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An innovative biotechnology for metal recovery from printed circuit boards

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ABSTRACT

The increase of waste from electric and electronic equipment has pushed the research towards the development of high sustainability treatments for their exploitation. The end-of-life printed circuit boards (PCBs) represent one of the most significant waste in this class. The interest for these scraps is due to the high Cu and Zn content, with concentrations around 25% and 2% respectively, combined with further precious metals (e.g. Au, Ag, Pd). Currently, the most common approaches developed for PCBs recycling include pyrometallurgical and hydrometallurgical treatments. On the other hand, biohydrometallurgical strategies are gaining increasing prominence, for the possibility to decrease both the environmental and the economic costs. Nevertheless, these techniques show the main limit due to the possibility to treat low quantities of waste, which makes unsustainable the further scale-up. To overcome this criticality, the present paper introduces an innovative bioleaching process carried out by *Acidithiobacillus ferrooxidans* (*At. ferrooxidans*) and *Leptospirillum ferrooxidans* (*L. ferrooxidans*). The developed technology allows to reach high PCB concentration, up to 5% (w/v), thanks to a high efficiency two-step design, able to reduce the metal toxicity on the bacteria metabolism. The treatment uses the ferric iron generated by bacterial oxidation, as oxidant, to leach Cu and Zn from PCBs. The possibility to overcome the solid concentration criticality is combined with high yield of 94% and 70% for Cu and Zn, respectively. The best selected conditions involve the *At. ferrooxidans* bacteria use at: 30 °C, solid concentration of 5% (w/v), 10 g/L of Fe^{2+}, time of treatment 9 days. The experimental results are further enhanced by the carbon footprint assessment which proved the environmental advantage, compared to both the reference chemical treatment through ferric iron and literature processes (hydrometallurgical and bioleaching approaches). The analysis explained as the PCBs concentration in the solution allows to decrease the bioreactor size with the consequent reduction of energy and raw material demand. This benefit can be translated into a 4 times reduction of the CO_2-eq./kg treated PCB emissions, compared to the best bioleaching processes, reported in the literature.

1. Introduction

In the last two decades, the amount of waste from electrical and electronic equipment (WEEE) had an exponential growth (about 45 million tons, in 2016), mainly due to both the new technologies, that have replaced the old and obsolete equipment, and their relatively short replacement cycles (Baldé et al., 2017; Zhang and Xu, 2016). In 2016, Asia was the region that produced most of the WEEE, with 18 Mt, followed by Europe (12 Mt) and USA (7 Mt). Nevertheless, only a scarce fraction, around the 20% of the whole stream, was documented to be collected and properly recycled (Baldé et al., 2017). The illegal WEEE management is mainly due to their content of hazardous components which makes impossible the implementation of traditional methods, such as disposal in landfilling sites or incinerator, for the human and the environmental protection (Leung et al., 2008; Musson et al., 2006; Shen et al., 2008). The end-of-life printed circuit board (PCBs) is a typical example of WEEE, which is the basal and essential component of the electronic industry, since it represents the brain of all the electronic products (Xiang et al., 2010). The PCBs constitute the 3–5% of the WEEE amount and they are one of the most relevant waste of the category since their composition combines many environmental restrictions with several economic advantages for the valuable metal content (Faraji et al., 2018; Priya and Hait, 2017). Furthermore, the quantity and the purity of these metals are higher than those in rich-content minerals (Li et al., 2007; Xiang et al., 2010). In this context, the metal recovery, mainly focused on Cu and precious elements, produces a

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Table 1
Literature about the average Cu concentration in the end-of-life PCBs.

Cu content (%)	References
< 20	(Arshadi and Mousavi, 2015a; Bas et al., 2014, 2013; Jadhav et al., 2016; Natarajan and Ting, 2014; Pant et al., 2012; Pradhan and Kumar, 2012; Priya and Hait, 2017; Veit et al., 2005; Vestola et al., 2010)
20–30	(Birloaga and Vegliò, 2016; Creamer et al., 2006; Ilyas and Lee, 2014; Wang et al., 2018; Xiang et al., 2010; Yang et al., 2009, 2014; Zhang and Xu, 2016)
> 30	(Faraji et al., 2018; Hong and Valix, 2014; Wang et al., 2009; Zhu et al., 2011)

double advantage: the avoided primary mining and the enhancement of a hazardous waste. The whole effect is a relevant advantage for: the environmental, the human health protection and the economic spheres (Ilyas et al., 2013; Natarajan and Ting, 2015; Veit et al., 2005).

Currently, the most common approaches used for the PCBs recycling include both pyrometallurgical and hydrometallurgical treatments. The first kind of process is mainly based on the smelting technologies that require temperatures between 300 and 900 °C (Rocchetti et al., 2018). On the other hand, the hydrometallurgical technologies use chemical solutions for metal dissolution, mainly hydrochloric, sulfuric and nitric acid or/and hydrogen peroxide (Bas et al., 2014; Rocchetti et al., 2018; Zhang and Xu, 2016). For the further Au recovery, the most common leaching agent is cyanide with the consequent production of contaminated wastewater (Behnamfard et al., 2013).

