



Electrokinetic recovery of Cd, Cr, As, Ni, Zn and Mn from waste printed circuit boards: Effect of assisting agents

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ABSTRACT

The printed circuit boards (PCBs) contains large number of heavy metal such as Cd, Cr, As, Ni, Zn and Mn. In this study, the use of electrokinetic (EK) treatment with different assisting agents has been investigated to recover the heavy metals from waste PCBs, and the effectiveness of different assisting agents (HNO₃, HCl, citric acid) was evaluated. The PCBs were first pre-treated by supercritical water oxidation (SCWO) process, then subjected to EK process. The heavy metal speciation, migration and recovery efficiency in the presence of different assisting agents during EK process were discussed. The mass loss of Cd, Cr, As and Zn during the SCWO process was negligible, but approximately 52% of Ni and 56% of Mn were lost in such a process. Experimental results showed that different assisting agents have significant effect on the behavior and recovery efficiency of different heavy metals. HCl was highly efficient for the recovery of Cd in waste PCBs due to the low pH and the stable complexation of Cl⁻. Citric acid was highly efficient for the recovery of Cr, Zn and Mn. HNO₃ was low efficient for recovery of most heavy metals except for Ni.

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1. Introduction

The production of printed circuit boards (PCBs) is the basis of the electronic industry as it is the essential part of almost all waste electrical and electronic equipment (WEEE) [1]. In recent years, the rate of PCBs manufacture increased by 14.4% in China, and the total production value of the PCBs manufacturing industry has already reached more than \$12 billion in 2006 [2]. Each year huge amounts of waste PCBs are generated in China, among which few are processed by any techniques [3].

PCBs are a mixture of glass fiber, polymer materials and multiple kinds of metals [4]. Their physical and chemical characteristics make it difficult to recycle these matters. In normal waste PCBs, plenty of toxic materials including heavy metals, polyvinyl chloride (PVC) plastic, and brominated flame retardants (BFR) can be easily found. Especially the toxic heavy metals, such as lead (Pb), cadmium (Cd), arsenic (As), zinc (Zn), nickel (Ni) and chromium (Cr), create serious pollution upon disposal [5]. Currently, many techniques, such as solidification/landfill and incineration, not only cannot remove those toxic matters efficiently, but also have many negative impacts to the environment. Heavy metals could be leached into groundwater in landfills and toxic pollutants, including dioxins, could be emitted into the air during incineration [6]. Therefore,

it is urgent and necessary to develop the benign techniques to treat those toxic organic matters and heavy metals.

Recently, Chien et al. [7] reported that toxic organic matters in waste PCBs could be degraded effectively in supercritical water oxidation (SCWO) process. At supercritical conditions ($T > 647.3$ K, $P > 22.1$ MPa), organic matters, oxygen and water form a single and homogeneous phase, which allows oxidation to proceed rapidly [8,9]. On the other hand, electrokinetic (EK) processes have been successfully applied for metal recovery and removal from solid wastes for many years [10–12]. In our previous study [13], a process combined SCWO and EK techniques was developed for the treatment of waste PCBs. Compared to other processes for waste PCBs treatment, it is feasible and benign to decompose toxic organic matters and recover heavy metals at the same time by the integrated SCWO + EK process. In the EK process, Cu was recovered as a deposit on the cathode and Pb was recovered as concentrated solutions. However, the behavior of many other heavy metals (Cd, Cr, As, Ni, Zn and Mn) in the PCBs during such combined process was still unclear.

Many studies [14,15] have shown that when combining the EK treatment with the addition of an appropriate assisting agent to the waste during treatment, it is effective to selectively improve the leaching and recovery of heavy metals in EK system, thereby improving the recovery efficiency considerably for targeting metals. And different assisting agents have significant different effect on the recovery of targeting heavy metals. According to Pedersen [16], the following factors are important when choosing an assisting agent:

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(1) which metal to extract; (2) which chemicals may suit the metal of concern (acid, bases, others); (3) the nature of the waste (pH, chemical composition, etc.). In the case of the SCWO-treated PCBs, which usually were slightly alkaline, it seems reasonable to use acids as assisting agents because many heavy metals are soluble at low pH. In addition, complexing or chelating agents also may be the best choice to enhance the metals leaching ability during EK process.

