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Research article

Bioleaching of metals from WEEE shredding dust

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ABSTRACT

A bioleaching process developed in two separate steps was investigated for the recovery of base metals, precious metals and rare earth elements from dusts generated by Waste Electrical and Electronic Equipment (WEEE) shredding. In the first step, base metals were almost completely leached from the dust in 8 days by *Acidithiobacillus thiooxidans* (DSM 9463) that lowered the pH of the leaching solution from 3.5 to 1.0. During this step, cerium, europium and neodymium were mobilized at high percentages (>99%), whereas lanthanum and yttrium reached an extraction yield of 80%. In the second step, the cyanide producing *Pseudomonas putida* WSC361 mobilized 48% of gold within 3 h from the *A. thiooxidans* leached shredding dust. This work demonstrated the potential application of biohydrometallurgy for resource recovery from WEEE shredding dust, destined to landfill disposal, and its effectiveness in the extraction of valuable substances, including elements at high supply risk as rare earths.

1. Introduction

As metals are largely used for manufacturing electronic products, the electrical and electronic equipment reaching the end-of-life, known as Waste Electrical and Electronic Equipment (WEEE) or e-waste, is regarded as a potential secondary source of these elements (Hageluken, 2006; Lee and Pandey, 2012; Tuncuk et al., 2012). The recycling of metals from WEEE represents an important opportunity to feed resources back to the economy, lowering the environmental impacts as well as the energy consumption associated with raw material supply, as promoted by the circular economy concept (Hageluken, 2006). In this view, WEEE recycling plays a key role as this waste stream has a fast growth rate per year (Robinson, 2009) and moreover contains strategic elements, such as precious metals and rare earth elements (REEs) (Schüler et al., 2011). Mining REEs from WEEE is particularly attractive since these elements have been listed by the European Commission as the most critical raw materials at supply risk due to their growing demand by several process industries and their scarce worldwide production, mainly restricted to a single country, i.e. China (Binnemans et al., 2013; European Commission, 2014, 2010). Furthermore, the recycling of WEEE can bring benefits not only in terms of raw resource supply, but it can also contribute to the reduction of potential environmental pollution and human health risks related to the release of the hazardous substances present in WEEE as a result of improper management practices (Zeng et al., 2017).

Mechanical, pyrometallurgical and hydrometallurgical processes are used for the recovery of metals from WEEE (Cui and Zhang, 2008; Khaliq et al., 2014; Priya and Hait, 2017). Besides these conventional techniques, emerging technologies in the field of electrochemistry, supercritical fluids, mechanochemistry and ionic liquids are receiving increased attention (Tan and Li, 2015; Wang et al., 2017). Over the recycling chain, mechanical treatments are employed in the early stage as pre-processing, aiming at separating the larger valuable and hazardous components and upgrading the metal fraction, which is further routed to end-refining processes (Cui and Forssberg, 2003). Pyrometallurgy and hydrometallurgy are metallurgical techniques coming from the mineral sector, employed as end-processes for the recovery of the metal of interest (Cui and Zhang, 2008). These processes are, however, not yet effectively targeted on the extraction of REEs, whose recycling rates are still low (Binnemans et al., 2013). During the mechanical treatment, REEs as well as large amounts of

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precious metals end up in the output fractions as dust or ferrous materials, usually not involved in the metallurgical refining process (Bachér and Kaartinen, 2016; Tsamis and Coyne, 2015). REEs are also easily lost during pyrometallurgical processes, since these elements are generally entrapped in the slag phase due to their strong affinity with oxygen (Binnemans et al., 2013; Haque et al., 2014). Conversely, hydrometallurgy has been considered as an effective method for the recovery of REEs, although its industrial application is still limited (Tunsu et al., 2015).

Current metallurgical treatments have, moreover, several impacts on the environment due to the generation of secondary pollutants. The necessity to set environmentally sound treatments, increasing the sustainability of recycling processes (Canal Marques et al., 2013), has thus moved the research interest towards biohydrometallurgy as a promising technique for metal recovery from WEEE (Cui and Zhang, 2008; Khaliq et al., 2014; Priya and Hait, 2017). Biohydrometallurgy exploits either the ability of microorganisms to solubilize the metals contained in the solid matrix (bioleaching) or the capacity of microbial biomass to sorb metals from the aqueous solution (biosorption) (Erüst et al., 2013; Hoque and Philip, 2011; Olson et al., 2003; Zhuang et al., 2015). The feasibility of biohydrometallurgy has been demonstrated for the extraction of metals from ores (Bosecker, 1997; Brierley and Brierley, 2013, 2001; Watling, 2016) and electronic waste (Beolchini et al., 2012; Cui and Zhang, 2008; Ilyas and Lee, 2014; Işıldar et al., 2016) as well as for the removal of metals from aqueous solutions (Andrès et al., 2003; Vijayaraghavan and Yun, 2008; Wang and Chen, 2009; Won et al., 2014). In this regard, several autotrophic and heterotrophic bacteria as well as fungi have been involved in WEEE bioleaching studies (Madrigal-Arias et al., 2015; Rozas et al., 2017; Zhang and Xu, 2016) which, however, mainly focused on the extraction of single metals, particularly copper and gold, from printed circuit boards (PCBs). Despite the increasing interest in biotechnology, the potential application of this technique needs to be further explored, especially for the recovery of critical metals from WEEE as the REE-microbe interactions are not clearly understood and the bio-recovery of REEs from secondary sources, including WEEE, is poorly documented (Barmettler et al., 2016; Ilyas et al., 2017).

