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Application of *Acidithiobacillus ferrooxidans* for recovering metals from electronic waste dust

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Abstract: The paper discusses metal recovery from metallic wastes by means of pure chemical and biological leaching in an acidic medium. The research tested whether it is possible to remove Cu, Al, Sn and Ag from the dust formed during processing of electronic waste by means of *Acidithiobacillus ferrooxidans*. A laboratory flow bioreactor was designed and assembled for the purpose of the experiments. The experiments confirmed the effect of the bacteria on the dissolution of Cu, Al, and Sn, as well as different efficiency levels depending on the type of the leached material. The experimental results confirmed the positive effect of the presence of *Acidithiobacillus ferrooxidans* bacteria on the recovery of Cu, Al and mainly Sn into the leaching solutions from the leached dust.

Keywords: *bacterial leaching, Acidithiobacillus ferrooxidans, electronic waste*

1. Introduction

The waste herein subjected to leaching arises from industrial processing of electronic waste. So far no utilisation has been found for this waste and it needs to be disposed generating considerable financial costs. The company, which provided a sample for the experiment, is engaged in the processing of electrical and electronic equipment. During the process the company produces about 403 Mg of "cyclone separated dust", 38 Mg of "internal exhausted dust" and 403 Mg of "external exhausted dust" per year. At the same time, increasing amounts of metals of a high market value are lost, particularly copper, silver and tin. The recovery of metals from electronic waste by either chemical or biological methods has been reported by a number of authors (Wang et al., 2009; Yang et al., 2009; Xiang et al., 2010; Liang et al., 2013; Willner and Fornalczyk, 2013; Yang et al., 2013; Xu et al., 2014; Yang at al., 2014). This study is different from other authors' experiments mainly because of the leached material and designed bioreactor. In our experiments, the dust from crushing and grinding was used, while other authors used selected circuit boards and batteries. Experiments in the 9K medium and leaching in the 9K medium under the abundance of bacteria were chosen to follow up onto the authors' experiments (Blazek, 2011) as the work empirically proved that in the presence of *Acidithiobacillus ferrooxidans* in the medium of sulfuric acid or in the medium of sulfuric acid without bacteria (pH 2), copper poorly leaches into the solution. The results of the paper reveal that copper was released in 9K medium and 9K medium with bacteria.

2. Materials and methods

Four samples of separated dust were leached, i.e. cyclone-separated dust, two fractions of dust from an external exhauster, and dust from an internal exhauster (Fig. 1). The dust from the external exhauster was separated into 2 fractions: 0-0.5 and 0.5-2 mm. The dust is formed in different sections of the electronic waste reduction process. The process flow chart is given in Fig. 2.

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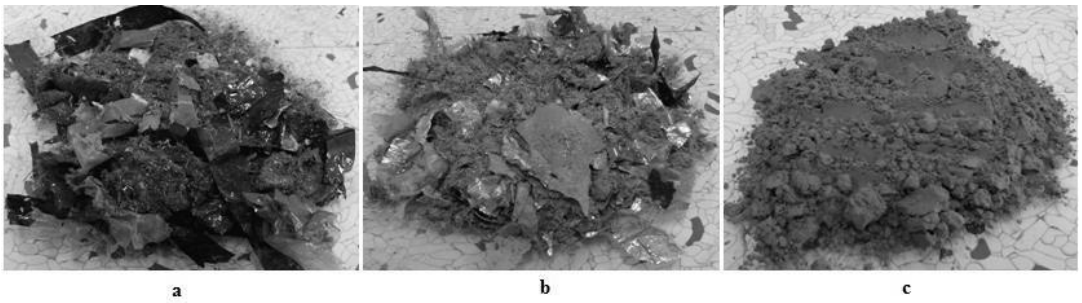

Fig. 1. Electronic dust: (a) cyclone-separated dust, (b) dust from the external exhauster, c) dust from the internal exhauster