The purpose of this study was to evaluate the enhancing effectiveness of different assisting agents on the recovery of different toxic heavy metals from PCBs scraps and investigate how different assisting agents influenced the different heavy metal speciation, migration and recovery during the EK process. The heavy metals studied were Cd, Cr, As, Ni, Zn and Mn. Three different assisting agents (HNO₃, HCl and citric acid) were employed. Inorganic acid HNO₃ is nonselective assisting agent, which dissolve heavy metals and other essential metals at the same time. Inorganic acid HCl can form complexes with many heavy metals and acidify the waste simultaneously. Organic acid citric acid, which is a chelating agent, can form stable chelates with many heavy metals and has previously been used for extraction of heavy metals from polluted soils [17,18].

2. Materials and methods

2.1. Materials

Waste PCBs used in this work were supplied by HUAXING Environmental Protection Company of Beijing. After the components (relays, capacitors, etc.) were disassembled, the PCBs were sent to comminute in a cutting mill until the fractions reached particle size smaller than 0.1 mm. All chemicals were purchased from Chemical Reagent Company of Beijing in analytical grade.

2.2. SCWO experiments

SCWO experiments were carried out according to a previous study [13]. In a typical treatment experiment, 5 g of the sample and 25 ml of distilled water were employed, while hydrogen peroxide (H₂O₂, 30 wt.%) was used as oxygen source. Experiment was conducted in a 200 ml high-pressure reactor made of 316 alloy, and SCWO treatment conditions were 713 K, 30 MPa and 1 h, since most of the toxic organic matters in PCBs could be decomposed to CO₂ and H₂O at these conditions [13]. Metal contents in initial PCBs and that in SCWO-treated PCBs were determined by inductively coupled plasma optical emission spectrometer (ICP-OES, OPTIMA2000, PerkinElmer, USA) after aqua regia digestion [19].

2.3. EK experiments with three different assisting agents

Fig. 1 shows a schematic drawing of the EK setup. The setup was made of glass, compartment II was 8 cm long and the distance between anode and cathode was 10 cm. After each SCWO treat-

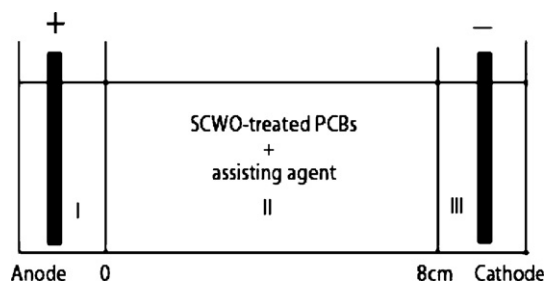


Fig. 1. Schematic drawing of the EK setup.

Table 1

Assisting agents, initial and final potential gradients in the EK experiments.

Experiment	Assisting agents	Initial voltage (V/cm)	Final voltage (V/cm)
1	1 M HCl	0.36	0.42
2	1 M HNO ₃	0.35	0.29
3	1 M Citric acid	2.09	1.87

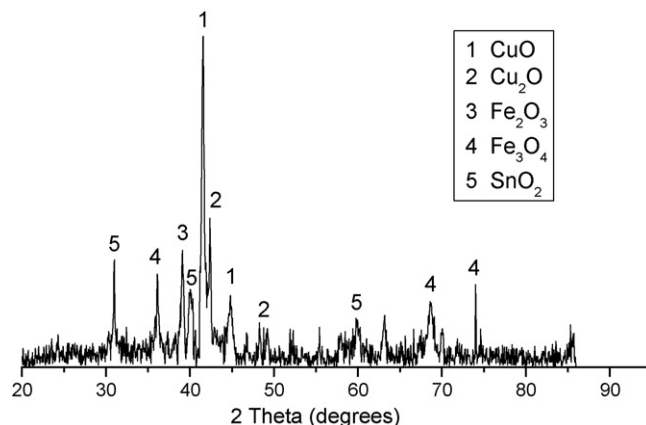


Fig. 2. XRD results of the SCWO-treated PCBs.