In the last decades, biohydrometallurgical strategies has gained increasing prominence in this field. Indeed, the microorganism use could be more cost efficient and environmentally friendly than the chemical approaches (Beolchini et al., 2012; Ilyas et al., 2013). Furthermore, the bio approaches allow to solve the main limits of pyrometallurgical methods due to the request of high temperatures. Several works investigated the acidophilic bacteria ability for metal extraction from WEEE, mainly using: *Acidithiobacillus thiooxidans* and *Acidithiobacillus ferrooxidans*. High efficiencies, greater than 95% were achieved by ferrous iron concentration between 4 and 10 g/L, a solid concentration between 0.78 and 2.8% (w/v), a leaching time from 2 to 10 days (Arshadi and Mousavi, 2015a; Choi et al., 2004; Liang et al., 2013; Wang et al., 2009; Wu et al., 2018; Yang et al., 2009, 2014; Zhu et al., 2011), also using pyrite, as ferrous iron energy source for bacteria metabolism (Bas et al., 2013). Comparable extraction results were obtained by the microbial community recovered from pyrite mine or activated sludge taken from municipal sewage treatment plant (Liang et al., 2010a; Xiang et al., 2010). On the other hand, cyanobacteria (*Chromobacterium violaceum, Pseudomonas fluorescens and P. aeruginosa*) showed an affinity to Au, with recovery efficiency of 10%, with a solid concentration between 0.5–1% (w/v) (Chi et al., 2011; Pham and Ting, 2009; Pradhan and Kumar, 2012; Yuan et al., 2019, 2018).

Fungi strains are investigated as an alternative for the metal extraction from WEEE, mainly *Aspergillus niger* and *Penicillium semplicissimum*. Nevertheless, the greatest Cu extraction efficiency was around 70% with PCB concentrations between 0.1 and 0.5% (w/v) (Brandl et al., 2001; Faraji et al., 2018). Jadhav et al. (2016), increased the solid content up to 1% (w/v) adding the hydrogen peroxide to the fermentation medium, to increase the metal extraction by organic acid produced by *A. niger*. In this context, the present work aims at the development of an innovative high efficiency biotechnology for Cu and Zn recovery from end-of-life PCBs. The target is the increase of the treated waste concentration, limiting the metal toxicity on both *At. ferrooxidans* and *L. ferrooxidans* metabolism. This novelty allows to get beyond the state of the art to solve the limits of the current technologies developed in the literature (Arshadi and Mousavi, 2015b; Liang et al., 2016, 2013; Nie et al., 2015a, 2015b; Priya and Hait, 2018; Rodrigues et al., 2015). Furthermore, following the same approach used in the literature for other kind of WEEE, the life cycle assessment (LCA) tool was used to evaluate the process sustainability (Amato et al., 2017, 2016; Amato and Beolchini, 2018; Latunussa et al., 2016; Rocchetti et al., 2013). More in detail, the carbon footprint of the best developed technology was estimated and compared to the chemical and hydrometallurgical approaches.

2. Materials and methods

2.1. Preparation of waste printed circuit boards (PCBs)

The PCBs used in this study were kindly provided by the University of L'Aquila, Italy. The samples of high grade PCBs, mainly from personal computers, were prepared as follows: the material was shredded using stainless steel blades and pliers after manually removing the main parts of electronic components (e.g. capacitors, batteries and resistors). Thereafter, the refuse was crushed to obtain a granulometry of less than 0.5 mm which was used for all the bioleaching experiments. A consecutive washing, with saturated water (with NaCl), removed the plastics, toxic for bacteria metabolism, improving the metal extraction efficiencies (Ruan et al., 2018). The literature characterization results report average values of 25% and 2%, for Cu and Zn, respectively. More in detail, the Zn content is in a 0.02–7% range, whereas the Cu data, with a significant variability due to the sample heterogeneity, are reported in Table 1.

2.2. Microorganisms and culture conditions

Leptospirillum ferrooxidans (DSMZ 2705^T) and *Acidithiobacillus ferrooxidans* (DSMZ 14882^T), used for the experiments, were obtained from the Deutche Sammlung von Mikroorganismenn und Zellkulturen GmbH, Germany. The DSMZ 882 medium, consisting of three solutions (A, B and C), was prepared as follows; Solution A: 0.132 g of $(NH_4)_2SO_4$, 0.053 g of $MgCl_2 \cdot 6H_2O$, 0.027 g of KH_2PO_4, 0.147 g of $CaCl_2 \cdot 2H_2O$ dissolved in 0.950 L of MilliQ water and pH adjusted to 1.6 with a 2 M H_2SO_4 solution. Solution B: 49.8 g of $FeSO_4 \cdot 7H_2O$, in 50 mL of 2 M H_2SO_4 adjusted to pH 1.2 with NaOH. Solution C (1 L): 0.062 g of $MnCl_2 \cdot 2H_2O$, 0.068 g of $ZnCl_2$, 0.064 g $CoCl_2 \cdot 6H_2O$, 0.031 g of H_3BO_3, 0.010 g of Na_2MoO_4, 0.067 g of $CuCl_2 \cdot 2H_2O$, brought to pH 1.8 with 2 M H_2SO_4 solution. Solution A and C were autoclaved separately, whereas solution B was sterilized by filtration (Sartolab Filter Systems, polyethersulfone PES 0.22 μm) in order to prevent the oxidation of Fe^{2+} to Fe^{3+}. The culture medium was obtained by adding 50 ml of solution B and 1 ml of solution C to the solution A, maintaining the sterility. The bacteria cultures grew in the DSMZ 882 medium and they were incubated, under stirring, at 120 rpm, at 30 °C, for 5 days (Stuart, orbital incubator S510).

