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The influence of pre-oxidation and leaching parameters on Iranian ilmenite concentrate leaching efficiency: Optimization and measurement

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ABSTRACT

This research was performed to model and optimize the response of Iranian ilmenite to pre-oxidation and leaching parameters using fractional factorial design. The Ti and Fe leaching percentages were considered as leaching response with respect to leaching parameters as the particle size, temperature, time, hydrochloric acid molarity, solid to liquid ratio and stirring speed. In order to detect the effect of pre-oxidation on leaching process half of the samples were oxidized at 900 °C for 120 min. Then experiments were performed at the designed condition and consequently Ti and Fe leaching percentages were measured using an atomic absorption spectroscopy. Finally the most significant factors affected on Fe and Ti leaching percentages and optimum conditions were obtained using analysis of variance. It was concluded that using the statistical method is useful not only for optimizing and modeling the leaching parameters but also for understanding the detrimental effect of pre-oxidation on Fe and Ti extraction.

1. Introduction

White titanium dioxide pigments have been produced by two processes, namely the sulfate process, and the dry chlorination process. The sulfate process utilizes ilmenite ($FeTiO_3$) as a raw material [1,2] while the dry chlorination process utilizes rutile (TiO_2) [3] or high titanium slag. The chloride process is a high energy consuming process [4].

There are several processes for the production of synthetic rutile from ilmenite, most of which uses upgrading of ilmenite by acid leaching which leads to a high quality product. For example in a process named Huelva factory process (a kind of sulfate process) concentrated sulfuric acid (H_2SO_4) is used to dissolve the titaniferous feedstock, ilmenite (around 50% of TiO_2) and slag (around 75% of TiO_2), which are milled and dried beforehand to aid the digestion process [5].

The literature reviews show that the pre-leach heat treatments increase the rate of extraction of iron [6,7]. Additions to the leach liquor of ferrous chloride [8] and alcohols [9] increase the rate of iron extraction [10]. Other authors [11] have suggested the use of a carbothermic reduction followed by hydrochloric acid leaching. A stage of oxidation prior to reduction has also been proposed in elsewhere [12,13]. Nevertheless, the use of two heat treatments before leaching made the process more energy consumer [14].

Numerous researchers discussed the effect of prior oxidation on the subsequent reduction or/and leaching efficiency and the morphology of oxidation, reduction products [13,15,16]. Some of them reported that pre-oxidation increases the maximum attainable extent of reduction [13,17], enhances the reduction rate of the iron oxide

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[18,19], decrease the reduction temperature [18] and enhances the rate of leaching of the reduced iron [14,20,21]. But Jones [17] and Janssen and Putnis [22] concluded contradictorily that oxidation prior to reduction has detrimental effect on the rates of subsequent reduction and/or leaching of iron respectively, while Ogasawara and Veloso de Araújo [23] reported that oxidation prior to reduction does not significantly alter the leaching results.

A large number of experimental investigations have been carried out relating iron and rutile recovery, but only few of them substantiated their observations with a mathematical model. However, present day industrial application demands comprehensive theoretical simulation before actual design [24–26]. There are very few studies about using factorial design in Fe and Ti leaching [27].

The kinetics of the Iranian ilmenite concentrate oxidation was studied in the previous work [28], therefore the objective of the present research is about studying Ti and Fe leaching efficiency of Iranian ilmenite concentrate (with TiO_2 content higher than 40%) with regard to the effect of pre-oxidation treatment and the effect of significant parameters using factorial design for the first time.

2. Experimental procedure

2.1. Material

Commercial Kahnouj ilmenite concentrate was used as the starting raw material. The chemical composition of this material was as shown in Table 1. The weight ratio of titanium to iron in ilmenite concentrate was 0.74:1.

2.2. Oxidation

Ilmenite concentrate powders were ground in a rotating disk mill and dried before investigation. Then classified in 54–83 and 80–100 μm particle size fractions by the standard DIN sieves (DIN 4188) and dried to constant mass at temperature 250 °C in order to remove the moisture and volatile compounds. Thermal oxidation of the samples to produce pre-oxidized samples were carried out in 900 °C for the maximum time of 120 min. Samples of ilmenite concentrate were placed inside the furnace having a specified temperature for 120 min. Each sample was then taken out of the furnace and cooled down under argon atmosphere. Then its weight was measured by Sartorius AG Gottingen scale (GC 1603 S-OCE, Germany). This procedure was performed to determine the maximum attainable oxidation for each particle size fraction.

2.3. Leaching

Leaching of the raw and pre-oxidized powders was carried out in a three-necked Pyrex glass reactor (250 mL) filled with 100 mL and 5.21–9.13 M hydrochloric acid. A thermometer was fitted to one of the openings. The reactor was heated in a mantle to the required temperature and controlled at the desired temperature using a power stat. The temperature during the leaching experiments could be controlled to within ±2 °C. The ilmenite sample was added to a preheated acid solution. The stirring speed was varied from 400 to 600 rpm. The leaching experiments were carried out isothermally at different temperatures (65, 80 and 95 °C) and for various durations (60–120 min) in separate batch experiments. At the end of each leaching experiment, the solutions were filtered. The Ti and Fe content in the solution were determined using an atomic absorption spectrophotometer (GBC Avanta). Calibration using standard solutions was done prior to the analysis every 10 analysis. For better understanding a schematic sketch of the process are illustrated in Fig. 1.

2.4. Experimental design

The fractional factorial method is a combination of mathematical and statistical technique. It is economical for characterizing a complicated process. It requires fewer experiments in order to study all levels of all input parameters, and filters out some effects due to statistical variation [29]. The 2^k factorial design is basically a series of experiments involving k factors that each of them has two levels. Fractional factorial designs are the most widely used designs in experimental investigations, and mainly used for the screening portion of experiments.

In the present work fractional factorial design consists of four center points for each design was applied for experiments with numerical parameters in order to evaluate mathematical models for responses and optimize the leaching parameters (Fig. 1). Here, two 2^{6-2} designs consist of 40 sets of runs; six selected independent process parameters (particle size, temperature, time, hydrochloric acid molarity, solid to liquid ratio, and stirring speed) were used for design of experiments (Tables 2 and 3). The measured response parameters were the Fe and Ti leaching percentages in both pre-oxidized and without pre-oxidized (regular leaching) conditions. The results obtained through the experiments and predicted models are summarized in Table 3 in both regular leaching and pre-oxidized concentrate leaching, respectively. According to Table 3 the experiments were performed randomly at the given runs and at the center point the experiments were repeated so as to avoid any systematic error entering into the system. The available data have been analyzed by analysis of variance (ANOVA) using Design-Expert 6.11 software, Stat-Ease, Inc., Minneapolis, USA.

3. Results and discussion

3.1. Regular leaching (without pre-oxidation)

3.1.1. Modeling and statistical analysis

Fully randomized factorial design, with various combinations of acid concentration (A), S/L ratio (B), temperature (C), stirring speed (D), particle size (E), and time (F), was used. The value of main factors, symbols, and their levels are listed in Table 2. Selected fractional factorial design (2^{6-2}) consists of 16 experiments and 4 center points. Center points are used to test the curvature of the model. According to this design two generators of E = ABC and F = BCD were defined. Using these generators, three