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ORIGINAL ARTICLE

Leaching and recovery of zinc and copper from brass slag by sulfuric acid

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Recovery;
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Abstract Leaching and recovery processes for zinc and copper from brass slag by sulfuric acid were carried out and iron and aluminum were also precipitated as hydroxides in addition to silica gel. The factors affecting the performance and efficiency of the leaching process; such as agitation rate, leaching time, acid concentration and temperature were separately investigated. The results obtained revealed that zinc and copper are successfully recovered from these secondary resources, where the percent recovery amounts to 95% and 99% for zinc and copper, respectively. The experimental data of this leaching process were well interpreted with the shrinking core model under chemically controlled processes. The apparent activation energy for the leaching of zinc has been evaluated using the Arrhenius expression. Based on the experimental results, a separation method and a flow sheet were developed and tested to separate zinc, copper, iron, aluminum and silica gel from the brass slag.

1. Introduction

The increasing demand of zinc and copper in the world has required intensive studies for the recovery and extraction of these metals from different sources. Also, the tremendous increase in the use of these metals over the past few decades has inevitably resulted in an increased flux of metallic substances in the environment. Recovery and extraction of these metals is necessary and important both from an economic point of view and due to the increased requirement for environmental protection. Therefore, it is essential to remove these elements from industrial effluents and radioactive wastes before discharging into natural water bodies or onto land (El Dessouky et al., 2008). Ores, secondary materials, wastes, etc. are the most important sources for some metals like copper, nickel, cobalt and zinc. The leaching of Cu and Zn from ground samples of spent particles of boat paint was studied by Jessop and Turner (2011). Huge quantities of slag, which is a waste by-product of smelting and converting operations in metallurgical plants, is posing a potential environmental threat due to entrained values of base metals and sulfur. High temperature pressure oxidative acid leaching of nickel smelter slags was investigated as a process to facilitate slag cleaning

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and selective dissolution of base metals for economic recovery (Yunjiao et al., 2008). Hydrometallurgical process to recover and recycle metals from the wastes of brass which contain copper, zinc and lead using different leaching agents including sulfuric acid, ammonia, hydrochloric acid, cyanide and acetic acid was developed by Nesbitt and Xue (1995). It is reported that sulfuric acid containing copper sulfate with dissolved oxygen is the most successful leaching agent (Sharma and Row, 1985).

The pyrometallurgical recovery of zinc from secondary resources using different waste materials has been studied under different conditions, the average process efficiency values were variable (Lopez et al., 1988) However, the hydrometallurgical recovery of zinc metal or its salts was also described in the literature by Barrett et al. (Barrett and Nenniger, 1992; Barrett et al., 1992). The leaching of zinc from a sphalerite concentrate using hydrogen peroxide as an oxidant in sulfuric acid solutions was examined by Pecina et al. (2008). A study was performed to assess the influence of different factors such as acid and peroxide concentrations, particle size, reaction time, and temperature on zinc dissolution kinetics and the global reaction rate is governed by the chemical reaction rate. Shibasaki and Hasegawa (1992) carried out a combined hydrometallurgical treatment of copper smelter dust and lead smelter copper dross. It was shown that about 80% of the copper in the dross could be extracted at 80°C with 200 kg/m^3 sulfuric acid; 150 kg $solid/m^3$ and 4–6 h of leaching under oxidizing conditions. In comparison, the leaching of the copper smelter dust was relatively easy. Bzura (1979), studied the recovery of all metal values from brass foundry skimmings containing > 85% metals, primarily copper and zinc in alloyed form and zinc oxide. The skimmings were crushed, ball milled and reacted with HCl and sulfuric acid at pH 0.1–1.5. The resulting solution of zinc salts was continuously separated.

In Egypt, several types of zinc-containing waste materials are available. These include zinc scrap, spent dry cell batteries; zinc dross, resulting from the zinc cathode industry, and zinc ash, obtained from galvanizing processes. The latter two materials contain chlorides, which render the recovery of zinc by sulfuric acid electrowinning unsuitable (Rabah and EI-Sayed, 1995).

Different processes, such as cementation, can be used to remove and separate Cu^{2+} from different solutions and from leach solutions.

The aim of this work is to recover and separate metal values such as zinc and copper from brass by applying a hydrometallurgical method using sulfuric acid as the leaching agent. Different parameters affecting the recovery processes such as agitation rate, leaching time, acid concentration, liquid/solid mass ratio, and temperature of the system were investigated. Kinetics of the system and preliminary economic study was performed to evaluate the cost of the products compared to market prices. A flow sheet is developed and tested to produce high purity metal oxides.

2. Experimental

2.1. Materials

The slag sample used in this investigation is given from a brass manufacturing plant in Cairo, Egypt, whose chemical composition is given in Table 1 determined by Energy Dispersive X-ray. The concentration of the metals were estimated using Shimadzu UV/VIS, double beam recording spectrophotometer, Model 160-A, Japan.

Table 1 Chemical composition of the brass slag.

Element	Zn*	Cu*	Fe*	Al*	Si*	Cl	Ca
Contents	69	13	1.3	4.4	6.0	4.6	1.7

* Present as oxides.

Mineralogical analysis of the sample was determined by XRD using a Shimadzu-6000) (Japan) diffractometer. The conditions of these measurements were obtained using Cu-K α radiation and Ni filter. Analysis of the diffraction patterns was made by the whole powder pattern-fitting computer program intra-connected to the system.

The hydrogen ion concentration for the solutions was measured using a digital pH meter of the Hanna instruments type at the ambient laboratory temperature of 25 ± 1°C.

2.2. Leaching of slag

In the leaching process, 5 g of ground slag that was below 100 μm and was not exposed to any metallurgical pre-treatment operation was added to 25 g of 30% H_2SO_4 for 10 min in 250 ml conical flask with an agitation rate of 150 rpm at 35°C unless otherwise said. One milliliter solution sample was taken using a syringe filter of 1 mm pore size. The samples were chemically analyzed for the determination of zinc and copper content. After that, the percentage recovery of zinc and copper was calculated.

3. Results and discussion

3.1. Leaching

3.1.1. Effect of agitation rate

The results of the effect of agitation rate on zinc and copper dissolution by 30% H_2SO_4 with leaching time 10 min and at 35°C investigated in the range of 60–400 rpm with liquid/solid mass ratio of 5/1 are shown in Fig. 1. The presented data show that the recovery of zinc was increased from 60% at 50 rpm to around 93% at 150 rpm and the recovery of copper was kept around 9% with various stirring speeds. Therefore, the dissolution process does not seem to be controlled by mass transfer through the liquid boundary layer, despite the change in solution viscosity caused by the formation of silica gel. As a result, the agitation rate was kept at 150 rpm, unless otherwise stated.

3.1.2. Effect of leaching time

The effect of leaching time on the leaching of 5 g of slag using 25 g of 30% H_2SO_4, keeping the agitation rate at 150 rpm and temperature at 35°C, was studied in the range of 5–80 min, Fig. 2. It is clear that the recovery of zinc increases with increasing time up to 10 min then decreases with a further increase in time. The recovery of copper increased with time up to 30 min where it reached 95% then decreased with a further increase in time. This behavior may be due to the competition between zinc and copper ions in solution. Besides, with