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Oxidative pressurized acid leaching of waste printed circuit boards

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Abstract: The amount of waste printed circuit board (WPCB) increases continuously. There is an economic and environmental need to recover valuable metals from WPCBs. In this study, the applicability of oxidative pressurized acid leaching of WPCBs rich in copper and tin was investigated. The effect of leaching parameters such as temperature, partial oxygen pressure and chloride concentration on metal dissolution from WPCBs was studied in sulfuric acid media. It was shown that non-metallic elements present in WPCBs initiated gas formation, namely CO₂ and CO, during oxidized pressure acid leach. Decomposition of plastic components already started at 90 °C with 1.5 MPa oxygen overpressure in the presence of 1 g/dm³ chlorides. Gas formation was shown to have a negative impact on the process since copper extraction was reduced to 27% compared to 80-90% extraction achieved from anode slimes used as a reference material. It was suggested that gas formation related to plastic components could have a severe impact on metal yields. The highest dissolution of tin equal to 36% was achieved at room temperature. It was found that higher temperature promoted tin hydrolysis.

Keywords: batch leaching, sulfuric acid, copper, tin, gas formation

Introduction

Electronic industry is one of the fastest-growing resulting in increasing amount of waste electrical and electronic equipment (WEEE) (Cui and Zhang, 2008; Wang and Xu, 2014). Due to (i) globally increasing amount of WEEE, (ii) significant valuable and even rare metal contents in WEEE, and (iii) pollution caused by artisanal recycling, there is increasing interest towards recycling of electrical and electronic components (Chen et al., 2015). In order to promote sustainable development, WEEE directive 2012/19 (EU Directive, 2012) in Europe requires reduction, reuse and

recycling of electrical components within the EU region and encourages development of new recovery, recycling and treatment technologies.

According to Haig et al. (2012), the highest value in personal computers (PCs) is contained in printed circuit boards (PCBs). It is important to be able to recycle and recover metallic fractions (MFs) from PCBs. In addition to the metallic fraction, with Cu, Sn, Au and Ag values, PCBs consists of a large non-metallic fraction (NMF). NMFs are complex. WEEE plastics can contain, for instance polymers, reinforcements (glass fibers, carbon fibers, natural fibers), fillers (talc, clay, carbon black), flame retardants, antioxidants, stabilizers and compatibilizers, which may complicate recycling and reuse of metallic components (Wang and Xu, 2014).

In order to recycle the valuable metals embedded in electrical and electronic equipment, various hydrometallurgical methods have been investigated, such as leaching with sulfate, chloride, aqua regia, iodide, ammonia, cyanide, thiourea, thiosulfate, ionic liquids, supercritical carbon dioxide and bioleaching (Sheng and Etsell, 2007; Cui and Zhang, 2008; Yazici and Deveci, 2014, 2015, Calgaro et al., 2015; Chen et al., 2015; Lambert et al., 2015; Sun et al., 2015; Xiu et al., 2015;). However, to the best of the authors knowledge, there is only limited amount of research (Jha et al., 2011) related to pressurized acid leaching of metals from waste electronic materials (PCBs) and no risks related to gas formation have been reported.

Pressurized oxidative acid leaching is industrially used in metallurgical industry for copper removal and recovery from anode slimes (Järvinen, 2000). This study focuses on investigating the applicability of oxidative pressurized acid leaching for a secondary raw material, i.e. copper-tin rich waste printed circuit boards (WPCB). Metal yields, leaching kinetics and gas formation phenomena during autoclave leaching are investigated.

Experimental

Materials and methods

WPCB and anode slime raw materials for leaching

Experiments were conducted with three raw materials: (i) WPCB sample, (ii) copper anode slime sample (CAS) and (iii) plastic sample (PS) containing only plastic components collected from WPCBs. The target was to investigate WPCB material leaching behaviour during pressurized acid leaching. The anode slime sample was used as a plastic-free reference material, whereas the plastic sample was used as a plastic-rich metal free reference material. WPCBs were disassembled and components were cut into small pieces. PS consisted only of plastic components present in WEEE. Pieces were then further shredded using a SK600 hammer crusher to under 8 mm size. The 8 mm samples were further milled all below <1 mm.

