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Selective leaching of zinc from hazardous As-bearing zinc plant purification filter cake

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ABSTRACT

This article is part of a continuing endeavor to decrease environmental conflicts and industrial hazards of As-bearing zinc plant hot purification filter cakes (ZPHPFC's) and to break through this problem and extract Zn selectively from filter cakes to obtain a filtrate containing Zn capable of feeding to zinc electrowinning stage. In this part of research, a process was investigated with a view to selective Zn leaching from ZPHPFC, as well as diminishing the quantity of the ZPHPFC to be dumped. Firstly, pH-dependent leaching tests were done by H_2SO_4 or NaOH solution with the pH ranging from 1 to 13 to monitor Zn and impurities leaching behaviors and finding suitable pH zone for selective Zn leaching. Then, orthogonal array (OA) of Taguchi's method was employed to arrange the experimental runs in order to distinguish the most notably affecting factors and the optimum leaching conditions for maximizing Zn extraction with minimum impurities dissolution. OA L_{16} (4^5) consisting of five factors, each with four levels, was utilized to scrutinize the effects of reaction temperature [T = 303, 313, 323, 333 (30, 40, 50, 60) K (°C)], reaction time (t = 30, 60, 90, 120 min), pH (pH = 4.5, 5, 5.5, 6), pulp density (S/L = 50, 100, 150, 200 g/L), and stirring speed (R = 375, 500, 625, 750 rpm), on leaching efficiency of the individual metals. Statistical analysis of variance (ANOVA) was done to conclude the relationship between experimental conditions and metals leaching efficiencies. The results showed that increasing pH reduced Zn, Fe and Cd leaching efficiencies. Likewise, increasing of temperature declined extraction of impurities such as Ni, Cd, and Co. Also, time increasing, especially in high temperature, decreased Ni and Co leaching efficiencies. This is due to that conditions in leaching media are suitable for cementation of Ni and Co by metallic zinc that exists in the ZPHPFC in the presence of As. The experimental results for selective leaching showed that under ultimate optimal leaching conditions (T_4: 333 K (60 °C), t_4: 120 min, pH_2: 5, $(S/L)_3$: 150 g/L, R_3: 625 rpm), the leaching efficiency of Zn could be almost 95%, with low dissolution of impurities.

1. Introduction

The amount of industrial residue has endlessly ascended in the last 40 years and a large amount of residue is a threatening menace everywhere. Minerals or metals production from ores using hydrometallurgical and/or pyrometallurgical processes typically causes a gigantic amount of residues. The special residue dumpsites and storage space of landfills are restricted, and the dumping expenditures become extremely elevated. Then, ideas of reusing and recovering of minerals or metals are emerging to lower the residues. A quantity of the residues of the primary extraction might include residual valuable minerals or metals which might be subjected to an additional process. The enormous amount of these residues give rises to wasting base metal values

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and results in environmental contamination. Consequently, there is a vast concentration to recycle the costly metal species as well as hazardous compounds from the residues, selectively. In fact, residues regularly propose cost-effective profits to industries through the recycle of costly materials present in them. This means that lowering process costs through replacement of primary materials by recoverable products causing to the protection of primary materials, and must be an aim for our world, since the sources in the world are restricted and recovering results their conservation as well as environmental protection. Finally, it can be mentioned that metals regeneration from hazardous residues is associated with the laws allocated to diminish adverse environmental effects and assist to satisfy the upcoming requirement of metals under lack of primary sources (Alizadeh et al., 2011; Chen et al., 2011; De Souza et al., 2001; Nusen et al., 2015).

