Hydrometallurgy 137 (2013) 92–100

Contents lists available at SciVerse ScienceDirect

Hydrometallurgy

journal homepage: www.elsevier.com/locate/hydromet

Electronic waste treatment: Part 1. Autoclave oxidation–leaching using pyrite waste from mine tailing

Atsushi Shibayama *, William Tongamp [1], Batnasan Altansukh [1], Kazutoshi Haga [1], Akira Hosoi [1]

Faculty of Engineering and Resource Science, Akita University, 1-1 Tegata-Gakuen cho, Akita 010-8502, Japan

ARTICLE INFO

Article history:
Received 5 July 2011
Received in revised form 22 April 2013
Accepted 23 May 2013
Available online 29 May 2013

Keywords:
Electronic waste
Mine tailings
Pyrite
Autoclave oxidation
Copper

ABSTRACT

A process to co-treat electronic and pyrite (FeS_2) wastes in an autoclave (high pressure and temperature) under oxygen gas atmosphere for extraction of metals from electronic waste was investigated in this study. The electronic waste sample contained high Cu (13–20%), Au (230–360 g/t), Ag (480–1020 g/t), including Pt, Pd and Ni among many other metals. Pyrite samples obtained as mine tailings for the study contained 69.1% FeS_2 (32.5% Fe and 36.6% S) and was used as sulfur source for the generation of H_2SO_4 during autoclave oxidation to subsequently extract Cu and other base metals in the electronic waste. Reaction temperature and time, autoclave pressure, pulp density and H_2SO_4 concentration were investigated to evaluate metals extraction. At temperatures ranging from 120 to 180 °C and autoclave pressure of 2 MPa, pyrite was completely oxidized. Subsequently, Cu extraction reached over 99% and very high extraction of Zn, Ni and Cr metals was achieved within 1 h. The solid products from autoclave oxidation containing precious metals (Au, Ag, Pt, Pd) can be treated separately for their recovery.

1. Introduction

Electronic wastes in general refer to end of life or discarded electronic appliances including computers, mobile telephones, televisions, etc., and their amount is continuously increasing due to increased global demand. These wastes contain high content of valuable metals such as Cu, Pt, Pd, Au, and Ag. An assessment of global electronic waste and its environmental impacts is discussed by Robinson (2009); Widmer et al. (2005), and the status of electronic waste recycling in the U.S. by Kang and Schoenung (2005). Many articles are widely available in literature such as Cui and Zhang (2008), giving a review of the metallurgical recovery of metals from electronic wastes, discussing in detail both pyrometallurgical and hydrometallurgical processes for the treatment of electronic wastes.

Pyrite (FeS_2) on the other hand is the dominant gangue mineral in sulfide ore deposits (of copper, lead, zinc, molybdenum), usually separated by flotation to improve the grade of the valuable ores for subsequent processing. Often pyrite wastes from mining operations are directly disposed into water systems or kept in open stockpiles which pose environmental concern for acid mine drainage (AMD) and heavy metal release. When exposed to air and moisture, pyrite and other sulfide containing minerals oxidize with both oxygen and ferric ions as oxidizers. Stringent environmental regulations now demand proper control and storage of such wastes and many papers discuss contamination issues related to pyrite and control options (García et al., 2005; Kock and Schippers, 2006; Moreno and Neretnieks, 2006; Simón et al., 2001, 2005).

The possibility of utilizing pyrite waste as a source of sulfur for the generation of sulfuric acid (H_2SO_4) during autoclave oxidation for extraction of metals from the electronic waste was investigated in this study. Pyrite ores and concentrates with high gold (Au) and silver (Ag) are usually treated in an autoclave under oxygen gas atmosphere and H_2SO_4 is generated as a by-product. Thomas (2005) provides an overview of autoclave oxidation and its application and specific reference to pyrite containing high Au and individually discussed by other researchers (Gudyanga et al., 1999; Long and Dixon, 2004; Vračar and Cerović, 1997). The high pressure and temperature environment allows destruction of the sulfide matrix and free gold for leaching and recovery and simultaneously generates large amounts of sulfuric acid (H_2SO_4).