PCBs are known to contain remarkable amount of metallic, plastic and ceramic fractions with a big variety of different elements. PCB substrates typically consist of

glass fiber reinforced with epoxy resin or paper reinforced with phenolic resin, both with brominated flame retardants (Bizzo et al., 2014). Table 1 presents the metal content in the WPCB material used in this study. Gold and iron concentrations are relatively low, while Zn is relatively high compared to reported minimum and maximum values by Yamane et al. (2011) and Bizzo et al. (2014). The amount of other components such as Sb, Cr, Na, Ca, Pt, Cd, K, In, Mn, Se, As, Mg, Pd, Co and Ti is rarely reported. Copper anode slimes used in this study were collected from the Boliden Harjavalta copper electrorefining process, in the anode slime filter press. The slime was divided and dried at 60 °C for 24 hours. The solid samples were digested in aqua regia and analyzed with ICP at Labtium (Espoo, Finland). The main components of anode slime are presented in Table 1 and compared with typical metal content ranges in WPCBs reported by Yamane et al. (2011) and Bizzo et al. (2014).

Table 1. Metal contents determined in WPCBs and Boliden Harjavalta anode slime (CAS)

	Cu (%)	Sn (%)	Au (%)	Ag (%)	Pb (%)	Fe (%)	Al (%)	Zn (%)	Ni (%)	Sb (%)	Bi (%)	Se (%)	As (%)
WPCB	19.3	3.2	0.01	0.07	1.8	0.3	2.5	2.6	0.23	0.21	n.a.	n.a.	n.a.
CAS	18.8	n.a.	n.a.	15.9	10.6	n.a.	n.a.	n.a.	1.5	2.2	3.6	7.3	6.3
Bizzo et al. (2014)	12.5–	1.0–	0.008–	0.008–	1.0–	0.08–	1.7–	0.04–	0.002–	n.a.	n.a.	n.a.	n.a.
Yamane et al. (2011)	28.7	5.6	0.1	0.5	3.9	11.5	7.1	2.7	2	n.a.	n.a.	n.a.	n.a.

Chemicals and utilities

The following chemicals were used: concentrated technical grade sulfuric acid (96% H₂SO₄) and technical grade NaCl. In addition, pressurized oxygen was used as an oxidizing agent. Sulfuric acid was diluted to the concentration of 100 g/dm³ used in all WPCB and PS leaching tests. CAS was leached with 144 g/dm³ sulfuric acid.

Leaching tests

The parameters used in the leaching tests are listed in Table 2. The leaching time in all WPCB leaching tests was 2 hours with a solid-to-liquid ratio of 1/40. WPCB leaching was carried out in atmospheric (25 and 85 °C) and pressurized (90–110 °C) environments. With anode slime reference tests (11 and 12) the same pressure leaching procedure was used at 100 and 160 °C. PS leaching was carried out in pressurized environment at 150 °C.

Oxidative pressure acid leaching tests were conducted in a Büchi Ecoclave 075 (Ti gr. 2) with a volume of 1.1 dm³ and with solution volume of 0.8 dm³. The reactor was fitted with a cyclone 075 magnetic agitation unit with a six-blade pitched blade propeller (400 rpm), oxygen inlet, manometer, temperature detector, bursting disk and nitrogen inlet with a sample system. A leak test was conducted for every experiment with nitrogen gas at 1.0 MPa overpressure; 0.01 MPa nitrogen overpressure was left in

the chamber to ensure an oxygen free atmosphere for the autoclave heating period. After warming, the chamber was left for 15 minutes to achieve a steady temperature and oxygen pressure was only applied after that. The starting time ($t = 0$ s) for the experiment was defined as the moment when oxygen pressure was raised to the set level. After each leaching experiment, the pressure was decreased, autoclave cooled down and the leach residue filtered, washed and dried.

