

Evaluation of electrokinetic removal of heavy metals from sewage sludge

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Received 8 December 2004; received in revised form 2 April 2005; accepted 20 April 2005

Available online 1 July 2005

Abstract

The presence of heavy metals is one of the main obstacles for agricultural use of million tonnes of dewatered sewage sludge produced in wastewater treatment plants. Electrokinetic (EK) treatment can be applied to remove heavy metals from sludge. The aim of this study was to increase the efficiency of electrokinetic removal of heavy metals from dewatered sewage sludge. EK experiments were carried out with and without pH adjustment in cathode chamber of acidified sewage sludge. The selective sequential extraction (SSE) was used to determine the fractionation of heavy metals in sewage sludge. The mobility of heavy metals in sludge significantly increased after its acidification at pH 2.7 and followed the order: Ni, Zn, Cu, As, Cr, Pb. Removal efficiencies of heavy metals in the experiment with acidified sewage sludge and pH adjustment at cathode chamber at 2.0 were: 95% for Zn, 96% for Cu, 90% for Ni, 68% for Cr, 31% for As and 19% for Pb. The concentrations of Zn, Cu, Ni, Cr and Pb after EK treatment were below the United States Environmental Protection Agency limits for biosolids applied to agricultural land, forest, public contact sites or reclamation sites.

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Keywords: Electrokinetic technology; Sewage sludge; Heavy metals; Sequential extraction

1. Introduction

Sewage sludge from anaerobic digester of municipal wastewater treatment plants contains essential for plant growth nutrients such as nitrogen, phosphorus, potassium, calcium, microelements, and can be reused in agriculture as fertilizer or soil conditioner without treatment [1], after composting [2,3] or after intensive aerobic bioconversion [4,5]. However, one of the main obstacles for agricultural use of sewage sludge is high content of heavy metals [6], which can cause environmental contamination after sewage sludge landfill application [7]. Therefore, removal of heavy metals from sewage sludge is a desirable pre-treatment before its use as fertilizer or soil conditioner.

The accumulation of heavy metals in sludge is due to uptake of heavy metal ions from domestic and industrial wastewaters by microbial biomass of activated sludge

(biosorption) and due to precipitation of sulfides in anaerobic digester. Chelation and subsequent intracellular accumulation of metals, adsorption onto the microbial cell walls, sheaths and capsules are the main mechanisms of biosorption [8–11].

To remove heavy metals from sewage sludge, chemical extraction [12,13] and bioleaching treatment [14–16] were proposed. Electrokinetic (EK) technology is considered as one of the most promising methods to remove heavy metals from the matter with low hydraulic permeability including contaminated soils and sludges [17–20]. The advantages of electrokinetic technology used for treatment of sewage sludge are the simultaneous removal of different heavy metals, high efficiency of removal and short time of the process. Heavy metals approaching electrodes could be removed or reclaimed by different methods, e.g., by adsorption, pumping out, precipitation or ion-selective membrane [20,21].

Heavy metals occur in sewage sludge in various forms [18]. These forms differ by their speciation and mobility, which have a significant impact on the removal efficiency of

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heavy metals [22]. The specifications of heavy metals can be determined with the selective sequential extraction analysis, which consists of several extraction steps based on using of different chemical reagents and conditions [23–26]. The detected fractions of heavy metals generally include soluble, exchangeable, adsorbed, bound to organics and sulfides, bound to carbonate and residual components. If heavy metals will be in less binding form such as soluble, exchangeable and adsorbed forms, they can be easily removed from sewage sludge. Therefore, it is possible to enhance EK technology by the pre-treatment of sewage sludge to transfer heavy metals in the forms with higher mobility.

The aim of this study was to increase the efficiency of electrokinetic heavy metal removal from dewatered sewage sludge by its acidification and pH adjustment in cathode chamber.

2. Materials and methods

2.1. Sewage sludge

Sewage sludge used in this study was anaerobically digested dewatered sludge collected from a local wastewater treatment plant, which treats mainly industrial wastewater with high concentrations of heavy metals.

2.2. Chemical analyses

The pH value of the sludge samples was measured in a suspension of 1 g of sewage sludge in 10 ml of distilled water using a pH meter (CORNING 145, UK). The content of dry matter (total solids) was determined by standard method [27]. The content of organic matter was determined as a percentage of volatile solids in total solids by the weight loss after ignition of approximately 1 g of dried sample at 500 °C for 20 min [27].

