



Synthesis of magnetite nanoparticles from iron ore tailings using a novel reduction–precipitation method



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ABSTRACT

Magnetite nanoparticles successfully produced from a pregnant leach solution (PLS) of iron ore tailings via a simple reduction–precipitation method using $\text{Na}_2\text{S}_2\text{O}_3 \cdot 5\text{H}_2\text{O}$ reducing agent while the ratio of $[\text{Fe}^{3+}]$ to $[\text{S}_2\text{O}_3^{2-}]$ were 3 and 2 in the presence and absence of nitrogen gas respectively. The produced nanoparticles were characterized by X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), scanning and transmission electron microscopy (SEM, TEM), Brunauer–Emmett–Teller (BET), and vibrating sample magnetometry (VSM). The achieved XRD and FTIR results confirmed the presence of Fe_3O_4 nanoparticles while SEM and TEM images displayed that the formed nanoparticles were spherical with an average diameter of 19 ± 3 nm. The average surface area and the pore volume of produced nanoparticles were calculated to be $64.04 \text{ m}^2 \text{ g}^{-1}$ and $0.079 \text{ cm}^3/\text{g}$, respectively using the BET method. By performing VSM analysis and estimation of the low magnetic remanence (M_r) and coercive field (H_c), it was concluded that the produced nanoparticles are highly ferromagnetic at the room temperature.

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1. Introduction

Each year, about 5 million tons of tailings containing hematite and goethite are dumped from concentrator plants in Gol-Gohar mine complex of Sirjan, Iran. At present, about 30 million tons of the dry tailings are present in the complex with an average iron content higher than 25%. By 2025, this amount will be increased to 80 million tons thus it is essential to find some alternatives uses for this significant source of minerals.

One approach is to solubilize the iron ions in a pregnant leach solution (PLS) to produce magnetite (Fe_3O_4) nanoparticles. Magnetite nanoparticles can be used in a number of applications including waste water treatment [1], magnetic resonance imaging [2], manufacture of catalyst [3], synthesis of pigments, and magnetic storage [4]. To prepare PLS, HCl is recommended as the main acid leaching agent in the literature [5–11]. However, using highly concentrated HCl may boost the formation of impurities such as CaCl_2 and MgCl_2 in the PLS [10]. Therefore, using other alternative combined lixivants (hybrid lixiviant) containing an appropriate concentration of H^+ and Cl^- may optimize the tailings leachability while improving the leaching productivity. Table 1 represents most

of the successful methods that produced magnetite nanoparticles from waste and acid mine drainage (AMD) [5–14].

We herein describe a novel and simple reduction–precipitation method for Fe_3O_4 nanoparticles preparation from PLS of iron ore tailings leached by $\text{H}_2\text{SO}_4(5 \text{ M})/\text{NaCl}(2 \text{ M})$ reagents. It is assumed that $\text{H}_2\text{SO}_4/\text{NaCl}$ hybrid lixiviant decreases Ca^{2+} ions impurity in PLS due to the precipitation of gypsum. Before increasing the pH of PLS by the addition of ammonia, the Fe^{3+} was reduced using sodium thiosulfate pentahydrate ($\text{Na}_2\text{S}_2\text{O}_3 \cdot 5\text{H}_2\text{O}$) in the stoichiometric amount. The synthesized Fe_3O_4 nanoparticles were characterized by X-ray diffraction (XRD), Fourier transform infrared spectrum (FTIR), inductively coupled plasma–optical emission spectroscopy (ICP–OES), scanning and transmission electron microscopy (SEM, TEM), vibrating sample magnetometry (VSM) and the Brunauer–Emmette–Teller (BET) method.

