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# Effect of Graphite on Copper Bioleaching from Waste Printed Circuit Boards

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**Abstract:** The efficient extraction of copper as a valuable metal from waste printed circuit boards (WPCBs) is currently attracting growing interest. Here, we systematically investigated the impact of bacteria on the efficiency of copper leaching from WPCBs, and evaluated the effect of graphite on bioleaching performance. The HQ0211 bacteria culture containing *Acidithiobacillus ferrooxidans*, *Ferroplasma acidiphilum*, and *Leptospirillum ferriphilum* enhanced Cu-leaching performance in either ferric sulfate and sulfuric acid leaching, so a final leaching of up to 76.2% was recorded after 5 days. With the addition of graphite, the percentage of copper leaching could be increased to 80.5%. Single-factor experiments confirmed the compatibility of graphite with the HQ0211 culture, and identified the optimal pulp density of WPCBs, the initial pH, and the graphite content to be 2% (*w*/*v*), 1.6, and 2.5 g/L, respectively.

**Keywords:** waste printed circuit boards; graphite; bacteria; leaching; copper

## 1. Introduction

The capability of bacteria in oxidizing pyrite and, hence, dissolving its copper content was first realized in the middle of the last century, opening a window to great biohydrometallurgical opportunities [1]. Modern bioleaching processes rely on the function of specialized micro-organisms to solubilize valuable target metals. Subsequently, pure metals can be extracted with techniques such as solvent and ion exchange. Bioleaching technology has the advantage of being energy-efficient without generating toxic gases. Therefore, it has been widely used to recover valuable metals from low-grade minerals, and to remove heavy metals from various sources such as sludge, fly ash, polluted soil, and sediment [2–6]. Furthermore, bioleaching was found to be a viable method to leach metals from electronic waste, such as used batteries and waste printed circuit boards (WPCBs) [7,8].

A huge amount of electrical and electronic equipment (EEE) reaches the end of its life every day and ends up as waste. With the constant improvement of EEE performance, an increasing amount of such waste is generated [9]. Only in 2016, 44.7 million metric tons of e-waste were globally generated, according to the Global E-Waste Monitoring Report. The report predicted that global

e-waste production will probably increase to 52.2 million metric tons by 2021. Electronic products such as TVs, computers, mobile phones, and tablets contain printed circuit boards (PCBs), accounting for about 3.1% of total EEE weight [10]. Because of the consumption of electronic products, the number of WPCBs is rapidly growing [11]. Therefore, metal pollution caused by WPCBs has become a serious environmental problem.

Bioleaching, which is mainly used for sulfidic or oxidized ores [12–14], has also attracted attention for WPCB recycling due to advantages of low cost, low energy consumption, simple operation, and environmental friendliness. Studies have shown that graphite can be used as a catalyst to increase the bioleaching rate of pyrite. In the case of sphalerite, graphite was found to influence microbial populations [15,16]. At present, the role and potential of graphite in WPCB bioleaching has not been reported. This study presents detailed information about the effect of graphite on bioleaching via the shake-flask bioleaching process. We studied the leaching behavior of WPCBs with and without graphite addition to elucidate the reaction mechanism of bioleaching. In addition, the effects of bacteria and chemical reagents on WPCB copper leaching were compared, and the mechanism of bacteria action was clarified. In the process of bacterial growth, the role of graphite was discussed in detail. Finally, the best graphite-leaching conditions for bioleaching copper from WPCBs were determined.

## 2. Materials and Methods

### *2.1. Materials*

WPCBs were extracted from desktop-computer motherboards that were purchased from an e-waste recycling company located in Shanghai, China. WPCBs were crushed using a pulverizer (SJ1000-1, Jiangxi tongyong chemical examination system type equipment Co., Ltd., Nanchang, China) and sieved to obtain a size range of less than 250 μm. The obtained powder was washed with distilled water and dried at 60 °C for 48 h. For chemical analysis, WPCB powder was digested using digesting equipment (aqua regia) followed by analysis using an inductively coupled plasma optical emission spectrometer (ICP-OES, Avio500, PerkinElmer, Waltham, MA, USA). Obtained results are shown in Table 1. The main metal in WPCBs was copper with a content of 23 wt %. In addition, Al, Sn, Ti, Mg, Pb and Zn were present. XRD analysis (Figure 1) confirmed that copper was the only detectable crystalline phase. Graphite powder with an average particle size of 28 μm was purchased from the Shenyang Fifth Reagent Company. Table 1 shows the ICP chemical analysis of WPCBs (wt %).