Sewage sludge was oven-dried at 60 °C for 48 h before the determination of heavy metals. Sample digestion was the microwave assisted acid digestion [28] using Microwave Sample Preparation System PAAR PHYSIA (Perkin-Elmer, UK). The liquid sample after digestion was diluted to 100 ml and filtered using 0.45 µm nitrocellulose membrane filters. The content of heavy metals was determined by inductively coupled plasma-optical emission spectrometry [29] using an Optima 2000DV spectrometer (Perkin-Elmer). All tests and analyses were conducted in triplicate, and the results were expressed as mean ± standard deviation.

2.3. Selective sequential extraction

The selective sequential extraction (SSE) method, used to determine the fractions of heavy metal in sludge, was modified from the method developed by Oake et al. [23]. The chemical reagents, used as extractants for heavy metals in SSE, are listed in Table 1. The sludge samples were

Table 1

Chemicals reagents and procedure used in the selective sequential extraction steps of heavy metals

Step	Fractions	Reagent added to sludge in ratio (v/w of dry matter)	Duration of extraction (h)
1	Soluble	Deionized water, 20:1	2
2	Exchangeable	1 M KNO ₃ , 50:1	16
3	Adsorbed	0.5 M KF, 80:1	16
4	Bound to organics or sulfides	0.1 M Na ₄ P ₂ O ₇ , 80:1	16
5	Bound to carbonate	0.1 M Na ₄ EDTA, 80:1	16

oven-dried at 105 °C and ground into powder with size particles smaller than 0.08 mm. Sludge in powder form, 1 g, was placed in 100 ml bottle. Chemical reagents were added in different ratios to sludge by steps and extraction was provided under agitation at 35 rpm (Table 1). Samples were centrifuged after each step at 3000 rpm for 20 min. The supernatants were transferred in volumetric flask and deionized water was added to adjust volume to 100 ml. Solution was filtered through 0.45 µm nitrocellulose membrane filter and analyzed using inductively coupled plasma-optical emission spectrometer. Following five fractions of heavy metals were analyzed: (1) soluble metals; (2) exchangeable metals; (3) adsorbed metals; (4), metals bound to organics or sulfides; (5) metals bound to carbonate. Fraction of heavy metals left in sewage sludge after selective sequential extraction was named residual fraction.

2.4. Acidification of sludge

Acidification of sewage sludge was provided at different pH 2.7, 3.9, 6.0 and 7.1. pH of sludge was adjusted by 3N nitric acid. Sludge was mixed using a mixer (SP-800, RHINO, Taiwan) at 100 rpm for 10 min and treated at 4 °C for 24 h. Initial sewage sludge with pH of 8.3 was used as a control. The acidified sludge was analyzed by SSE method to evaluate the changes in heavy metal mobility.

2.5. Design of reactor for electrokinetic treatment of sewage sludge

The schematic diagram of the electrokinetic reactor is shown (Fig. 1). Both the anode and cathode chambers had the same size, 8 cm in diameter and 3 cm in length. The reaction cylinder had a diameter of 8 cm and a length of 20 cm. Three orifices were drilled evenly on top of the reaction cylinder to monitor voltage distribution during the experiment. Graphite electrodes with a diameter of 7 cm and a thickness of 1 cm were used in the chambers. Forty orifices with 2 mm diameter were drilled at each graphite electrode to allow liquid to flow freely. Two pieces of glass fiber filter paper were used to separate treated sludge and electrodes. Processing fluid was continuously supplied using peristaltic pump to maintain constant level in anode chamber. One normal nitric acid

