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### Management strategies and valorization for waste sludge from active treatment of extremely metal-polluted acid mine drainage: A contribution for sustainable mining





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### ABSTRACT

This study assesses the environmental impact and the potential valorization of metal-sludge waste generated by the active neutralization of extremely metal-polluted acid mine drainage (AMD). To this end, two regulated leaching tests (EN 12457-2 and 1311 USEPA TCLP), a standardized sequential extraction protocol (BCR sequential extraction) and single leaching tests were performed using dilute common industrial acids. The results of the two standardized leaching tests showed a complete discrepancy, classifying the waste as both inert (according to the TCLP) and not suitable for disposal at landfills for hazardous waste (according to EN 12457-2). In this regard, the environmental characterization of the waste using the BCR sequential extraction lined up with interpretations made by the EN 12457-2 leaching test, reinforcing the hazardousness of this type of residue. This waste requires careful management, as evidenced by the release of high concentrations of metals (e.g., Cd, Zn, Al) when interaction with rainfall and organic acids take place, exceeding the risk threshold values for aquatic life. The easy extraction of base, industrial- and tech-metals that is possible with dilute acids encourages the consideration of this type of sludge as an interesting alternative metal source with great economic potential. The joint application of remediation treatments and metal recovery schemes could contribute to the goal of zero waste production in mining activities, which would help to develop sustainable mining practices worldwide.

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#### 1. Introduction

Water resources can be severely damaged by mining activity, especially when no action is taken to prevent pollution. In working and orphan sulfide-bearing mining sites, exposure of mining waste to oxygen and water leads to the generation of acidic waters with high concentrations of sulfate and metal(loid)s, known as acid mine drainage (AMD). These highly polluted waters become a serious environmental concern (Akcil and Koldas, 2006). AMD generation processes are very long-lived and can persist for hundreds and even thousands of years after cessation of the mining activities (Younger, 1997). Mine waters can be treated by two generic approaches,

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active or passive treatment systems (Johnson and Hallberg, 2005). Both in active and passive treatment systems, acidity and metals are removed from the solutions using different (bio)geochemical reactions that lead to the generation of a solid waste (generally as a metal-rich sludge) (Johnson and Hallberg, 2005).

Zinck and Griffith (2013) conducted a detailed study on the waste production from conventional active and passive treatment technologies at 108 working mine sites in Canada and other countries. On average, these sites produced about 9500 tons of dry sludge per year, with production ranging from 20 to 135,000 dry tons per year depending of the site. These figures are not exhaustive but indicative of the huge worldwide production of waste from AMD neutralization. In addition, this production is expected to rise due to the foreseen increase in metal mining and the expected generation and/or implementation of stricter environmental regulations around the world.

This waste should be properly disposed of in order to avoid environmental impacts. To this end, the stability of this waste under different disposal and weathering scenarios must be addressed. In this regard, several scenarios have been proposed (e.g., Zinck, 2005; Zinck and Griffith, 2013); however, to date no studies on leaching behavior under different weathering conditions have been performed: thus, there are no information of the suitability of disposal according to environmental regulations. In this sense, static and dynamic, single and sequential leaching tests are well established and widely accepted methods to determine the potential metal release of mining waste (Hageman et al., 2015). However, to our knowledge, these types of leaching tests have not been properly performed in waste that derives from the neutralization of acidic waters, mainly in conventional active treatment plants, which are the most common technology used for AMD depuration.

The recycling and reuse of mining and mineral processing waste can be considered one of the main challenges for future waste management (Lottermoser, 2011). A few investigations focused on the reusability of this waste have been reported, mainly in construction, agriculture or carbon dioxide sequestration (Zinck, 2005), also as neutralizing material for AMD-generating waste rocks and tailings (Demers et al., 2015) or directly for mine waters remediation (Wolkersdorfer and Baierer, 2013). Nevertheless, such practices could be limited by the high content of toxic impurities, and so most waste remains stored in disposal areas, causing environmental problems. Recent researches report a fractional precipitation of metals of economic interest by active neutralization of AMD which could be a promising metal source from AMD (Chen et al., 2014), which potential recovery is being still tested at pilot scale (Yan et al., 2015).

The mining industry currently faces a challenge of global significance: the development of sustainable mining (Laurence, 2011; Moran et al., 2014). Developed countries such as those of the European Union (EU) or United States of America emphasize the need of supplying the growing demand for base-, industrial- and techmetals, by reducing its dependence on external suppliers (Silberglitt et al., 2013), while ensuring the environmental safety (EU European Commission, 2016). Among them, the reuse of mining and mineral-processing wastes is one of the most promising secondary sources (Bian et al., 2012). To our knowledge, the reuse of conventional active treatment wastes as a potential source of metals has not been properly assessed until now.