Zinc concentrate leaching is a famous process for producing zinc electrolyte. In general, metallic zinc production from zinc concentrate, especially sphalerite, includes of roast–leach–electrowinning (RLE) steps. At the leaching stage, in addition to zinc, impurities such as iron, arsenic, germanium, antimony, copper, cadmium, cobalt and nickel are liberated into the leaching liquor that shall be purified before electrowinning. These impurities are removed by selective precipitation and cementation. For example, Iron is removed in the form of three major types of residues: (a) goethite, (b) jarosite, or (c) hematite that called iron cake or zinc plant leach residue (ZPLR). In other words, the ZPLR is obtained during the leaching of the zinc concentrate in sulphuric acid and selective iron precipitation stage. This constitutes mainly iron and impregnated zinc sulphate and zinc ferrite besides other impurities, such as arsenic, antimony, germanium that co-precipitated with iron. Also, lead and silver were reported in the ZPLR which can be utilized for the recovery of these two metals (Behnajady and Moghaddam, 2011, 2014; Behnajady et al., 2012). Further liquor purification was done by cementation in two-step; copper, cadmium and some of nickel were precipitated by zinc powder in the first step according to Reactions (1)–(3) and this precipitate called as zinc plant cold purification filter cakes (ZPCPFC) in literature (Behnajady et al., 2014a). Cobalt and nickel were removed from leach solution using $CuSO_4{\cdot}5H_2O$, As_2O_3 and zinc powder in the second step according to Reactions (4)–(8), the filter cake obtained from second step called as As-bearing zinc plant hot purification filter cake (ZPHPFC) (Behnajady and Moghaddam, 2015) that containing a high quantity of Zn in addition to other metals like Fe, Pb, Cd, Cu, Ni, Co, As. After separation of ZPHPFC, the pregnant leach solution (PLS) is used to make up zinc electrowinning solution cycle.

$$CuSO_4(aq) + Zn(s) \rightarrow Cu(s) + ZnSO_4(aq) \quad (1)$$

$$CuSO_4(aq) + Cu(s) + H_2O \rightarrow Cu_2O(s) + H_2SO_4(aq) \quad (2)$$

$$CdSO_4(aq) + Zn(s) \rightarrow Cd(s) + ZnSO_4(aq) \quad (3)$$

$$2Co^{2+}(aq) + 2As^{3+}(aq) + 5Zn(s) \rightarrow 2CoAs(s) + 5Zn^{2+}(aq) \quad (4)$$

$$2Ni^{2+}(aq) + 2As^{3+}(aq) + 5Zn(s) \rightarrow 2NiAs(s) + 5Zn^{2+}(aq) \quad (5)$$

$$6Cu^{2+}(aq) + 2As^{3+}(aq) + 9Zn(s) \rightarrow 2Cu_3As(s) + 9Zn^{2+}(aq) \quad (6)$$

$$Co^{2+}(aq) + Cu_3As(s) + Zn(s) \rightarrow 3Cu(s) + CoAs(s) + Zn^{2+}(aq) \quad (7)$$

$$Ni^{2+}(aq) + Cu_3As(s) + Zn(s) \rightarrow 3Cu(s) + NiAs(s) + Zn^{2+}(aq) \quad (8)$$

