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Field application of selective precipitation for recovering Cu and Zn in drainage discharged from an operating mine

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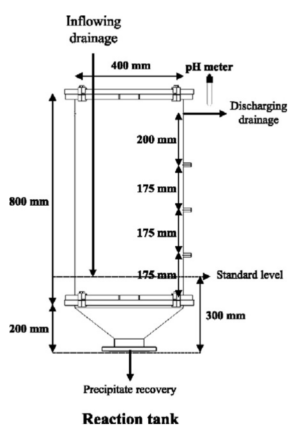
HIGHLIGHTS

- Selective precipitation process was directly applied to an operating mine.
- Cu and Zn precipitate of 80% purity with 90% precipitation rate can be obtained.
- The strategies to reduce impurities were discussed.

GRAPHICAL ABSTRACT



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ABSTRACT

Acid mine drainage (AMD) generated from mining activities has been recognized as a serious problem due to its increased acidity and high concentration of heavy metals. In this research, a feasibility test of the selective precipitation (SP) process was performed using AMD discharged from a currently operating mine in Korea for the purpose of minimizing the environmental impact of AMD. For the SP process, a pilot scale equipment (100 L reaction tank) was used in field and among various metals, Cu and Zn were the target metals. Through the research, it was confirmed that AMD from an operating mine has two disadvantages of being applied to the SP: altering water quality and unexpected inclusion of clay debris. Despite unfavorable conditions, Cu and Zn precipitate of 80% purity with 90% precipitation rate was able to be obtained from 1.4 L/min (2.0 tons/day) AMD. The recovered precipitates were identified as amorphous CuS and ZnS with small amounts of impurities (Si minerals, $CuFeS_2$, and Fe/Al hydroxide). The strategies to reduce these impurities were also discussed. Recovery rate, which is the amount of precipitate collected per unit volume of AMD, was proposed as an indicator to evaluate the working efficiency of the SP process. It was confirmed that the recovery rate was strongly dependent on flow rate and dose of coagulant. The results of this study may be helpful in reducing the potential complications which occurs when SP is applied on field.

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1. Introduction

The exposure of rocks to water and air caused by mining and construction activities can generate unfavorable acid mine drainage (AMD) (Achterberg et al., 2003; Bodlák et al., 2012; Khan et al., 2013). AMD, due to its increased acidity and high concentration of sulfate and heavy metals, is regarded as a toxic pollutant itself and also causes secondary pollution of contaminating adjacent stream or groundwater by dissolving toxic metals from surrounding rocks and ores (Azapagic, 2004; Han et al., 2015; Ji et al., 2008; Oh et al., 2012). AMD can also cause yellow boy which aggravate the scenery near mining sites (Brady et al., 1986; Ji et al., 2007; Oh et al., 2015). Among the various AMD treatment techniques such as oxidation, neutralization, adsorption, reverse osmosis, ion exchange, and electrolysis (Akcil and Koldas, 2006; Ghorbanzadeh et al., 2015; Johnson and Hallberg, 2005; Oh et al., 2016), the neutralization and precipitation under oxidative conditions is one of the most economically preferred methods (Matlock et al., 2002). This technique is beneficial because it is easily applicable and does not require a complex treatment facility while still securing relatively high remediation efficiency. A major disadvantage of this technique however, is the production of secondary wastes such as metal sludge: a heterogeneous mixture of Fe, Al, Mn, Ca oxides/hydroxides (Park et al., 2015). The sludge is often a serious problem in the field treatment of AMD, as it inevitably causes a continual need for dredging of downstream water basins and settling ponds (Park et al., 2016; Wang et al., 1996).

Selective precipitation (SP) is one of the most promising ways to overcome the problems of the conventional precipitation technique. SP recovers individual metal precipitates from AMD by determining optimal precipitation conditions for each target metal using a connected series of reaction tanks (Wei et al., 2005). The technique is advantageous in terms of liability mitigation by decreasing the amount of sludge and also beneficial since it can generate valuable products to compensate for treatment costs, while meeting the effluent discharge limitation (Rao and Finch, 1992). Particularly, using sulfide as a neutralizing agent in SP has various advantages compared to using hydroxide (Lewis, 2010). First of all, a high possibility of metal separation can be achieved using sulfide, owing to the more distinct solubility of the various metal sulfides (Sampaio et al., 2009). Metal sulfides also have other benefits such as better settling properties, higher potential for reuse by smelting, and faster reaction rates (Gharabaghi et al., 2012).