A mine tailings sample containing 69% FeS_2 was used in the current study to provide H_2SO_4 during autoclave oxidation for dissolution of metals contained in the electronic waste. In a separate study by Vračar and Cerović (2000), FeS_2 was used as a source of sulfur for H_2SO_4 formation in an autoclave for subsequent leaching of manganese from MnO_2.

The objective of this study was to evaluate extraction behavior of metals contained in electronic wastes using pyrite as source of sulfuric acid under varying experimental conditions and establish optimum conditions for metal extraction and recovery. Reaction temperature and time, autoclave pressure, pulp density, acid type and concentration were used as major parameters to evaluate metal extraction.

---

* Corresponding author. Tel./fax: +81 18 889 3051, +81 18 889 3052.
E-mail address: sibayama@ipc.akita-u.ac.jp (A. Shibayama).
[1] Tel./fax: +81 18 889 3052.

0304-386X/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.hydromet.2013.05.007

2. Experimental

2.1. Samples

Two separate waste samples from different sources were used in this study; an electronics waste sample containing high copper and precious metals and a mine tailings waste sample containing high pyrite. The electronic waste sample was prepared as follows; (a) shredded to 2–3 cm, (b) shredded and milled to −106 μm and (c) shredded and incinerated at 800 °C followed by milling to −100 μm. Chemical analysis of the incinerated sample indicated a high Cu content at 20.4% with Au and Ag at 360 g/t and 1020 g/t respectively. The sample also contained many other metals as shown in Table 1, indicating that incineration allowed removal of the volatiles and reduction of sample weight resulting in an increase of metal concentration. Phase analysis by XRD showed CuO, SnO_2, and SiO_2 as the main constituents (Fig. 1).

The second waste material was a mine flotation tailings sample (lead–zinc mine), also obtained as fine powder with size ranging between 0.5 and 100 μm ($d_{50} = 13$ μm). Dominant phases in the sample were observed as FeS_2, and SiO_2 (Fig. 2) and its chemical analysis showed high Fe content at 32.5% and other constituents as shown in Table 2. Assuming Fe content is from FeS_2, the tailing sample contained about 69% FeS_2 and had a sulfur content of 36.6%. The electronic waste and pyrite tailing samples were mixed at varying weight ratios of 1:1, 1:2, and 1:4 (e-waste:pyrite tailings) to prepare as feed for autoclave oxidation study. Characterization by XRD and chemical analyses by ICP of the mixed samples is shown in Fig. 3 and Table 3 respectively. FeS_2 in the sample mixtures serves as the sulfur source for generation of H_2SO_4.

2.2. Autoclave

A 300 mL (0.3 L) high pressure micro-reactor laboratory autoclave (MMJ–300, Oemu Labotec–Japan) was used in this study and a schematic illustration is shown in Fig. 4. The setup has a control panel for setting temperature and a thermocouple inserted into the reaction vessel for monitoring temperature, an inlet line for gas feed (O_2 in this case), control valves for controlling autoclave pressure and a gas discharge line for the removal of excess gas.

2.3. Oxidation and leaching experiment

E-waste + Pyrite tailing sample mixtures were first conditioned in 100 mL of water and transferred to a 220 mL Teflon tube and fitted/inserted into the 300 mL autoclave vessel. For all experiments, impeller rotation was fixed at 1000 rpm and temperature carefully observed until 10 °C before target temperature when oxygen gas was introduced into the slurry. Reaction was allowed to proceed for set reaction times after which the autoclave was cooled to below 50 °C. Reaction tube was then removed from the autoclave and filtered to obtain filtrate and solids for analyses.

Table 1
Chemical contents of the electronic waste ash sample analyzed both before and after incineration at 800 °C.