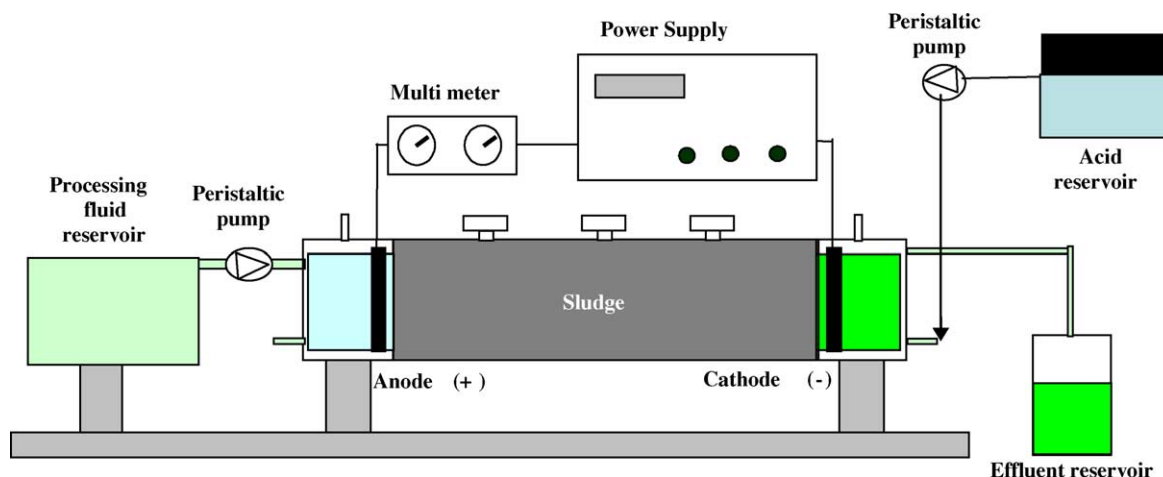


Fig. 1. Schematic diagram of experimental reactor for electrokinetic treatment of sewage sludge.

was added continuously into cathode chamber using peristaltic pump to maintain pH 2. The voltage across EK reactor was measured using a Multimeter CD800 (Sanwa, Japan).

2.6. Experimental set up

Dewatered sewage sludge with pH 8.3 was used in the control (E1). Acidified sludge with pH 2.5 was used in two other EK experiments E2 and E3. The pH in cathode chamber was changed during E2 but was maintained at 2 by the addition of 1N nitric acid in E3 (Table 2).

About 1.2 kg of sludge was placed into the reactor for each experiment. All treatments were conducted with a constant voltage gradient of 1.25 V cm^{-1} for 7 days. Tap water was used as the processing fluid for E1 and E2. Nitric acid solution with pH 2 was used as the processing fluid in E3. The levels of the processing fluid in the anode and cathode chambers were kept the same to prevent hydraulic gradient across sludge in the reactor.

After EK treatment, the cylinder of sewage sludge was cut into five equal parts. The part of sludge nearest to the cathode chamber was further divided into two equal sub-parts. The sludge parts numbered as part 1, 2, 3, 4, 5 and 6 were located at a distance 0–4, 4–8, 8–12, 12–16, 16–18 and 18–20 cm from anode, respectively. Each part was thoroughly mixed and then kept at 4°C for further analysis. The pH values

and the content of heavy metals in the sludge samples were determined.

The quantity of dry sludge after EK treatment (Q_2) was calculated from the initial (VS_1) and final (VS_2) values of volatile solids content assuming that the quantity of organic matter in sewage sludge was not changed during EK treatment.

$$Q_{VS_1} = Q_1 \cdot VS_1 \quad (1)$$

and

$$Q_{VS_2} = Q_2 \cdot VS_2 \quad (2)$$

thus

$$Q_2 = Q_1 \cdot \left(\frac{VS_1}{VS_2} \right) \quad (3)$$

where Q_1 is the quantity of dry sludge before EK treatment.

3. Results and discussion

3.1. Characteristics of sewage sludge

The characteristics of sewage sludge were analyzed as: moisture content, $76.8 \pm 1.4\%$; volatile solids content, $65 \pm 0.2\%$ of total solids; pH, 8.3 ± 0.1 . The initial contents

Table 2
Program of the experiments for EK treatment of sewage sludge

Parameters	Experiments		
	E1 (control)	E2	E3
Sewage sludge	Dewatered sewage sludge	Acidified dewatered sewage sludge	Acidified dewatered sewage sludge
pH of sludge	8.3	2.5	2.5
Amount (kg)	1.2	1.2	1.2
Voltage gradient (V cm^{-1})	1.25	1.25	1.25
Time (days)	7	7	7
Processing fluid	Tap water	Tap water	0.01N nitric acid
pH in cathode chamber	Changed from 7 to 12	Changed from 7 to 12	Maintained at 2 using titration with 1N nitric acid

Table 3
Content of heavy metals in US EPA limits for biosolids, dewatered sewage sludge and sewage sludge after electrokinetic treatment in experiment 3

