease of skeletal fluorosis. The condition manifests itself as an increase in bone density, leading to a thickness of long bones and calcification of ligaments [4].

The available methods to control the concentration of fluoride ions in drinking water include electrochemical processes, coagulation–precipitation, ion exchange, and sorption [5–8]. Sorption is one of the most important methods employed for fluoride removal from groundwater because it is relatively inexpensive and simple to implement [9]. The parameters used to select a suitable adsorbent to remove fluoride are adsorption capacity, ease of operation, potential for reuse, and the feasibility for regeneration [10].

* Corresponding author.

E-mail address: jjuangsl@gmail.com (J.J. García-Sánchez).

Many adsorbents have been used to remove fluoride ions, such as metal oxides and hydroxides, biosorbents, carbonaceous materials, ion-exchange resins, zeolites, alumina, iron and calcium based adsorbents, bioadsorbents and minerals [11–13].

The physico-chemical properties of minerals such as surface area, porosity, crystal morphology, structure and composition are the basis for their technological applications, and thermal, mechanical or chemical treatment can modify these properties [14]. These treatments could trigger modifications to the surface characteristics, porosity and partially destroy the chemical structure [15]. Among the potential minerals for fluoride removal are iron ores such as hematite, siderite or magnetite. Patel et al. [16] prepared magnetite in the laboratory to evaluate its potential use for fluoride adsorption from water. In this case, an 84% removal of fluoride from a 6 mg L^{-1} fluoride solution was reported. Although magnetite is a potentially effective material for fluoride removal, relatively few studies have been reported.

Several types of iron ores have been chemically modified to enhance fluoride removal. For example, Teutli et al. [17–19] modified hematite with aluminum hydroxide to investigate its capacity to remove fluoride ions. It was reported that $\text{Al}(\text{OH})_3$ adhered as small particles on hematite and this modification increased the superficial area by 18%. Moreover, the treated mineral did not display any notable morphological differences with respect to unmodified hematite. Shan et al. [20] modified natural siderite using aluminum via a thermal treatment. It was found that the removal of fluoride increased because of the formation of hematite and $\gamma\text{-Al}_2\text{O}_3$.

Other modifications of iron minerals have been performed, for example, with lanthanum ions. Published results [20] indicate that lanthanum-modified hematite displays superior adsorption capacity than natural hematite, with an increase of the removal percentage from 30 to 60%; however, lanthanum could not be detected in the spectroscopic characterization.

In this paper, aluminum and lanthanum ions were incorporated as hydroxides to natural magnetite and their effects on the adsorption of fluoride ions were evaluated. The composition and structure of the natural and modified minerals were studied using X-ray diffraction (XRD), infrared spectroscopy (IR), scanning electron microscope (SEM) and energy dispersive spectroscopy (EDS). Potential zeta measurements were performed to identify the isoelectric point (IEP) of the modified materials and BET (Brunauer-Emmett-Teller) experiments were performed to determine the size of their surface areas.

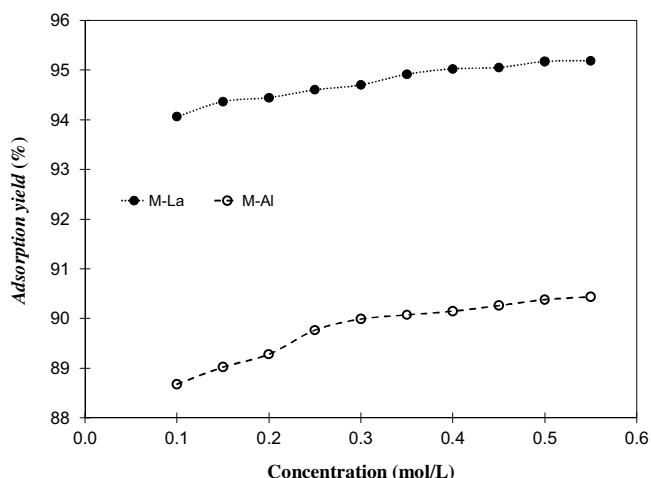


Fig. 1. Concentrations of Al or La versus adsorption capacities.