Element	Before	After	Element	Before	After
Cu (wt.%)	13.41	20.43	Co (g/t)	230	240
Fe (wt.%)	1.33	1.63	Mn (g/t)	740	1030
Al (wt.%)	2.84	3.13	Ga (g/t)	65	63
Pb (wt.%)	1.79	1.30	In (g/t)	27	58
Zn (wt.%)	0.92	1.30	Zr (g/t)	1400	300
Au (g/t)	230	360	W (g/t)	100	100
Ag (g/t)	480	1020	Mo (g/t)	170	180
Pt (g/t)	63	110	La (g/t)	106	136
Pd (g/t)	170	250	Ce (g/t)	29	19
Ni (g/t)	4200	5900	Pr (g/t)	22	14
Sb (g/t)	3800	5100	Nd (g/t)	124	178
B (g/t)	5400	6600	Sm (g/t)	10	14
Ti (g/t)	2100	2200	Gd (g/t)	5	10
Cr (g/t)	1700	700	Dy (g/t)	15	21

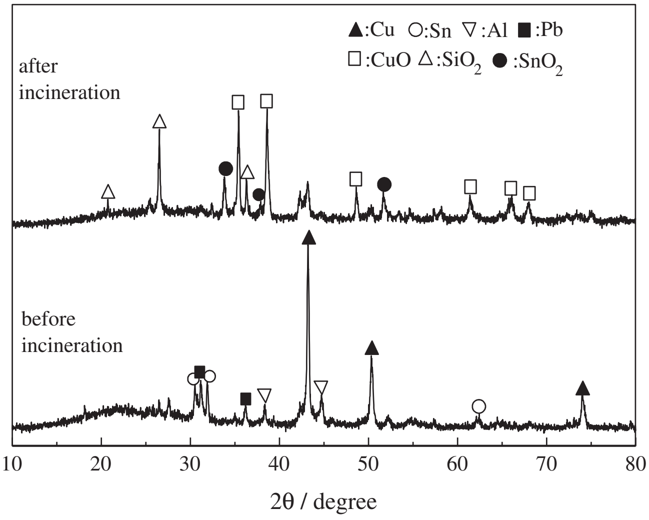

Fig. 1. Characteristic X-ray diffraction pattern of the electronic waste ash before and after incineration at 800 °C.

Parameters investigated include; pulp density (50–1000 g/L), reaction temperature (120–180 °C), autoclave pressure (1.0–4.0 MPa), H_2SO_4 concentration (0–1.0 mol/L), reaction time (0.5–3 h), E-waste: pyrite tailings weight ratio (1:1, 1:2, 1:4). The experimental results were used to evaluate oxidation of pyrite in the mine tailings and subsequent dissolution of metals from the electronic waste ash. Sulfuric acid (H_2SO_4), hydrochloric acid (HCl), nitric acid (HNO_3) and sodium hydroxide (NaOH) as chemical grade reagents were used as leaching solutions/reagents to evaluate dissolution behavior of the electronic waste.

2.4. Characterization and chemical analyses

X-ray diffraction (XRD) measurements were performed, using Rigaku, RINT–2200/PC system with a Cu Kα irradiation source (λ = 1.5405 Å) at 40 kV and 50 mA. Both starting samples and their mixtures and leach residues from the tests were analyzed in a continuous scan mode between 10° and 80° 2θ. All solution analysis was conducted using an Inductively Coupled Plasma (ICP-AES/OES) atomic/optical

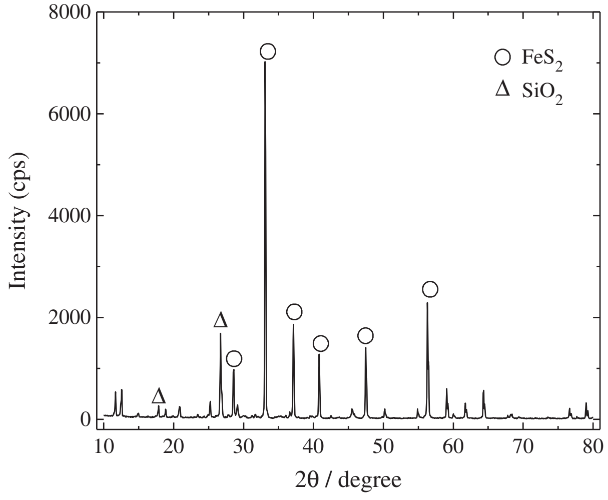

Fig. 2. Characteristic X-ray diffraction pattern of the lead–zinc mine tailings waste.