ment, the suspension obtained was filtrated using a vacuum filter and the residue was transferred into compartment II of EK cell after dryness, while 20 ml of assisting agent solution was added. Three different assisting agents (HCl, HNO₃, citric acid) were applied in the EK experiments (see Table 1). The SCWO-treated PCBs was kept in the suspension by constant stirring (1600 rpm) with magnetic stirrer (SH-3 from JBD, Beijing, China). The anolyte and catholyte were separated by two porous glass frits with 1 mm thickness and pore size below 50 μm. Platinum-coated electrodes from LEICI Company (Shanghai, China) were used as working electrodes. The electrodes were plated with an active area of 2 cm², and the power supply was a TPR-6405D (LONGWEI, H.K). Either anolyte or catholyte initially consisted of 20 ml assisting agent solution without adjusting all along the experiment. The constant current density applied in every experiment was 20 mA/cm², where higher Cu recovery percentage and current efficiency can be obtained [13]. Potential gradients in all experiments were recorded hourly. The duration of each experiment was 10 h.

After each EK experiment, electrodes were dipped in 5 M HNO₃ overnight. Volumes of the cleaning acids and the electrolytes were measured followed by analysis of the metal concentrations by ICP-OES. The pH of SCWO-treated PCBs before and after EK treatment was measured in 1 M KCl at liquid-to-solid ratio (L/S) 5 and after 1 h of agitation [20]. The pH was measured by a METTLER TOLEDO pH electrode. Organic matter content was measured by loss of ignition at 550 °C for 1 h [21].

The structure of PCBs solid residue after SCWO treatment was characterized by X-ray diffraction spectroscopy (XRD, Philips PW1700) at 32.5 kV and 30 mA using Co Kα radiation (λ = 1.79025 Å).

3. Results and discussion

3.1. SCWO experiments

The XRD results of the SCWO-treated PCBs residue are shown in Fig. 2. CuO, Cu₂O, SnO₂, Fe₂O₃ and Fe₃O₄ were the main crystalline phases in the SCWO-treated PCBs residue.

Table 2 shows heavy metal contents of the SCWO-treated PCBs and the conversion percentages of heavy metal during SCWO

Table 2

Metal contents of SCWO-treated PCBs and metal conversion percentages after SCWO treatment.

Metal	Content (mg/kg)	Conversion (%)
Cr	1320	100.1
Cd	1290	100.0
As	1246	102.0
Zn	14320	99.8
Ni	3749	48.1
Mn	1178	44.2

process. The metal conversion was calculated as follows:

$$\text{metal conversion (\%)} = M_2 \times 100 / M_1 \quad (1)$$

where M_1 is the metal content in initial PCBs and M_2 the metal content in SCWO-treated PCBs. The mass loss of Cd, Cr, As and Zn during the SCWO process was negligible. Similar result about Cu and Pb was observed under the same SCWO conditions in a previous study [13]. Whereas, the conversion percentages of Ni and Mn were 48.1% and 44.2%, which were particularly low compared to other heavy metals. The possible reason is that Ni and Mn existed mainly as salts, which are more easy to dissolve than metal oxides in aqueous system after SCWO reaction. Furthermore, the recovery percentages of Ni and Mn were also higher than many other heavy metals such as Cr, As and Zn in the subsequent EK process (see Section 3.2.2.).

It can also be seen in Table 3 that the SCWO-treated PCBs residue was slightly alkaline (pH 7.6), with 2.2% organic matter.