Table 2. Test conditions for pressurized WPCB leaching (tests 1–10), pressurized anode slime leaching (tests 11, 12), atmospheric WPCB leaching (tests 13–16) and pressurized PS leaching (tests 17–20)

Test number	T (°C)	Partial oxygen pressure (MPa) or O_2 feed (dm^3/h)	[Cl] (g/dm^3)	[H_2SO_4] (g/dm^3)	Additional
Pressurized acid leaching of WPCB					
1	90	0.5	0	100	–
2	110	0.5	0	100	–
3	90	1.5	0	100	–
4	110	1.5	0	100	–
5	90	0.5	1	100	–
6	110	0.5	1	100	–
7	90	1.5	1	100	–
8	110	1.5	1	100	–
9	100	1.0	0.5	100	–
10	100	1.0	0.5	100	–
Pressurized acid leaching with plastic-free CAS					
11	100	0.8	0	144	–
12	160	0.8	0	144	–
Atmospheric leaching of WPCB					
13	25	125 dm^3/h	0	100	–
14	25	125 dm^3/h	1	100	–
15	85	125 dm^3/h	0	100	–
16	85	125 dm^3/h	1	100	–
Pressurized acid leaching with metal-free PS					
17	150	0.5	0	100	–
18	150	1.0	0	100	–
19	150	0.5	0	100	+Sn 1.2 g
20	150	0.5	1	100	+Sn 1.2g +Cu 3.8 g

Atmospheric oxidative leaching was conducted in 0.8 dm^3 reactors with 0.4 dm^3 of solution, embedded in a water bath (Kavalier EL-20D). The reactor was equipped with an agitation unit with a two-blade pitched blade propeller (400 rpm), oxygen inlet (125

dm³/h) and temperature meter. The WPCB material was added after solution temperature increased to the set value, that time being defined as $t = 0$. After leaching the residue was filtered, washed and dried.

Solution, solid and gas analysis

Solution sampling (10 cm³) was carried out at 0, 5, 10, 20, 30, 60 and 120 min in all experiments. Samples were taken through the dip tube used for gas feed and pressurizing. First, the gas feed valve was closed, then the inlet valve to sample vessel was opened and the pressure in the reactor pushed sample to the vessel. Then, the inlet valve was closed and the sample was taken from the vessel through its outlet line. Finally, the gas feed line was flushed with oxygen. The samples were filtered. The samples were analyzed by inductively coupled plasma (ICP-OES), Perkin Elmer 7100 DV after autoclave leaching and by the atomic absorption spectrometer (AAS) Varian AA20+ after atmospheric leaching (tests 13–16). The WPCB sample and leach residues after tests 1, 3 and 8 were subjected to X-ray (XRD) qualitative diffraction analysis on an X-ray diffractometer PANalytical X'Pert PRO MRD using CoK α radiation. The gas composition was analyzed after PS leaching tests (17–20) by the GASTEC gas sampling analyzer GV-100S.

Results and discussion

Gas formation initiated by WPCBs in oxidative pressurized acid leaching

Gas formation was observed to be initiated after WPCB leaching experiments (1–10) at temperatures 90–110 °C (Fig. 1). When the oxygen pressure was released and chamber temperature decreased (at $t > 150$ min), the total pressure in the autoclave increased due to gas formation after leaching (the strongest for $p(\text{O}_2) = 1.5$ MPa in the presence of chlorides). Strong gas formation was also observed after leaching at $p(\text{O}_2) = 10$. High temperature (110 °C) also seemed to favor gas formation more than low temperature (90 °C), while low oxygen partial pressure and the absence of chlorides seemed to decrease gas formation.

In order to establish the cause of gas formation, the metallic rich plastic free anode slime material (CAS, tests 11–12) and metallic free samples (PS, tests 17–20) were subjected to oxidative pressure acid leach under different leaching conditions (Fig. 2). No gas formation was observed, when the plastic free CAS material was leached, even though more aggressive conditions were used to stimulate gas formation ($T = 100$ and 160 °C and $[\text{H}_2\text{SO}_4] = 144$ g/dm³). With plastic samples, more intensive gas formation occurred at higher partial oxygen pressure. Figure 1 shows that the determining parameter for gas formation was the partial oxygen pressure rather than temperature, since in test 7 ($p(\text{O}_2) = 1.5$ MPa, $T = 90$ °C with 1 g/dm³ chloride addition) increase in the oxygen partial pressure initiated more intensive gas formation compared to test 18 ($p(\text{O}_2) = 1.0$ MPa, $T = 150$ °C) regardless of remarkably lower temperature. Also the

presence of chlorides was shown to support gas formation. Addition of metallic components (Sn or Sn and Cu) to the plastic sample in tests 19 and 20, respectively, did not enhance gas formation. This suggests that the non-metallic components present in WPCBs can initiate gas formation already at low temperature during WPCB oxidative pressure acid leaching at high oxygen partial pressure in the presence of chlorides.

