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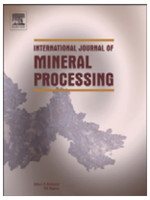

High efficiency iron removal from quartz sand using phosphoric acid

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ABSTRACT

Phosphoric acid (H_3PO_4) was firstly used to remove iron impurities at very low concentrations from quartz sand under ultrasound irradiation with higher efficiency, simpler technology and is less harmful to the target product compared with the other comparable methods that use industrial strong acids such as sulfuric (H_2SO_4), hydrochloric (HCl), and even hydrofluoric (HF) acids. Our results have revealed that phosphoric acid is a good leaching agent for iron removal from quartz sand because of its leaching efficiency up to 77.1%. It is about 30%–40% higher than the other comparable methods, and leaching time consumption is less than the others. The optimal values of operating parameters, such as types and concentrations of the leaching agents, leaching temperature, particle sizes of quartz sand, s/l mass ratio, and with/without ultrasound irradiation, are also examined. The optimal leaching rate (the maximum in the conditions considered in this context is at least 81.0%) of iron removal is given by a consideration of higher leaching efficiency, lower concentrations of acids used, less energy consumed and less leaching time used.

1. Introduction

Iron is one of the most troublesome impurities in silica sand; the presence of iron compounds leads to an unacceptable color and damages its property greatly. At the present, one of the main tasks in the purification of silica sand is to further remove iron compounds to a required level by a highly efficient method. A wide variety of methods have been examined including physical (Mowla et al., 2008; Palaniandy et al., 2007), biological (Styriaková et al., 2003; Sarvamangala and Natarajan, 2011), and chemical methods (Zhou, 2005; Liu et al., 1996; Zhang, 2001; Shen and Peng, 2008; Vegliό et al., 1998; Jin et al., 2004; Vegliό et al., 1999; Farmer et al., 2000; Snoswell et al., 2005). In conventional methods, HF and non-HF inorganic acids are used to remove iron. In many cases, HF has to be mixed with inorganic strong acids such as H_2SO_4, HNO_3 and HCl to raise leaching efficiency of the impurities and to reduce reaction loss of the target product. For example, Zhou (2005) used 28.6% HF acid at 120 °C for leaching for 4 h; they obtained a leaching percentage of 74.7% of the total eight impurities, including aluminum, iron, titanium, etc. Liu et al. (1996) used a mixture of 10% HF acid and 90% H_2SO_4, HCl and HNO_3 for 3–12 h; they obtained 96.3% of iron removal. These leaching efficiencies appear to be very good, however, because of the target product lost, high energy consumed and a worsened working environment; the use of this method is now declining. To search for a new leaching method with high efficiency and of non-fluoride acid, researchers have to either extend leaching time or to raise the concentrations of industrial strong acids, or to lift the leaching temperature to remedy the loss of the leaching efficiency due to the absence of HF acid. For example, Zhang (2001) reported that he removed 39% of iron impurities from the silica sand by 10% HCl at 80 °C for 8 h. Further, he removed iron impurities from the silica sand for 2 h, and obtained a leaching percentage of 75.6% by 1200 °C of hot pure HCl gas. Although the latter looked good, it consumed a lot of energy and caused more pollution because of the high concentration and temperature of HCl gas. Shen and Peng (2008) studied the removal of iron impurities from Gaoping silica sand using 0.5 M H_2SO_4 and leached for 96 h; 48.2% of iron impurities in the silica sand were removed from their experiments. Vegliό et al. (1998) leached iron with H_2SO_4 at 95 °C for 4.0–5.0 h, and obtained 3.0–9.0% iron removal. It is clear that though inorganic acids as leaching agents in removing iron impurities do not hurt the target product, the residual liquors of these strong inorganic acids are often unacceptable to environment and are corrosive to the leaching equipment. Differing from the above methods, which usually use sample powders of quartz sand from the market, Jin et al. (2004) started from dressing mineral rock to raise the leaching efficiency, by first roasting the rock to 850 °C, Then it was quenched by cold water to make the rock break into many pieces in a loose way; the broken rocks were ground to 200 μm, equivalent to 70 mesh on average, then leached by H_2SO_4 at 250 °C for 40 min; and they finally obtained the leaching percentage of 96.5% of iron oxide. This result looked excellent, however, existing equipments and flow sheet have to be changed exclusively for this method. The method is unable to use existing commercial powders of quartz sand; lengthy flow sheet and high-energy consumption are also unacceptable. Recently, organic acids have been used in iron removal and have shown a greater potential. Among these, oxalic acid is certified to be the best one of all the acids owing to its better complexing ability and higher reducing power. Vegliό et al. (1999) leached iron from the silica sand by oxalic acid and obtained

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45–50% iron removal in the conditions of temperature 80 °C, average size 160 μm, 3 g/l oxalic acid and 10% (w/v) ratio of solid to liquid for 3 h. Du et al. (2011) used the oxalic acid in leaching iron and obtained 75.4% leaching rate in the conditions of 4 g/l oxalic acid, 95 °C, 500 rpm/150 W. Li et al. (2010) used a mixture of a large quantity of a very diluted HCl and a small quantity of $H_2C_2O_4$ to remove aluminum. Chiarizia and Horwitz (1991) used phosphoric acid to dissolve α-FeOOH and concluded that HEDPA (a derivative of the phosphoric acid) was extremely effective in leaching iron because of its strong complexing ability. Though these methods used the renewable oxalic acid, to take a whole, the leach efficiency is unsatisfactory and the leaching time is usually long. At the same time, ultrasound-assisted leaching processes have demonstrated its superiority in removing impurities from different types of minerals (Wilson et al., 2006, 2007). To date in the published literature, little information is available in the application of phosphoric acid to remove iron impurities from silica sand.