Samples	Heavy metals (mg kg ⁻¹ of dry weight)					
	As	Cr	Ni	Pb	Cu	Zn
Sewage sludge ^a						
Initial	191 ± 24	1901 ± 11	2053 ± 140	584 ± 41	7764 ± 278	18062 ± 304
After E3	131 ± 19	612 ± 21	211 ± 12	475 ± 14	334 ± 22	921 ± 53
US EPA limits for biosolids						
Limits	75	3000	420	840	4300	7500

^a The results are expressed as mean ± standard deviation.

of heavy metals in sewage sludge were (mg kg⁻¹ of dry weight): As, 191 ± 24; Cr, 1901 ± 11; Ni, 2053 ± 140; Pb, 584 ± 41; Cu, 7764 ± 278; Zn, 18,062 ± 304 (Table 3). The limits for these elements for biosolids applied to agricultural land, forest, public contact sites, or reclamation sites recommended by the United States Environmental Protection Agency (US EPA), in terms of mg kg⁻¹ of dry weight, are as follows: As, 75; Cr, 3000; Ni, 420; Pb, 840; Cu, 4300; Zn, 7500 [30] (Table 3). The content of heavy metals in sewage sludge used in experiments exceeded the limits in 2.5 times for As; 1.6 for Cr; 4.9 for Ni; 1.8 for Cu; 2.4 for Zn.

3.2. The influence of acidification of sewage sludge on the heavy metals mobility

The order of fractions of heavy metals according to their mobility is as follows: soluble > exchangeable > adsorbed > bound to organics/sulfides > bound to carbonate > residual. It is known that release of heavy metals from anaerobically digested sludges is pH dependent [31]. Fractionations of heavy metals in initial sewage sludge and sewage sludge after acidification at different pH are shown in Fig. 2. The mobility of heavy metals increased when pH of the acidification decreased. Within the pH range from 2.7 to 7.1, mobility of heavy metals followed the sequence: Ni, Zn, Cu, As, Cr, Pb. These data are in line with observation that affinity of heavy metals for biomass of white-rot basidiomycete, *Phanerochaete chrysosporium*, under competitive conditions decreased in the following order: Cu(II), Pb(II), Cd(II) [32] and Pb(II), Cu(II), Zn(II) [33]. Heavy metals uptake by biomass of Gram-negative bacterium *Sphaerotilus natans*, isolated from the waste streams of a water treatment plant, decreased also in the order: Pb, Cu, Zn [34]. The metal-affinity for biosorption of heavy metals by bacterial cells of *Pseudomonas aeruginosa*, *Esherichia coli* and *Micrococcus luteus* was in order: Cr³⁺, Cu²⁺, Ni²⁺ [35].

According to the results, from 50 to 60% of arsenic (As) remained in the residual fraction after sludge acidification at all pH used (Fig. 2). Arsenic is a metalloide, not a metal. Microbial transformation of arsenic in anaerobic sewage sludge is well known [36]. In methanogenic bacteria, methylation of inorganic arsenic under anaerobic conditions is coupled to the methane biosynthesis [37]. Several bacterial species able to methylate arsenic compounds to volatile

dimethyl- or trimethylarsine. Therefore, gas released from anaerobic wastewater treatment facilities contains a considerable amount of volatile arsine (AsH₃) [38]. Probably, the fraction of arsenic, remaining after anaerobic treatment, is a stable fraction bound with sewage sludge. The existence of such stable arsenic bound with SH-groups and organoarsenical compounds under reducing conditions was reported [39].

Almost all Cr and Pb remained as organic/sulfides, carbonate and residual fractions in acidified sewage sludge (Fig. 2). Acidification at pH higher than 6 did not significantly change fractionations of Cr. The conversion of carbonate and residual fractions of Cr and Pb into the organic/sulfides fraction correlated with the pH decrease. Compared to the residual fraction, decrease of the carbonate fraction was more intensive when pH decreased. Thus, the fraction bound to carbonate was more sensitive to pH change. The negligible content of soluble and exchangeable fractions of Cr and Pb indicated their poor mobility in sludge.

The percentage of soluble and exchangeable fractions of Ni increased with the pH decrease of sludge acidification (Fig. 2). The mobility of Ni reached its lowest level at pH 6. The contents of organic/sulfides, carbonate, and residual fractions decreased when pH was lower than 6. Residual and organic/sulfides fractions predominated for Cu and Zn at pH 6. The content of soluble and exchangeable fractions of Zn increased significantly at pH below 6. For Cu, large part of residual fraction was transformed into soluble, exchangeable, adsorbed, and organic/sulfides fractions, which can be removed by EK treatment. In general, the effect of pH on mobility of Zn was higher than that of Cu.