2.3. Bioleaching experiments

The bioleaching process (Fig. 1) was conducted in different phases: the growth phase, the first step and the second step processes. The experiments were carried out in 250 mL flasks containing 80 mL of DSMZ 882 medium (10 g/L of Fe^{2+}) and 20 mL of bacterial stock culture. After 48 h of bacterial growth (growth phase), time necessary for the oxidation of Fe^{2+} to Fe^{3+}, 5% (w/v) of PCBs were added to the solution of *At. ferrooxidans*. In the first step the bacterial growth was carried out in the presence of PCBs for 48 h. In the second step, the 80% of medium was removed to reduce the metal toxicity on the bacteria metabolism and the remaining 20%, with the PCBs, was refreshed by

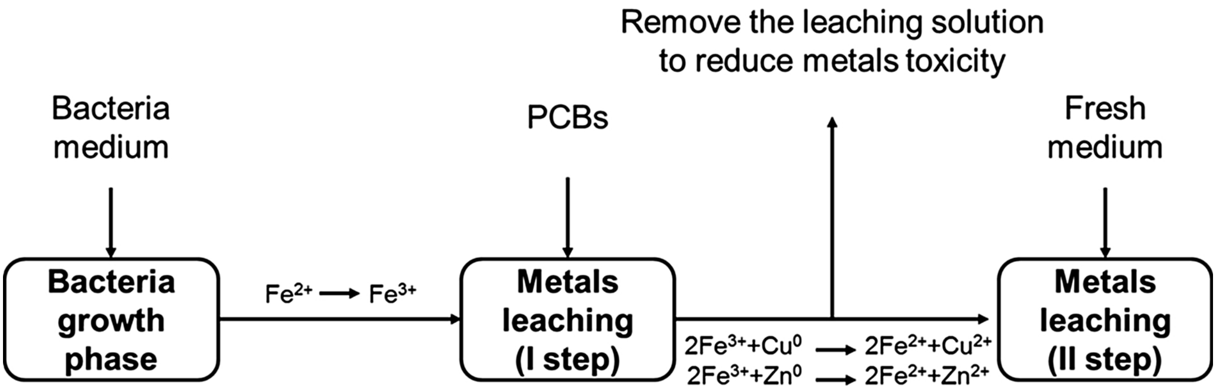

Fig. 1. Bioleaching scheme.

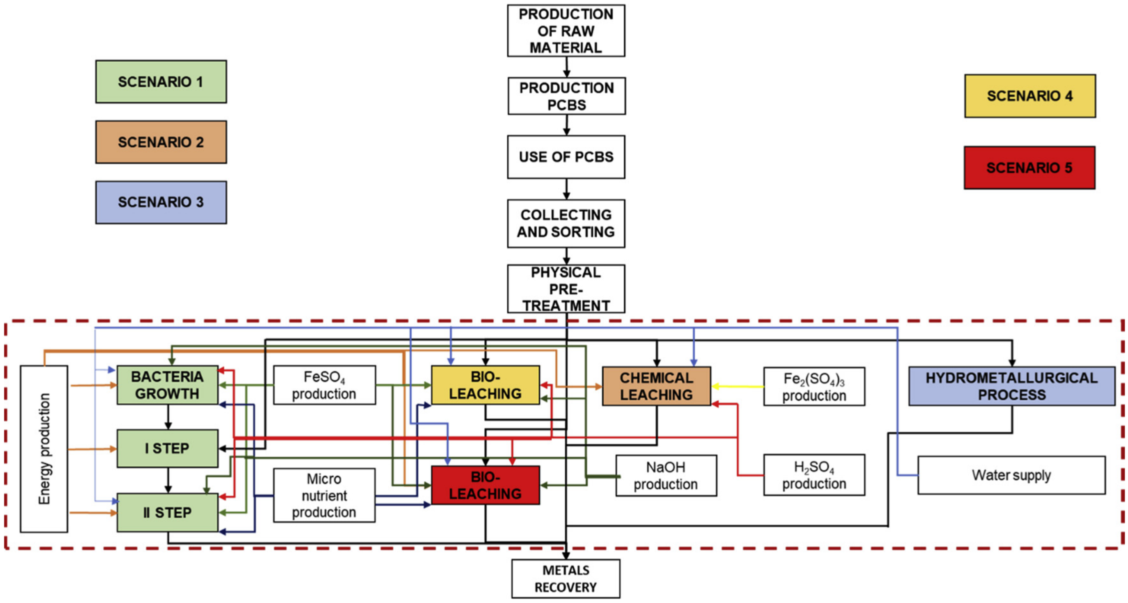

Fig. 2. System boundaries considered for the carbon footprint assessment.

fresh medium, to restore the starting bioleaching volume and the Fe concentration, for further 7 days (Cabrera et al., 2005a, 2005b; Tuovinen et al., 1971). After the inoculation, the flasks were kept in the Stuart incubator (orbital incubator S510) at 30 °C and 120 rpm, for 11 days. The pH was adjusted, every day, at 1.6 value for both *At. ferrooxidans* and *L. ferrooxidans* by the dropwise addition of 1 M H_2SO_4. This condition is optimal for the bacterial growth and to prevent the formation of jarosite precipitation on PCBs, that reduce the reaction kinetic with metals (Boon et al., 1999; Crundwell, 1997; Daoud and Karamanev, 2006). Furthermore, this acidity level does not represent an obstacle in the perspective of an industrial application, as confirmed by Yong Liu et al. (2011) in the registered patent CN10221843. For *L. ferrooxidans*, which has a slower growth rate than *At. ferrooxidans*, the bacterial growth phase was 4 days, after which a 5% (w/v) of the PCBs was added to the solution, carrying out the bioleaching experiment in two steps, at the same conditions of *At. ferrooxidans*.