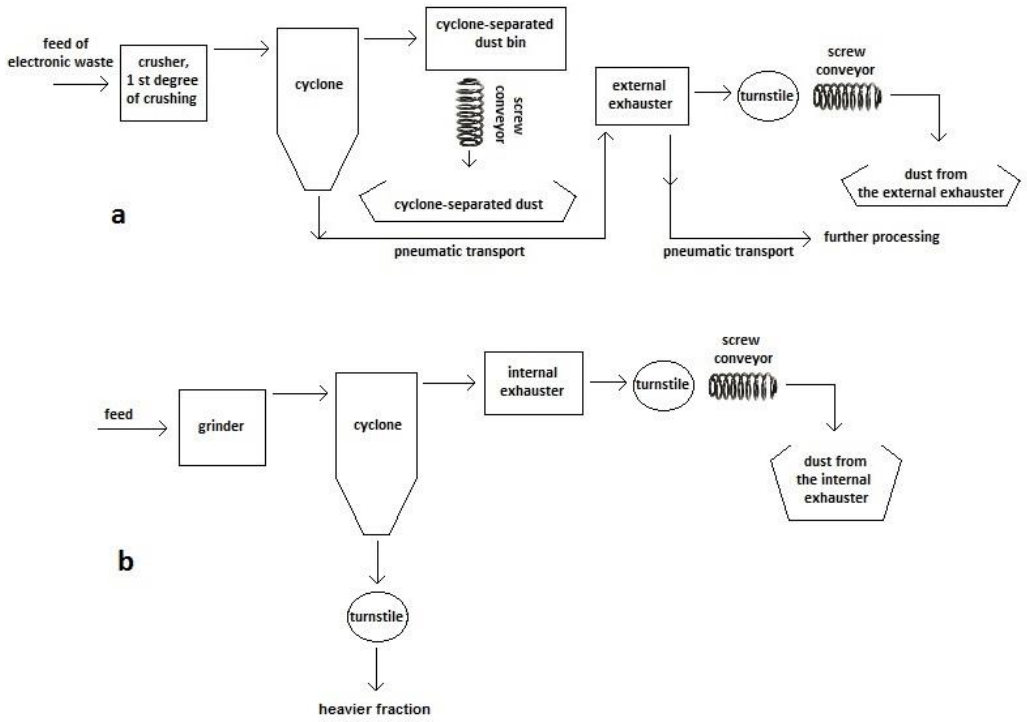

Fig. 2. Flow chart of dust formation in (a) external dust exhauster and cyclone, (b) the internal dust exhauster

2.1 Cyclone-separated dust

This waste is formed when dust is separated during the first electronic waste crushing. Dust removal from the crushed material is ensured by a pneumatic cyclone which is a downstream crusher. A sieve analysis was carried out and the particle size distribution ranged from 0.5 to 2 mm. The particles over 25 mm had a high volume but low mass. They were mostly larger pieces of foils. The particle size distribution of cyclone-separated dust was as follows: 0-0.5 mm (1.67%), 0.5-2 (58.5%), 2-7 (7.02%), 7-16 (18.73%), 16-25 (8.87%), over 25 mm (5.22%). The cyclone-separated dust had the particle size distribution below 2 mm (because of the low proportion of particles below 0.5 mm).

Visually it was possible to determine that particles having size from 2 to 25 mm had almost no metals. Because of low-metal content, this fraction was not leached. Nevertheless, the fraction -2 mm was analyzed (ICP-MS) and the results are shown in Table 1.

2.2 Dust from the external exhauster

The so-called dust from the external exhauster arises during dedusting of the pneumatic transport of electronic waste from a cyclone. Via the turnstile, which prevents the dust reflux, it continues towards a screw conveyor which doses the dust into big-bags. The dust from the external exhauster contains much finer particles than the cyclone-separated dust. The particle size distribution of the dust from

the external exhauster was: 0-0.5 mm (52.65%), 0.5-2 (27.31%), 2-7 (4.16%), 7-16 (11.6%), 16-25 (2.55%), over 25 mm (1.73%).

