

## Iron removal in production of purified quartz by hydrometallurgical process

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### ABSTRACT

Iron, the predominant impurity in quartz ores, can be substantially removed via aqueous chemical processes using organic and inorganic acids. Quartz was highly purified by leaching with sulfuric acid ( $\text{H}_2\text{SO}_4$ ) in the presence of reducing agents (oxalic acid, citric acid, and glucose). The effects of the parameters in the leaching tests were evaluated with the full factorial design method and the analysis of variance (ANOVA) method. Higher amounts of ferric oxide ( $\text{Fe}_2\text{O}_3$ ) were removed in the presence of reducing agents during  $\text{H}_2\text{SO}_4$  leaching. Under optimum conditions (20% pulp density, 0.5 M  $\text{H}_2\text{SO}_4$ , 10 g/L of oxalic acid, 90 °C, and 120 min), 98.9% of  $\text{Fe}_2\text{O}_3$  was removed, resulting in a final quartz product with 1 ppm of  $\text{Fe}_2\text{O}_3$  in the presence of oxalic acid in  $\text{H}_2\text{SO}_4$  leaching. Although oxalic acid showed particularly significant effects, citric acid and glucose also produced quartz products with low  $\text{Fe}_2\text{O}_3$  content (1.8 and 3.1 ppm of  $\text{Fe}_2\text{O}_3$ , respectively). The models obtained from the data of  $\text{H}_2\text{SO}_4$  leaching with the addition of oxalic acid, citric acid, and glucose, based on the ANOVA results, were suitable for estimating the leaching yield. The process was simulated within the scope of economic modeling studies. These studies showed the economic feasibility of quartz purification by hydrometallurgical processes in obtaining quartz products with high market value and high quality.

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### 1. Introduction

The physical, chemical, and mineralogical characteristics of iron-containing minerals; appropriate reagents (organic and/or inorganic acid); and optimum conditions must be determined in leaching procedures (Vegliò et al., 1998; Banza et al., 2006; Tuncuk and Akcil, 2014). Vegliò et al. (1998) stated that sulfuric acid ( $\text{H}_2\text{SO}_4$ ) leaching alone is not very effective in removing iron from a quartz ore, although the addition of oxalic acid increases its efficiency (35–45% Fe). They reasoned that the high concentration of iron in mica lowered the efficiency of its removal. Lee et al. (2007) demonstrated that goethite ( $\text{FeOOH}$ ) dissolves more rapidly and efficiently than ferric oxide ( $\text{Fe}_2\text{O}_3$ ). In a study on the dissolution of some iron minerals in hydrochloric acid and oxalic acid, iron oxyhydroxides such as  $\text{FeOOH}$  were found to dissolve under oxidizing conditions (0.6–0.8 V (Ag/AgCl ref.)),  $\text{Fe}^{+3}$  by forming oxalate complexes, and minerals such as hematite ( $\alpha\text{-Fe}_2\text{O}_3$ ) and maghemite ( $\gamma\text{-Fe}_2\text{O}_3$ ) only by reduction (Cepriá et al., 2003). These studies demonstrate the significance of the structure of iron in its removal from quartz.

Nowadays, purified quartz is widely used in several high technology applications; such as industry of optical fibers, construction of silicon cells for use in photovoltaic systems, semiconductors for the electronic industry, industrial catalytic chemistry for the synthesis of catalysts,

zeolites and adsorbent materials (Vatalis et al., 2015). When quartz contains less than 50  $\mu\text{g/g}$  of impurities, which mainly contaminating trace elements (especially Fe and B, Li, Al, Ge, Ti, Mn, Ca, K, Na, P) in the quartz lattice, quartz is described as purified (Müller et al., 2007). Generally purified quartz has  $\text{Fe}_2\text{O}_3 < 15$  ppm,  $\text{Al}_2\text{O}_3 < 300$  ppm and alkali earth oxides  $< 150$  ppm (Richard Flook; Vatalis et al., 2015). In several high technology applications, silica glass needs to have adequate properties, such as chemical purity, optical transparency and radiation resistance (Santos et al., 2015). Also impurities especially iron occurring in these quartz productions is harmful as it impairs transmission in optical fibers and the transparency of glasses, it discolors ceramic products and lowers the melting point of refractory materials. (Taxiarchou et al., 1997a).