3.2. EK experiments with three different assisting agents

After the EK treatments, the final pHs of the SCWO-treated PCBs were quite different according to assisting agents, but in all acidic condition (Table 3). The most significant change in pH occurred in the treatment with HCl as assisting agent. Decreases of the mass in SCWO-treated PCBs after EK treatments were observed for all the experiments, which could be attributed to the acidification and dissolution of the SCWO-treated PCBs. The organic matter contents decreased after the EK treatments due to the removal of easily dissolved organic acids. The exception was the treatment with citric acid, where the mass increased a little, probably due to the excess citrate. Similar result was also obtained by Nystroem et al. [22], who reported that the mass increased in electro-dialytic experiment of harbour sediment with citric acid as the desorbing agent. The dissolvable organic acids in SCWO-treated PCBs were derived from the incomplete decomposition of organic compounds in initial PCBs during SCWO process [6].

3.2.1. Variation of potential gradient during the EK process with different assisting agents

Fig. 3 shows the variation of potential gradient during EK process with different assisting agents: HCl, HNO₃, citric acid. The initial potential gradient of the treatment with citric acid was much higher than that of the treatments with HCl and HNO₃, which could be attributed to the lower dissociation ability of citric acid compared to HCl and HNO₃. The potential gradient in treatment of citric acid

Table 3

Final pH, organic matter content and mass change in SCWO-treated PCBs after 10 h EK process.

Experiment	pH	Organic matter (%)	Mass change (%)
Original SCWO-treated PCBs	7.6	2.2	
1 (HCl)	3.2	n.d.	-30.34
2 (HNO ₃)	4.6	0.4	-22.26
3 (Citric acid)	5.1	2.5	-9.42

The decrease of mass in SCWO-treated PCBs is marked with (-); n.d.: non-detectable.

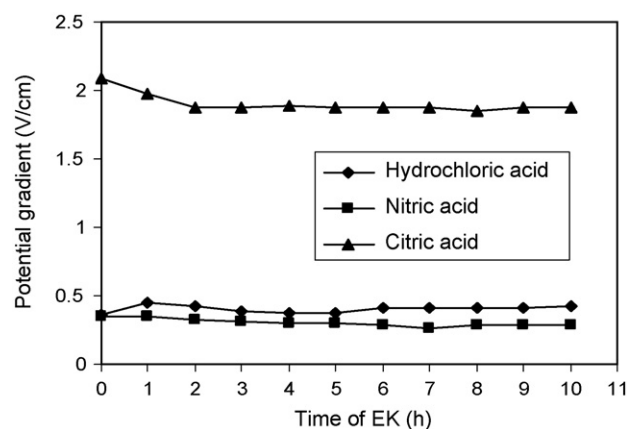
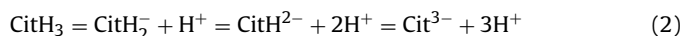


Fig. 3. Variation of potential gradient during the EK process.

decreased significantly at the first 2 h, then kept a stable level. This was also different from the treatments of HCl and HNO₃, where the variations of potential gradient were minor during the whole EK processes. Especially the potential gradient in HCl treatment increased a little at the initial stage, which was discussed elsewhere [13]. In citric acid treatment, the rapid decrease of potential gradient at the first 2 h could be attributed to that the citric acid could form chelates easily with heavy metals and large numbers of charged species were produced, then more and more H⁺ was generated due to the moving of dissociation equilibrium (2).



3.2.2. Recovery of heavy metals in the EK process with different assisting agents

Recovery of heavy metals in 10 h EK experiment is shown in Fig. 4. The recovery was defined as follows:

$$\text{Heavy metal recovery (\%)} = (A + B + C) \times 100 / D \quad (3)$$

where A , B and C are the contents of heavy metal found in compartment I, II (liquid) and III, respectively, and D the initial heavy metal content of SCWO-treated PCBs.

The highest recovery of Cd was obtained when HCl was used as the assisting agent, e.g., 70% Cd were recovered. The recovery percentages of other heavy metals in HCl treatment were lower than those in citric acid treatment. In addition, the recovery of Cd was higher than other metals in HCl treatment probably due to the combination effect of low pH (see Table 3) and complexation of Cl⁻. According to Martell [24], Cd could form more stable complexes with Cl⁻ compared to other heavy metals.