Various researches on effective control of SP process retrieving metal ions as sulfide minerals have been conducted. Since sulfide concentration strongly affects precipitation efficiency of each metal sulfide, researches regarding the controlling of sulfide concentration in SP to separate Cu from Zn (Sampaio et al., 2009) or Ni from Zn (Sampaio et al., 2010) were conducted. Gharabaghi et al. (2012) tried to maintain a constant sulfide concentration through accurate pH adjustment and by adding thioacetamide as a sulfide source. Reducing the usage of chemicals for the SP process has also been recognized as one of the important topics for research. Sahinkaya et al. (2009) have attempted to generate H_2S gas for the sulfide source of SP using sulfate-reducing bacteria. Park et al. (2015) have investigated the potential of electrochemical methods to produce oxidizing and neutralizing agents.

Although the topic of SP has been extensively explored in various researches, most of the studies were conducted with artificial AMDs or with laboratory scale tests (mostly in the size of few liters). Therefore, a pilot scale application should be carried out in order to verify the feasibility of SP application in actual site with complex environments. As a part of this effort, SP experiments targeting actual AMD from a pit lake (Wei et al., 2005) and a coal mine (Tabak et al., 2003) have been conducted. These experiments, however, are still insufficient in simulating the actual environment because in the experiments, a certain amount of AMD is collected at once, and therefore the resulting data does not reflect the fluctuating nature of the water quality in operating mines (Limpitlaw et al., 2005). To the best of our knowledge, only a few pilot scale experiments on field have been conducted which investigates the influence of operation method or the design of the equipment on the efficiency of SP. Environmental management, especially water treatment (Warhurst and Mitchell, 2000), in mine operation is one of the most critical factors in minimizing the production cost of minerals (Warhurst, 1992). Therefore, researches regarding the optimization of operation parameters of SP should be focusing on securing economic feasibility of mining activities.

To fulfill the ultimate goal of this research of applying SP to actual mining sites and minimizing both environmental impact and operating cost, SP targeting Cu and Zn recovery was performed directly at an operating mine site with a pilot scale equipment (100 L reaction tank). The specific objectives of this research can be outlined as follows: 1) to determine the precipitation and purity rate of SP when being applied directly to operating mines, 2) to understand the origination of impurities and figure out the way to minimize it, 3) to propose an indicator of evaluating the working efficiency of SP and examine factors that affect the indicator.

2. Materials and methods

2.1. Site and acid mine drainage description

All experiments were performed at an operating gold and silver mine in Haenam, South Korea (Fig. 1). The geology of the area consists of alluvial layer of the Quaternary, plutonic rock, volcanic rock, and sedimentary layer of the Mesozoic based on gneiss complex of the Precambrian eon. The deposit of this mine is an epithermal gold-silver one which is embedded in quartz veinlet filling fissure of fine lapilli, welded or non-welded tuff, and tuff breccia. The gold and silver are yielded as electrum together with sulfide minerals in a size of tens of μm. The contaminated drainage generated during mining is discharged to a settling pond outside of the mine and the discharged AMD is reused for watering an operating mine. A certain amount (0.20–2.12 L/min) of AMD in the pond was pumped into a reaction tank during the SP field experiment in this research (Fig. 1). Data for 9 months confirmed that the water quality of AMD varies widely depending on mining schedule and rainfall (Table 1). The main metals usually included in AMD are Cu, Zn, Fe, Al, and Mn (Tabak et al., 2003). Among the five metals, Cu and Zn which are relatively more valuable were selected as target metals for selective separation (Sahinkaya et al., 2009).

2.2. Experimental equipment

Cu and Zn in the drainage which was pumped from the settling pond were selectively precipitated together in a reaction tank. The reaction tank used in this research was made in acrylic material and was designed to hold approximately 100 L of AMD. The dimension of the reaction tank is depicted in Fig. 1. The forms in which the metals flowed in and discharged via the drainage were as follows: Forms during inflow were dissolved ion (D_{in}) and floating solid substance (F_{in}). Forms during discharge were dissolved ion (D_{out}), floating solid substance (F_{out}), and solid substance settled down at the bottom (SM Fig. 1). The experiment began by flowing in the AMD towards the reaction tank in a constant flow rate using a volumetric pump. Neutralizing agent (Na_2S), which is also used as a source of sulfide, was added to the AMD before being injected into the reaction tank. The mixed AMD was then flown into the reaction tank through the delivery tube which induced the water height at 300 mm from the bottom (standard level in Fig. 1). The reaction tank was designed so that the produced precipitate settles at the bottom while the supernatant gets discharged through the outlet. After each experiment, the settled precipitate along with a certain amount of AMD was sampled by opening the valve installed at the bottom of the reaction tank. This concentrated AMD then went through processes of additional settlement and freeze drying so that completely dried solid precipitate could be obtained for analysis. The reaction pH in