Chemical methods include leaching of minerals using organic and inorganic acids (Panias et al., 1996). The most commonly used inorganic acids are hydrofluoric, hydrochloric, sulfuric, and perchloric acids. These acids, especially HF, can be extremely hazardous to health and the environment, and they should eventually be recycled; HF, in particular, causes undesirable quartz mass losses (Santos et al., 2015). However, the most important advantage of inorganic acids is their low cost compared with organic acids (Taxiarchou et al., 1997b). However, products treated with inorganic acids such as  $\text{H}_2\text{SO}_4$  and hydrochloric acid (HCl) must be washed/cleaned thoroughly after the leaching procedure, because it is contaminated with sulfate ( $\text{SO}_4^{2-}$ ) and chloride ( $\text{Cl}^-$ ) ions. Organic acids are more commonly preferred than inorganic acids as

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iron is dissolved more rapidly and efficiently (especially with oxalic acid and citric acid). They also prevent the precipitation of iron (increasing its solubility) by forming a complex with the dissolved iron such that leaching can proceed in a wide pH range.

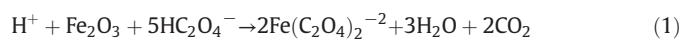
Oxalic, citric, ascorbic, acetic, fumaric, and tartaric acids are used for their ability to dissolve iron and other metal oxides. Among these organic acids, oxalic, citric, and ascorbic acids are carboxylic acids, which are used as alternatives to other inorganic acids that are most commonly used as effective dissolvent reagents and low-cost leaching reagents (Panijs et al., 1996; Ambikadevi and Lalithambika, 2000). In addition, the rate of impurity removal (Fe, Al, Na, K, Ca, Mg) can be increased partly with the use of complexing agents (citric acid, oxalic acid, acetic acid, humic acid, ethylenediaminetetraacetic acid (EDTA), and thiourea) (Zhong et al., 2014).

Among other organic acids, oxalic acid can be used as a leaching reagent to clean industrial minerals, because of its high efficiency, good complexation property, and good reducing activity. In addition, oxalic acid can be economically obtained as a by-product from other industrial procedures. In leaching procedures with added oxalic acid, the dissolved iron waste can be precipitated from the leaching solution as iron oxalate, which can then be transformed into pure Fe<sub>2</sub>O<sub>3</sub> by calcination. Similarly, the oxalate that remains in the mineral phase is removed in the form of carbon dioxide during the thermal procedure (for example, during the kiln drying stage in ceramic production) (Lee et al., 2006). Oxalic acid was deemed the most appropriate leaching reagent in a study conducted by Ambikadevi and Lalithambika (2000), evaluating the performance of different organic acids (acetic, formic, citric, ascorbic, succinic, tartaric, and oxalic acids) in removing iron from a kaolin clay sample used as a raw material for ceramic.

The reaction mechanism of iron oxide dissolution by organic acids was explained in the following three steps: adsorption of organic ligands in the solution on the surface of the iron oxide, dissolution, and reducing dissolution during the autocatalytic process. This mechanism shows that protons participate in the dissolution process (Panijs et al., 1996). The temperature and pH of the solution affect the dissolution of Fe<sub>2</sub>O<sub>3</sub> in acidic oxalate solutions (Taxiarchou et al., 1997a). The dissolution of Fe<sub>2</sub>O<sub>3</sub> in oxalic acid can be prevented by the formation of a Fe<sup>+2</sup>-oxalate product layer at a pH value between 1.6 and 3.2 (Lee et al., 2007; Salmimies et al., 2012).

Many environmental factors such as pH, organic compounds, and the intensity and wavelength of the sunlight have been found to influence the dissolution of Fe<sup>+3</sup> in aqueous solution systems and the balance between Fe<sup>+2</sup> and Fe<sup>+3</sup> (Voelker et al., 1997; Song et al., 2005). Formate, acetate, citrate, and oxalate (Graedel et al., 1986; Zuo and Holgne, 1992) are common organic ligands, of which oxalate was found to be most effective ligand in the dissolution of iron. Fe<sup>+3</sup> was observed to dissolve and adsorb onto oxalate ions at a pH of ~2.5 (Xu and Gao, 2008). Many researchers have used oxalic acid in the dissolution of iron oxides (Segal and Sellers, 1984; Cornell and Schindler, 1987; Jepson, 1988; Panijs et al., 1996; Vegliò et al., 1998; Lee et al., 2007).