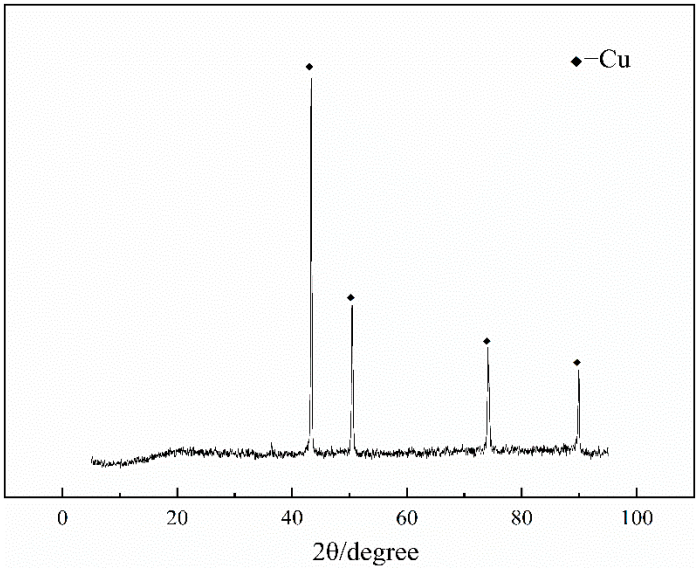


**Figure 1.** X-ray diffraction pattern of WPCB powder (−250 μm) before bioleaching.

**Table 1.** ICP chemical analysis of waste printed circuit boards (WPCBs; wt %).

| Component | $SiO_2$ | CuO | CaO | Br | $Al_2O_3$ | $SnO_2$ | $Fe_2O_3$ | $TiO_2$ | MgO | PbO | SrO | ZnO | Others |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Content | 33.07 | 23.41 | 19.11 | 12.13 | 7.32 | 1.02 | 0.65 | 0.42 | 0.39 | 0.28 | 0.13 | 0.11 | 1.96 |

### 2.2. Micro-Organisms and Cultivation

The HQ0211 culture (*Acidithiobacillus ferrooxidans*, *Ferroplasma acidiphilum*, *Leptospirillum ferriphilum*) was cultured at 9 K in a medium containing 3.0 g $(NH_4)_2SO_4$, 0.1 g KCL, 0.5 g $K_2HPO_4$, 0.5 g $MgSO_4{\cdot}7H_2O$, 0.01 g $Ca(NO_3)_2$, and 44.2 g $FeSO_4{\cdot}7H_2O$ in 1 L distilled water. The initial pH of the medium was adjusted to 1.6 using concentrated sulfuric acid. Then, 10% (*v*/*v*) of bacterial suspension was inoculated in 200 mL growth medium and incubated in an orbital shaker (HZQ-QX, Harbin Donglian Electronic Technology Development Co., Ltd., Harbin, China) at 170 rpm with a constant temperature of 45 °C. Bacterial growth was quantitatively analyzed by optical microscopy and the blood-cell-counting chamber method until cell concentration reached $1.0 \times 10^8$ cells per mL. All reagents were analytical-grade (AR).

### 2.3. Chemical Leaching

To determine the role of bacteria in leaching process, WPCB powder was first leached with sulfuric acid and ferric sulfate.

For this, the initial pH, pulp density, and the concentration of ferric ions were adjusted to 1.6, 5% (*w*/*v*), and 9 g/L, respectively. Leaching experiments were carried out at 45 °C and 170 rpm in an orbital shaker. WPCBs contain brominated flame retardants, and these compounds, together with pH value, can affect the growth of bacteria. The pH value of the leaching samples was adjusted using 1:1 sulphuric acid to between 1.6 and 1.8 throughout the process. All experiments were conducted in triplicate.