For the particle size from 2 to 25 mm the metal content was determined for samples prepared by manual selection. In this fraction there were mainly plastics, rubber and wood. The proportion of copper wires and aluminium foil was negligible with regard to the mass of the fraction. Leaching was not executed because of the low metal content. The experiments focused on two size fractions: 0.5-2 mm and below 0.5 mm. The particle size fractions 0-0.5 and 0.5-2 mm were subjected to an analysis (ICP-MS). The results are shown in Table 1.

Table 1. Mass proportions of the different metals in the dust from electronic waste processing

Sample	Metal content in mg/kg							
	Ag	Al	Au	Cu	Pb	Pd	Pt	Sn
Internal exhauster dust fractions	52.8	26800	23.8	38500	32400	7.11	0.23	8480
External exhauster dust fraction below 0.5 mm	73.9	20400	7.99	19400	11100	12.3	0.21	4780
External exhauster dust fraction 0.5-2 mm	142	12300	5.12	7910	6410	6.56	0.28	3520
Cyclone separated dust fraction below 0.5 mm	124	21700	3.02	30800	9080	4.68	0.09	1120

2.3 Dust from the internal exhauster

This dust is formed in the internal exhauster during grinding. The heavier fractions from the cyclone pass via the turnstile towards further processing. Lighter fractions are sucked by the exhauster via the turnstile towards the screw conveyor that doses the dust into big-bags. Fig. 2b shows the flow chart of the process. The dust from the internal exhauster was the finest and contained the highest percentage of metals (Table 1). Considering its very fine particle size distribution, the dust from the internal exhauster was leached without prior screening.

Contrary to crushing, during grinding the finest electronic parts containing Au, Pd, Pt, Pb, Cu, etc. are liberated. The dust particles are shown in Fig. 1. Their particle size is equal to 11.37 μm X-ray diffraction was used to preliminarily determine the amount of oxidised and elementary metals in the fine fractions of the dust from the internal exhauster. The ratio of the oxidised metals is higher in this sample due to the larger specific surface of the finer material. Results of the analysis are given in Table 2.

Table 2. Forms of the metals in the dust from the internal exhauster

Form of metal	Formula	Parts by weight in %
Elementary copper	Cu^0	12.5
Cuprite	Cu_2O	2.82
Elementary aluminium	Al^0	6.09
Hydrocalumite	$Ca_2Al(OH)_7{\cdot}3H_2O$	2.40
Microcline	$KAlSi_3O_8$	15.1
Bromargyrite	AgBr	1.50

2.4 Leaching experiments

The aim of the experiments was to verify the potential of bacterial leaching of waste dust from electronic waste processing to recover metals, especially Cu, Al, Ag and Sn. To determine the influence of *Acidithiobacillus ferrooxidans* on the leaching process, the experiments were also repeated without the use of bacteria. In both cases the 9K medium was used. The medium intended for the biological leaching was inoculated with 100 cm^3/dm^3 of bacterial culture 14 days prior to its use in order to simulate the conditions of industrial leaching. The bacterial culture came from the Institute of Environmental Engineering at VSB-Technical University of Ostrava. The bacterial culture was not adapted to the presence of leached material or increased concentrations of metals in solution before

leaching. The samples were leached in a flow reactor (Fig. 3). The reactor can operate with a volume of 1500-3000 cm^3 and is designed for 5-30 g of the leached material.