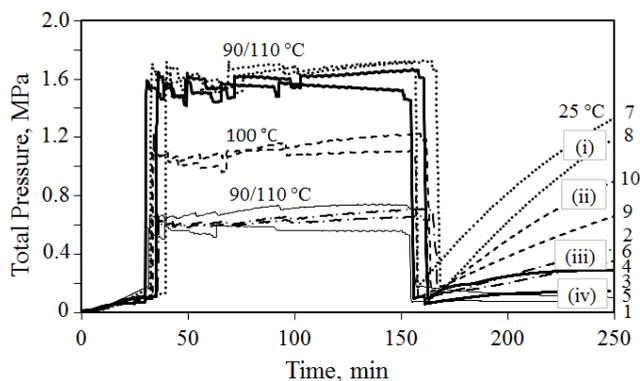


Fig. 1. Autoclave pressure profile during leaching tests 1-10 (numbers on the right). Gas formation classified according to leaching parameters: (i) strong gas formation after leaching with $p(\text{O}_2) = 1.5$ MPa in the presence of chlorides, (ii) relatively strong gas formation after leaching at $p(\text{O}_2) = 1.0$ MPa, (iii) moderate gas formation at 110°C and (iv) low gas formation at 90°C

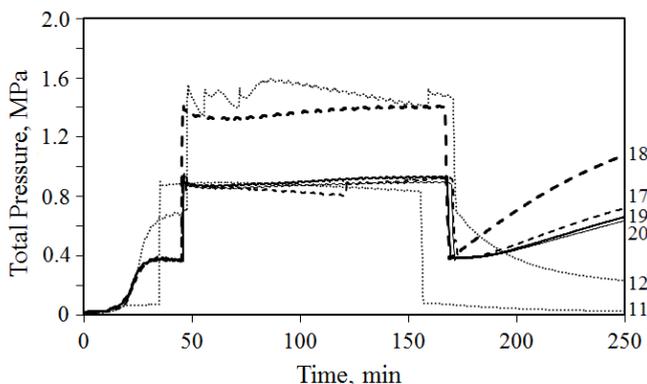


Fig. 2. Autoclave pressure profile during leaching tests 11-12 (numbers on the right) with anode slime (CAS) and with plastic sample (PS) tests 17-20 (numbers on the right)

Table 3 shows that the main gas initiated by the non-metallic fraction of WPCBs was CO_2 , while traces of CO were also present, but no SO_2 was found there. This indicates that carbonaceous plastic components present in the WPCBs started to decompose to CO and CO_2 during pressure leaching, with high oxygen pressure

enhancing carbon oxidation. This decomposition was shown to occur already at $T = 90\text{ }^{\circ}\text{C}$ (test 7) with sufficient oxygen pressure ($p(\text{O}_2) = 1.5\text{ MPa}$, $[\text{Cl}^-] = 1\text{ g/dm}^3$).

Table 3. Gas analyzed from autoclave chamber after PS leaching tests (17-20)

Test number	CO ₂ (ppm)	CO (ppm)
17	1600	12
18	900	2
19	100	15
20	1300	30

Copper yield

The copper leaching kinetics was shown to be faster during oxidative pressurized acid leaching (Fig. 3) than during atmospheric one (Table 4). Copper dissolution during atmospheric leaching at room temperature was minimal (<1%) and at 85 °C in the absence of chlorides equally low. However, the presence of chlorides supported copper leaching at 85 °C (yield 6.9%) in atmospheric leaching. The maximum Cu yield was achieved by pressurized acid leaching at 110 °C, at $p(\text{O}_2)$ equal to 1.0 MPa and 1 g/dm³ chloride addition (test 8, yield 27%). In test 8 the copper yield was shown to increase almost linearly with time indicating that the maximum yield was not reached within two hours of the leaching experiment. The major parameter affecting copper leaching in the studied conditions (tests 1–10) was partial oxygen pressure. The copper yield increased of ca. 15%-units with 1.0 MPa increase in the oxygen partial pressure under each studied conditions (Fig. 4). In addition, increase in the temperature and chloride concentration had a positive impact.