### 2.4. Graphite Compatibility with Bacteria

Various quantities (0.1 and 0.5 g) of graphite were added to 500 mL Erlenmeyer flasks that contained 200 mL bacterial culture, and were incubated in an orbital shaker at 170 rpm with a constant temperature of 45 °C for 5 days. In addition, a blank control group without graphite was established. Planktonic cell concentration and of the attached form were periodically monitored to determine the number of bacteria and thus to evaluate possible graphite toxicity to the bacteria. The bacterial suspension was centrifuged at low speed, and the supernatant was used for the determination of planktonic bacteria. The same volume of PBS buffer was added to the centrifuge tube after the supernatant was removed, followed by centrifugation by a vortex oscillator (Vortex Genie 2) for several minutes. The supernatant was absorbed by centrifugation for the determination the attached bacteria.

### 2.5. Pulp-Density Effect

Various quantities of WPCBs (0, 2, 4, 10, and 20 g) were added to 500 mL Erlenmeyer flasks containing 200 mL bacterial culture for the biological experiments. The flasks were incubated at 45 °C and 170 rpm in an orbital shaker for 5 days. Cell concentration, copper-leaching percentage, and redox potential ($E_h$), which was measured with a Pt electrode by using a calomel electrode ($Hg/Hg_2Cl_2$), were periodically monitored to determine the tolerance of bacteria to WPCBs. All experiments were done in triplicate.

### 2.6. Graphite Effect

To evaluate the impact of graphite, experiments were conducted in 500 mL flasks containing 200 mL bacterial culture with the addition of various amounts of graphite (0, 0.1, 0.3, and 0.5 g). The initial redox potential and inoculum were 650 mV and $3 \times 10^8$ cells per mL, respectively. The flasks were preincubated at 45 °C and 170 rpm in an orbital shaker for 3 days, and then 4 g dried WPCB

powder for leaching was added for 5 days. All experiments were carried out in triplicate on the basis of the flowchart shown in Figure 2.

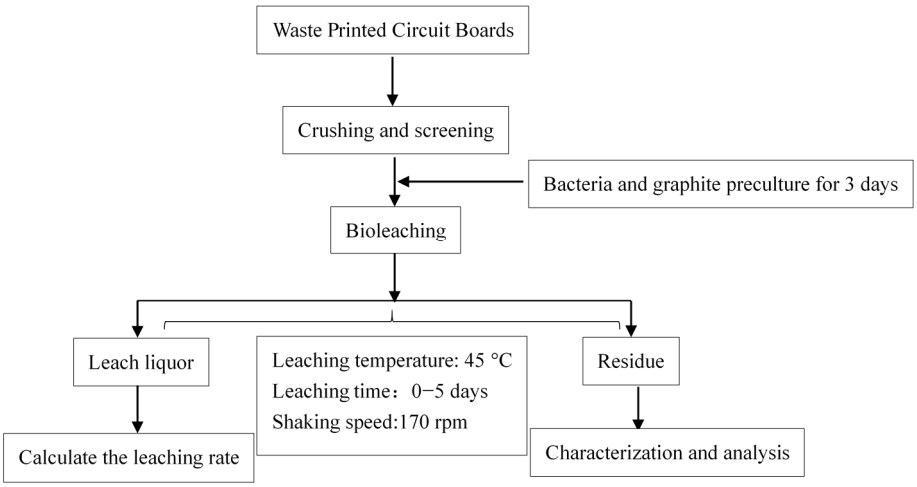


**Figure 2.** Flow diagram of conducted experiments to evaluate graphite function in WPCB copper bioleaching.

### *2.7. Analytical Methods*

The pH of the leachate was measured by a pH meter (pHS-25, INESA Scientific Instrument Co., Shanghai, China). $E_h$ was monitored by calomel electrode (TM39, INESA Scientific Instrument Co., Ltd., Shanghai, China). The phase composition of the samples was studied by X-ray diffraction (XRD, Cu target, Kα rays, Philips X' pert, PANalytical, Westborough, MA, USA). The morphology and chemical composition of powders were analyzed by scanning electron microscopy (SEM, SU-8010, Hitachi high technology Co., Ltd., Tokyo, Japan), equipped with an energy dispersive spectroscope (EDS). Thermo Nicolet 380 Fourier transform infrared spectrometer (Thermo Fisher, Cambridge, MA, USA) was used to measure the infrared spectrum (FT-IR). $Fe^{2+}$ and $Cu^{2+}$ concentration was measured by potassium dichromate titration and iodometry, respectively. Copper leaching was evaluated using Equation (1):

$$\text{copper leaching percent } = C_1/C_2 \times 100\%, \quad (1)$$

where $C_1$ is copper concentration in the leachate, and $C_2$ is the initial WPCB copper concentration.