2. Materials and methods

2.1. Materials

The iron ore tailings obtained from Gol-Gohar magnetite tailings dump in Sirjan, Iran. The elemental analysis of the tailings comprised 43.47% Fe, 7.99% Si, 9.07% Ca, 3.8% Mg, and 2.4% Al. We used 5 M H_2SO_4 and 2 M NaCl for the leaching process while it is assumed that NaCl will improve the H^+ activity by the addition of a

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Table 1
Various synthesis methods of magnetite nanoparticles from iron tailings and AMD.

| Studying procedure | Ref. |
|--|------------|
| $4\text{Fe}_2\text{O}_3$ (Iron ore tailings = $\text{Fe}_2\text{O}_3 + \text{SiO}_2 + \text{Al}_2\text{O}_3$) + Fe $\xrightarrow{\text{milling for } 2 \text{ h}}$ $3\text{Fe}_3\text{O}_4$ | [5] |
| $\text{Fe}_3\text{O}_4 + 8\text{HCl}$ + metallic Fe mixed in the tailings $\xrightarrow{95^\circ\text{C for } 4-5 \text{ h}}$ $2\text{FeCl}_3 + \text{FeCl}_2 + 4\text{H}_2\text{O}$ $2\text{FeCl}_3 + \text{FeCl}_2 \xrightarrow{\text{urea}(\text{CH}_4\text{N}_2\text{O}) \text{ at } 95^\circ\text{C}}$ Fe_3O_4 | [6] |
| iron ore tailings $\xrightarrow{\text{HCl}(37.5\%)}$ $\text{Fe}^{3+} + \text{Fe}^{2+}$ $\text{Fe}^{3+} + \text{Fe}^{2+} \xrightarrow{\text{H}_2\text{O}_2}$ Fe^{3+} Fe^{3+} heated to 60°C /pH adjusted to 3.2 $\xrightarrow{\text{Fe}(\text{OH})_3(\text{s})}$ Fe_2O_3 | [7] |
| $\text{Fe}_2\text{O}_3 \xrightarrow{\text{HCl}}$ $\text{FeCl}_3(\text{aq})$ $2\text{FeCl}_3(\text{aq}) + \text{FeSO}_4(\text{aq}) \xrightarrow{\downarrow \text{NaOH}(\text{drop slowly})}$ Fe_3O_4 | [8] |
| The tailing sample $\xrightarrow{\text{HCl at } 150^\circ\text{C for } 5-6 \text{ h}}$ $\text{FeCl}_3 + \text{SiO}_2 + \text{Al}_2\text{O}_3$ $4\text{FeCl}_3 + 3\text{NaBH}_4(\text{acid ascorbic}) + 9\text{H}_2\text{O} \rightarrow 4\text{Fe}(\text{metallic iron}) \downarrow + 3\text{NaH}_2\text{BO}_3 + 12\text{HCl} + 6\text{H}_2 \uparrow$ $2\text{FeCl}_3 + \text{Fe}(\text{metallic iron}) \rightarrow 3\text{FeCl}_2$ $2\text{FeCl}_3 + \text{FeCl}_2 \xrightarrow{\text{NH}_4\text{OH}(\text{pH} = 10) \downarrow}$ Fe_3O_4 | [9] |
| iron ore tailings _(iron, silica and alumina) $\xrightarrow{\text{HCl at } 80^\circ\text{C for } 1 \text{ h}}$ $\text{FeCl}_3(\text{aq})$ $\text{FeCl}_3(\text{aq}) + 3\text{NH}_4\text{OH} \rightarrow \text{Fe}(\text{OH})_3 + 3\text{NH}_4\text{Cl}$ washed with 1.0% HCl and distilled water $\text{Fe}(\text{OH})_3$ $\text{Fe}(\text{OH})_3 + \text{C} \xrightarrow{900-1300^\circ\text{C for } 4 \text{ h}(\text{reductive - roasting})}$ Fe_3O_4 | [10] |
| iron ore tailings _(iron, silica and alumina) $\xrightarrow{\text{HCl at } 80^\circ\text{C for } 1 \text{ h}}$ $\text{FeCl}_3(\text{aq})$ $\text{FeCl}_3(\text{aq}) + 3\text{NH}_4\text{OH} \rightarrow \text{Fe}(\text{OH})_3 + 3\text{NH}_4\text{Cl}$ Washed with 1.0% HCl and distilled water $\text{Fe}(\text{OH})_3$ $2\text{Fe}(\text{OH})_3 + \text{FeCl}_2 \xrightarrow{\text{in } 50\% \text{HCl}}$ $2\text{Fe}^{3+} + \text{Fe}^{2+}$ $2\text{Fe}^{3+} + \text{Fe}^{2+} \xrightarrow{\text{NaOH (1.5M) at } 70^\circ\text{C in } \text{N}_2 \text{ gas}}$ Fe_3O_4 | [11] |