In order to analyze the bacteria effect on the bioleaching mechanism, a chemical oxidation and an abiotic control test were carried out. More in detail, in the chemical oxidation test, 5% (w/v) of PCBs were added to 100 mL of $Fe_2(SO_4)_3*H_2O$ solution (50 g/L of Fe^{3+}) which is incubated, under stirring at 120 rpm, for several hours, until the whole Fe^{3+} was reduced to Fe^{2+}. The test was repeated at two different temperatures (30 °C and 50 °C) to study the effect of this parameter on the kinetic reaction. On the other hand, the abiotic control was carried in two steps using a $Fe_2(SO_4)_3*H_2O$ solution (10 g/L of Fe^{3+}) to simulate the biological oxidation, under the same conditions of bioleaching treatment. This control aims at the quantification of the positive effect of the bacteria action.

Samples were collected to analyze the concentrations of Fe^{2+}, Fe^{3+}, Cu and Zn. Each treatment was carried out in duplicate.

2.4. Analytical determination

The leaching solutions were periodically analyzed for the determination of both Zn and Cu concentration, carried out by an atomic absorption spectrophotometer (AAS) (Varian spectrometer SpectrAA 200). Each sample of 0.5 mL was diluted in agreement with the detection limit of 0.1 mg/L. On the other hand, the quantification of the Fe content was performed by an UV/VIS spectrophotometer (Jasco Model 7850). The determination was obtained by potassium thiocyanate (KSCN) which allows the quantification of Fe^{3+} (at the absorption wavelength of 480 nm). For the measure of the total Fe, the solution reacts with potassium permanganate to oxidize the Fe^{2+} in the sample. Thereafter, the Fe^{2+} concentration was determined as the difference between the total Fe and the Fe^{3+} concentration. The detection limit of this method was 1 mg/L of Fe, and the adsorption measurement was between 0.000 and 1.000 with an accuracy of ± 0.005.

The pH was recorded by a pH metro inoLab Multi 720 (WTW) and it was monitored over the bioleaching period.

Table 2
Input and output flows considered for the carbon footprint assessment of the five scenarios (Functional unit: 1 kg of shredded PCBs).

	Input flow	Products
Biotechnological treatment (Scenario 1)	Electricity 2.6 kW h $FeSO_4$ 50 g Water 3.4 kg H_2SO_4 0.30 kg NaOH 0.20 kg Micronutrients 8.0 g	Cu 240 g Zn 30 g
Chemical treatment (Scenario 2)	Electricity 4.5 kWh $FeSO_4$ 120 g Water 2.0 kg H_2SO_4 0.30 kg NaOH 0.60 kg	Cu 230 g Zn 30 g
Hydrometallurgical treatment (Scenario 3)	Electricity 0.30 kWh Water 5.0 kg H_2SO_4 1.1 kg H_2O_2 (50% v/v) 0.70 kg	Cu 140 g
Biological Treatment (Liang et al., 2013) (Scenario 4)	Electricity 4.2 kWh $FeSO_4$ 20 g Water 35 kg H_2SO_4 0.17 kg NaOH 0.19 kg Micronutrients 120 g	Cu 120 g
Biological treatment (Wu et al., 2018) (Scenario 5)	Electricity 2.6 kW h Water 200 kg H_2SO_4 0.98 kg NaOH 1.1 kg Micronutrients 1600 g	Cu 650 g

2.5. *Assessment of the carbon footprint*

The determination of the carbon footprint took into account five different scenarios for the treatment of PCBs. The target is the comparison of the impact due to the bioleaching (scenario 1, Fig. 2) and the chemical approaches (scenario 2, Fig. 2), described in the present paper, with a hydrometallurgical treatment (scenario 3, Fig. 2) and two bioleaching approaches (scenarios 4 and 5, Fig. 2), reported in the literature. More in detail, the hydrometallurgical data referred to the process developed by Birloaga and Vegliò (2016), which includes the use of a solution of sulfuric acid and hydrogen peroxide, at room temperature for 1 h, with a solid-liquid ratio of 15% (w/v). On the other hand, the two referenced bioleaching treatments, report a maximum PCB concentration of 2.8% w/v (Liang et al., 2013) and the shortest time of 26 h (Wu et al., 2018), with Cu extraction efficiency higher than 90%". Table 2 summarizes the input and the output flows considered for the assessment, assuming a possible recirculation of water and Fe sulphate of 95%. The micronutrients of scenario 1-4-5 include the agents described in the paragraph 2.2. Considering the lack of data related to the electric consumption of the scenario 2, we referred to Ippolito et al. (2017), which described the exploitation of fluorescent powder at comparable conditions. The values were adapted, on the basis of the reaction time and the solid concentration.

The overall system boundaries (Fig. 2), exclude the steps of: PCB production, use and the pre-treatment of the end-of-life product. All the phases for the Cu and Zn extraction are considered for the analysis, except for the recovery. The functional unit selected for the analysis is 1 kg of shredded PCB.