Therefore, acidification of sewage sludge changed the distribution of heavy metals by transfer of their different fractions into more mobile forms.

3.3. pH and VS profiles of sewage sludge after electrokinetic treatment

In electrokinetic treatment of sewage sludge without acidification (E1), the acid flow, generated at anode, moved towards cathode under electrical field, as well as advection and diffusion. The pH of sludge parts from 1 to 4 (acidic) was basically lower than that of sludge parts 5 and 6 (alkaline). However, the pH value did not reach 2 in acidic and 12 in alkaline parts (Fig. 3a).

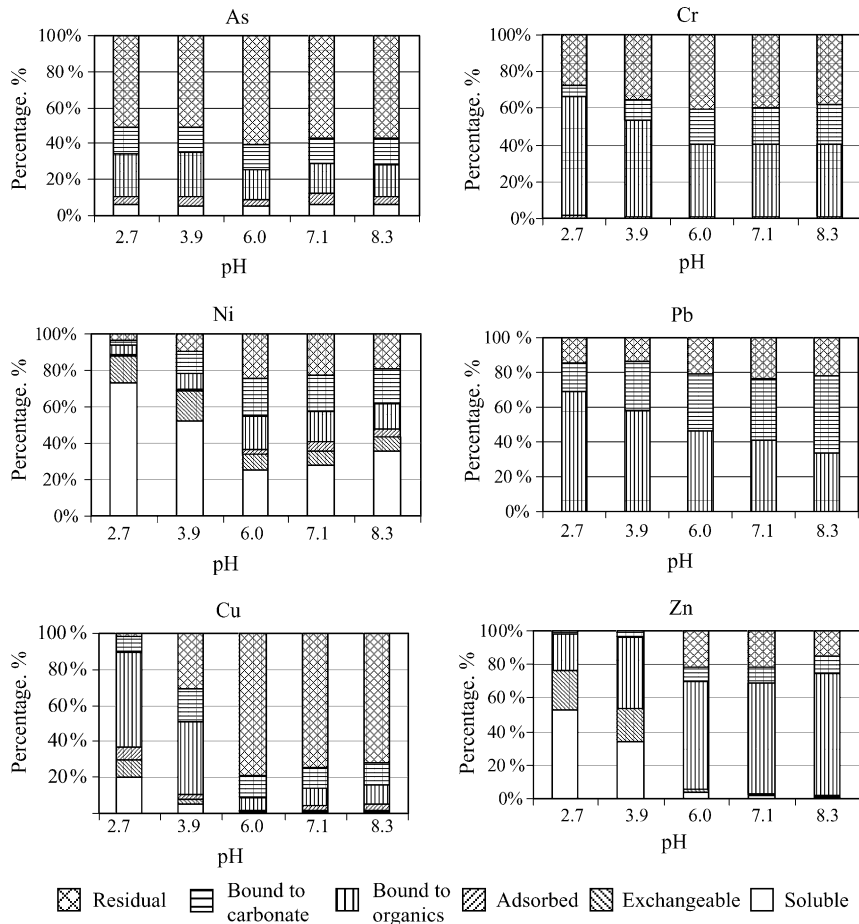


Fig. 2. Fractionations of heavy metals in initial sewage sludge (pH 8.3) and sewage sludge after acidification at different pH. The standard deviations are represented by error bars.

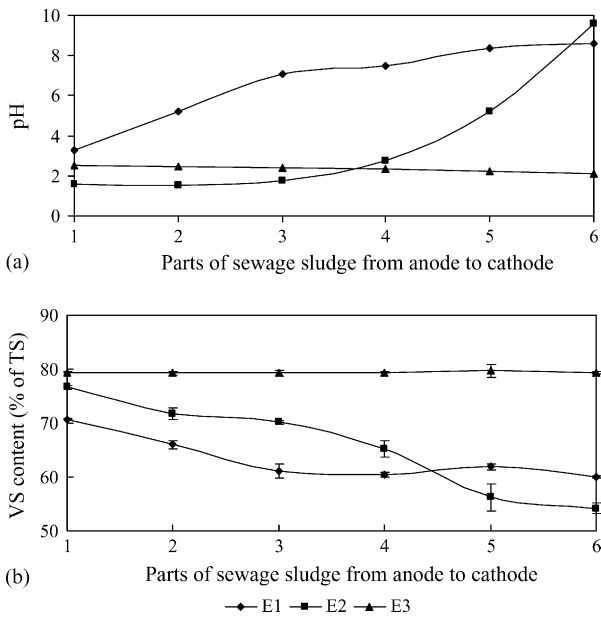


Fig. 3. pH (a) and VS (b) profiles in sewage sludge parts after electrokinetic treatment in E1, E2 and E3. The standard deviations are represented by error bars.

