Electrodialytic removal of heavy metals from municipal solid waste incineration fly ash using ammonium citrate as assisting agent

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Abstract

Electrodialytic remediation, an electrochemically assisted separation method, has previously shown potential for removal of heavy metals from municipal solid waste incineration (MSWI) fly ashes. In this work electrodialytic remediation of MSWI fly ash using ammonium citrate as assisting agent was studied, and the results were compared with traditional batch extraction experiments. The application of electric current was found to increase the heavy metal release significantly compared to batch extraction experiments at comparable conditions (same liquid-to-solid ratio, same assisting agent, and same extraction time). Up to 86% Cd, 20% Pb, 62% Zn, 81% Cu and 44% Cr was removed from 75 g of MSWI fly ash in electrodialytic remediation experiments using ammonium citrate as assisting agent. The time range for the experiments varied between 5 and 70 days.

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1. Introduction

Fly ashes from municipal solid waste incineration (MSWI) contain high concentrations of heavy metals and other toxic components Due to the potential leaching of these components when the ashes are land filled, the MSWI fly ashes are regulated as hazardous waste in many countries, and the ashes should be treated for detoxification and/or for the recovery of metals prior to disposal. Electrodialytic remediation, an electrochemically assisted separation method which has previously shown capability for removing heavy metals from different fly ashes [1–3], could be a potential treatment method. In previous work different assisting agents have been evaluated in connection with electrodialytic remediation of MSWI fly ash, and it has been found that of four different assisting agents, alkaline ammonium citrate solution was the most promising assisting agent for simultaneous removal of Cd, Pb, Zn Cu and Cr [1]. In this present work electrodialytic

remediation of MSWI fly ash using ammonium citrate solution as assisting agent is studied further by conducting new series of electrodialytic remediation experiments. The time dependent heavy metal removal is evaluated, and the results are compared with traditional extraction methods.

2. The principle of electrodialytic remediation

Electrodialytic remediation, which was originally developed for remediation of heavy metal polluted soil, is a combination of electrokinetic remediation and electrodialysis [4,5]. Recently, the method has been further developed for remediation of other porous materials as well, e.g. CCA-impregnated wood waste [6] and, as reported in this present work, fly ash [1–3]. The principle of electrodialytic remediation of fly ash is shown in Fig. 1.

A stirred suspension of fly ash and an assisting agent (e.g. ammonium citrate solution) is placed in compartment III. Aqueous solutions (electrolytes) are circulated in compartments I–V. When a low voltage dc current is applied, the ions in solution will electromigrate in the electric field either

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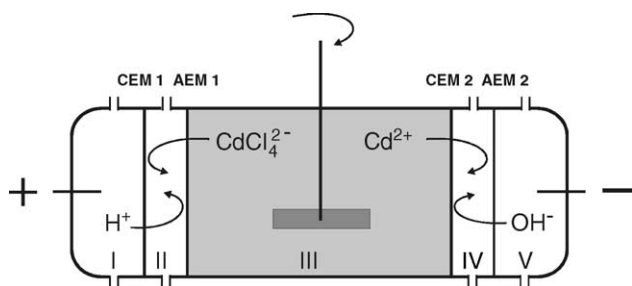*E-mail address:* ajp@byg.dtu.dk (A.J. Pedersen).

Fig. 1. Electrodialytic remediation of fly ash in principle. CEM: cation exchange membrane, AEM: anion exchange membrane.

toward the anode (+) or toward the cathode (−) depending on their speciation. As a result, the ions are transported out of the fly ash and into the electrolytes in compartment II or IV where they can be separated or precipitated by conventional methods. Ion exchange membranes are used to separate the fly ash suspension (or other polluted medium) from the electrolytes. The use of ion exchange membranes ensures current efficiency by allowing ions from the ash suspension to electromigrate to the electrolytes, but preventing ions (with opposite charge) from the electrolytes from entering the ash suspension. Both electrodes are made of inert material.