| Crushing of the magnetite iron ore _{(less than 149μm)}} $\xrightarrow{\text{HCl}(0.66 \text{ g ore/ml HCl) at } 100^\circ\text{C for } 2 \text{ h}}$ $\text{Fe}^{3+} + \text{Fe}^{2+} + \text{other ions}$ $\text{Fe}^{3+} + \text{Fe}^{2+} + \text{other ions} \xrightarrow{\text{H}_2\text{O}_2}$ Fe^{3+} $\text{Fe}^{3+} + \text{NaBH}_4 \rightarrow 2\text{Fe}^{3+} + \text{Fe}^{2+}$ $2\text{Fe}^{3+} + \text{Fe}^{2+} \xrightarrow{2\text{M NaOH at } 40^\circ\text{C}}$ Fe_3O_4 | [12] |
| IOT _(iron ore tailings) $\xrightarrow{\text{HCl at } 90^\circ\text{C (w/w} = 1)}$ $\text{Fe}^{3+} + \text{Fe}^{2+} + \text{other ions}$ $\text{Fe}^{3+} + \text{Fe}^{2+} + \text{other ions} \xrightarrow{\text{H}_2\text{O}_2 \text{ at } 60^\circ\text{C}}$ $\text{Fe}^{3+} + \text{other ions}$ $\text{Fe}^{3+} + \text{NH}_4\text{OH} \rightarrow 2\text{Fe}(\text{OH})_3 \xrightarrow{\text{HCl}(1:1)}$ Fe^{3+} $2\text{Fe}^{3+} + \text{FeSO}_4 \cdot 7\text{H}_2\text{O} + \text{GO}_{(\text{graphen oxide } 3\text{gr/l})} \xrightarrow{\text{Na}_4\text{OH}}$ Suspension $\xrightarrow{160^\circ\text{C for } 12 \text{ h}}$ Fe_3O_4 | [13] |
| $\text{FeSO}_4 \cdot 7\text{H}_2\text{O} + \text{FeS}_2 \xrightarrow{400^\circ\text{C for } 10 \text{ min}}$ $3\text{Fe}_3\text{O}_4 + 10\text{SO}_2(\text{g}) + 56\text{H}_2\text{O}(\text{g})$ | [14] |
| acid mine drainage (AMD) $\xrightarrow{\text{Oxidation}(\text{pH}=2.3)}$ $\text{Fe}(\text{OH})_3$ $\text{Fe}(\text{OH})_3 + \text{HCl} \rightarrow \text{FeCl}_3$ $2\text{FeCl}_3 + \text{FeSO}_4 \cdot 7\text{H}_2\text{O}(\text{Merk}) \xrightarrow{\text{NaOH}\&\text{NH}_4\text{OH}(\text{pH} = 11.5)}$ $\text{Fe}_3\text{O}_4(51\% \text{maghemite}, 26\% \text{Magnetite}, 22\% \text{Hematite}, 1\% \text{other forms of iron oxide})$ | [15] |
| Acid mine drainage (AMD) _(Coal Mine) $\xrightarrow{\text{H}_2\text{O}_2(\text{pH} = 3.5-4 \text{ with the addition of } 4\text{N sodium hydroxide})}$ $\text{Fe}(\text{OH})_3$ $2\text{Fe}(\text{OH})_3 + 3\text{H}_2\text{SO}_4 \rightarrow \text{Fe}_2(\text{SO}_4)_3 + 6\text{H}_2\text{O}$ $2\text{Fe}_2(\text{SO}_4)_3 + \text{FeSO}_4 \cdot 7\text{H}_2\text{O}(\text{Merk}) \xrightarrow{6.4\text{M}(\text{NH}_4\text{OH})(\text{pH}=9.5) \text{ without the presence of oxygen}}$ Fe_3O_4 | This study |
| crushing of the iron ore tailings _{(D₈₀=150μm)}} $\xrightarrow{5\text{M H}_2\text{SO}_4/2\text{M NaCl at } 85^\circ\text{C for } 2 \text{ h}}$ $\text{Fe}^{3+} + \text{other ions}$ $\text{Fe}^{3+} + \text{Na}_2\text{S}_2\text{O}_3 \cdot 5\text{H}_2\text{O}(\text{reducing agent}) \xrightarrow{[\text{Fe}^{3+}/\text{S}_2\text{O}_3^{2-}] = 3}$ $2\text{Fe}^{3+} + \text{Fe}^{2+}$ $2\text{Fe}^{3+} + \text{Fe}^{2+} \xrightarrow{\text{NH}_4\text{OH}(2.5\text{M}, \text{pH} = 9.5) \text{ in the presence of } \text{N}_2 \text{ gas at } 25^\circ\text{C}}$ $(\text{Fe}^{3+})_2\text{Fe}^{2+}(\text{OH})_8$ or $\text{Fe}_3(\text{OH})_{8(\text{s})}$ $\text{Fe}_3(\text{OH})_{8(\text{s})} \xrightarrow{85^\circ\text{C} - 100^\circ\text{C}}$ $\text{Fe}_3\text{O}_4 + 4\text{H}_2\text{O}$ | |