### *2.8. Statistical Experiment Design*

All experiments were done in triplicate. The data are presented as means ± SD to evaluate the statistical significance between the 3 parametric groups. Statistical analysis was conducted with Origin 9.0 software.

## 3. Results and Discussion

### *3.1. Chemical Leaching*

The performance of the leaching and bioleaching processes to extract copper from WPCBs depended on experiment conditions, as exhibited in Figure 3. Copper-bioleaching extraction had higher performance than that of chemical leaching. With the increase of leaching time, copper leaching in a sterile ferric sulfate increased to a maximum value of 44% after around 3 days, and then fluctuated around this value at longer leaching periods. The efficiency of copper leaching in the acid solution was far less than that of the ferric sulfate, so that only 9% copper leaching was recorded after 3 days. At this time, the copper leaching of the bacterial group was found to be 60%, which was further increased to

76% after 5 days. Interestingly, with the presence of 0.12 g graphite, leaching performance was increased to a value of 80%, demonstrating the positive impact of graphite in enhancing bioleaching performance.

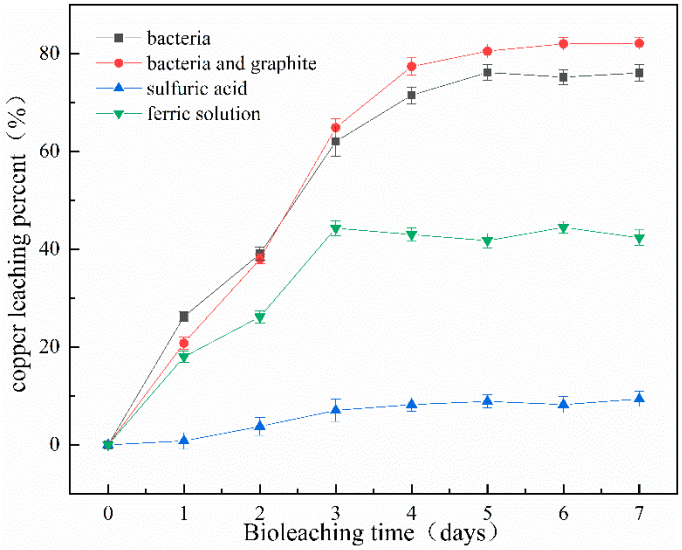


**Figure 3.** Copper-leaching percentage (mean ± SD) versus leaching time under different conditions (bacteria without graphite; bacteria with 0.6 g/L graphite; pH = 1.6 sulfuric acid and 9 g/L ferric sulfate, respectively; other conditions: 45 °C, 170 rpm).

### *3.2. Graphite Compatibility with Bacteria*

Graphite is often used in various applications and types of equipment. However, the use of graphite for the microbial leaching of WPCBs has not been reported in the literature. Before the catalytic experiment, compatibility between bacterial culture and graphite was tested. As can be seen in Figure 4, the bacteria in the assays with the graphite grew in free form and were adsorbed. The number of free and adsorbed bacteria was $1.7 \times 10^8$ and $1.3 \times 10^8$, respectively. The total number was equal to the control assay, which was $2.9 \times 10^8$. Results indicated that graphite did not inhibit the growth of bacteria.

