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Leaching of a complex sulfidic, silicate-containing zinc ore in sulfuric acid solution under oxygen pressure

Hongsheng Xu, Chang Wei *, Cunxiong Li, Gang Fan, Zhigan Deng, Xuejiao Zhou, Shuang Qiu

Faculty of Metallurgical and Energy Engineering, Kunming University of Science and Technology, Kunming 650093, China

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ABSTRACT

This paper reports an acid leaching process study on the selective extraction of base metals from complex sulfidic, silicate-containing zinc ore containing sphalerite, hemimorphite, smithsonite, quartz, galena, pyrite, troilite, muscovite, calcium aluminate, calcite, gypsum and cerussite with traces of other minerals under oxygen pressure. The effects of influential factors (leaching temperature, sulfuric acid concentration, leaching time, ore particle size distribution, liquid/solid ratio and pressure) on the dissolution efficiencies of zinc, iron and silica from said ore were investigated. The optimum conditions, which were sought for reducing downstream purification requirements by limiting iron and silica dissolution, were found to be a leaching temperature of 160 °C, a liquid/solid ratio of 6 mL/g, a sulfuric acid concentration of 0.42 mol/L, a leaching time of 150 min, a particle size of −98 + 74 μm and 1.6 MPa partial pressure. Under these conditions, the zinc extraction was above 97%, with an iron and silica dissolution of less than 25% and 0.3%, respectively. The reduction of silica dissolution during leaching also promotes easier solid–liquid separation.

1. Introduction

Zinc is the third most common nonferrous metal after aluminum and copper as its global annual output stood at nearly 11.28 million tons in 2009, accounting for 132 ppm of the Earth's crust [1–3]. Most of the zinc produced worldwide from zinc-bearing sulfurous and oxidized ores by pyro- and/or hydrometallurgical processes is used to galvanize steel and iron products to prevent corrosion [4]. However, more than 80% of the global zinc metal output is produced from zinc sulfide concentrates using the roast–leach–electrowinning process [5].

With the exhaustion of "ideal" zinc concentrates, such as high-grade sphalerite ores, and with an increasing demand for zinc metals, new alternative processes to produce zinc metal from low grade ores, complex ores and secondary resources (such as industrial wastes) need to be proposed for application in the zinc industry in future. Typical processes in current use as reported in the literature for primary zinc metal recovery include the pyrometallurgical (roasting) approach which requires high energy consumption from the heating of gangue materials [6] and direct chemical leaching which is limited in that the zinc ores have a low solubility in most reagents and the process requires purification of the resulting complex multimetallic ion-containing solution [7–9]. Bioleaching processes have also been economically profitable [10].

Generally speaking, hydrometallurgical processes without pretreatment are more advantageous and less harmful than pyrometallurgical processes from an economic and environmental point of view, especially for complex ores. Amongst the alternative processes to treat complex zinc ores, oxygen pressure acid leaching as a new paradigm in zinc production is considered to be attractive owing to the characteristic of shorter leaching time required, more precise control, higher selective extraction and metal recovery [11–13], as well as better flexibility for different grades and compositions ores [14–17]. A hydrometallurgical process based on the oxidative pressure acid leaching of zinc sulfide concentrates for the direct production of zinc sulfate solution (which is suitable for conventional recovery of zinc by electrowinning) was first successfully applied commercially by Sherritt Gordon [18]. Since then, the importance of oxygen pressure acid leaching as an environmentally friendly process has grown and this technique has been applied to the recovery of zinc as well as other metals (such as silver, copper, lead and indium) from low grade ores, complex ores and secondary resources by some researchers [16,19–21].

For complex minerals containing high amounts of silicate such as willemite or hemimorphite, the formation of silica gel makes it difficult to separate the zinc sulfate solution from the slurry during direct sulfuric acid leaching. Many researchers have attempted to avoid the formation of silica gel, such as by using flocculants [22–24] and microwave-assisted leaching [6], to good effect and with some application value. The sulfuric acid pressure leaching of zinc silicate ore is also a feasible process in that it effectively extracts most of the zinc into solution and rejects silica while providing good characteristics for solid–liquid separation [25].

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* Corresponding author. Tel./fax: +86 871 5188819.
E-mail address: weichang502@sina.cn (C. Wei).

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Complex sulfidic, silicate-containing zinc ores containing sphalerite, hemimorphite, smithsonite, quartz, galena, pyrite, troilite, muscovite, calcium aluminate, calcite, gypsum and cerussite with traces of other finely disseminated minerals, found in large quantities in the northwestern part of China's Yunnan province, are important zinc metal sources in China [26,27]. However, few efforts have been made to develop a direct leaching process suitable for zinc recovery from these ores owing to the high content of silica which readily enters the solution as silica gel together with zinc and the complexity of the said ore itself which may impose too great a burden on the downstream purification of the leaching solution, and previous studies have rather emphasized the importance of flotation technologies [26,27].