Iron oxide is dissolved during leaching with oxalic acid via a photoelectrochemical reduction process that includes the load transfer mechanism between the dominant types of ferric oxalate (Fe(C<sub>2</sub>O<sub>4</sub>)<sub>3</sub><sup>-3</sup>) and ferrous oxalate, which serves as an autocatalyst (Fe(C<sub>2</sub>O<sub>4</sub>)<sub>2</sub><sup>-2</sup>) (Blesa et al., 1987; Taxiarchou et al., 1997b). The reducing mechanism in this case is a redox reaction, with oxalate being oxidized to form carbonic acid or carbon dioxide, which occurs in two half-cells, and Fe<sub>2</sub>O<sub>3</sub> being reduced to form Fe<sup>+2</sup>-oxalate (Eq. 1). This redox reaction indicates that hydrogen ions, oxalate, and iron oxide (Fe<sub>2</sub>O<sub>3</sub> particles) are involved in the leaching procedure. The kinetics of the reaction is determined by the optimum pH (2.5–3.0), temperature, oxalate concentration and particle size of Fe<sub>2</sub>O<sub>3</sub> (Lee et al., 2006):



Carbohydrates such as glucose, sucrose, and lactose are also used as reducing agents in acidic solutions. Glucose (Trifoni et al., 2001; Ghafarizadeh et al., 2011) was used as a reducing agent in typical acid leaching procedures to recover manganese from manganese dioxide (MnO<sub>2</sub>)-containing ores/wastes. Within the scope of these studies, dissolution of Fe<sub>2</sub>O<sub>3</sub> contained in the ore/waste as Fe<sup>+2</sup> into the solution was observed, with possible recovery.

The present study aims to investigate quartz purification by leaching and process simulation within the scope of economic modeling. Parameters such as reagent concentration, temperature, pulp density, effect of different reducing agents, and leaching duration were studied in detail to determine optimum conditions for purifying quartz via the factorial design method.

## 2. Experimental

### 2.1. Experimental procedure

The quartz ore used in the experiments was provided from Kaltun Madencilik San. ve Tic. A.Ş. (Cine, Aydın, Turkey). The reserve of quartz ore deposit is more than eight million tons, and the average Fe<sub>2</sub>O<sub>3</sub> content of the samples used in this study is 310 ppm. The quartz samples were analyzed using an X-ray fluorescence (XRF, Spectro Xepos) method.

For all experiments, 95–98% H<sub>2</sub>SO<sub>4</sub> solution (Merck) was used as a stock solution and very pure (>99%) oxalic acid (Merck), citric acid (Merck), and glucose (Merck) were used as reducing agents. Distilled water was used for the stock solutions and dilutions.

To prepare the quartz sample for the leaching procedure, quartz ore of approximately 5-cm size was subjected to various size reduction procedures (crushing and grinding), with a final size of ~500 µm. In particular, the Fe<sub>2</sub>O<sub>3</sub> content of the quartz samples of each size was analyzed based on sieve fractions after grinding; the subfractions with high Fe<sub>2</sub>O<sub>3</sub> content in all quartz samples were, in turn, separated using sieves (Retsch) before leaching and enriched according to particle size.

All leaching tests were performed in 600-mL reactors with glass lids in an analysis volume of 300 mL. The experiment samples were mixed using a magnetic mixer with heater (Velp, Arc) and a digital overhead mixer (Teflon-coated impeller tip) (Heidolph, RZR 2021) at a mixing rate of 200 rpm. At the end of the leaching procedure, the filtered and washed samples were dried at 105 °C in the drying oven for 24 h, and the chemical composition of the samples was determined with XRF.

The leaching tests were based on 2<sup>n</sup> full factorial experimental design by considering four different factors and using the Yates experimental design technique. The factors tested for all leaching tests and their levels are presented in Table 1. The levels were chosen at specified rates in order to calculate a wide reflection surface area with analysis of variance (ANOVA) (Montgomery, 1991). The models were derived from the evaluated data using the multiple linear regression method with Minitab 14 Statistical Software based on the Yates experimental design technique.

### 2.2. Economic modeling

The factorial design for the leaching tests is generally used as a statistical approach to increase the efficiency of the production process and quality of the product and to determine the effect of product inputs on the product. Therefore, data from leaching tests were found to be suitable for use in economic modeling studies. For this purpose, SuperPro Designer Version 2.71 Software, which was developed by Intelligen Inc., was used. The facility was simulated with the software; the units required for the facility, the operating conditions of these units, the reactions realized in the units, and the required chemicals were determined and entered in the system; and the facility flow diagram, costs, expenditures, the amount of product obtained, and its economic value were stated.

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