Zinc plant purification filter cakes (ZPPFC) are classified as hazardous residues mainly due to the presence of toxic metals e.g. Pb, Cd, As, etc (Alizadeh et al., 2011). Such high metal contents turn these deposits into precious metal sources which are known as secondary sources. A large amount of these residues remains stored waiting for recycling procedures that are industrially and cost-effectively feasible. In other words, discarding of ZPPFC is becoming costly due to environmental regulations, so there has been a rising attention to recover metallic values from these intermediate residues. There has been a continuing attempt to discover an economic procedure to extract metals from ZPPFC without undergoing solvent extraction, ion exchange or pyrometallurgical processing. Hydrometallurgy, as an extensively utilized technology in today's base and costly metals production, is more environmentally appropriate and economic to treat materials containing low valuable metals and has received wide interest by researchers involved in recovering base metals from residues. Hydrometallurgical efforts have been made on recovering some important metals from ZPPFC e.g. cobalt (Haghshenas et al., 2007; Moradkhani et al., 2014; Qian et al., 2013; Safarzadeh et al., 2011; Stanojević et al., 2000; Wang and Zhou, 2002), cadmium (Gharabaghi et al., 2011, 2012, 2014; Gouvea and Morais, 2007; Safarzadeh et al., 2008, 2009a), zinc (Gouvea and Morais, 2007; Haghshenas et al., 2007; Qian et al., 2013; Safarzadeh et al., 2008, 2009b), nickel (Gharabaghi et al., 2013; Safarzadeh et al., 2008; Safarzadeh and Moradkhani, 2010) and Indium (Koleini et al., 2010; Nusen et al., 2015; Yao et al., 2011; Zhang et al., 2010). However, several studies (Gharabaghi et al., 2011, 2012, 2013, 2014; Gouvea and Morais, 2007; Haghshenas et al., 2007; Koleini et al., 2010; Moradkhani et al., 2014; Nusen et al., 2015; Qian et al., 2013; Safarzadeh et al., 2008, 2009a, 2009b, 2011; Safarzadeh and Moradkhani, 2010; Stanojević et al., 2000; Wang and Zhou, 2002; Yao et al., 2011; Zhang et al., 2010) carried out about valuable metals extraction from ZPPFC but no one was focused to selective leaching of zinc.

Today, a division of zinc is recycled from different secondary resources such as zinc ash, flue dust, zinc dross, etc. which include different levels of zinc and impurities depending on their resources. Conversely, Zn is leached following by dissolution of many other impurities such as Co, Ni, Cd, Fe, As, Ge, Ca, Sb, Mg, etc. As a result, the acid consumption is elevated, and multipart purification processes are mandatory (Alizadeh et al., 2011). Although, previous studies have dealt with metals extraction, no one has reported a study on the selective extraction of Zn from ZPPFC. Hydrometallurgical operations can be utilized to achieve high and selective Zn leaching while synergistically declining the volume and improving the disposability of the ZPPFC. The present study was designed to investigate selective Zn extraction from ZPHPFC, which is obtained in zinc sulphate solution hot purification process by arsenic activation of zinc powder. As above mentioned, ZPHPFC is a mixture of Zn and various metal contaminants (e.g. Ni, Cd, Co, etc.) in the form of metal, oxides, sulphates, and/or alloy (Gharabaghi et al., 2011, 2012, 2013, 2014; Gouvea and Morais, 2007; Koleini et al., 2010; Nusen et al., 2015; Qian et al., 2013; Safarzadeh et al., 2008, 2009a, 2009b; Safarzadeh and Moradkhani, 2010; Wang and Zhou, 2002; Yao et al., 2011; Zhang et al., 2010) and especially arsenic compounds from the use of zinc powder activator in the hot purification process. At the first stage, the ZPHPFC was leached in different pH by leaching in H_2SO_4 or NaOH solution with pH ranging from 1 to 13 to monitor Zn and impurities leaching behaviors in acidic, neutralized and alkaline environments. In the second stage, design of experiment (DOE) was utilized to explore the effect of factors on Zn and impurities leaching behavior. In fact, Taguchi method was utilized to distinguish the most significantly affecting parameters and optimize leaching conditions for maximizing Zn extraction with minimum impurities dissolution. Accordingly, the impact of operating factors containing reaction temperature (T), reaction time (t), pH (pH), pulp density (S/L), and stirring speed (R) on the extraction efficiencies of Zn and impurities were scrutinized using an L_{16} (4^5) orthogonal array.

2. Experiments and methods

2.1. *Characterization of the ZPHPFC*

The ZPHPFC used in the experiments was kindly supplied from R&D pilot plant of Zanjan Zinc Khales sazan Industries Company (ZZKICo), located at Zanjan, Iran. The ZPHPFC was dried at 378 K (105 °C) for 24 h and then crushed for further analysis. Mineralogical structure of ZPHPFC was identified by X-ray diffraction (Philips PW1800) analysis. Atomic absorption spec-