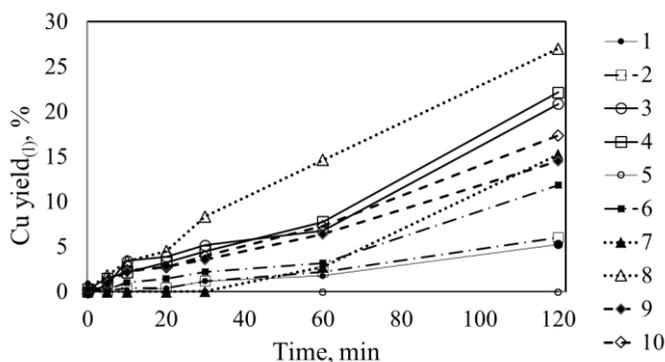


Fig. 3. Copper yield as a function of time during pressurized acid WPCB leaching (tests 1–10, numbers on the right) at 90, 100 and 110 °C, and $p(\text{O}_2) = 0.5, 1.0$ and 1.5 MPa with and without chloride addition (conditions given in Table 2)

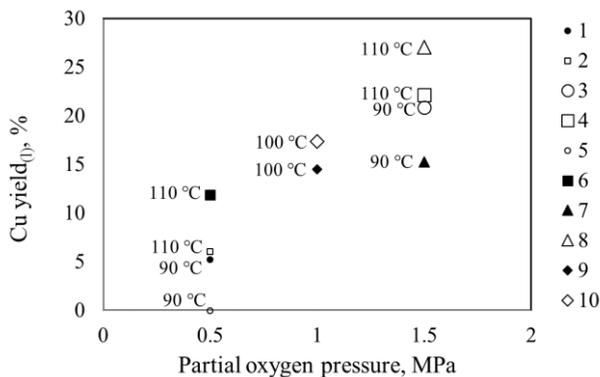


Fig. 4. Copper yield as a function of partial oxygen pressure during oxidative pressurized acid WPCB leaching (tests 1-10, numbers on the right) at 90, 100 and 110 °C, and at $p(\text{O}_2) = 0.5, 1.0$ and 1.5 MPa with and without chloride addition (conditions given in Table 2)

Table 4. Metal yield to solution after WPCB and anode slime oxidative pressure acid leaching and WPCB oxidative atmospheric acid leaching, based on solution analysis. Experimental conditions listed in Table 2

Experiment	Conditions: $T, p(\text{O}_2), [\text{Cl}], [\text{H}_2\text{SO}_4]$	Cu yield, %	Sn yield, %
1	90 °C, 0.5 MPa, 0 g/dm ³ , 100 g/dm ³	5.3	6.0
2	110 °C, 0.5 MPa, 0 g/dm ³ , 100 g/dm ³	6.1	1.3
3	90 °C, 1.5 MPa, 0 g/dm ³ , 100 g/dm ³	20.9	2.5
4	110 °C, 1.5 MPa, 0 g/dm ³ , 100 g/dm ³	22.2	1.2
5	90 °C, 0.5 MPa, 1 g/dm ³ , 100 g/dm ³	0.0	6.8
6	110 °C, 0.5 MPa, 1 g/dm ³ , 100 g/dm ³	11.9	2.6
7	90 °C, 1.5 MPa, 1 g/dm ³ , 100 g/dm ³	15.9	6.1
8	110 °C, 1.5 MPa, 1 g/dm ³ , 100 g/dm ³	27.0	2.3
9	100 °C, 1.0 MPa, 0.5 g/dm ³ , 100 g/dm ³	14.5	2.6
10	100 °C, 1.0 MPa, 0.5 g/dm ³ , 100 g/dm ³	17.4	2.5
11	100 °C, 0.8 MPa, 0 g/dm ³ , 144 g/dm ³	91.6	-
12	160 °C, 0.8 MPa, 0 g/dm ³ , 144 g/dm ³	80.1	-
13	25 °C, atm (125 dm ³ /h), 0 g/dm ³ , 100 g/dm ³	0.9	35.8
14	25 °C, atm (125 dm ³ /h), 1 g/dm ³ , 100 g/dm ³	0.9	29.2
15	85 °C, atm (125 dm ³ /h), 0 g/dm ³ , 100 g/dm ³	0.5	8.6
16	85 °C, atm (125 dm ³ /h), 1 g/dm ³ , 100 g/dm ³	6.9	4.8