The thinkstep GaBi software-System and the Database for Life Cycle Engineering (compilation 7.3.3.153; DB version 6.115) were used for the production processes of energy and raw materials and the quantification of the carbon footprint of the assessed scenarios.

3. Results and discussion

3.1. *The chemical leaching*

The results in Fig. 3 show the leaching profiles for time-dependent extraction of Cu and Zn from PCBs and the variability of Fe^{3+} and Fe^{2+} ratio, at the two selected temperatures: 30 °C and 50 °C. The highest Cu extraction (91%) was achieved at 50 °C, after 6 h of leaching, whereas only the 73% of Zn was leached at the same conditions (Fig. 3b). It is evident that the temperature rise exerts a significant effect on the leaching kinetics, reducing the time from 24 to 6 h, and it accelerates the reduction of Fe^{3+} to Fe^{2+}. The constant conversion of Fe, during the chemical oxidation process, proves its essential role for the Cu and Zn dissolution. The Cu and Zn mobilization consumes Fe^{3+}, a strong oxidant, and the rate of dissolved metals is proportional to the reduced Fe, following the Eqs. (1) and (2) (Hong and Valix, 2014; Lee and Dhar, 2012; Liang et al., 2010b; Wu et al., 2018):

$$Cu^0 + 2Fe^{3+} \rightarrow Cu^{2+} + 2Fe^{2+} \tag{1}$$

$$Zn^0 + 2Fe^{3+} \rightarrow Zn^{2+} + 2Fe^{2+} \tag{2}$$

3.2. *The bioleaching process*

The bioleaching takes advantage of the role of bacteria for the Fe^{3+} regeneration, as oxidant, following the Eq. (3) (Lee and Dhar, 2012; Rawlings et al., 2017; Xiang et al., 2010). The bioleaching process permitted to use less Fe than chemical reaction, obtaining the same extraction yield. After Fe^{3+} is reduced in the metal dissolution, Fe^{2+} is again available by bacteria metabolism, used as energy source to increase the bacterial population and the metabolism, ready for further metal oxidation (Fig. 4). This bioleaching process is cyclical and Fe^{3+} has its importance for the leaching reaction.

$$Fe^{2+} + O_2 + H^+ \rightarrow Fe^{3+} + H_2O \tag{3}$$

In spite of the chemical results, which supported the highest

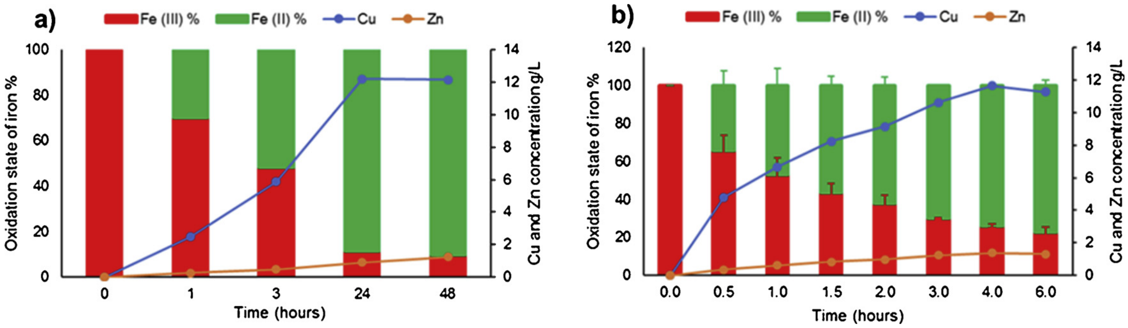

Fig. 3. Influence of the temperature on the kinetic extraction of Cu, Zn from PCBs and oxidation state of Fe in the chemical control: 50 g/L Fe^{3+}, 50 g PCBs at (a) 30 °C, (b) 50 °C.

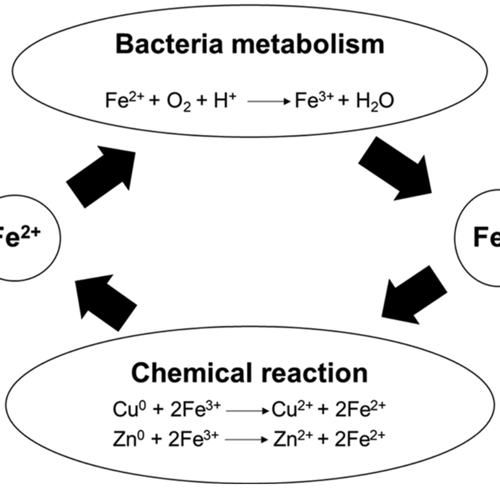

Fig. 4. Fe cyclical process due to the bacteria metabolism action as supporting to the chemical reaction, showed in the abiotic control.

temperature, the incubation was carried out at 30 °C for both the bacteria (*At. ferrooxidans* and *L. ferrooxidans*), since it is the optimal growth condition (Ojumu et al., 2006; Rawlings et al., 2017).

