



Ferric leaching of the sphalerite contained in a bulk concentrate: Kinetic study

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ABSTRACT

A novel process for the integral treatment of polymetallic sulphide ores is proposed. The process consists of a global flotation, two stages of ferric leaching, the first stage dissolves the sphalerite and the rest of secondary sulphides, and the second dissolves the chalcopyrite with a silver catalyst, and finally a brine leaching to recover lead and silver (added as catalyst). The proposed process offers several advantages comparing to traditional pyrometallurgical techniques. This study is focused on the sphalerite dissolution with ferric sulphate, as the first stage of the treatment of Cu-Zn-Pb concentrates. The ferric ion concentration, the temperature and the particle size have an important role in the sphalerite oxidation. However, the initial sulphuric acid, ferrous ion and sulphate ion concentrations have no influence in the process rate. The formation of an elemental non-porous layer of sulphur along the reaction hinders the ferric ion transport to the sphalerite surface. Two kinetics regimes are proposed, in the beginning the chemical reaction is the rate controlling step, and, at 30% Zn extraction, the rate controlling step changes to diffusion through a non-porous film of elemental sulphur. The activation energy obtained for the chemical reaction has a value of 51.3 kJ/mol, and the apparent activation energy in the diffusional stage is the 47.7 kJ/mol. The reaction order with respect ferric ion is 0.26 and the reaction rate is proportional to the zinc sulphide amount in the first stage. The behaviour of sphalerite, contained in a bulk concentrate, is similar to the pure sphalerite or the sphalerite contained in a differential concentrate.

1. Introduction

Almost all of the world's primary zinc production is derived from the treatment of sulphide concentrate, obtained through differential flotation, in which sphalerite is the dominant zinc mineral (Dutrizac, 2006). The *roast-leach-Electrowinning Process* (R-L-E) is currently responsible for more than 85% of the total zinc production (Souza et al., 2007b). This process has several troubles, such as the restrictions to treat sphalerite concentrate with silica, calcium, copper and iron, the emissions of SO₂ and marketing of sulphuric acid (Deller, 2005). Several processes have been studied, in the last years, to extract Zn from differential concentrates, such as atmospheric pressure leaching (Salmi et al., 2010; Souza et al., 2007a; Babu et al., 2002), bioleaching (Haghshenas et al., 2012) or heap-Bioleaching (Lizama et al., 2003).

Most of these processes need a previous concentration of zinc sulphide through a differential flotation. Differential flotation has several disadvantages regarding global flotation, where a concentrate of all non-ferrous metals is obtained, such as a lower recuperation of non-

ferrous metals, high content of impurities in the concentrates or a greater grinding energetic consumption (Carranza, 1985; Majima, 1969; Tipre et al., 1999; Carranza et al., 1993). Global concentrates must be treated through hydrometallurgical processes because these processes offer a greater versatility to extract the valuable metals from different ores, and low-grade concentrates, and are more environmentally friendly than pyrometallurgical processes (Córdoba et al., 2008; Carranza et al., 1997b; Conic et al., 2014). Zinc, and the rest of metals, can be extracted from global concentrates through bacterial leaching (Tipre and Dave, 2004; Conic et al., 2014; Gómez et al., 1999; Gómez et al., 1997), pressure leaching with O₂ (Xu et al., 2011; Xu et al., 2016) and atmospheric pressure leaching with ferric ion, as ferric sulphate, only studied with Cu-Zn concentrates (Carranza et al., 1997a; Palencia et al., 1990). Ferric chloride reaches higher reaction rates, but this leaching agent increases industrial costs due to equipment corrosion. Conversely, ferric sulphate is a cheap and efficient oxidant agent to dissolve non-ferrous metals and can be regenerated by bio-oxidation (Mazuelos et al., 2000; Carranza et al., 1993; Palencia

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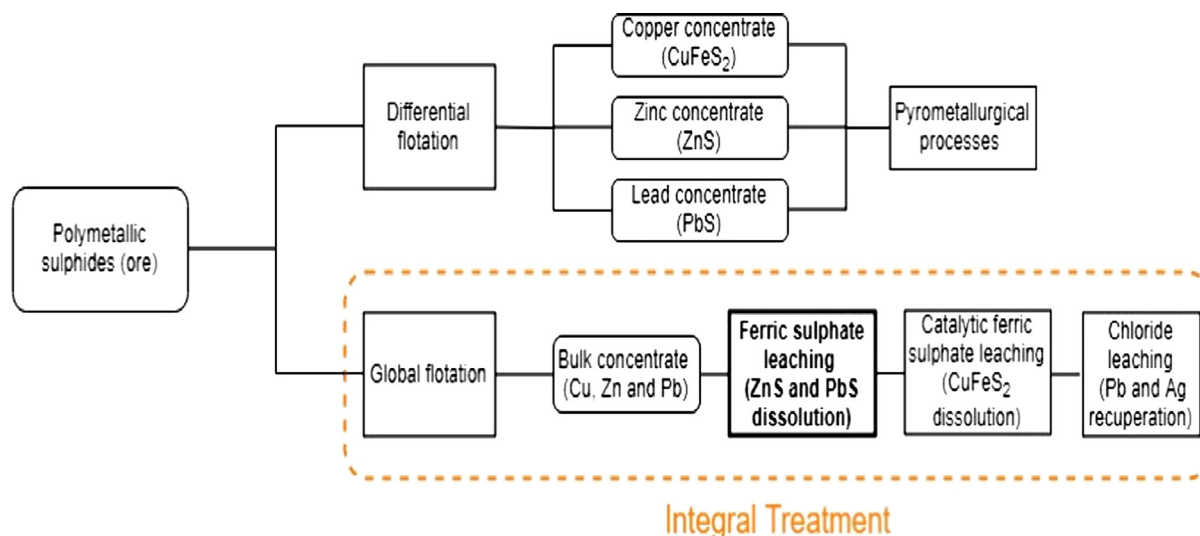
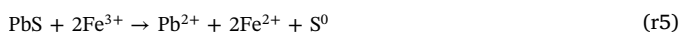
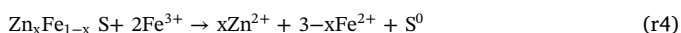
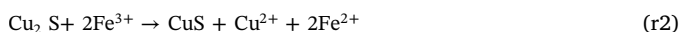
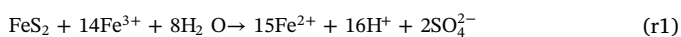


Fig. 1. Diagram of hydrometallurgical process vs. traditional processes to polymetallic sulphides exploitation.