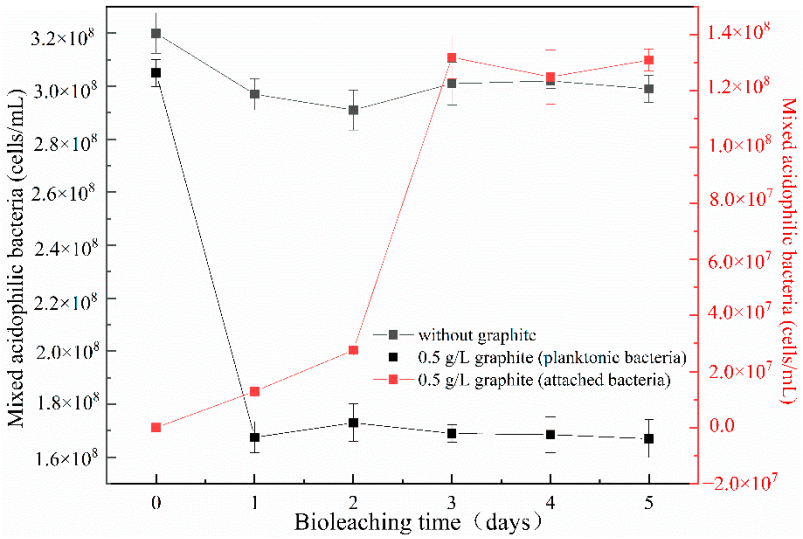


**Figure 4.** Planktonic- and attached-cell concentration (mean ± SD) during bacterial growth with graphite addition (other conditions: 1.6 pH, 0.5 g/L graphite, 45 °C, 170 rpm).

The infrared spectra of the original graphite sample and of the graphite sample treated by bacteria are shown in Figure 5. The graphite involved in bacterial growth exhibited $\upsilon_3$ stretching vibration peaks for $SO_4^{2-}$ at 1183 and 1082 $cm^{-1}$. $FeO_6$ octahedral vibration appeared at 468 $cm^{-1}$, indicating that jarosite was attached to the surface of the graphite material [17]. In addition, a stretching vibration peak related to the saturated C=O bond appeared at 1737 $cm^{-1}$. Other peaks that were detected in the spectrum include a $-NO_2$ peak at 1510 $cm^{-1}$, benzene ring C=C at 1463 $cm^{-1}$, aliphatic C=O at 1387 $cm^{-1}$, acetate C=O or aryl ether at 1239 $cm^{-1}$, and aromatic compounds at 833 and 738 $cm^{-1}$ [18].

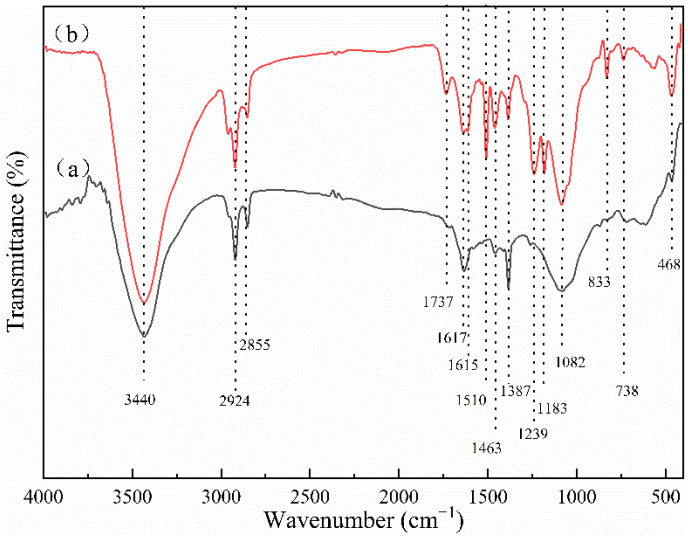


**Figure 5.** FT-IR spectra of graphite in bacterial growth after coculture for 5 days: (**a**) before co-culture; (**b**) after co-culture (other conditions: 1.6 pH, 0.5 g/L graphite, 45 °C, 170 rpm).

Graphite morphology before and after being used in the leaching process is shown in Figure 6. As can be seen in Figure 6a, the raw graphite material contained curved graphite flakes with a layered structure. The morphology of the graphite sample after interaction with the bacteria is shown in Figure 6b. Obviously, graphite flakes accumulated, reducing their surface area. On the other hand, such accumulated carbon particles can form flat substrates on which bacteria can be attached.

