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A novel approach for recovery of metals from waste printed circuit boards and simultaneous removal of iron from steel pickling waste liquor by two-step hydrometallurgical method

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ABSTRACT

Waste printed circuit boards (WPCBs) and steel pickling waste liquor (SPWL) have received extensive attention in recent years because of its harmfulness and resource. In this work, two-step leaching process was carried out by using SPWL as the leaching agent. A series of continuously domesticated bacteria were used for bioleaching and the bacterial strain was identified as Acidithiobacillus ferrooxidans (A. ferrooxidans) by 16S rDNA gene sequence analysis. The vast majority of the metals in WPCBs were recovered by two-step leaching, such as Cu, Pb, Zn, Sn, Al, Ni. Meanwhile, a large amount of iron was removed from SPWL, which greatly reduces the burden of the subsequent treatment. Pulp density and pH were optimized to achieve maximum recovery of copper and simultaneous removal of iron in bioleaching. It was found that the optimum conditions were pulp density 60 g/L and pH 0.5–1.0. Nearly 100% of copper was recovered and 51.94% of iron was removed under optimum conditions. The kinetic experiments showed that the rate of copper leaching was controlled by external diffusion rather than internal diffusion, because the acidic environment of the leachate prevented the formation of the precipitate and maintained it in a smaller size.

1. Introduction

In recent years, the production of electrical and electronic equipments (EEEs) has substantially increased with the development of science and technology. Printed circuit boards (PCBs) are the basal and essential components of electrical and electronic equipments (EEEs), widely used in computers, personal computers, television, mobile phones and other electronic products (Huang et al., 2009; Silvas et al., 2015). The current electronic products are being replaced with faster rate and the lifecycles are getting shorter and shorter. The amount of waste electrical and electronic equipments (WEEE) is increasing dramatically (Flandinet et al., 2012). As a result, electronic pollution caused by WPCBs has become a serious environmental problem.

WPCBs contain polymers, ceramics and metals. The proportion is 23%, 49%, 28%. The typical metals, including Cu (20%), Fe (8%), Sn (4%), Ni (2%), Pb (2%), Zn (1%), Ag(0.2%), Au (0.1%), and Pd (0.005%), are very valuable in recycling (Chen et al., 2015a). The purity and amount of metals contained in WPCBs are higher than those in rich-content minerals, which could be considered as an "Urban Mine". Therefore, it is of great significance for environmental protection and energy value to seek a scientific and environment-friendly method to recycle the metals from WPCBs. At present, the existing methods of recycling metals from WPCBs include mechanical separation, pyrometallurgical, hydrometallurgical and biohydrometallurgical techniques (Birloaga et al., 2013). Mechanical separation is usually used in the pretreatment process to increase the metal content of WPCBs to be processed, which includes crushing, vibration and electrostatic separation (Zhu et al., 2013). Pyrometallurgy requires high investment and produces large amounts of toxic and harmful substances, which makes it less applications in the recycling of metals in WPCBs. Hydrometallurgical processes also used to be expensive as it involved long procedures, lower efficiency of metal recovery, heavy consumption of chemicals and secondary pollution. Biohydrometallurgy is the process of dissolving metal from ore or recovering valuable metals from water, using certain microorganisms or their metabolites on

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the oxidation, reduction, dissolution and absorption of certain minerals and elements. Compared with the traditional metallurgical technology, biohydrometallurgy has the advantages of low cost, low energy consumption, no pollution, simple operation, etc. (Arshadi and Mousavi, 2015; Bryan et al., 2015; Chi et al., 2011; Yang et al., 2014). The advanced technology of biohydrometallurgy has remarkable economic and environmental benefits, which has been popularized, perfected and improved in the world.

At present, the most commonly used microorganisms are A. ferrooxidans in leaching metals contained in WPCBs (Arshadi and Mousavi, 2014). This bacterium is mesophilic, chemolithotrophic, gram-negative, acidophilic that obtains energy by oxidizing Fe^{2+} to Fe^{3+} and sulfur to sulfate, usually present in acidic environments with pH values of 1–3 (Kurade et al., 2016). It was frequently used in biological leaching process of copper. The process of copper dissolution by bioleaching can be divided into two consecutive periods. Fe^{2+} is oxidized to Fe^{3+} under the action of microorganism, and Fe^{3+} reacts with copper to make it into the solution. Copper is leached and Fe^{3+} is reduced to Fe^{2+}. This cycle is repeated, which is similar to the indirect mechanism of microbial leaching (Chen et al., 2015b).