Fig. 5 shows the metal (Cu and Zn) concentrations and the variability of Fe^{2+} and Fe^{3+} (%) with the time, using pure culture of *At. ferrooxidans* and *L. ferrooxidans*. During the first two days (bacteria growth phase), the bacteria *At. ferrooxidans* were cultivated under their optimized conditions to obtain a high activity. At the end of this phase, the Fe^{3+} concentration in solution increased, up to 89%, suitable for Cu and Zn leaching, following the oxidation reaction of Eq. (3). At the end of the first step, the amount of Fe^{3+} decreased until the 50% of the total Fe, compared to the abiotic control, where, the total Fe^{3+} was converted to Fe^{2+}. The amount of Cu recovery was 3.7 ± 0.1 g/L, meanwhile the Zn concentration was 0.38 ± 0.04 g/L in the treatment with *At. ferrooxidans*, more than abiotic control where the Cu and Zn concentration at the end of the first step were 1.78 ± 0.04 and 0.14 ± 0.01 g/L, respectively. These data show a higher efficiency of bioleaching than the chemical technique, thanks to the bacteria activity which oxidized Fe^{2+} to Fe^{3+}, during the first step. The removal of 80% of the medium, replaced with fresh solution, causes the further reduction of Fe^{3+}, until 12% of the whole Fe (II step). At the end of this step, the Fe is completely converted to Fe^{3+}. On the other hand, the abiotic control shows a constant amount of Fe^{3+}, 7% of the total, after 24 h from the beginning of the second step, when fresh leaching solution (10 g/L of Fe^{3+}) was added. Nevertheless, as highlighted in Fig. 5, the additional Cu and Zn leaching is due to both the iron oxidation for the temperature and shaking action and the H_2SO_4 activity (Yang et al., 2014). After 7 days, at the end of the two bioleaching steps, 8.9 ± 0.9 g/L and 1.1 ± 0.3 g/L of the Cu and Zn, respectively, were recovered from PCBs samples, with a whole efficiency higher than the abiotic control. Furthermore, the Fe^{3+} concentration in the treatment with *At. ferrooxidans* shows that the Fe oxidation by bacteria activity (Eq. (3)) was faster than chemical action of Fe, due to the chemical reaction with Cu and Zn (Eqs. (1), (2)). In addition, the maintenance of the pH at 1.6 limits the Fe precipitation as jarosite (Xiang et al., 2010; Yang et al., 2014). The innovative strategy of medium refresh allowed to increase the treated PCBs amount without a toxicity effect on the bacteria metabolism, solving one of the main criticality of the current bioleaching described in the literature (Liang et al., 2010b; Xiang et al., 2010; Yang et al., 2014; Zhu et al., 2011). Furthermore, an external addition of 10 g/L of Fe^{2+} (II step), produced a positive effect on the dissolution rate of Cu and Zn.

The second bacteria used for the bioleaching is *L. ferrooxidans*, under the same conditions of *At. ferrooxidans*. The only difference, compared to the first bacteria, is the PCB addition, carried out after 4 days of bacterial growth. As showed in Fig. 6, *L. ferrooxidans* is very sensitive to the toxic effect of metals and the highest achieved recovery value is around 40% and 20% for Cu and Zn, respectively, as a result of low conversion of Fe^{2+} into Fe^{3+} from bacteria oxidation (Fig. 5).

This result is also confirmed by the literature, which only reports the *L. ferrooxidans* use in mix bacteria culture (Joshi et al., 2017).

Furthermore, the results in Fig. 6 confirm the highest effectiveness of the microbial leaching (using *At. ferrooxidans* as bacteria) compared to the abiotic control, obtaining an extraction yield around 95% and 70% of Cu and Zn, respectively.

The additional quantification of both As and Cr concentration in the leach liquor showed values below the limits established by European Commission for the direct discharges to a receiving water body (The European Commission, 2018). This relevant information supports the low toxicity of the resulting reflue, in the perspective of the treatment implementation on a real scale.

3.3. Kinetic of metal recovery

To better understand the achieved results, the kinetic of metal recovery was studied for all the treatments to highlight the differences between the chemical and the bioleaching approaches. In this regard, the shrinking core model (Eq. (4)), based on a mechanistic study approach, was used to describe the kinetic reaction for the diffusion through the solid pores (Goto et al., 1996; Yang et al., 2014):

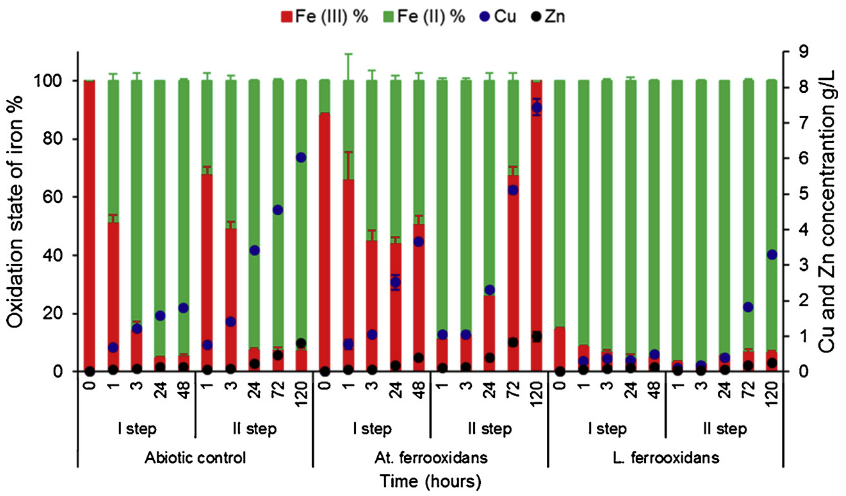

Fig. 5. The leaching kinetics of Cu and Zn recovery from PCB and oxidation of Fe^{3+} in the different treatment (abiotic control, *At. ferrooxidans* and *L. ferrooxidans*).