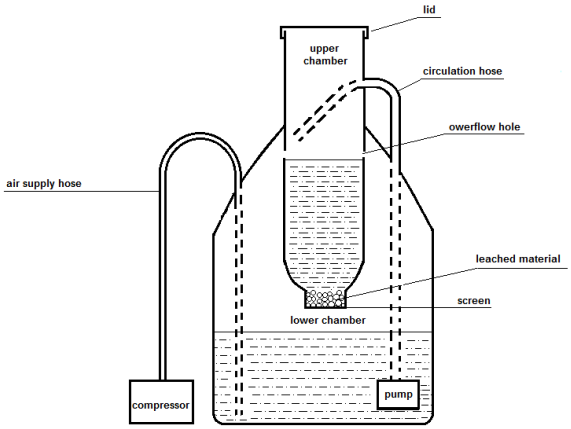

Fig. 3. Scheme of flow reactor

Overall, there were 8 reactors used because of two types of leaching methods, i.e. chemical and bacterial leaching, and 7.5 g/dm^3 of four types of dust (dust from the external exhauster – fraction below 0.5 mm, dust from the external exhauster – fraction from 0.5 to 2 mm, cyclone-separated dust – fraction below 2 mm, dust from the internal exhauster). The raw material concentration in the solution was deliberately chosen as 7.5 g/dm^3 to avoid potential toxicity for bacterial culture.

The samples were placed in nylon nets in the upper chambers of the reactors. Each reactor was filled with 2 dm^3 of leaching solution and the 9K nutrient medium. In bacterial leaching, the medium was inoculated with $4 \cdot 10^8$ of bacterial cells (*Acidithiobacillus ferrooxidans*) in 1 cm^3 of solution. The seeding method of grown colonies on solid agar M788 Thiobacillus - TA (HiMedia Laboratories, Mumbai, India) was used to determine the concentration of bacterial cells in a leaching medium. The reactors were separated from one another to prevent mutual contamination by bacteria. Immersion pumps and compressors ensured aeration. During 28-day leaching, the colour and turbidity of the leaching solutions were visually checked every 48 hours. The temperature and pH of the suspensions were measured also every 48 hours, while samples for metal analyses were taken once a week. During the leaching experiments, the pH value was not modified in any of the reactors. Metal concentrations in the solutions were determined using ICP-OES.

3. Results and discussion

3.1 Dust from the external exhauster – fraction below 0.5 mm

After 28 days of bacterial leaching in the reactors, an increase in the concentration of live cells from $4 \cdot 10^8$ (at the beginning of the leaching process) to $1.2 \cdot 10^9$ in 1 cm^3 of solution was noted. It disproves the idea that the leached material could contain substances able to inhibit the replication of *Acidithiobacillus ferrooxidans*. The experiments reveal that, except for silver, the abundance of bacteria has a decisive influence on the leaching process and that the metals are released faster from the electronic waste than during chemical leaching. For example, tin was poorly released from the dust without the presence of bacteria. The results are shown in Figs. 4-6.

Under the given conditions neither biological nor chemical leaching had a significant effect on the release of silver from the electronic waste.

3.2 Dust from the external exhauster – fraction from 0.5 mm to 2 mm

The dust from the external exhauster after processing of electronic waste of the particle size 0.5 to 2 mm was also leached in the reactors. During the biological leaching process, the concentration of live cells grew to $6.6 \cdot 10^8$ in 1 cm^3 of the leaching solution. This again confirms the fact that the leached material (external exhauster dust from processing of electronic waste) did not inhibit the replication of the bacteria in question. The results are given in Figs. 7-9.

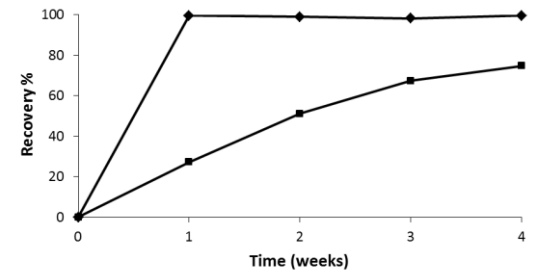

Fig. 4. Copper recovery as a function of time of (◆) biological and (■) chemical leaching