The maximum copper yield (27%) in oxidative pressurized acid leach was not as good as the copper yield from anode slimes (tests 11 and 12: 91.6 and 80.1%, respectively) (Table 4). It was shown by Yazici and Deveci (2014) that high Cu yields (>90%) can be reached already during two hours of atmospheric ferric sulfate leaching

of WPCBs with air or oxygen as oxidants ($T = 80\text{ }^{\circ}\text{C}$ and with 5% w/v solids). During pressure oxidized leaching, higher pressures ($p(\text{O}_2) = 0.5\text{--}1.5\text{ MPa}$), temperature of $90\text{--}110\text{ }^{\circ}\text{C}$ and addition of chlorides (1 g/dm^3) did not enhance copper yield from WPCBs above 27% (test 8, at $110\text{ }^{\circ}\text{C}$ and at 1.5 MPa oxygen overpressure). This suggests that the characteristic CO_2/CO gas formation during WPCB leaching does have a severe impact on the copper yield during pressurized acid leaching. The final Cu and Sn extraction (%) to solution after each leaching experiment (1–16) is presented in Table 4.

Tin yield

The kinetics of autoclave and atmospheric tin leaching are presented in Figs. 5 and 6. It can be seen from Fig. 5 that during all pressurized leaching tests, the rate of tin dissolution increased sharply during the first 5–60 minutes but decreased after that time. The ultimate tin yield was lower compared to oxidative atmospheric WPCB leaching at room temperature. Similar leaching behavior was evident during atmospheric leaching at $85\text{ }^{\circ}\text{C}$ (Fig. 6). In the early minutes of leaching at the atmospheric pressure, higher temperature ($85\text{ }^{\circ}\text{C}$) gave faster tin dissolution kinetics. However, the rate of tin dissolution slowed down after 30 minutes of leaching. The extractions of tin during two hours of oxidative atmospheric leaching at room temperature were 35.8 and 29.2% in the absence and presence of chlorides, respectively. The dissolution increased almost linearly as a function of time suggesting even higher yields at higher leaching times, as shown in Fig. 6.

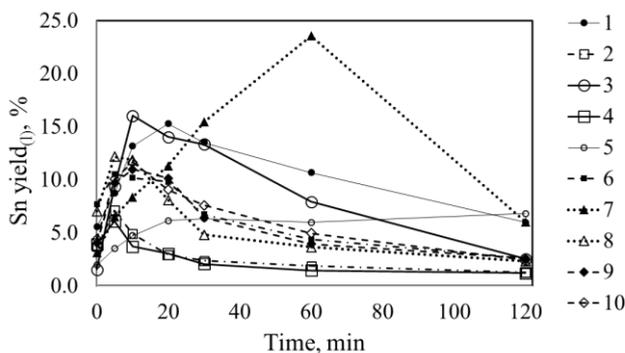


Fig. 5. Tin yield as a function of time of oxidative pressurized acid WPCB leaching (tests 1–10) at 90, 100 and $110\text{ }^{\circ}\text{C}$, $p(\text{O}_2) = 0.5, 1.0$ and 1.5 MPa with and without chloride addition (conditions described in Table 2)

Thermodynamic calculations performed with HSC 8.0 suggest that, at room temperature, tin is soluble in sulfuric acid. With increasing temperature, crystallization of hydrated tin oxide becomes favored (HSC Sim 8, 2014). Figure 7 shows the magnified XRD pattern for the WPCB sample before and after leaching in tests 1, 3

and 8. The peak formed indicated that a crystallized phase of SnO_2 (cassiterite) was formed during WPCB pressure leaching.

