



Leaching of copper oxide with different acid solutions

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ABSTRACT

In this study, the dissolution of CuO in HCl, H₂SO₄, HNO₃ and citric acid solutions was investigated in a batch reactor employing parameters expected to affect the dissolution rate of copper such as stirring speed, temperature and acid concentration. It was found that 99.95% of copper was dissolved after 14 min with inorganic acids at 0.5 M, 25 °C and l/s = 10 ml/g while more drastic operation conditions were needed to reach the same dissolution efficiency with citric acid solution. Anions seem to be involved in the surface reaction. The dissolution kinetics of CuO was examined according to heterogeneous model and it was found that the dissolution rate was controlled by surface chemical process in all cases.

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1. Introduction

Hydrometallurgical extraction of metals is a branch of industry for which the research work is ongoing to develop processes which are less costly, more environmentally friendly and acceptable economically. In hydrometallurgical extraction, many leaching agents are used such as HCl, HNO₃ and H₂SO₄. However, most of these leachants cause environmental pollutions. Organic acids such as citric, acetic or oxalic acids can be attractive leaching agents as the leaching is carried out at moderate acidic conditions [1–3]. However, they cannot be used as leaching agents for hard dissolving compounds due of their weak acidities, low boiling temperatures and decompositions.

Copper is used in vast variety of products in domestic and industrial domains as thermal and electrical conductor and as a constituent of various metal alloys. Copper is also used in chemical industry as catalyst in the oxidative conversion of ethyl acetate in water [4], hydrogen production by partial oxidation of methanol [5], liquid-phase oxidation of benzene to phenol [6], carbon monoxide oxidation [7] and in removal of NO_x and SO_x from flue gases [8]. After deactivation, the components of the catalyst can be reused as secondary sources of metals. This is considered as more beneficial from environmental and economical point of views than landfill depositing.

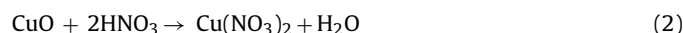
Leaching processes of metallic copper or ores containing copper in the divalent state have been the subject of many research works in recent years [9–13]. However, no study was found concerning the

dissolution kinetics of CuO/Al₂O₃ catalyst in inorganic or organic acid solutions.

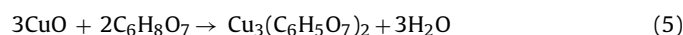
Copper (II) oxide is a basic oxide, so it dissolves in mineral acids according to the general reaction [14]:



In case of hydrochloric acid, sulfuric acid or nitric acid the reaction becomes:



Copper oxide can also be dissolved by a weak acid such as citric acid:



This article presents the results on copper and alumina separation from CuO/Al₂O₃ catalyst treated in fresh form with HCl, HNO₃, H₂SO₄ and citric acid solutions. The effects of acid concentration, stirring speed and temperature on the dissolution rate were evaluated with the four acid solutions. The dissolution kinetics were examined according to the heterogeneous reaction models and the best fitted equation to the experimental data was determined.

2. Experimental

The catalyst used in this study was a 10%CuO/α-Al₂O₃ (n/n) prepared with incipient wetness method. In this method, a defined volume of Cu(NO₃)₂·3H₂O (99.6%, Fluka) with known concentration was used to impregnate a given mass of α-Al₂O₃. With this

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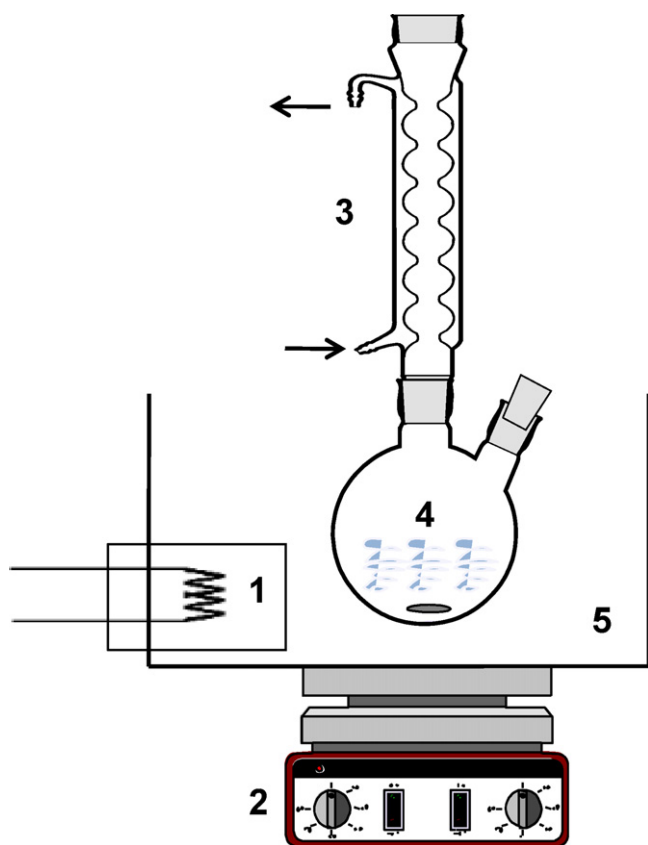


Fig. 1. Dissolution experiment setup: (1) temperature-controller; (2) magnetic stirrer; (3) glass condenser; (4) glass reactor; (5) water bath.