This study focused on the recovery of base metals, precious metals and REEs from WEEE shredding dust through biohydrometallurgical processes. The work aimed to investigate (i) the application of bioleaching to dusts originating from a WEEE mechanical treatment facility and (ii) its effectiveness for the recovery of different metals contained in WEEE. To this end, the bioleaching experiments were carried out in two separate steps in order to assess if a selective recovery of the metals could be achieved. The first step aimed for the extraction of base metals and REEs exploiting acidophilic bacteria (*Acidithiobacillus thiooxidans)*, whereas the second stage was performed for precious metal mobilization by cyanogenic bacteria (*Pseudomonas putida* WSC361). Moreover, wider technical considerations on the development of biohydrometallurgical treatments and on the management of WEEE shredding dust in a circular economy view have been presented.

2. Materials and methods

2.1. Material characterization

A full scale plant located in Southern Italy and providing the mechanical treatment of small electronic equipment, information technology (IT) and consumer appliances was chosen for this study (Cesaro et al., 2017). Over the treatment line, representative samples of dust materials resulting from the shredding of WEEE, namely WEEE shredding dusts, were collected.

The carbon content of the sampled dust was determined by means of an elemental analyser (OEA Flash 2000; Thermo Finningan). For metal analysis, samples were digested using the standard aqua regia extraction procedure (ISO 11466:1995). Thereafter, the concentrations of dissolved base metals (aluminium, cadmium, copper, iron, nickel, lead and zinc), precious metals (gold, silver, palladium, platinum) and REEs (cerium, europium, lanthanum, neodymium and yttrium) were analysed by means of inductively coupled plasma-optical emission spectroscopy (ICP-OES, Thermo iCap 6000 series).

The particle size distribution was obtained by a sieve analysis, considering the following size fractions: <0.5 mm, 0.5–1 mm, 1–2 mm, >2 mm. Mineral phase analysis was performed using a X-ray powder diffractometer (XRD, Bruker D8 advance). The following conditions were applied: Cu Kα radiation, 35 keV accelerating voltage, 40 mA current, 10–80° scanning range and 0.5 s/step (0.0296°/step) scan speed.

2.2. Microorganisms and growth conditions

Bioleaching was performed using the acidophilic and cyanogenic bacterial strains as described by Işıldar et al. (2016). *A. thiooxidans* (DSM 9463) was purchased from the Leibniz Institute (DSMZ), Braunschweig (Germany), whereas *P. putida* WSC361 was kindly provided by Peter Bakker from Utrecht University (the Netherlands).

A. thiooxidans was grown in a mineral medium containing (g/L): $(NH_4)_2SO_4$ (2.0), $MgSO_4 \cdot 7H_2O$ (0.25), K_2HPO_4 (0.1), KCl (0.1) and S^0 (5.0). The pH was adjusted to 3.5 using H_2SO_4. Cultures were inoculated with 10% (*v/v*) inoculum size in 100 mL growth medium and incubated at 30 °C and 150 rpm for 10 days prior to the bioleaching experiments. The pH and oxidation reduction potential (ORP) were periodically recorded by Ag/AgCl reference electrodes (SenTix 21, WTW, Germany and QR481X, Qis, the Netherlands) for monitoring indirectly the bacterial growth.

A. thiooxidans cells were enumerated using the spread plate method (Starosvetsky et al., 2013). *Thiobacillus* agar, containing 0.4 g $(NH_4)_2SO_4$, 0.5 g $MgSO_4 \cdot 7H_2O$, 0.25 g $CaCl_2$, 4 g KH_2PO_4, 0.01 g $FeSO_4$, 5 g $Na_2S_2O_3$, 12.5 g agar in 1 L distilled water, was used. Samples were serially diluted up to 10^{-7} using a saline solution (0.85% NaCl) and plated on agar. The Petri dishes were then incubated for 7–10 days at 31 (±0.5) °C.

P. putida was grown in nutrient broth containing (g/L): meat extract (1.0), yeast extract (2.0), peptone (2.0) and NaCl (5.0). The pH was adjusted to 7.3 using NaOH. The cultures were inoculated with 1% (*v/v*) in 150 mL growth medium supplemented with 10 g/L of glycine (Işıldar et al., 2016) and incubated at 30 °C and 150 rpm for 18 h prior to the bioleaching experiments. The bacterial growth was monitored by the measurement of both pH and optical density (OD) at 600 nm using a Perkin Elmer Lambda 20 Spectrophotometer.

The ability of *P. putida* to produce CN^- was checked by a modified colorimetric method (Işıldar et al., 2016; Ruan et al., 2014). *P. putida* was plated on nutrient agar supplemented with glycine. A sterile filter paper soaked in a solution of 0.5% picric acid and 2% sodium carbonate was fixed to the inner side of the Petri dish lid. The dishes were covered with paraffin film and incubated at 30 °C (Işıldar et al., 2016). CN^- production induces a colour reaction on the filter paper which turns from yellow to red, proportionally to the CN^-concentration (Ruan et al., 2014).