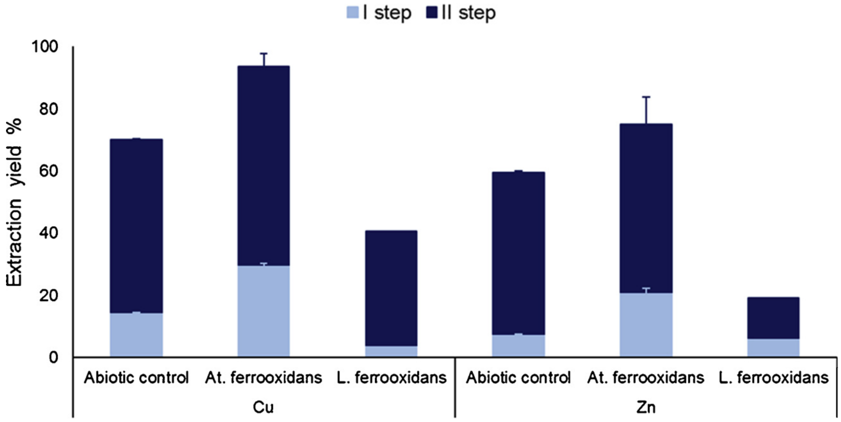

Fig. 6. Cu and Zn extraction yield from PCB treated with *At. ferrooxidans* and *L. ferrooxidans* by a two steps bioleaching process.

$$kt = 1 - \frac{2}{3}M_t - (1 - M_t)^{\frac{2}{3}} \tag{4}$$

Where t is the time, M_t represents the fraction of mobilized metal and k is the constant rate for the shrinking core model. The obtained results (Fig. 7a and b), show a reaction kinetic increase of 5 and 12.5 times for Cu and Zn leaching, respectively, in the chemical approaches with 50 g/L of Fe^{3+}, thanks to the temperature growth from 30 °C to 50 °C. The confirmation of the temperature positive effect justifies the time decrease, from 24 (at 30 °C) to 6 h (at 50 °C), required for the chemical leaching conclusion. On the other hand, Fig. 7c and d compare the behavior of bioleaching and abiotic control proving the fastest kinetic of the first one, during the two steps of the process. More in detail, the k related to the I step of the biological treatment shows values 4 and 8 times higher than the abiotic control for Cu and Zn leaching, respectively. The difference is less evident during the II step, where the kinetic reactions for Cu and Zn are 1.1 and 2 times faster than abiotic control. The different result obtained for the two steps can be explained by a double reason: 1) in the I step the PCBs were added at the end of bacteria growth phase, when microorganism count and metabolism has already reached the exponential phase. The decrease of the rate during the II step is due to the partial medium replacement, which slows down the whole kinetic for the necessity of a preliminary bacteria growth, essential to restore the same conditions of the I step; 2) the highest metal concentrations in the II steps decreased the specific growth rate and metabolism of the *At. ferrooxidans* (Cabrera et al., 2005a, 2005b; Chisholm et al., 1998; Dopson, 2003).

As concern the comparison between the chemical and bioleaching (at 30 °C, chosen as experimental condition), the estimated k values are 4 and 9%, during the I step and 5 and 25% during the second one for Cu and Zn, respectively. The reported percentages are expressed as ratio between the bioleaching and the chemical approaches.

3.4. Assessment of the carbon footprint

The results reported in Fig. 8 describe the carbon footprint of the five scenarios selected for the analysis. As concerns the innovative bioleaching (scenario 1), the treatment with *A. ferrooxidans* bacteria was selected for the best achieved efficiencies, comparable with those of the other assessed options. Overall, the scenarios 1 and 3 show comparable impacts, with a minimal advantage of the biotechnological approach (0.4 and 0.7 kg CO_2-eq./kg treated PCBs, respectively,

