



# Enhanced bioleaching efficiency of metals from E-wastes driven by biochar



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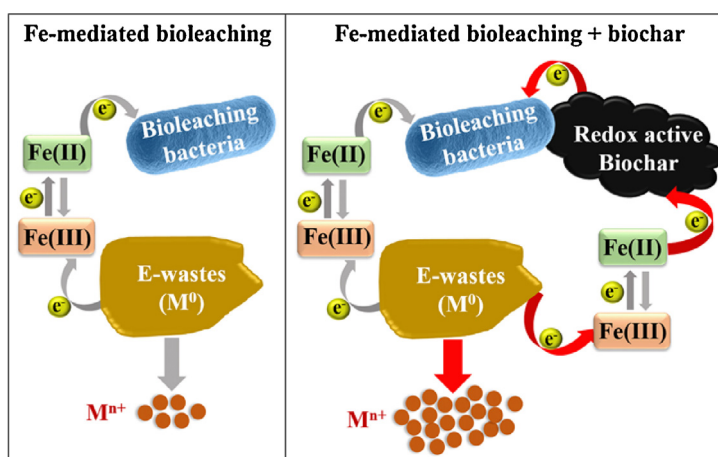
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## HIGHLIGHTS

- Developing a strategy to enhance the efficiency of bioleaching by biochar.
- The role of biochar is discussed from C-, S- and Fe-mediated bioleaching.
- The composition of function microbes is regulated by biochar for bioleaching.
- *Alicyclobacillus* spp. and *Sulfobacillus* spp. cooperate in Fe-mediated bioleaching.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Electronic wastes (E-wastes) contain a huge amount of valuable metals that are worth recovering. Bioleaching has attracted widespread attention as an environment-friendly and low-cost technology for the recycling of E-wastes. To avoid the disadvantages of being time-consuming or having a relatively low efficiency, biochar with redox activity was used to enhance bioleaching efficiency of metals from a basic E-waste (i.e., printed circuit boards in this study). The role of biochar was examined through three basic processes: Carbon-mediated, Sulfur-mediated and Iron-mediated bioleaching pathways. Although no obvious enhancement of bioleaching performance was observed in the C-mediated and S-mediated systems, Fe-mediated bioleaching was significantly promoted by the participation of biochar, and its leaching time was decreased by one-third compared with that of a biochar-free system. By mapping the dynamic concentration of Fe(II) and Cu(II), biochar was proved to facilitate the redox action between Fe(II) to Fe(III), which resulted in effective leaching of Cu. Two dominant functional species consisting of *Alicyclobacillus* spp. and *Sulfobacillus* spp. may cooperate in the Fe-mediated bioleaching system, and the ratio of these two species was regulated by biochar for enhancing the efficiency of bioleaching. Hence, this work provides a method to improve bioleaching efficiency with low-cost solid redox media.

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## 1. Introduction

With the coming of the Information Age, E-waste has become the fastest growing solid waste stream worldwide [1]. According to a survey by United Nations University, the total amount of E-waste reached 41.8 million tons in 2015 and is projected to increase by 21% to 50 million tons by 2018 [2]. The improper disposal of E-waste would trigger a huge increase in toxic metal and organic contaminant pollution [3]. However, the study also noted that 16 million tons of iron, 1.9 million tons of copper and other precious metals could be found in large amounts in E-waste [2]. E-waste can be considered to be a potential man-made mine for turning “wastes” into wealth [3,4].

Printed circuit boards (PCBs) are basic electronic equipments that are part of various electronic instruments [5]. The traditional treatments of discarded PCBs can be categorized as follows [5–7]: mechanical processing, pyrometallurgy, hydrometallurgy and biometallurgy. Compared with other technologies, bioleaching is a promising and sustainable technology that is low cost and eco-friendly. To summarize the previous research, there are three primary methods of bioleaching via different bioleaching bacteria with various substrates, including the Carbon-mediated pathway [8], Sulfur-mediated pathway [9] and Iron-mediated pathway [10,11]. C-mediated bioleaching can be achieved through the reaction between E-wastes and microbial metabolites (e.g., cyanide, citrate and oxalate) produced by microbes through carbon metabolism. S-mediated bioleaching is based on reduced sulfur-oxidizing microbes to produce sulfuric acid, which leads to acidizing metals from E-wastes. Fe-mediated bioleaching includes two steps: Fe(II), serving as an electron donor to Fe-oxidizing microbes, is oxidized to Fe(III), and then the metal  $M^0$  is sequentially oxidized to ionic  $M^{n+}$  by donating electrons to Fe(III). There is one common key point for C-mediated, S-mediated and Fe-mediated bioleaching systems, which is that bioleaching is achieved via metal redox reactions and acidizing which are affected by the complexity and diversity of microbes. Hence, it is worthwhile to explore the contributions of three basic pathways on the performance of bioleaching and to further propose corresponding regulatory strategies.