2. Experimental

2.1. Materials and solutions

The adsorbent used in this study was natural magnetite from Colima, México (Peña Colorada Co). The solid was crushed and sieved to the desired particle size of 0.029 mm. Aluminum sulfate (99.99% trace metals basis, Sigma–Aldrich), lanthanum chloride heptahydrate (99.9%, Fisher Scientific) and potassium hydroxide (ACS reagent, $\geq 97.0\%$, pellets, Sigma–Aldrich) were used to prepare the solutions.

The working standards of F^- were prepared from a fluoride standard solution (100 mg/L) of sodium fluoride (NaF) (Thermo Fisher Scientific, USA). An aqueous solution of fluoride ions (5 mg L^{-1}) was used for the F^- removal experiments. All solutions were prepared using deionized water.

2.2. Magnetite modification

Aluminum hydroxide or lanthanum hydroxide were prepared by adding a 2 M KOH solution to a 0.1 M aluminum sulfate or 0.1 M lanthanum chloride solution (100 mL), respectively, to obtain a solution mixture with a pH of 7.5. The precipitates were washed in triplicate with deionized water to eliminate K^+ , Cl^- and SO_4^{2-} ions from the solid. The aluminum or lanthanum hydroxides were mixed with magnetite (10 g) in 200 mL of deionized water, and the mixtures were shaken for 4 h. Finally, the water was decanted and the oxides were dried at 50°C for 24 h. The samples were labeled as M-Al and M-La for the aluminum hydroxide and lanthanum

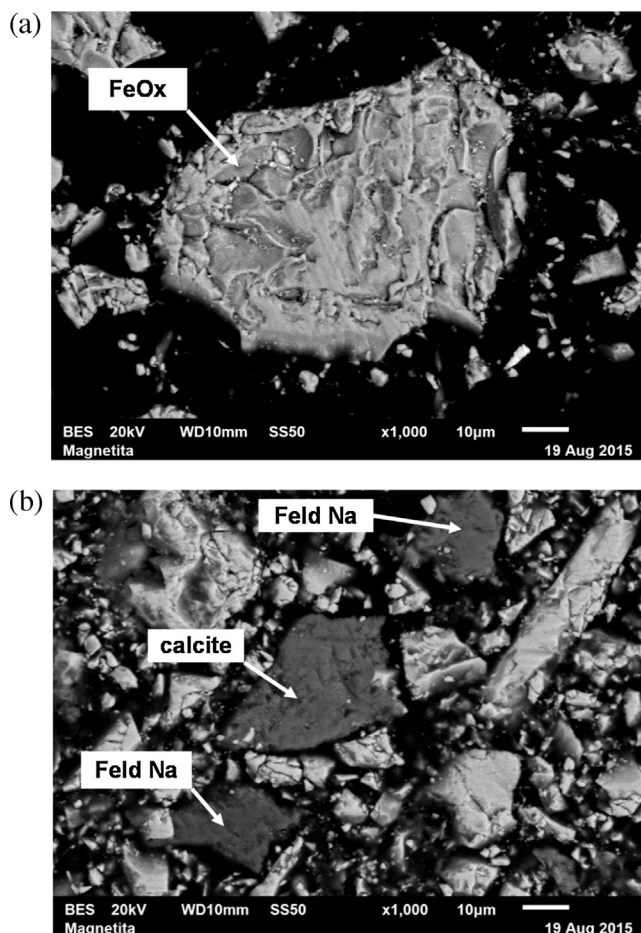


Fig. 2. (a) SEM images of natural magnetite. (b) Presence of feldspars and calcite.

Download English Version:

<https://daneshyari.com/en/article/1313579>

Download Persian Version:

<https://daneshyari.com/article/1313579>

[Daneshyari.com](https://daneshyari.com)