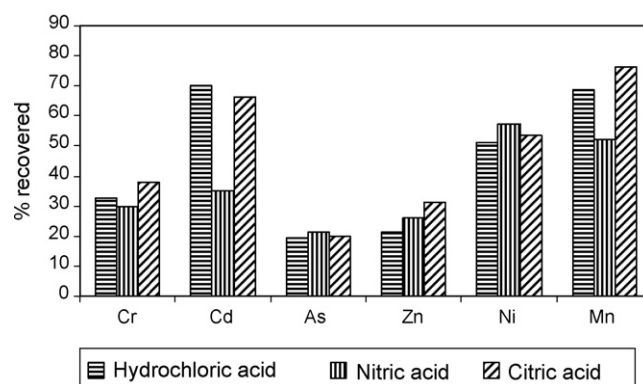


Fig. 4. Total recovery of heavy metal in 10 h EK experiments.

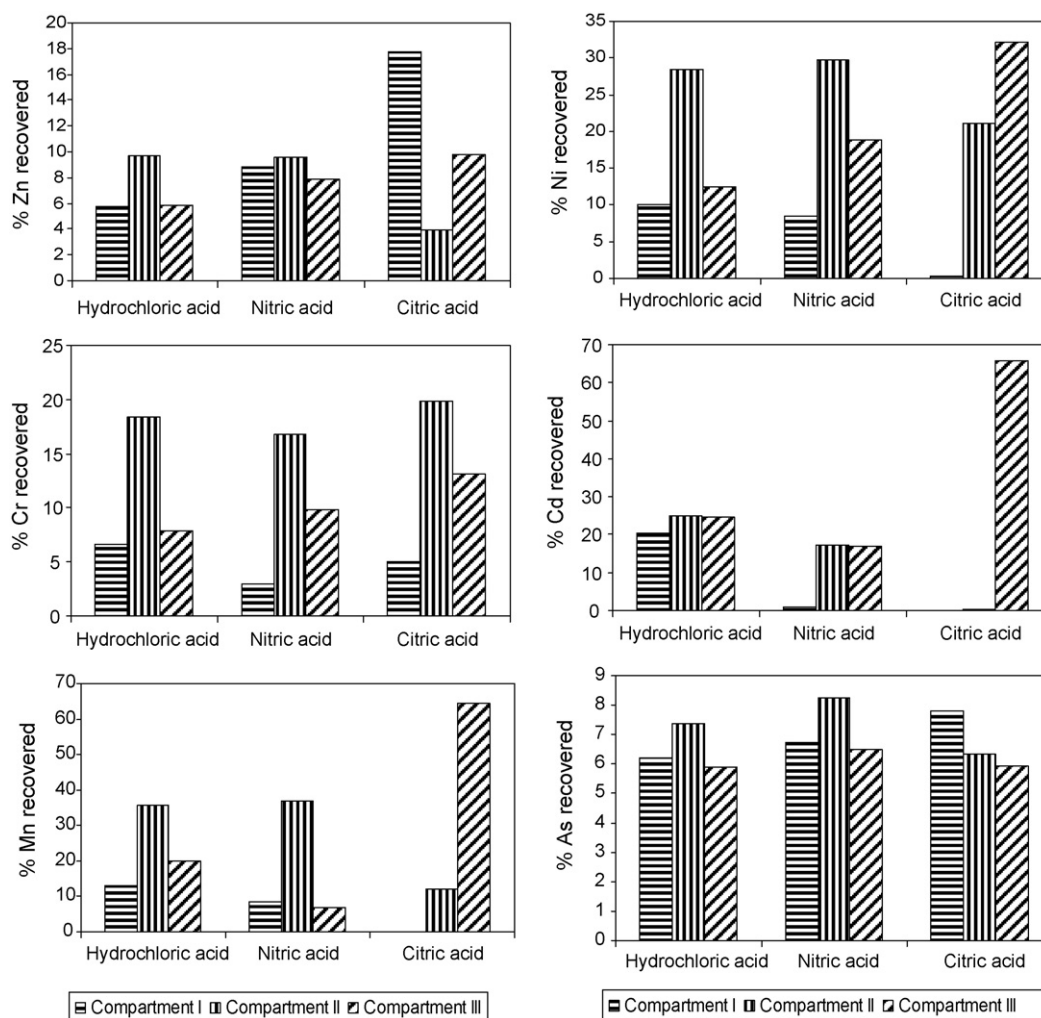


Fig. 5. Recovery of heavy metal in compartment I, II and III.