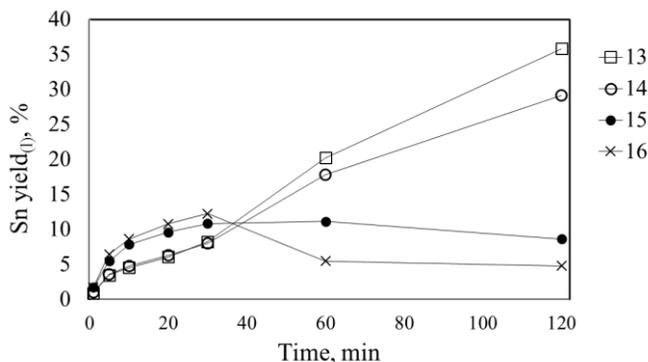


Fig. 6. Tin yield as a function of time of atmospheric WPCB leaching at 25 °C (test 13 without chloride addition and test 14 with 1 g/dm³ chloride addition) and at 85 °C (test 15 without chloride addition and test 16 with 1 g/dm³ chloride addition). Sulfuric acid concentration was 100 g/dm³

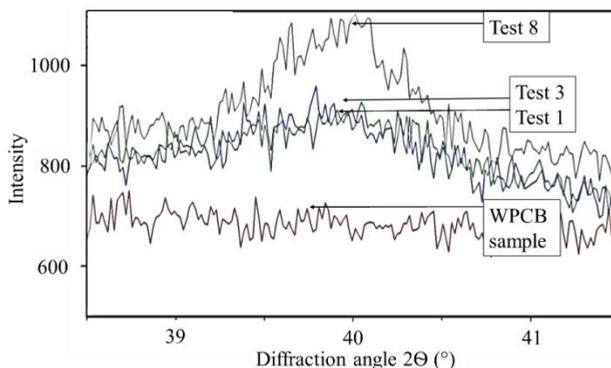


Fig. 7. Magnified XRD patterns of WPCB and leach residue after oxidative pressure acid leaching of WPCB in tests 1 (90 °C, $p(\text{O}_2) = 0.5$ MPa), 3 (90 °C, $p(\text{O}_2) = 1.5$ MPa) and 8 (110 °C, $p(\text{O}_2) = 0.5$ MPa, chloride addition 1 g/dm³). Sulfuric acid concentration was 100 g/dm³

This study shows the contradictory influence of temperature on tin dissolution. Increase in temperature (*i*) increases the tin dissolution rate but, at the same time, also (*ii*) increases the driving force for tin hydrolysis. The phenomenon observed has actually been utilized for tin purification from impure raw materials by a patented process (Forward and Vizsolyi, 1966). In the process tin is leached in sulfuric acid at room temperature (10–30 °C). The dissolved tin sulfates, SnSO_4 and $\text{Sn}(\text{SO}_4)_2$, are then further hydrolyzed in the sulfate media to SnO_2 at 90–110 °C with the oxygen pressure 0.5–1.0 MPa (or to $\text{Sn}(\text{OH})_4$ at 50–90 °C).

The major parameter affecting the final tin extraction to solution in the studied conditions was temperature, the best tin yield during two hours of the leaching being achieved at room temperature (35.8%) and the lowest tin yield (1.2%) at 110 °C.

Conclusions

The applicability of a hydrometallurgical process, oxidative pressurized acid leaching, for treating a secondary raw material, waste printed circuit boards (WPCBs) rich in copper and tin, was investigated. This process is industrially used for copper yield from copper electrorefining anode slimes. The copper content in WPCBs and in anode slimes is of the same magnitude which makes the comparison reasonable.

In the current study, it was shown that non-metallic components present in WPCB initiated CO₂ and CO gas formation. This gas formation was favored by high oxygen partial pressure, occurring already at $T = 90$ °C with $p(\text{O}_2) = 1.5$ MPa in the presence of chlorides (1 g/dm³). Gas formation was shown to have a severe impact on copper extraction to solution. The maximum copper extraction amounted to 27%, much less than copper extraction during oxidative pressurized acid leaching of anode slimes (80-90%). Gas formation in autoclave environment limited the applicability of oxidative pressurized acid leaching for copper recovery from WPCBs. In atmospheric leaching, the presence of chlorides ($T=85$ °C) slightly improved copper extraction. However, copper extraction to solution was equal to 6.9%.

The tin dissolution kinetics was fast during the first minutes of leaching. However, the tin concentration in the leaching solution decreased as a function of time in atmospheric and autoclave experiments at 85-110 °C. The effect of temperature on final tin yield was contradictory. Increase in temperature increased the reaction rate of tin dissolution but also tin hydrolysis. The best tin yield equal to 35.8% was achieved at room temperature in the absence of chlorides.

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