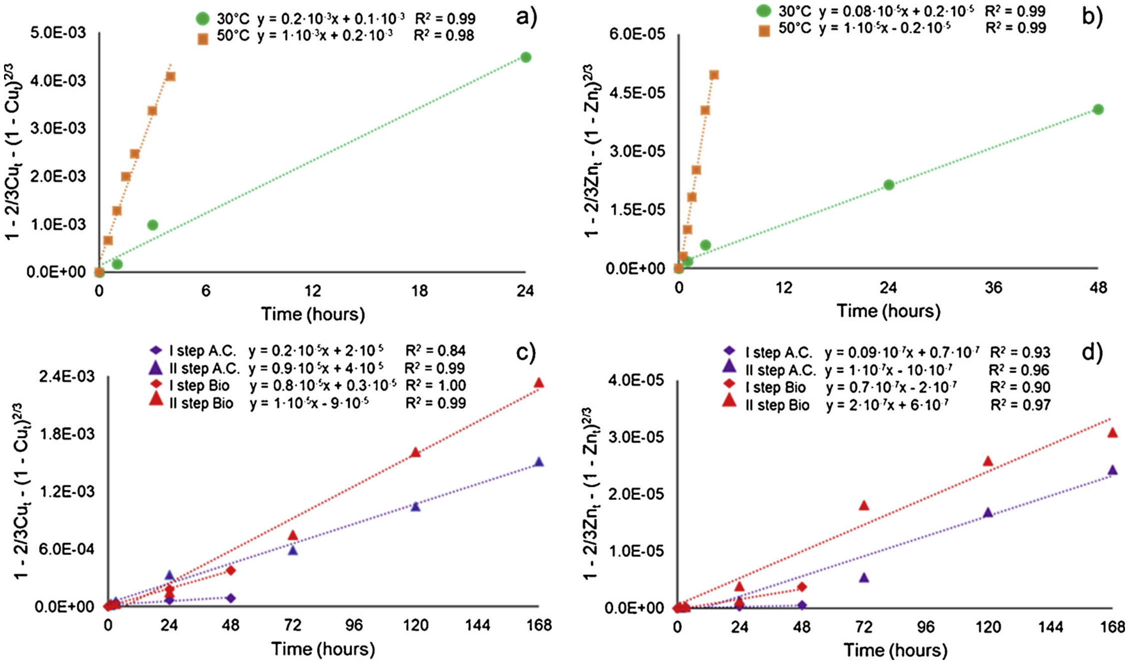

Fig. 7. Implementation of the shrinking core model for the kinetic study of the recovery of: a) Cu b) Zn by the chemical approach and c) Cu and d) Zn by the bioleaching (Bio) and abiotic control (A.C.).

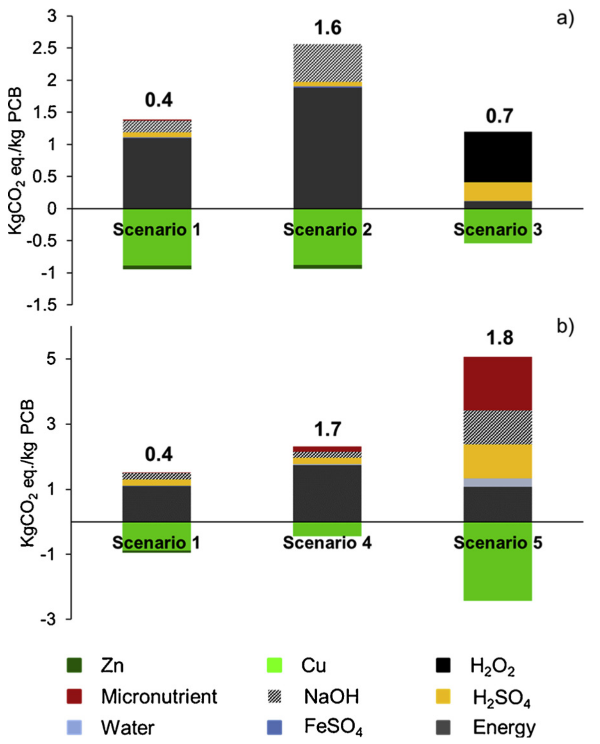

Fig. 8. Carbon footprint of the five considered approaches. (Function unit: 1 kg of shredded PCBs).

Fig. 8a). The positive effect of the innovative process design, proposed in the present paper, is highlighted in Fig. 8b, where the environmental impact decreases up to 4 times, compared to the best bioleaching approaches reported in the literature. Indeed, the increase in PCB concentration allows the reduction of both micronutrient and water demands, with the consequent decrease of the bioreactor size and the energy request. This result is confirmed by Wu et al. (2018), which reports an energy requirement of 2.6 kW h (Table 2), comparable with that of the proposed innovative process, for the treatment of the same PCB amount, with a bioreactor 10 times greater. In the five options the positive effect of the Cu and Zn recovery (quantified as avoided primary production) allows to partially balance the environmental load due to the energy and raw materials consumption. Nevertheless, this effect is less evident for the third and fourth scenarios since the hydrometallurgical and biotreatment option focuses on the only Cu with a lower efficiency than the options 1 and 2. The effect of electricity consumption is highlighted for both the chemical treatment and the biotechnological approach, due to the high required temperature (scenario 2) and the long reaction time of scenario 1 and 4.

Overall, the environmental credit associated to the target metals is not enough to balance the process impact. Nevertheless, the resulting sludge exploitation could further increase the environmental benefit. In this regard, chemical characterization identified the presence of rare earths (e.g. Sc, Y, La, Ce, Nd, Gd, Lu) and precious metals (e.g. Au, Ag, Pd) (Hubau et al., 2019; Tkaczyk et al., 2018; Yamane et al., 2011), which could further increase the environmental benefit, in the perspective of the process scale-up.

4. Conclusion

In a context of continuous development of biotechnological approaches, the present study describes a high efficiency and high sustainability treatment for the metal extraction from PCBs.

A two-step bioleaching process, using *A. ferrooxidans* bacteria, at the best selected conditions: 30 °C, solid concentration of 5% (w/v), 10 g/L of Fe^{2+} conc., for 9 days, allowed to achieve 94% and 70% of extraction yield for Cu and Zn, respectively, with a further decrease of Fe demand higher than the 60%, compared to the chemical treatment. The carbon footprint assessment (0.4 kg CO_2-eq./kg treated PCBs) proved the process advantage, with an impact comparable with the hydrometallurgical option. The main biotechnology criticality is the high electricity demand, which could be solved by the implementation of a heap leaching design, able to considerably decrease the necessary energy. This possibility, combined with the high process efficiency and an already low climate change impact, proves the advantage of the biotechnological approach for the exploitation of a WEEE which becomes a secondary source of raw materials, in agreement with the circular economy pillars.

Declaration of Competing Interest

None.

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