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Enhancement of copper, nickel, and gallium recovery from LED waste by adaptation of *Acidithiobacillus ferrooxidans*

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ABSTRACT

This paper is the first study on the extraction of Cu, Ni, and Ga from Light Emitting Diode (LED) waste by bio-hydrometallurgy technology. LEDs have a high concentration of metals and various types of brominated flame retardants (BFRs). This study demonstrates the need for strains with resistance to high concentrations of LED powder. The adaptation of *Acidithiobacillus ferrooxidans* to LED powder was done through a serial acclimatisation procedure in five steps of 5, 10, 15, 20, and 25 g/l. The results indicated that the heavy metals tolerance of *Acidithiobacillus ferrooxidans* decreased as the pulp density increased from 5 to 20 g/l. The pulp density > 20 g/l of LED powder caused a toxic response resulting in an evident inhibitory effect on bacterial activity. In the presence of 20 g/l of LED powder, adapted *Acidithiobacillus ferrooxidans* exhibits higher Fe^{3+} level, cell amount, ORP, and lower pH than the non-adapted cells. The recovery of copper, nickel, and gallium were higher by adapted bacteria compared to non-adapted bacteria. The adapted *A. ferrooxidans* leached approximately 84%, 96%, and 60%, copper, nickel, and gallium, respectively. It could be concluded that adaptation can be an effective tool for enhancement of copper, nickel, and gallium bioleaching from LED powder and adapted *Acidithiobacillus ferrooxidans* would be a suitable strain in LED waste bioleaching.

1. Introduction

The total global production of electronic waste (E-waste) in 2014 was estimated to be 41.8 million tons (Balde et al., 2014). The behavior of electrical equipment waste has caused many concerns in both developed and developing countries (Veit and Bernardes, 2015).

The generation rate of lamp waste is growing at an alarming rate. In 2014, the total amount of lamp waste was 1 million tons globally and most of this waste was produced in Asia (Balde et al., 2014).

The amount of waste from light-emitting diodes (LEDs) requires attention. LEDs can be used for a range of automotive, electronics, and lighting display applications. They have been replacing other lighting sources because they are environmentally friendly, offer better energy efficiency and are cost-effective (Nardelli et al., 2017; Rahman et al., 2017; Wilburn, 2012).

Several environmental protection agencies have defined LEDs as hazardous waste because they contain a mixture of compounds such as copper, iron, aluminum, chromium, nickel, zinc, arsenic and various types of brominated flame retardants (BFRs) are beyond safe limits that are harmful to humans and the ecosystem (Veit and Bernardes, 2015; Rahman et al., 2017; Wilburn, 2012). Tetrabromobisphenol-A (TBBPA) is a widely-applied BFR in LEDs. At higher concentrations, TBBPA binds to estrogen hormone receptors and causes other effects on hormone-sensitive parameters (Gosavi et al., 2013). In recent decades, the effect of toxic and dangerous heavy metals on humans and the environment has become an important issue. In biological systems, metal ions react with cell components such as DNA and can cause structural changes (Tchounwou et al., 2012). Improper disposal of LED waste has an adverse impact on human health and the environment.

Bioleaching is a green technique, which has been successfully applied for metal recovery from electronic waste. Bioleaching is the dissolution of solid metallic compounds from their mineral sources through the natural ability of microbes to generate oxidizing agents such as iron (III) ions and/or protons in the system (Erüst et al., 2013). The iron (II) oxidizing bacteria *Acidithiobacillus ferrooxidans* (*A. ferrooxidans*) is used for metal recovery from electronic waste. *A. ferrooxidans* reoxidates ferrous iron to ferric iron by chemical action under acidic conditions in low oxygen. The insoluble metals are oxidized to soluble metal ions by biologically generated ferric iron (Mishra et al., 2005).

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Microbiology management is one of the most important parts of bioleaching (Jiang et al., 2015). One weak point in bioleaching is bacterial sensitivity to metallic ion concentrations (Astudillo and Acevedo, 2008). The microorganisms will have a biphasic response to heavy metals. Heavy metals such as Ni, Cu, and Zn are essential for bacterial growth, but beyond a certain concentration they become toxic and have a negative effect on bacterial activity and metal leaching efficiency (Valix and Loon, 2003). When the metal concentration increases beyond acceptable levels, the bacteria use a resistance mechanism for survival because the high metal concentration interrupts cell function by damaging the bacterial cell membrane and changing the nucleic acid structure and enzymes (Orell et al., 2010). The exposure of *A. ferrooxidans* to LED waste containing elevated mixed heavy metal concentrations result in the inhibition of bacterial growth, especially at high pulp densities; however, *A. ferrooxidans* can be adapted to tolerate several heavy metals. Adapted *A. ferrooxidans* can grow at a significantly higher pulp density due to the active biochemical pathway, which allows the bacterial cells to continue to grow when compared with the same non-adapted bacteria.

Zhang et al. (2015) found that TBBPA had an adverse impact on the structure and function of the microbial community. Their results suggested that the soil community decreased as the TBPPA concentration increased and a high concentration of these substances had an inhibitory effect (Zhang et al., 2015). The TBBPA exist at much electronic waste (at special LED); thus far, no study has reported their effect on bioleaching microbes.