To bridge these gaps, the present study aimed to explore environmental risk, management strategies for land disposal and the valorization of metal sludge waste generated by AMD neutralization at active treatment plants. With this purpose, sludge formed by the active neutralization of an extreme metal polluted AMD from the Almagrera mining industrial complex (Iberian Pyrite Belt, IPB) was characterized. This characterization included different leaching tests evaluating the land disposal options proposed by the current European (EN 12457-2, 2002) and US environmental regulations (US EPA, 1998). The potential mobility of metals contained in the sludge was assessed by the standardized sequential extraction procedure proposed by the European Community Bureau of References (BCR, Ure et al., 1993a), while the exposure of aquatic life to metals by an uncontrolled leachate from the sludge was studied under several scenarios (e.g., rainfall, co-disposal with municipal wastes). Finally, the potential valorization of the sludge was investigated by leaching experiments to extract elements of technological and industrial interest. This novel approach based on the joint application of environmental tests (single and sequential, static and dynamic), and metal recovery schemes on these metalrich residues may contribute to a sustainable mining of sulfide ore deposits.

#### 2. Materials and methods

# 2.1. Metal sludge from the AMD neutralization at Almagrera industrial complex

From 1982 to 2001 polymetallic sulfide ores were processed in Almagrera Complex (SW Spain) by flotation to obtain Cu, Pb and Zn concentrates. In addition, crude pyrite refuses were processed (by roasting and SO<sub>2</sub> recovering) to produce sulfuric acid, oleum and Cu sulfate. As a consequence of these mineral processing activities two tailing ponds were built to accommodate the generated wastes (sulfide tailing pond and roasted pyrite pond, Fig. 1A). Despite restoration in 2006, two AMD discharges (AMD1 and AMD2, Fig. 1A) emerged from both impoundments. AMD1 exhibits remarkable extreme metal concentrations (especially Al, Cu, Fe, Mn and Zn; Table 1), whereas AMD2 could be considered a typical moderately polluted AMD within a sulfide mining district like the IPB.

Due to the severity of the mentioned AMD pollution, the regional authorities implemented a conventional active treatment plant which comprised a first step of alkaline dosing (CaO and/or MgO) followed by an agitation/sedimentation tank (Fig. 1B). Due to the low flow rates of AMD1 (annual mean of 1.5 L/s) and AMD2 (0.4 L/s), both discharges are pumped out to a storage pond near to the treatment plant (Fig. 1A). Thus, the treatment plant works temporally when enough combined AMD has been accumulated. The resulting metal-rich waste sludge is pumped out to a pond built over the surface of the tailing ponds, where around 39,000 tons are stored (Fig. 1A and C). In the metal-sludge pond, a representative sample (approximately 2 kg) of the first 20 cm from the shallowest part was collected close to the discharging point during an inactive period of the treatment plant, using a polypropylene shovel previously washed with distilled water, and transferred to polypropylene sterile bags. In the laboratory, the sludge was oven-dried (30 °C), ground and stored in sterile polypropylene containers until analysis. Subsamples were used for the different analytical procedures.

## 2.2. Leaching protocols for management and hazardousness assessment

As stated before, management assessment and hazardousness classification of the sludge were addressed according to the EU standard EN 12457-2 (2002) leaching test and the US standard TCLP leaching test (US EPA, 1998). The EN 12457-2 leaching test assesses the suitability of wastes for disposal in a landfill site. The experimental concentrations obtained in the test can be compared with the limit threshold values established by the European Council (EC Decision, 2003) for the acceptance of wastes in three types of landfill sites: inert, non-hazardous and hazardous wastes. Given that it is the first time that static tests are used in sludge from active AMD treatment, and following the suggestions of the European rule (EC Decision, 2003), the EN 12457-2 leaching test was repeated three consecutive times in a same sample aliquot to have information on the effluent quality and extractability over time.

The Toxicity Characteristic Leaching Procedure (TCLP) (US EPA, 1998) was originally designed to simulate co-disposal with municipal wastes but also used for the hazardousness classification of mineral-processing wastes (e.g., Vemic et al., 2015). Additionally, metal concentrations in TCLP leachates can be also employed as limits to determine if a specific waste needs to be submitted to a universal treatment standard (UTS) to accomplish with Land Disposal Restrictions (LDR, EPA 530-R-01-007) (US EPA, 2012). A detailed description of both tests can be found in Supplementary materials.

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