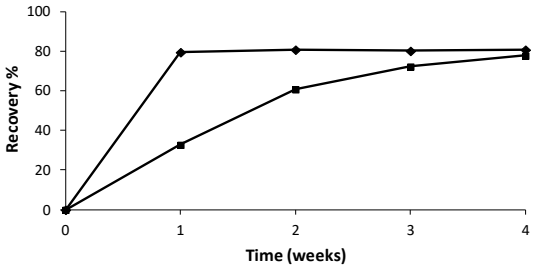

Fig. 5. Aluminium recovery as a function of time of (◆) biological and (■) chemical leaching

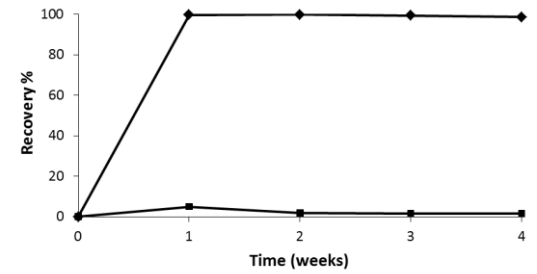

Fig. 6. Tin recovery as a function of time of (◆) biological and (■) chemical leaching

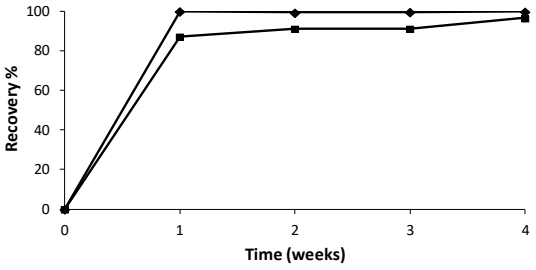

Fig. 7. Copper recovery as a function of time (◆) of biological and (■) chemical leaching

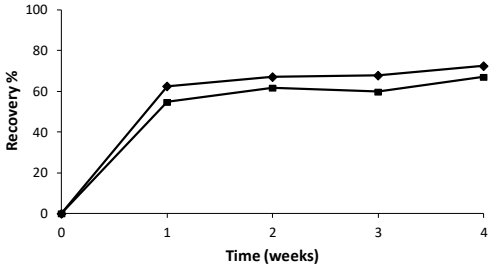

Fig. 8. Aluminium recovery as a function of time of (◆) biological and (■) chemical leaching

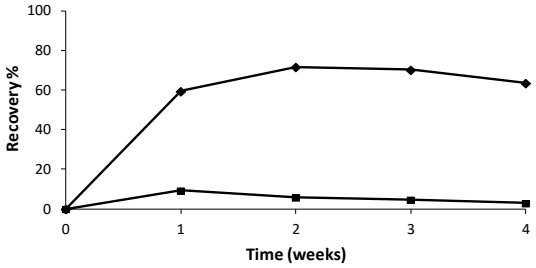

Fig. 9. Tin recovery as a function of time of (◆) biological and (■) chemical leaching

3.3 Cyclone-separated dust – fraction below 2 mm

Particles of size below 2 mm, which were separated by a cyclone during electronic waste processing, were also leached. As stated above, one reactor was used for biological leaching in the presence of *Acidithiobacillus ferrooxidans,* and another one for chemical leaching. In the case of chemical leaching, the pH value was around 2.5, at the beginning of the process, and gradually decreased to 2.3. During biological leaching, the pH value started at about 2.2 and gradually decreased to 1.9 due to the action of bacteria. The pH value, thus, influenced the metal release into the solution. Additionally, it was noted that after 28 days of leaching, the concentration of cells increased to $6.9 \cdot 10^8$ in 1 cm^3 of the leaching solution. The results are given in Figs. 10-12.