Acidification of sewage sludge at pH 2.5 was provided before EK treatment in E2. The moisture and VS contents of acidified sludge increased a little from 76.8 ± 1.4 to $78.1 \pm 0.58\%$ and from 65 ± 0.2 to $66.4 \pm 0.1\%$ of total solids, respectively. The texture of sewage sludge did not change after acidification. The pH of sludge parts 1–4 was equal to or below 2. The pH of sludge increased at distance more than 13 cm from anode and reached 12 at cathode. In addition to acidification of sludge, high pH in cathode chamber was adjusted using nitric acid during the EK process in experiment 3. The pH of all sludge remained constant at 2 in E3.

The change of VS content in sludge was probably because of the migration of inorganic matter during EK process (Fig. 3b). Soluble inorganic ions moved in the electrical field to the opposite polar electrode, while the organic matter migration was relatively negligible. Extensive precipitation, co-precipitation, complexation and adsorption could happen in the zone near cathode under high pH and reducing conditions. The decrease of the content of inorganic matter resulted in increase of the VS content. In E1 and E2, inorganic matter migrated from anode, which caused the increase of VS content in the part of sludge near anode. When metal ions

reached cathode, most of them were precipitated. VS content there decreased in both E1 and E2, but the content was lower in sludge in E2 due to acidification and higher mobility of metals. VS contents in sludge were 71 and 78% at anode and 60 and 55% at cathode in E1 and E2, respectively, compared to the initial content of 65%. These results indicated that migration of heavy metals was more intensive in E2 than that in E1. There was no precipitation in E3 and inorganic matter was directly extracted from sludge. Therefore, considerable heavy metals removal can be expected in E3. The VS content of sludge in E3 remained constant (80%) and did not change with the distance to anode. Therefore, it was 20% reduction of sludge weight. This was coincided with the observation of significant sludge volume reduction after the EK process.

3.4. Electroosmotic flow in the EK treatment

Recorded electroosmotic flow in experiment E1, with initial dewatered sewage sludge, was approximately 13 ml h^{-1} at the beginning of EK treatment. It was decreased to 2 ml h^{-1} after 8 h. However, the electroosmotic flow in experiments E2 and E3 was too slow for measurement due to acidification of sludge. Acidification of sludge caused decrease of zeta potential of sludge's particles, which resulted in slow electroosmotic flow.

3.5. Changes of heavy metals contents in sludge after EK

The changes of the content of heavy metals in different sludge parts as a percentage from their initial content after EK treatment are presented in Fig. 4.

The removal of heavy metals from non-acidified sewage sludge was not significant for all detected elements in E1 (Fig. 4a). The highest removal efficiency, 14.5%, was detected for Ni. This result was in line with the low mobility of elements, except Ni, as shown in Fig. 2. The content of heavy metals did not significantly change in the examined sludge parts. The content of As increased in sewage sludge parts towards anode. This could be because As tends to exist in the form of H_2AsO_3^- or HAsO_3^{2-} under reducing condition and at pH higher than 7.8. When H_2AsO_3^- or HAsO_3^{2-} reached the zone near anode, where pH was generally low, they were turned into H_3AsO_3 , hence lost its mobility.

The removal of heavy metals from acidified sewage sludge in E2 was the same as in E1. However, slight migration of heavy metals in sludge parts was observed (Fig. 4b). For example, the contents of Ni and Pb decreased to 25 and 79% from the initial value in the first part of sewage sludge near anode and to 55 and 87% from the initial value in the second part. Starting from the third sludge part, the contents of Cu, As, Cr and Pb were higher than the original values. In the parts 4–6 of the treated sludge, the contents of heavy metals increased from the initial values by 27% for As, 25% for Cr, 62% for Ni, 11% for Pb 48% for Cu and 95% for Zn. The results indicated that the heavy metals migrated towards