According to previous investigations, there are several methods for acidophilic bacterial adaptation to sulfide minerals and concentrates containing high concentrations of heavy metals. Haghshenas et al. (2009) reported successful adaptation of *A. ferrooxidans* to 4.5% wt/vol pulp density high-grade sphalerite concentrate using the serial culture method (Haghshenas et al., 2009).

The adaptation criteria are critical to the adaptation process. Wang et al. (2014) used as the adaptation criterion the bacterial cell number when the cell density of a culture at the end of culturing at a given pulp density reaches 2×10^8 cells/ml (Wang et al., 2014). Hong et al. (2016) reported on the adaptation of a mixed culture of acidophilic bacteria from refectory gold concentrate containing a high concentration of arsenic. The results showed that the adapted bacteria showed a peak maximum percentage of biooxidation and could overcome the negative impact of increases in pulp density (Hong et al., 2016).

This paper is the first study on the extraction of Cu, Ni, and Ga from Light Emitting Diodes (LED) waste using bioleaching technology. This study presents *A. ferrooxidans* adaptation behavior in the simultaneous presence of two toxic components, heavy metals and BFRs, which exist in all E-waste (especially LEDs) and the effect of adaptation on metals extraction. A large portion (>70%) of LED waste includes BFRs (Lim et al., 2010). The unpleasant aspect of heavy metals and BFRs in LEDs is a major impediment during bioleaching and can reduce metal recycling efficiency. Further systemic studies on the adaptation of *A. ferrooxidans* to electronic waste are required. The aim of this study was to examine the effect of *A. ferrooxidans* adaptation on copper, nickel and gallium bioleaching from LED waste. This was carried out to obtain the strain with potential resistance to high concentrations of the toxic substance in its growth medium.

2. LED waste management strategy

Several management tools have been applied to E-waste management to improve waste disposal. European Commission Directive (2002/95/EC), known as RoHS (Restriction of Certain Hazardous Substances) has been established to prevent the use of toxic substances in electronic equipment by manufacturing in the European Union. In recent years, Life cycle analysis (LCA) and end of life (EOL) analysis used to assessment of the environmental impacts and economic losses associated with electronics equipment production, consumption and disposal. They also enable to design the strategies to reuse and recycle electronic equipment components (Veit and Bernardes, 2015).

The E-waste disposal in landfills or by incineration can cause serious environmental contamination. Flame-retardants containing bromine are present in most E-waste compositions, where burning can generate harmful emissions such as dioxins and furans. Research and industrial processes are being planned to recover the metal fraction from E-waste due to its economic benefits (Kaya, 2016; Veit and Bernardes, 2015).

LED lamps are going to be entered into the E-waste stream after the end of their lifecycles and are specifically referred to as light waste. The management of LED waste is important to decrease its environmental effects and increase sources of raw materials. Because lighting plays an important role in society, extensive research has been done about this waste management on both national and international levels to remove the toxic impact to human health and the environment. LED waste has a heterogeneous nature that has a major effect on processing, recycling, and other aspects of the waste management, but the management of this waste is still deficient in the majority of countries.

3. Materials and methods

3.1. LED waste preparation

Several discarded pin-type LEDs were collected from various electronic repair shops in Tehran, Iran (Fig. 1(a)). The LED samples were of different colors and luminous intensities and were used in different applications. The discarded pin-type LEDs were in two sizes of 3 mm and 5 mm, with different colors (white, red, green, blue, and purple). The pin-type LED is composed of several parts, including cathode, metal frame and anode, package resin, chip, bond wire (Fig. 1(b)). The mixture of discarded pin-type LEDs were shredded with an industrial crusher (IKA-Werke; Germany), then milled using a ball mill (Fristch; Germany) to achieve a fine powder. They were ultimately screened through standard 200 mesh using electric vibration. The sieved particle size was $\leq 70\ \mu$m.

3.2. Microorganism and culture condition

A. ferrooxidans iron-oxidizing bacteria were obtained from the Iranian Research Organization for Science and Technology (IROST) in Tehran, Iran. *A. ferrooxidans* was cultured in 9 K medium containing 3 g $(NH_4)_2SO_4$, 0.5 g K_2HPO_4, 0.5 g $MgSO_4 \cdot 7H_2O$, 0.1 g KCl, 0.01 g $Ca(NO_3)_2$ and 44.22 g $FeSO_4 \cdot 7H_2O$ and 1000 ml deionized (DI) water. The medium pH was adjusted to 2 using H_2SO_4 (98%). The experiments were conducted in a 250 ml Erlenmeyer flask. The flask containing 98 ml of the 9 K medium was inoculated with 2 ml of inoculum and was shaken at 140 rpm in an orbital shaker at 29 °C for 30 days.

3.3. Bacterial adaptation protocol and bioleaching experiment

The bioleaching experiment was done in two different modes with and without serial acclimatisation. The *A. ferrooxidans* needed to be adapted to increase its tolerance to the high concentration of LED powder. Adaptation of the bacterial strain was performed through serial acclimatisation method. Two-factor bacterial cell and ferric ion concentration in bioleaching solution were considered for adaptation criterion. In each step, when the bacteria have