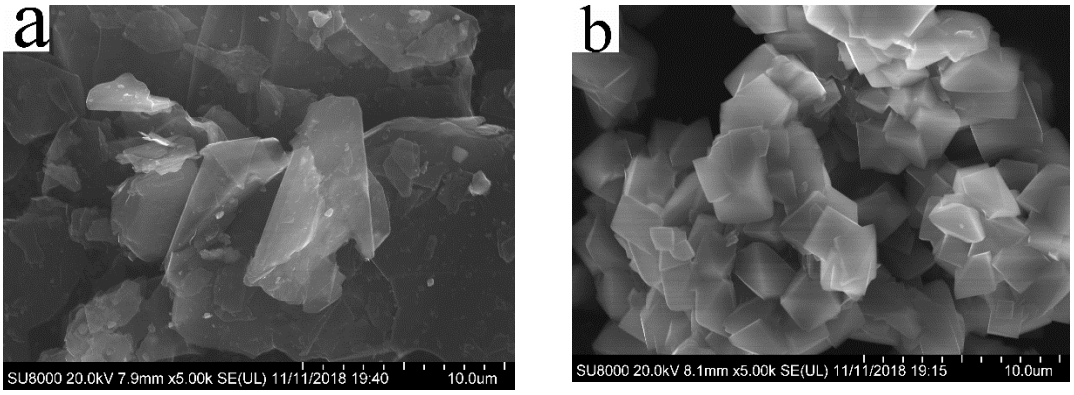


**Figure 6.** SEM graphite micrographs in bacterial growth after coculture for 5 days: (**a**) before co-culture; (**b**) after co-culture (other conditions: 1.6 pH, 0.5 g/L graphite, 45 °C, 170 rpm).

### *3.3. Pulp-Density Effect*

Figure 7 shows the bacterial-growth curve, redox potential ($E_h$), and copper leaching for assays with up to 10% (*w*/*v*) pulp density. With the increase of pulp density, the growth of bacteria decreased, so that, at 10% pulp density, cell concentration was far less than $1.0 \times 10^8$ cells per mL. This is because

friction between particles was promoted with the increase of pulp concentration, hindering bacterial growth. In fact, the bacteria's tolerance was reduced in the presence of a large quantity of heavy-metal species [19,20]. On the other hand, Figure 7b and Equations (2) and (3) [21] indicate that the redox potential initially decreased with increasing bioleaching time due to the presence of trivalent iron oxide copper. Then, potential increased at longer periods since the bacteria adapted to the environment [19]. Copper-leaching percentage decreased with increasing WPCB dosage (Figure 7c). At 1% (*w*/*v*) and 2% (*w*/*v*) pulp density, bacteria adapted well in terms of the leaching environment. In this study, maximum leaching for copper was obtained at 1% (*w*/*v*) pulp density. The leaching percentage at 1% (*w*/*v*) and 2% (*w*/*v*) pulp density was almost the same after 5 days, and copper leaching reached 90% after 3 days. When continuing to increase pulp density to 10%, copper-leaching percentage was only 35%. High WPCB dosage resulted in lower copper extraction partly due to limitations in air distribution and oxygen mass transfer, which inhibited the oxidation efficiency of $Fe^{2+}$ by the bacteria [8,22,23]. In addition, copper concentration increased with WPCB dosage. It can be seen from the Figure 7a that the ability of bacteria to resist copper decreased. Considering the obtained results for bacterial growth and copper leaching, a pulp density of 2% was selected for the subsequent experiments.