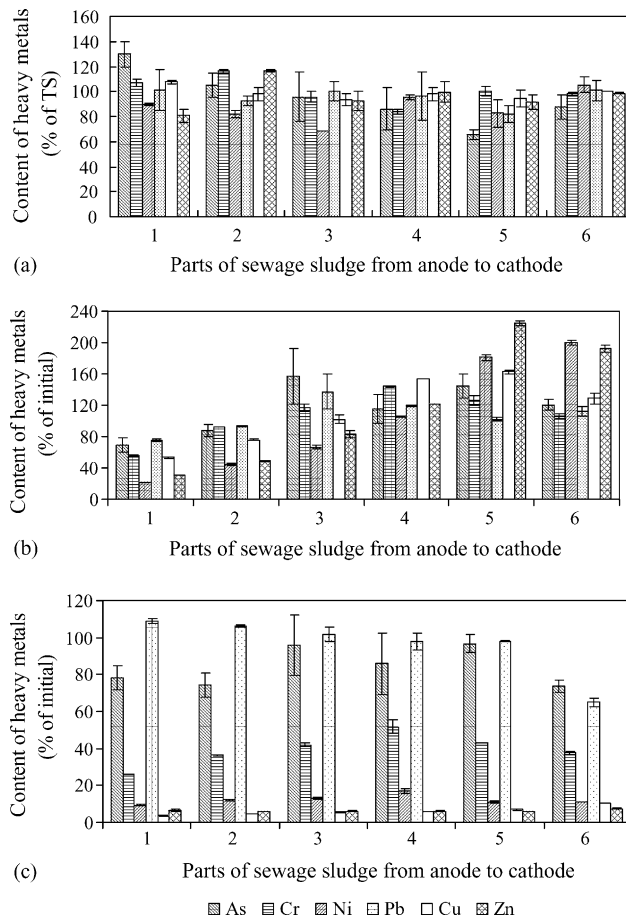


Fig. 4. Content of heavy metals, % of initial, in parts of sewage sludge after its electrokinetic treatment in: E1 (a), E2 (b) and E3 (c).

cathode, but were not accumulated into cathode chamber or electroplated onto cathode. Probably, this could be due to the high pH near the cathode electrode, which caused the lower heavy metals mobility or even their precipitation. These results were in line with the finding that the content of volatile solids decreased in the sludge parts near cathode (Fig. 3b).

Significant removal of heavy metals from sewage sludge was obtained in E3, when pH at cathode chamber was controlled at 2 in addition to acidification of sludge (Fig. 4c). Removal efficiencies of electrokinetic treatment with pH adjustment to 2.0 in cathode chamber were 95% for Zn, 96% for Cu, 90% for Ni, 68% for Cr, 31% for As and 19% for Pb.

The contents of heavy metals in sewage sludge after EK treatment in E3 were (mg kg^{-1}): As, 131 ± 19 ; Cr, 612 ± 21 ; Ni, 211 ± 12 ; Pb, 475 ± 14 ; Cu, 334 ± 22 ; Zn, 921 ± 53 (Table 3) and were (%): 69 for As; 32 for Cr, 10 for Ni; 81 for Pb; 4 for Cu; 5 for Zn of initial values. The low removal efficiency of Pb indicated its strong binding with sludge even at pH 2. The contents of Cr, Ni, Pb, Cu, Zn, but not As in sludge after EK treatment in E3, were below the limits for biosolids applied to agricultural land, forest, public contact sites, or reclamation sites recommended by the US EPA [30].