In this study, taking into consideration the unique structure of complex sulfidic, silicate-containing zinc ore where both sulfides and silicates co-exist at high silica levels, combined with the existing established application of pressure leaching technology to other minerals (such as sulfide minerals or oxidized ores with high silica content), an attempt was made to develop a suitable process to extract optimally the zinc present in complex sulfidic, silicate-containing zinc ores under oxygen pressure acid leaching conditions. A discussion of the use of this process as an improved method for obtaining good characteristics for solid–liquid separation or the minimization of downstream purification requirements has been also made. Such a process would be technically viable provided maximum zinc extraction, oxidation of sulfide sulfur to elemental sulfur and minimal dissolution of silica and iron can be obtained.

2. Experimental

2.1. Materials

The complex sulfidic, silicate-containing zinc ore investigated in this study was from the Lanping Mine in the northwest region of the Yunnan province (China) and was used without pretreatment, with the exception of grinding in a pestle and mortar and obtaining particle size distributions by sieving. Its chemical composition, which is primarily zinc, sulfur, silicon, iron, lead, calcium and shows traces of cadmium, magnesium, aluminum, potassium and copper, is given in Table 1. The major phases identified by XRD analysis were: sphalerite, hemimorphite, smithsonite, quartz, galena, pyrite, troilite, muscovite, calcium aluminate, calcite, gypsum and cerussite, as shown in Fig. 1. For the determination of the amount of acid soluble phases, the mineralogical composition of the studied sample was confirmed by semiquantitative XRD with results as listed in Table 2.

To investigate the effect of particle size distribution on leaching efficiency, 1 kg of the ore was sieved and divided into several size fractions using Tyler standard sieve. The main chemical analysis of each size fraction is listed in Table 3. Analytical grade sulfuric acid (98 wt.% H_2SO_4) was used as leaching agent in all experiments. Reagent grade sodium lignosulfonate was added as a surfactant to prevent the inactivation of unreacted ore particle surfaces by precipitated sulfur [28].

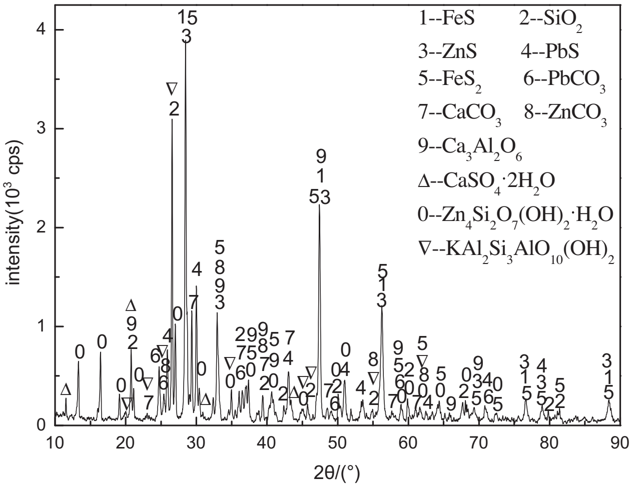

Fig. 1. XRD pattern of the complex sulfidic, silicate-containing zinc ore.

Table 2
Mineralogical composition of the complex sulfidic, silicate-containing zinc ore.

Mineral phases	Formula	Content (wt.%)
Sphalerite	ZnS	19.53
Hemimorphite	$Zn_4Si_2O_7(OH)_2·H_2O$	13.29
Smithsonite	$ZnCO_3$	1.71
Quartz	SiO_2	16.75
Galena	PbS	5.31
Pyrite	FeS_2	19.51
Troilite	FeS	9.31
Muscovite	$KAl_2Si_3AlO_{10}(OH)_2$	2.54
Calcium aluminate	$Ca_3Al_2O_6$	1.54
Calcite	$CaCO_3$	2.46
Gypsum	$CaSO_4·2H_2O$	1.99
Cerussite	$PbCO_3$	4.06
Others		2.00

Table 3
The chemical analysis of each size fraction from complex sulfidic, silicate-containing zinc ore (wt.%).

Particle size (μm)	Zn (%)	Fe (%)	S (%)	SiO_2 (%)
−246 + 147	19.93	14.07	18.10	19.31
−147 + 98	19.59	14.73	20.26	19.40
−98 + 74	19.7	14.24	20.28	20.79
−74 + 57	20.02	14.65	19.81	21.34
−57 + 53	19.92	14.07	20.21	21.36

2.2. Equipment and procedure

The leaching experiments were performed in a 500 mL vertical titanium-lined autoclave (Model 09041904-2/2 Reactor, Weihai Chemical Machinery Instrument Limited Company, Shandong Province, China), fitted with an internal titanium spiral-shaped water-cooling tube, and sparging tube through which oxygen was bubbled into the leaching solution. The autoclave was agitated using a titanium propeller on a shaft connected to a variable-speed magnetic stirrer with digital controller unit and heated by an electrical heating mantle. The temperature of the reaction medium was controlled by a platinum resistance temperature sensor with an accuracy of ±0.1 °C.

For each leaching experiment, 200 mL of leaching solution containing predetermined amounts of H_2SO_4 and ore mixed with a suitable amount of sodium lignosulfonate were charged into the autoclave and heated to the required temperature whilst being

Table 1
The chemical composition of the complex sulfidic, silicate-containing zinc ore.

Component	Zn	Fe	SiO_2	Pb	Cu	S	As	Cd	CaO	MgO	Al_2O_3	K_2O
Wt.%	19.7	14.24	20.79	7.51	0.05	20.28	<0.39	0.22	2.8	0.23	1.91	1.19