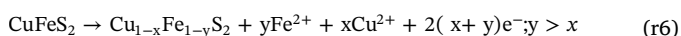
et al., 1990; Gómez et al., 1997; Dutrizac et al., 2003; Aydogan et al., 2005).

Global concentrates obtained from polymetallic sulphides of the Iberian Pyritic Belt (IPB) are composed, basically, of sphalerite, chalcocopyrite, galena and pyrite, as majority sulphide (Ortega and Bonilla, 1983). Fig. 1 shows some possibilities to benefit these polymetallic sulphides: the traditional processes, by pyrometallurgical methods, and the hydrometallurgical processes. The hydrometallurgical treatment, shown in Fig. 1, of bulk concentrates (Cu, Zn and Pb) is proposed as an integral treatment where Cu, Zn and Pb are recuperated from one sole concentrate and through a unique process. This hydrometallurgical plant can be located within the mining facilities, changing the production model of concentrates sales to production and sales of metals.

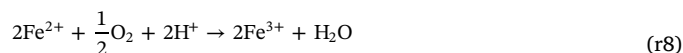
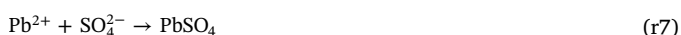
From a conceptual point of view (see Fig. 1), the ferric sulphate leaching, to benefit a concentrate that contains Zn, Cu, Pb and Ag, could consist of a first stage where zinc sulphide, and the rest of secondary sulphides, are dissolved and a catalytic second stage in presence of silver salts, where chalcocopyrite is oxidised. First stage is necessary because silver catalyst is not selective and passives the dissolution of the rest of non-ferrous sulphides. Silver and lead are extracted through a chloride leaching of solid residue (Palencia et al., 1998; Bahram and Javad, 2011). In this stage, the silver added as catalyst is recovered (Carranza et al., 2004; Carranza et al., 1997a; Barriga Mateos et al., 1993). Chemical reactions in the ferric leaching of a global concentrate, containing FeS_2 , $Zn_xFe_{1-x}S$, CuS , Cu_2S , PbS and $CuFeS_2$ are (r1)(r5):



In this medium, chalcocopyrite is passivated due to the formation of a metal deficient film according to (r6) (Ghahremaninezhad et al., 2015).



Various secondary reactions take place, as the lead sulphate precipitation (r7) or the possibility to regenerate the leaching agent (r8), which can be catalysed by bacteria. A possible mechanism to sphalerite dissolution found in literature, where sulphuric acid and ferric ion concentrations affect the reaction rate, is given by (r9), (r10).



Sphalerite leaching reaction with ferric ion has been extensively studied in zinc sulphide concentrate and natural crystals of pure sphalerite (Dutrizac, 2006; Salmi et al., 2010; Palencia and Dutrizac, 1991; Souza et al., 2007a; Crundwell, 1987b; Dutrizac and Macdonald, 1978; Estrada-de los Santos et al., 2016; Chang et al., 1994; Weisener et al., 2003; Da Silva, 2004). These studies can be summarized in the following points:

- Fe content in sphalerite structure has a great influence on sphalerite leaching reaction rate. The increment of iron impurities in sphalerite enhances the sphalerite leaching rate, decreasing the activation energy. An activation energy of 70 kJ/mol was observed for a sphalerite with 0.04% wt of iron, instead, for a sphalerite with 12.5% wt of iron the activation energy was around of 40 kJ/mol (Palencia and Dutrizac, 1991; Crundwell, 1988).
- The presence of other sulphides can change the reactivity of sphalerite due to the formation of galvanic couples. Sulphides with a higher rest potential than sphalerite enhance the sphalerite dissolution, and the sulphides with a lower rest potential, such as galena and chalcocite, decrease the sphalerite leaching rate (Mizoguchi and Habashi, 1983; Lo et al., 1985; Elsherief, 1994; Estrada de los Santos et al., 2016; Da Silva et al., 2003; Mehta and Murr, 1982).
- Many authors have found a great influence of sulphuric acid concentration on sphalerite leaching rate, according to (r9), (r10), where H_2S is formed as a reaction intermediate (Dutrizac, 2006; Dutrizac and Macdonald, 1978; Souza et al., 2007a; Verbaan and Crundwell, 1986). Dutrizac (2006) observed that the reaction rate depends on sulphuric acid concentration only when this concentration is higher than 0.1 M. However, other authors found no influence of sulphuric acid on sphalerite oxidation rate (Salmi et al., 2010; Santos et al., 2010).
- The rate controlling step is a reason for discrepancy between authors. Some authors have observed that chemical reaction is the rate controlling step (Dutrizac, 2006; Palencia and Dutrizac, 1991; Markus et al., 2004; Salmi et al., 2010). While, in other cases, the rate controlling step is the diffusion through a passive film (Dutrizac and Macdonald, 1978; Palencia et al., 1990; Lampinen et al., 2015).

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