technique the load of copper can be calculated in a simple way. The sample was dried overnight at 110 °C and calcined in oven at 500 °C for 2 h. Copper composition was checked after that by atomic absorption. The obtained powder was crushed into particle size of 80 μm. H₂SO₄ (96–98%, Biochem), HNO₃ (60%, Cheminova), HCl (37%, Carlo Erba) and citric acid monohydrate (C₆H₈O₇, 99%, Alfa Aesar) were used as leaching reagents. Leaching experiments were carried out in a spherical glass batch reactor of 100 ml heated by a temperature-controlled water bath and equipped with a return-flow cooler (glass condenser) to minimize solution losses due to evaporation (Fig. 1). The solutions were mixed using a magnetic stirrer to eliminate the influence of mixing and mass transfer on the kinetic results. A typical experiment was conducted as follows: 0.2 g of the sample was placed into the glass flask with a desired volume of the selected acid. After the leaching process, the reaction mixture was filtered and Cu²⁺ was analyzed by titration with EDTA using murexide as an indicator. The percentage of dissolution was calculated from the following equation:

$$\frac{\text{Mol number of copper in the solution}}{\text{Mol number of copper in the catalyst leached}} \times 100 \quad (6)$$

The data presented are an average of two test replicates with an error of 5%.

Leaching behavior of CuO was tested under reaction conditions which were characterized by a relatively high excess of acids to eliminate possible effects of the changes in lixiviant composition during individual runs on the rate of leaching. The main parameters that affect the dissolution of copper oxide such as the nature of acid (HCl, H₂SO₄, HNO₃ and citric acid), acid concentration (0.1, 0.5, 1, 2 M), stirring speed (50, 200, 400 rpm) and temperature (30, 40, 50, 60 and 80 °C) were considered and the best conditions for the maximum recovery were established.

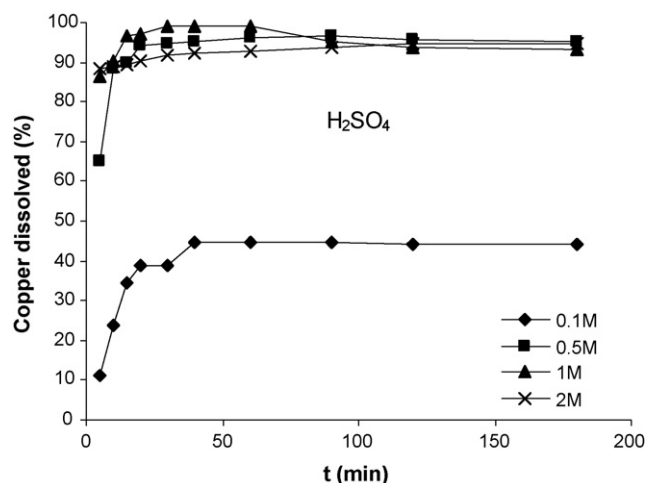


Fig. 2. Effect of H₂SO₄ concentration on copper dissolution. Operation conditions: T = 25 °C, l/s = 10 ml/g, without agitation.

3. Results and discussion

3.1. Inorganic acids

3.1.1. Effect of acid concentration

In order to investigate the effect of acid concentration, experiments were performed first with H₂SO₄ solution used as a reference with different concentrations (0.1, 0.5, 1, 2 M) without agitation, at 25 °C and l/s ratio of 10 ml/g. The results are shown in Fig. 2 as a fraction of copper reacted vs. time. It can be seen that the fraction of copper dissolved was not changed when acid concentration was increased from 0.5 to 1 M. The concentration of 0.5 M was selected for investigating the effect of other parameters. The same experiments were carried out after that with HCl and HNO₃ solutions under the same operation conditions and 0.5 M of acid concentration to compare their dissolution efficiencies (Fig. 3). It can be seen that copper dissolution was very fast with HCl. It attained 85% at the first minute and reached 99.95% at 14 min. The lowest kinetic dissolution was obtained with HNO₃ at the start of the reaction (1–6 min) but it increased gradually attaining 99.95% at 14 min. Alumina dissolution was checked with the four acid solutions several times at different concentrations. No dissolution was observed due to its chemical inertia and its high stability against acids [15]. In

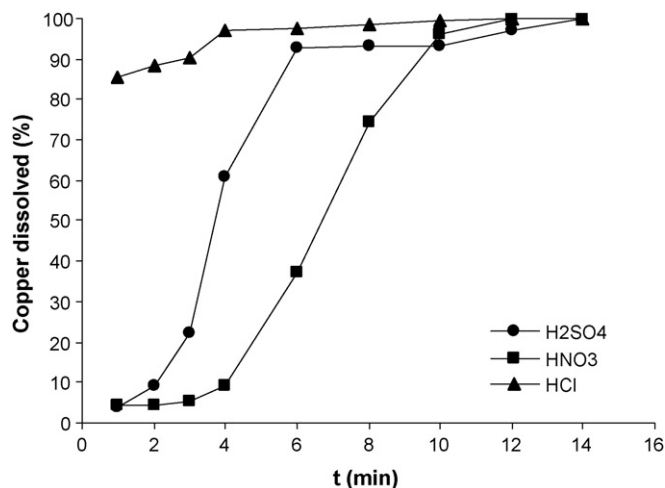


Fig. 3. Effect of the nature of acid on copper dissolution. Operation conditions: T = 25 °C, acid concentration = 0.5 M, l/s = 10 ml/g, without agitation.

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