$$Cu + 2Fe^{3+} \rightarrow Cu^{2+} + 2Fe^{2+} \quad (2)$$

$$4Fe^{2+} + 4H^{+} + O_2 \xrightarrow{HQ0211} 4Fe^{3+} + 2H_2O \quad (3)$$

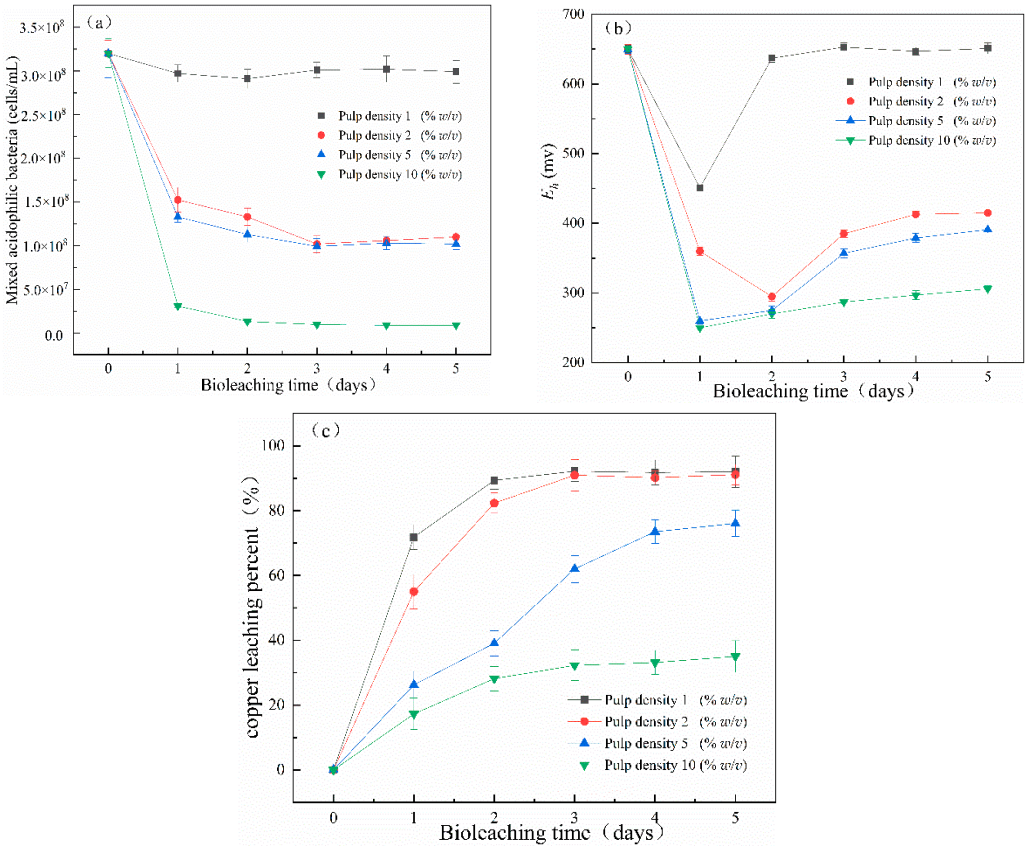


**Figure 7.** (**a**) Cell concentration, (**b**) redox potential, and (**c**) copper-leaching percentage (mean ± SD) during copper bioleaching at different WPCB pulp densities (other conditions: 1.6 pH, 45 °C, 170 rpm).

### *3.4. Graphite Effect on Copper Bioleaching*

The effect of graphite on copper ion concentration in the bacterial leaching assays is shown in Figure 8. As depicted, in the absence of graphite, the leaching of copper from WPCBs amounted to 90%

after 5 days. Leaching performance increased with the increase of graphite addition, so that the final copper concentration in the bioleaching assays, with the presence of 0.5, 1.5, and 2.5 g/L of graphite, was detected to be 93.66%, 95.22%, and 100%, respectively. As shown in Figure 4, some bacteria were adsorbed on the graphite surface. As discussed, the main mechanism involved in the bioleaching of copper from WPCBs is $Fe^{3+}$ oxidation. Graphite can act as the cathode accelerating the dissolution of copper anodes [16].

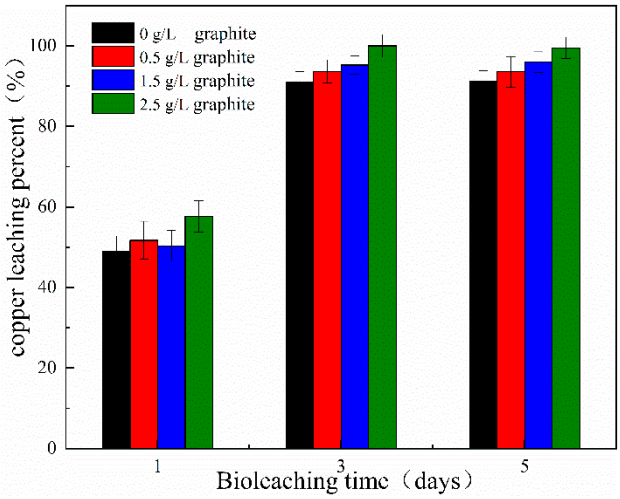


**Figure 8.** Copper-leaching percentage (mean ± SD) during WPCB bioleaching at different graphite concentrations (other conditions: 1.6 pH, 2% (*w*/*v*) pulp density, 45 °C, 170 rpm).

As observed in Figure 8, a 10% increase in the copper leaching was recorded by adding 0.5 g graphite to the system. In order to study the action mechanism of graphite, leaching residues were analyzed by infrared spectroscopy and scanning electron microscopy for assays with a graphite addition of 2.5 g/L.