This study constructed a functional mediator to develop a method for faster bioleaching in order to overcome shortcomings such as being time-consuming or having relatively low efficiency. Compared with liquid mediators, solid mediators are more prone to recycling in water treatment systems. Biochar, produced from biomass via pyrolysis, has been widely used to store carbon, remediate pollution, reduce greenhouse gas and promote soil quality in innumerable agricultural and environmental applications [12–14]. Biochar can also provide a biocompatible surface for microbes to attach [12] and has been well-known for abundant surface functional groups such as quinone and aromatic structures [15,16]. One excellent benefit is that biochar can facilitate the electron transfer in both abiotic [17] and biotic processes [13,14]. The essence of the bioleaching process is achieved via redox reactions based on electron transfer. Faster bioleaching of metals from E-wastes is likely to be achieved by accelerating the electron transfer.

In this work, biochar was used as the exogenous mediator to regulate the redox reactions of bioleaching from PCBs. The role of biochar was evaluated from three typical pathways (C-mediated, S-mediated and Fe-mediated bioleaching). The corresponding leaching dynamics were curved to reflect their leaching efficiency. To understand the function of microbes, the responses of microbial communities were explored using amplification sequencing.

## 2. Experiments

### 2.1. Preparation and characterization of biochar

The biochar used in this study was produced from activated sludge (provided by a sewage treatment plant in Xiamen Jimei, Fujian province, China) via slow pyrolysis. The temperature of the furnace with a limited oxygen supply was programmed to increase at 10 °C/min up to 500 °C. The biochar was powdered to a particle size of 100-mesh. We characterized the biochar with Brunauer-Emmett-Teller (BET) surface area analysis and Fourier transform infrared (FTIR) spectroscopy. Biochar properties are listed in Fig. S1 of the supporting information. The maximum adsorption capacity per gram of biochar for Cu(II) was approximately 2 mg, which served as a control.

### 2.2. Printed circuit boards (PCBs)

The commercially available PCBs used in the experiment were obtained from an online e-shop based in China. For experimental use, PCBs were shredded by a cutting machine and then crushed by a multi-function pulverizer with a speed of 31000 r/min for 10 min. The powders were sieved and collected with sizes below 80-mesh. The collected powders were dried in a vacuum oven at 105 °C to a constant weight before they were employed in bioleaching.

For metal determination, 0.10 g of a PCB sample was accurately weighted and then digested with a  $\text{HNO}_3$ -HCl-HF- $\text{H}_2\text{O}_2$  mixture in a microwave digestion system. The digestion solution was filtered through a 0.22- $\mu\text{m}$  membrane and diluted to the appropriate concentration with 2%  $\text{HNO}_3$ . Then, the samples were subjected to inductively coupled plasma-optical emission spectrometry (ICP-OES) for the determination of the metal contents; the results are given in Fig. S2. The average data comes from three replicates.

### 2.3. Microorganisms and culture conditions

Aerobic activated sludge (provided by a sewage treatment plant in Xiamen Jimei, Fujian province, China) was used as the source of microorganisms. M9, Starkey and 9K media were used to enrich three different types of microbial communities. The M9 medium contained the following (g/L):  $\text{NH}_4\text{Cl}$ , 1;  $\text{Na}_2\text{HPO}_4$ , 6;  $\text{K}_2\text{HPO}_4$ , 3;  $\text{NaCl}$ , 0.5. The solution pH was about 7.2 without adjustment and was autoclaved at 121 °C for 20 min. Then 1 mL each of 1 M  $\text{MgSO}_4$  and 0.1 M  $\text{CaCl}_2$  which have been autoclaved at 121 °C for 20 min were added to the solution in a sterile environment. Finally, the glucose was added to the solution to a final concentration of 4 g/L after autoclaved at 115 °C for 15 min [18]. The Starkey medium contained the following (g/L):  $(\text{NH}_4)_2\text{SO}_4$ , 0.4;  $\text{K}_2\text{HPO}_4$ , 3;  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ , 0.5;  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ , 0.01;  $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ , 0.25. The solution pH was adjusted to 2.0 using sulfuric acid and was autoclaved at 121 °C for 20 min. The energy source sulfur (10 g/L) was added to the solution in a sterile environment after intermittent sterilization [19,20]. The 9K medium contained the following (g/L):  $(\text{NH}_4)_2\text{SO}_4$ , 3;  $\text{KCl}$ , 0.1;  $\text{K}_2\text{HPO}_4$ , 0.5;  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ , 0.5;  $\text{Ca}(\text{NO}_3)_2$ , 0.01. The solution pH was adjusted to 2.0 using sulfuric acid and was autoclaved at 121 °C for 20 min. Filter sterilized ferrous sulfate solution was added to the solution to a final concentration of 44.7 g/L in a sterile environment before inoculation [1,20].

To obtain a bacteria consortium, 10 mL of sludge was added to 250 mL flasks with 100 mL M9, Starkey and 9K media, respectively. The flasks were cultured at 30 °C with shaking of 150 r/min for 3–8 days. To domesticate microbial leaching capacity, 10 mL of culture medium was transferred to 100 mL of M9, Starkey and 9K media with different amounts of PCBs.

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