The high recovery efficiencies for Cr, Zn and Mn (38% Cr, 32% Zn and 77% Mn) could be obtained using citric acid as the assisting agent in comparison with the HCl and HNO₃ treatments. Furthermore, the recovery of Cr and Mn was more efficient compared to other heavy metals when using citric acid as the assisting agent, which could be attributed to that Cr and Mn can exist as trivalence or higher valence ion such as Cr(III) and Mn(III), and the higher valence of metal ions, the more stable are the complexes and chelates formed with citric acid [16].

It is surprising that the recovery of As and Zn in HCl and HNO₃ EK treatments was very low, almost all of the recovery percentages were less than 30% when 10 h EK treatment was used. One possible explanation is that the leaching efficiencies of As and Zn were low using HCl and HNO₃ as the assisting agents. It is possible that Zn and As could form alloy substances during SCWO process and the alloy substances were more difficult to be leached from the SCWO-treated PCBs. However, the leaching and recovery of Zn could be enhanced when using citric acid as the assisting agent, which was discussed in Section 3.2.3.

3.2.3. Migration of heavy metals during the EK process with different assisting agents

Fig. 5 shows the recovery of different heavy metals in compartment I, II (liquid) and III in the presence of different assisting agents. Migration of heavy metals during the EK process is a result

of transfer competitions between different free metal ions, metal complexes and metal chelates [23].

In HCl treatment, it can be seen from Fig. 5 that Zn, Ni and Cd were recovered either in anode region (compartment I) or cathode region (compartment III) due to the forming of stable charged complexes: MeCl⁺, MeCl₃⁻ and MeCl₄²⁻. However, Cr, Mn and As also could migrate to anode and cathode simultaneously, which could be attributed to that they can exist as either free metal ions or oxysalt ions species such as CrO_xⁿ⁻, MnO_xⁿ⁻ and AsO_xⁿ⁻. It is possible that these oxysalt species were formed during the SCWO process because metals such as Cr and Mn could be oxidized easily to their high valence format in such an oxidizing atmosphere. The recovery of Cd in anode region was about 20%, which was much higher than other metals due to the MeCl₃²⁻ and MeCl₄²⁻ (Me = Cd) are much more stable compared to other metal chloride complexes [22]. The formation of dissolved heavy metal complexes could increase the solubility and the mobility of the metal in the EK process.

In general, Zn and Ni exist as free metal ions in HNO₃ system. However, it is interesting to found that about 9% of Zn and 10% of Ni were recovered in anode region when using HNO₃ as the assisting agent, indicating formation of negatively charged metal species. The possible explanation is as follows: it can be found in Table 3 that about 1.8% organic matters in SCWO-treated PCBs, mainly the organic acids [13], could be removed during the EK process with HNO₃. These dissolved organic acids probably could

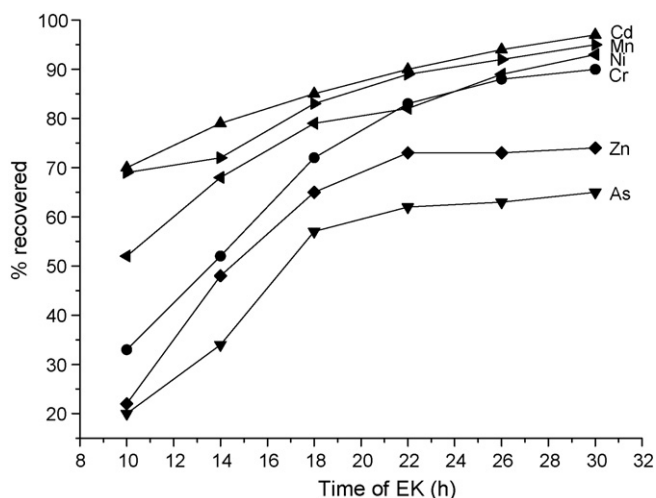
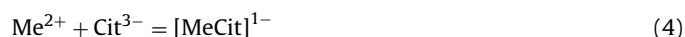