The aim of this project is to examine the high effectiveness of H_3PO_4 in removing iron impurities from silica sand by a simple method from the Ningliu quartz sand. Further, an attempt is also made to optimize the process parameters and to examine the effects of combined ultrasound, conventional stirring and the new leaching agent on leaching percentages.

2. Experimental

2.1. Materials

The silica sand in these experiments was purchased from Ningliu Quartz Filter Co. Ltd., China. The chemical compositions of the silica sand sample are presented in Table 1. The silica sand sample was ground and sieved to less than 100 mesh in size (equivalent to 150 μm) before use. All reagents in these experiments are of analytical grade. Distilled and deionized water were used for all experiments.

Table 1
Chemical compositions (%) of the silica sand sample.[a]

Components	Amount (%)
SiO_2	98.5674
Al_2O_3	0.5590
Na_2O	0.3969
K_2O	0.3052
CaO	0.0546
Fe_2O_3	0.0481
TiO_2	0.0194
MgO	0.0167
Sum	99.9703

[a] The oxides of less than 0.0167% are not shown.

2.2. Experimental set-up

A batch reactor used in this experiment is an ultrasonic cleaner bath (KQ-250B), which was made by Ultrasonic Instrument Company of Kunshan in China with dimensions of (L×W×H) 35×30×40 (cm) and mounted on an ultrasound generator with a frequency of 40 kHz and power of 150 W. The ultrasonic cleaning bath is equipped with a thermocouple connected to a digital instrument for measuring temperatures of reaction systems; a continuous circulation system of cooling water was used to control the pre-selected temperatures of the systems (Zhao et al., 2007; Ji et al., 2006). A schematic illustration of the experimental set-up is shown in Fig. 1.

2.3. Methods

For each experiment, 100 ml of leaching acid was poured into a 250 ml rounded three-necked flask, which was covered by a plug with a mechanical stirrer at 500 rpm in all the experiments so that the sample was sufficiently contacted with acids, and the mouth of the flask was covered by two lids in case of acidic evaporation. The flask was placed in the ultrasonic cleaning bath and irradiated by an ultrasonic generator of 150 W power throughout the experiments unless otherwise. After the pre-selected temperature was reached and maintained for at least 30 min, then the required amount of dry silica sand was introduced into the flask. At the end of the experiments, the samples were filtered, the residues were washed by deionized water for 2–3 times and the filtered liquors, together with washed water, were used to determine the extraction efficiency of the iron impurities by ICP-OES (Vista-MPX, Varian, USA). The content of iron impurities in both residues and filtered liquors was measured to guarantee the reliability of the analysis data. The percentage of iron removal E was calculated according to the following equation (Zhao et al., 2007):

$$E = \frac{\alpha - \beta}{\gamma - \beta} \times 100\% \qquad (1)$$

where E (%) is the percentage of iron removal; α is the iron content in leached solution; β is the background iron content carried by leaching agents (acid, water); γ is the iron content of aqua regia liquor after 4 h of leaching in the presence of an ultrasound which is considered to be the maximum value of iron removal (Zhao et al., 2007).

3. Results and discussion

3.1. Effects of different types of acids on leaching efficiency of iron impurities

The effects of acids H_3PO_4, $H_2C_2O_4$, HNO_3, H_2SO_4 and HCl on percentages of iron removal at 80 °C with a mass ratio of solid to liquid 1:9 (i.e., s/l = 10.0%) for 60 min in the presence of ultrasound are shown in Fig. 2. It is seen from Fig. 2 that percentages of iron removal correspondingly increase with increasing concentrations of the different acids, but the increments are quite different: of which the leaching increment of phosphoric acid is the largest, i.e., iron removal yields (up to 81.0%) by H_3PO_4 are far superior to those by the other acids. The removing percentages by HNO_3, H_2SO_4 and HCl are not only lower than that by H_3PO_4, but also lower than that by $H_2C_2O_4$ under the same conditions. For example, when the acidic concentration increased from 0 M to 3 M, the percentages of iron removal by H_3PO_4 increased greatly from 1.22% to 81.00%; but only 67.12% by $H_2C_2O_4$, 53.26% by HNO_3, 49.83% by HCl, and 42.53% by H_2SO_4. Further, the rates of iron removal of all acids increased rapidly with an increase of acidic concentrations from 0 to 0.5 M, after which the increments of iron removal seemed to be slowed. The explanation of this observation can be attributed to the fundamental molecular theory of liquids, i.e., the most dilute acid has the largest activity, so the reaction speed is fastest at the initial stage, after that, the activity of the acids is gradually dropping with increasing concentrations, so that the removing curves gradually flatten out. The leaching curves at 60-mesh silica sand show a similar trend as that at 100 mesh (so the figure is not given for conciseness), i.e. iron removal rate by H_3PO_4 is still obviously larger than those by the other acids at all acidic concentrations considered. However, the maximum value of iron removal by H_3PO_4 dropped to 61.67% from 81.00% as the particle size increased from 100 to 60 mesh, and iron removal rates by all the other acids were also decreased. This indicates that the particle sizes of the silica sand have a remarkable effect on the efficiency of iron impurity removal. It decreased the leaching percentage of about 20% in this case.

We understand that larger solid particle sizes in leaching processes usually lead to poorer yields, however, to search for an optimal particle size technically, larger and smaller particle sizes around 100 mesh have to be examined to show that the selected particle size is to be the best one by experimental data or the best one after a compromise between higher leaching yields (corresponding smaller solid particle) and less