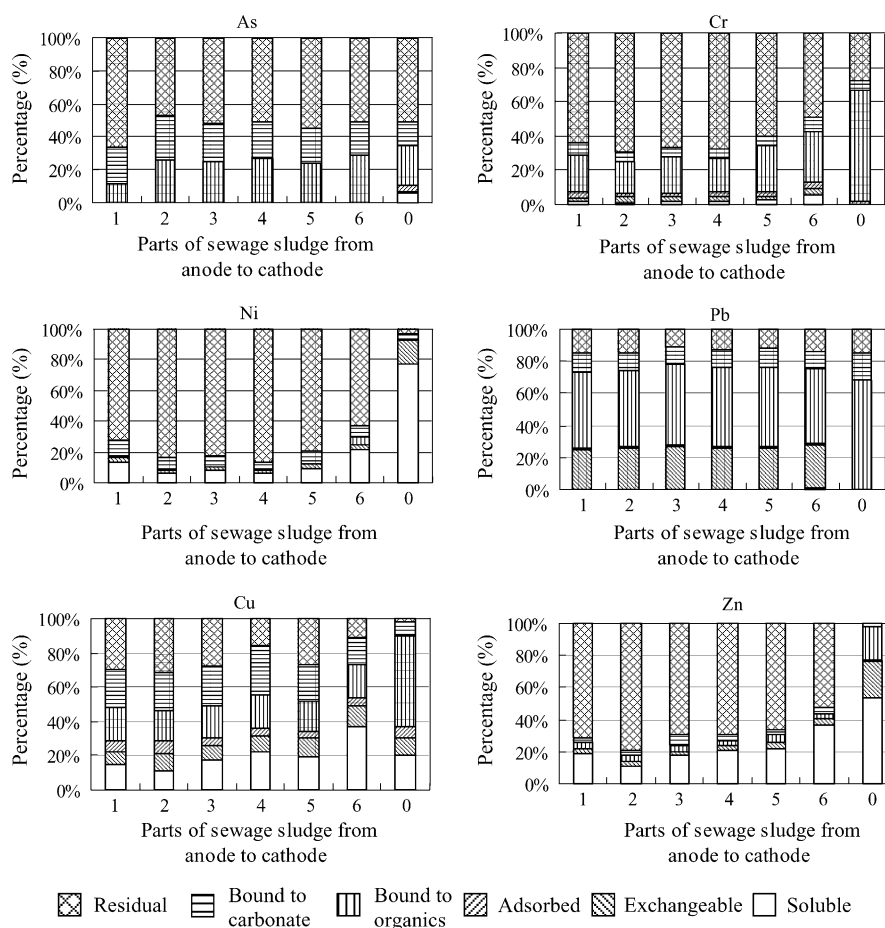


Fig. 5. Fractionations of heavy metals in initial sewage sludge (part 0) and in parts of sewage sludge after its electrokinetic treatment in E3. Initial sewage sludge was sludge acidified at pH 2.4 at 4 °C for 24 h.

3.6. Fractional distributions of heavy metal in acidified sewage sludge after EK treatment with pH 2.0 in the cathode chamber

The fractional distributions of heavy metals of acidified sewage sludge (expressed as part 0) and in different parts of acidified sewage sludge after EK treatment with pH adjusted to 2.0 in the cathode chamber (E3) are shown in Fig. 5.

It was found that inter-fractional transformations of heavy metals in sludge occurred during EK treatment in E3. Hardly removed fractions of heavy metals, such as bound to organics/sulfides or carbonate and residual fractions were the predominant forms in sludge after EK treatment. The percentage of residual fraction increased in sludge after EK treatment, especially for heavy metals with high removal efficiency, such as Cr, Ni, Cu and Zn, but it was practically stable for Pb and As. Exchangeable fraction of Pb in sludge after EK treatment consisted of 25%, however, the total removal of Pb was only 18.7%. For Cu, the soluble and exchangeable fractions were the main forms left in EK treated sludge though the removal efficiency was 95.7%. It was probably because certain level of dynamic equilibrium of loosely bound and strongly bound heavy metals existed in sludge. When more loosely bound

heavy metals were removed, the strongly bound heavy metals were partly turned into more loosely bound heavy metals to maintain the equilibrium. It was found that the percentage of residual fraction of Ni was higher than that of Cu and Zn after EK treatment in E3. It indicated that small part of residual fraction of Ni was transformed into loosely bound fractions due to EK treatment. This explained why the overall removal efficiency of Ni was lower than that of Cu and Zn though it was concluded from Fig. 2 that the mobility of Ni was higher than Zn and Cu.

4. Conclusions

Electrokinetic treatment of acidified sewage sludge with pH adjustment to 2.0 in cathode chamber can be used for removal of heavy metals. Acidification of sewage sludge at pH 2.4 at 4 °C for 24 h transferred heavy metals from forms with low mobility to forms with high mobility. Removal efficiencies of electrokinetic treatment with pH adjustment to 2.0 in cathode chamber were 95% for Zn, 96% for Cu, 90% for Ni, 68% for Cr, 31% for As and 19% for Pb. The concentrations of Zn, Cu, Ni, Cr and Pb after EK treatment were

below US EPA limits for biosolids applied to agricultural land, forest, public contact sites, or reclamation sites.

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