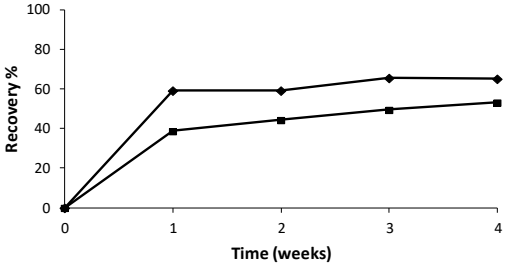

Fig. 10. Copper recovery as a function of (◆) biological or (■) chemical leaching time

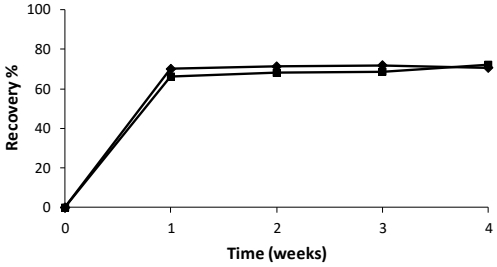

Fig. 11. Aluminium recovery as a function of (◆) biological or (■) chemical leaching time

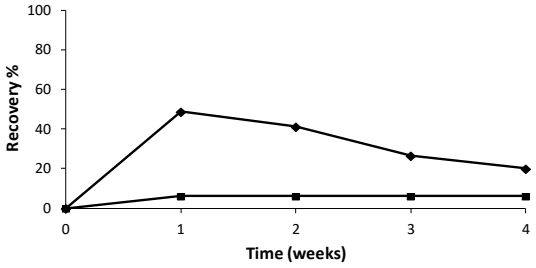

Fig. 12. Tin recovery as a function of (◆) biological or (■) chemical leaching time

The most significant differences in the metal recovery levels between the biological and chemical leaching were obtained in the case of tin dissolving from the electronic waste processing dust, especially during the dust leaching from the external exhauster. Almost 100% and only 5% of available tin was released into the solution during the first week of biological leaching and chemical leaching, respectively. During the next three weeks the values hardly changed. Significant reduction in the biological leaching of Sn from other samples was noted. Maximum of tin, 59.45%, was leached after the first week. Still, biological leaching was more effective than the chemical one. The reason for the reduced recovery of tin in some reactors could be worse access of the leaching agent to the tin particles. Plastics or any other material could have stuck onto the particles during the waste processing. Still, it was clearly demonstrated that the bacterial culture *Acidithiobacillus ferrooxidans* had a decisive effect on the tin liberation into the solution.

3.4 Dust from internal exhauster

The dust from the internal exhauster arising during processing of electronic waste was also leached in two reactors. During the chemical leaching the pH value was higher than during the biological leaching process. The pH value during the chemical leaching reached around 2.6, during the biological leaching the pH value started at around 2.3 and dropped finally to 1.9. Therefore, the pH value in the biological leaching facilitated the release of metals into the solution. The results are given in Figs. 13-15.

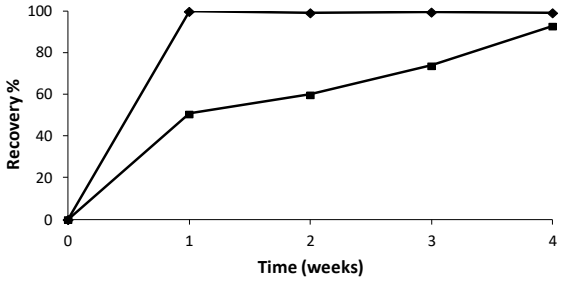

Fig. 13. Copper recovery as a function of (◆) biological or (■) chemical leaching time

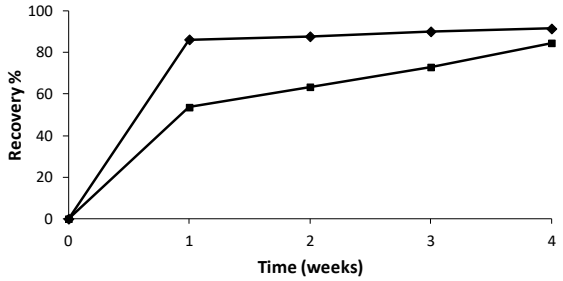

Fig. 14. Aluminium recovery as a function of (◆) biological or (■) chemical leaching time