necessary amount of Cl^- . Sodium thiosulfate pentahydrate (99.5%, $\text{Na}_2\text{S}_2\text{O}_3 \cdot 5\text{H}_2\text{O}$), ammonium hydroxide (25–30%, NH_4OH), deionized water, and ethanol ($\text{CH}_3\text{CH}_2\text{OH}$, 99.93%) were used in the synthesis of nanoparticles.

2.2. Leaching process

Prior to the acid leaching, the samples were ball milled to d_{80} 75 μm and immersed in a glass reactor (1000 ml) in a water bath at 85°C with a constant stirring condition (700 rpm). After 120 min, the leachate was filtered and the solution was chemically analyzed by ion chromatography (Waters Company) and ICP-OES (Model 730-ES, Varian Company, USA). The specification of prepared PLS is

mentioned in Table 2.

2.3. Nanoparticles' preparation procedure

$\text{Na}_2\text{S}_2\text{O}_3$ and NH_4OH were used to reduce the Fe^{3+} to Fe^{2+} and precipitate the PLS solution respectively. Thus, 5.3 ml of 1 M $\text{Na}_2\text{S}_2\text{O}_3 \cdot 5\text{H}_2\text{O}$ was mixed with 25 ml of the PLS solution to maintain the $\text{Fe}^{3+}/\text{Fe}^{2+} = 2$. The initial color of the mixture was changed from yellow to violet and turned back to yellow after $\text{Na}_2\text{S}_2\text{O}_3$ was added to PLS. The solution thereafter was mechanically stirred at 700 rpm while the nitrogen gas was circulated into the flask at the room temperature (25°C). Furthermore, 8 ml of 1 M $\text{Na}_2\text{S}_2\text{O}_3 \cdot 5\text{H}_2\text{O}$ was mixed with 25 ml of PLS solution in the absence of nitrogen gas

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