Numerous studies have shown that WPCBs contain certain other active metals (Chen et al., 2015a), which may compete with copper leaching, resulting in lower leaching efficiency of copper. Thus, this study intends to adopt chemical-biological two-step leaching process to recovery metals from WPCBs. The primary chemical leaching was treated with SPWL as leaching agent and bioleaching employed A. ferrooxidans as leaching microorganisms. SPWL is discharged from steel industry during pickling process, which is acidic and contains a mass of Fe^{2+} (Leonzio, 2016; Pathak et al., 2016; Rögener et al., 2012). The composition of the SPWL used in this study is shown in Table 1. Then, the leaching technology takes full advantage of Fe^{2+} of SPWL used as the energy source of the microorganism in the bioleaching, which avoids the addition of the bioleaching iron source and greatly saves the leaching cost. Simultaneously, the great majority of the iron in SPWL is removed in the form of iron precipitate, which greatly reduces the burden of iron removal of SPWL. More importantly, the iron precipitate by drying treatment can also be used for concrete admixture, catalyst, absorbent agent, etc. (Chen et al., 2017; Li et al., 2015; Sheydaei and Khataee, 2015; Wang et al., 2017; Zheng et al., 2016), so as to realize the reutilization of waste. In this study, the chemical leaching reduce the pressure of subsequent bioleaching after the preliminary leaching of copper in WPCBs, which makes the overall leaching cycle greatly shortened. To sum up, the application of SPWL makes the whole leaching process more economical, significant savings in leaching costs, while the SPWL has been a large degree of treatment.

The present study aims to realize the resource utilization of waste. On the one hand, the metal in the WPCBs is recovered. On the other hand, the iron in SPWL is removed, and the generated iron precipitate is further utilized. Taking into account the operating environment of this experiment, a strain resistant to WPCBs and SPWL was domesticated and its genes were identified. Simultaneously, the leaching parameters were optimized and the kinetics of the bioleaching process were analyzed to improve bioleaching performance. It is proved to be an effective method for recovery of metals from WPCBs and simultaneous removal of iron from SPWL.

2. Materials and methods

2.1. Preparation of WPCB sample

WPCBs used in this study were obtained from a local e-waste collection center in Nanjing, China. No physical or mechanical separation process was used before transportation to laboratory. Before the PCBs powder preparation, printed circuit board components, such as capacitors, resistors, transistors and cables, were manually separated from the PCBs. Then, the PCBs powder samples were prepared in three-stage crushing operation. Initially, the PCBs were manually cut into small pieces (diameter <40 mm) using stainless steel blades. Next, the samples were chopped by a high-speed universal pulverizer (FW-400A). Finally, the PCB powders were sieved using a vibrator shifter through #16 mesh and dried in an oven (Wiseven, Won 50) at 105 °C for 2 h, which were selected and used for all the leaching experiments.

2.2. Microorganisms and domestication conditions

Bacterial strain used in this study was A. ferrooxidans ATCC 23270. This strain was provided by Jiangsu Key Laboratory of Chemical Pollution Control and Resources Reuse, School of Environmental and Biological Engineering, Nanjing University of Science and Technology, Nanjing, China. The selected strain was cultured in the 9 K medium. The composition was as follows: $(NH4)_2SO_4$ 3.00 g/L, KCl 0.10 g/L, K_2HPO_4 0.50 g/L, $MgSO_4{\cdot}7H_2O$ 0.50 g/L, Ca $(NO_3)_2$ 0.01 g/L, $FeSO_4{\cdot}7H_2O$ 44.6 g/L. The pH of medium was adjusted to 2.0 by 5.0 M sulfuric acid. The culture of A. ferrooxidans was incubated in 250 mL Erlenmeyer flask containing 200 mL of the medium and 10% (v/v) inoculums, on a rotary shaker at 180 rpm and 30 °C (Ma et al., 2017; Rastegar et al., 2015; Yan et al., 2016). Culture conditions for further experiment were the same as described here. The series of domestication was carried out to obtain the bacteria required for bioleaching by successive transfer method, including resistance to chlorine, SPWL, and WPCBs. The results suggested that the highest tolerance of bacteria to WPCBs was 60 g/L and the time required for bacteria to adapt was 7 d, which is far superior to the previous reported results (Arshadi and Mousavi, 2014, 2015).

2.3. Molecular analysis of domesticated bacteria NJLGS18

The bacterial community after domestication was identified using 16S rDNA sequencing and phylogenetic analysis according to previous study. For the DNA analysis, the domesticated bacteria were centrifuged for 5 min at 8000 rpm (Huang et al., 2016). Then the DNA was extracted using the FastDNA SPIN kit according to the manufacturer's instructions (MP Bio-medicals, CA, USA). The DNA samples were stored at −20 °C until further analysis (Jiang et al., 2016). The extracted DNA was used as template of polymerase chain reaction (PCR) to amplify 16S rDNA using the universal primers 27F (5′-AGAGTTTGATCCTGGCTCAG-3′) and 1492R (5′-GGTT ACCTTGTTACGACTT-3′) (Patel et al., 2011). The PCR amplification program was carried out at an initial denaturation at 95 °C for 5 min, and 30 cycles at 95 °C for 30 s, 55 °C for 30 s, 72 °C for 120 s and a final extension at 72 °C for 2 min. The quality of the amplified DNA was confirmed through 1.5% agarose gel electrophoresis. Then the PCR product was purified using a QIAquick-spin PCR

Table 1
Chemical analysis of SPWL.

Composition	Total Fe	Fe^{2+}	Fe^{3+}	Zn	Al	Pb	Cu	H^+	Cl^-
Concentration/(g/L)	173.11	165.77	7.34	1.84	0.07	0.01	0.04	0.66	252.59

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