The FT-IR spectra of the residual materials retrieved from the bioleaching bath with and without the involvement of graphite are shown in Figure 9, in which peak absorption frequencies associated with chemical bonds or functional groups can also be seen. The absorption peak at 3396 $cm^{-1}$ belongs to the stretching vibration of O–H. Antisymmetric and symmetric stretching vibrations of $-CH_2$ appeared at 2930 and 2854 $cm^{-1}$, respectively. C=O stretching vibrations for carboxyl and carbonyl groups appeared at 1720 $cm^{-1}$. The bending vibration of water molecule –OH was located at 1640 $cm^{-1}$, and the stretching vibration of C=C at 1507 $cm^{-1}$. The FTIR spectrum of the residual WPCB material obtained after the bioleaching process with the presence of graphite is exhibited in Figure 9b. The spectrum shows the $\upsilon_3$ stretching vibration peaks for $SO_4^{2-}$ at 1190 and 1079 $cm^{-1}$. The deformation vibration of O–H and H–O–H could be observed at 1004 and 636 $cm^{-1}$, respectively. The octahedral vibrations of $FeO_6$ appeared at 506 and 473 $cm^{-1}$. The existence of characteristic peaks of $SO_4^{2-}$ and $FeO_6$ in the spectrum indicated the formation of jarosites. Furthermore, the presence of the absorption peak of $NH^{4+}$ at around 1430 $cm^{-1}$ demonstrated the precipitation of such jarosite species [17].

Figure 10 shows the scanning-electron-microscope images combined with EDS map analysis of the raw WPCB powder material, and of the biological leaching residue from the optimized assay in which graphite content was 2.5 g/L. The copper presence in the samples is highlighted by arrows in the micrographs. As can be observed in Figure 10a, in the raw WPCB, a relatively high amount of copper was distributed on the plastic substrate as distinguished clusters. According to Figure 10b, the total amount of existing copper in the sample obviously decreased after the bioleaching process without the presence of copper. On the other hand, no obvious copper could be detected in the WPCB sample after bioleaching with the presence of graphite (Figure 10c). These results further confirm the positive effect of graphite on the bioleaching assays.

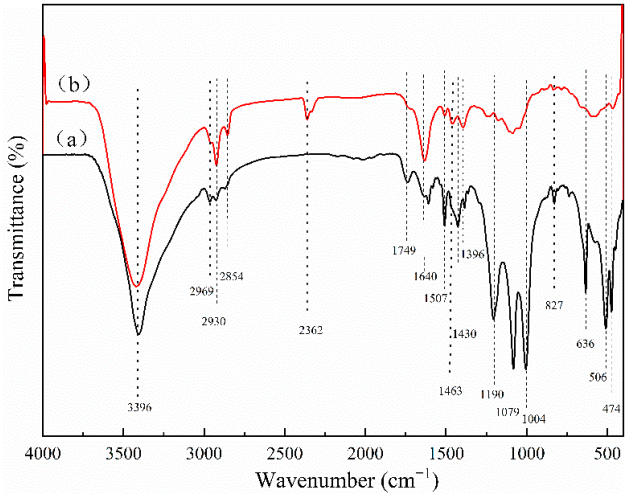


**Figure 9.** FT-IR of residual material retrieved after copper bioleaching from WPCBs. (**a**) Original WPCB powder and (**b**) WPCBs for residues containing 2.5 g/L graphite.


**Figure 10.** SEM and EDS map analysis of WPCBs (**a**) before bioleaching; WPCB surface morphology for residues (**b**) after bioleaching and (**c**) containing 2.5 g/L graphite under bioleaching condition.

## 4. Conclusions

A systematic investigation was conducted to evaluate the effect of bacteria and graphite on copper leaching from WPCBs. Results showed that: (1) The presence of bacteria improved the leaching of copper from WPCBs. (2) The graphite addition enhanced the dissolution of copper from WPCBs. (3) Bacteria could grow in the presence of graphite planktonically and in an attached form. (4) An optimal graphite addition could be recorded. In the presence of 2.5 g/L graphite, the leaching percentage of $Cu^{2+}$ reached 100%.

**Author Contributions:** Conceptualization, L.T.; methodology, L.T. and Q.Z.; validation, H.Y.; formal analysis, L.T.; investigation, Q.Z.; data curation, Q.Z.; writing—original-draft preparation, Q.Z.; writing—review and editing, L.T., W.S., A.R.K. and Q.Z.; funding acquisition, H.Y. All authors have read and agreed to the published version of the manuscript.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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