Fig. 6. Effect of EK time on total recovery of heavy metal after 10 h EK treatment with assisting agent HCl.

form negatively charged complexes or chelates with heavy metals such as Zn and Ni, and then migrate to anode region.

The chelation between citric acid and heavy metal in EK process can be expressed as follows:



When citric acid was used as the assisting agent, negatively monovalent chelate species could be formed, causing the increase of metals solubility. However, experimental results showed that the recovery of most heavy metals in anode region was negligible in citric acid treatment compared to that in HCl and HNO₃ treatments. The only exception was Zn. This could be attributed to the complexations of citric acid and heavy metals were limited by complexation competition of Zn. According to Martel [24], large number of Ca could inhibit the complexations of other heavy metals and citric acid. In SCWO-treated PCBs, the content of Zn (see Table 2) was much higher than that of other heavy metals. Hence, Zn could inhibit the chelations of other heavy metals and citric acid. In fact, it can be seen in Fig. 5 that the recovery of Zn in anode region in citric acid treatment was close to 18%, which was much higher than Zn recovered in cathode region, indicating Zn existed mainly as negatively monovalent chelate species in citric acid experiment. Another possible reason for the very low recovery of many metals in anode in citric acid treatment is that large size negatively monovalent chelate species may have very weak ionic mobility compared to the smaller, divalent metal ions Me²⁺ and citrate ions Cit³⁻. It was suggested that even as little as 10–20% dissociation of the chelate would be sufficient to prevent migration of [MeCit]¹⁻ to the anode region [22]. Therefore, many heavy metals like Ni, Cd and Mn migrated mainly to cathode region as free metal ions in citric acid experiment, and the recovery in anode was negligible.

It can also be found in Fig. 5 that the heavy metal contents were still high in the solution of compartment II after 10 h EK treatment in almost all experiments. Metal found in the solution of compartment II has been released from the SCWO-treated PCBs. Hence, 10 h EK treatment was not sufficient for the complete recovery of all heavy metals, and more EK treatment time was needed. Fig. 6 shows the effect of EK time on the total recovery of heavy metals after 10 h EK treatment in the presence of assisting agent HCl. The recovery of Cd, Cr, Ni, and Mn was more than 90% after 30 h EK treatment, and the recovery of As and Zn also increased to 65% and 74%, respectively. Hence, 30 h EK treatment was sufficient for the recovery of the heavy metals in waste PCBs when HCl was used as the assisting agent.

4. Conclusions

- (1) Heavy metals in waste PCBs could be oxidized into corresponding oxides during the SCWO treatment process. The conversion percentages were nearly 100% for most of the heavy metals by SCWO process, with the exception for Ni and Mn, since >50% of the two metals were transferred into the liquid phase of the SCWO-treated PCBs.
- (2) The efficiency of three different assisting agents (HCl, HNO₃, citric acid) for the recovery of six heavy metals (Cd, Cr, As, Ni, Zn and Mn) during electrokinetic treatment of SCWO-treated waste PCBs has been evaluated. It was found that for the recovery of Cd, HCl was the best assisting agent due to the formation of more stable complexation compounds. Citric acid was highly effective for the recovery of Cr, Zn and Mn. HNO₃ was not a satisfactory assisting agent because of the too low recovery efficiencies for Cd and Mn.
- (3) Almost all of the heavy metals migrated to either anode side or cathode side in the EK process, indicating the formation of positively or negatively charged metal species. However, Ni, Cd and Mn mostly migrated to the cathode region in citric acid experiment, where metals existed mainly as positively charged species such as free metal ions due to the complexation competition of Zn.

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