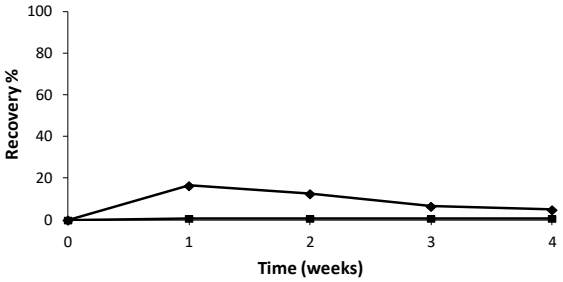

Fig. 15. Tin recovery as a function of (◆) biological or (■) chemical leaching time

The copper recovery into the solution during biological leaching was comparable to the experimental recovery values reported by other authors (90 to 100% copper recovery) (Wang et al., 2009; Yang et al., 2009, 2013; Xiang et al., 2010; Liang et al., 2013; Willner and Fornalczyk, 2013; Xu et al., 2014). The leaching of the cyclone-separated dust, neither biological (65.17%) nor chemical (53.18%), was very efficient, which may be caused by certain copper particles being trapped in plastics. The most efficient release of copper occurred in the case of pure chemical leaching of the dust from the external exhauster (particles from 0.5 to 2 mm), which can be explained by a better accessibility of the leaching medium to the copper particles in opposition to materials made up from finer particles (below 0.5 mm).

Aluminium was released into the solution the fastest during the biological leaching of the finest dust (particles below 0.5 mm) with the recovery values of 79.56 and 86.26% in the first week. During the following three weeks the values remained almost unchanged. It is also worth mentioning that during the pure chemical leaching of the identical material, analogous Al recovery levels were obtained almost after four-week leaching. On the contrary, during biological leaching of the dust of 0.5 to 2 mm particle size there was a slight decrease in the recovery of aluminium and an increase during the pure chemical leaching at the very beginning of the leaching process. This is likely to be caused by the varying permeability of the leached material (particle size 0.5 to 2 mm). Finer material (particles below 0.5 mm) had a larger specific surface but was less permeable than the coarser material. Also Xu et al. (2014) and Yang et al. (2014) leached the size-reduced electronic waste in order to recover aluminium applying *Acidithiobacillus ferrooxidans* and recovered 75 and 85.59% of the available aluminium from the electronic waste. These results are, thus, comparable to the results obtained during biological leaching of the dust (below 0.5 mm) from electronic waste processing.

As for silver, its release into the dust suspensions was observed but the bacteria *Acidithiobacillus ferrooxidans* had no influence on the leaching process. The concentrations of Ag ions in the solution were below the threshold of detectability. On the other hand, Jadhav and Hocheng (2013) succeeded in 98% release of the available silver from leaching powder from button cells (silver oxide) after three-hour leaching with the identical bacterial culture. The differences in the leaching results obtained within the work are likely to be attributed to the differences in the leached material.

4. Summary and conclusions

Three types of dust formed during processing of electronic waste were leached for 28 days. The experimental results confirmed the positive effect of the presence of *Acidithiobacillus ferrooxidans* bacteria on the recovery of Cu, Al and mainly Sn into the leaching solutions from the leached dust. Under the action of bacteria, the concerned metals were oxidized and released into the solutions much faster than during the chemical leaching in the 9K nutrient medium. It may be assumed that the bacteria influenced the leaching process both directly as well as indirectly. However, silver was not released from the dust formed during electronic waste processing, neither using the biological nor chemical leaching.

Almost 100% (except the cyclone-separated dust) of available copper in the dust was recovered during the first week of biological leaching. The recovery levels of chemical leaching did not reach the levels obtained by the biological method even during additional three weeks.

The recovery levels of Al either during the biological or pure chemical leaching of the suspension of dust from electronic waste processing were almost identical in values. However, they varied in the recovery rate.

The most significant differences in the recovery levels between the biological and chemical leaching were obtained in the case of tin dissolving from the electronic waste processing dust. The concentrations of Ag ions in the solution were below the threshold of detectability.

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