Since only the free ions in the solution are transported in the electric field, precipitated and adsorbed heavy metals must be converted to mobile ionic forms before they can be removed. Fortunately, the mobilisation is enhanced by pH changes occurring in the fly ash suspension during the treatment. During the electrodialytic remediation process the polluted medium (e.g. fly ash) is acidified, even though ion exchanges membranes are placed as shown in Fig. 1. Anion exchange membranes are not 100% ideal, and thus some of the acid may originate from the anolyte. However, it is likely that water splitting at the anion exchange membrane (AEM1) is of great importance to the acidification. Water splitting may occur when a limiting current is exceeded. In systems with NaCl as the only salt nearly all the current is carried by salt cations in cation exchange membranes even at very high current densities. In contrast pH changes are observed at the anion exchange membranes, thus the limiting current is lower for an anion exchange membrane than for a cation exchange membrane [7]. It has been found that the water splitting is highly determined by the nature of the charged groups in the membranes [8–10]. In most anion exchange membranes the charged groups are quarternary ammonium groups, whereas most cation exchange membranes contain sulfonic acid groups. In the case of cation exchange membranes no significant water splitting is reported as long as the solution contain only NaCl, whereas the introduction of various cations such ad Ca^{2+}, Mg^{2+} and NH_4^+, as well as certain organic species, has been found to induce water splitting [7,8]. Thus, water splitting during electrodialytic remediation of fly as may be induced not only by the functional groups in the ion exchange membranes, but also by the presence

and concentration of the different ions in the solution. Water splitting at ion exchange membranes during electrodialytic remediation of soil has been studied by Ottosen et al. [7]. They found that water splitting can be expected at the anion exchange membrane even at very low current densities (0.1 mA/cm^2 being the lowest current density investigated). Water splitting at the cation exchange membrane (leading to increased pH in the soil) was observed at 0.5 mA/cm^2 but not at 0.3 mA/cm^2 in the soil system [7]. In another study on electrodialytic remediation of soil, the limiting current was found to be between 0.4 and 0.75 mA/cm^2 in a similar soil system [11].

Heavy metal desorption can also be enhanced by adding assisting agents, e.g. acids, bases, surfactants or complexing agents, to the medium during electrodialytic remediation, for instance if the polluted medium has a high buffering capacity as is often the case with fly ashes, or if the metals of concern are not mobile at low pH values, as under certain conditions, e.g. As or Cr(VI).

Different assisting agents have previously been evaluated in connection with electrodialytic removal of Cd, Pb, Zn, Cu and Cr from MSWI fly ashes [1]. Of four assisting agents investigated (i.e. ammonium citrate, sodium citrate, ammonia solution and distilled water), it was found that ammonium citrate (a 0.25 M di-ammonium citrate/1.25% NH$_3$ solution) was the most efficient assisting agent for removal of all five metals at the same time. Hence, in this present study, a similar ammonium citrate solution was chosen as assisting agent in further electrodialytic remediation experiments on MSWI fly ash to evaluate the time-dependent removal.

Citrate is a naturally occurring chelating agent which forms soluble, multidentate complexes with various metals, and it has shown potential for extraction of heavy metals from polluted soil [12,13]. Ammonia (NH$_3$) can form stable tetra ammine complexes with, e.g. Cu, Zn and Cd at high pH. In contradiction to, e.g. EDTA, ammonium citrate is biodegradable and not toxic to the environment.

3. Experimental

3.1. Analytical methods

The following analytical methods were used in this work: metal concentrations were measured in aqueous phase by atomic absorption spectroscopy (AAS) in flame using a "GBC-923" or "Perkin Elmer 5000" atomic absorption spectrophotometer. Ash samples for metal analyses were pretreated by digesting 0.4 g dry ash and 10 ml concentrated HNO$_3$ for 30 min at 135 psi using microwave (CEM MDS-2000), followed by vacuum filtration (45 μm) and dilution to 50 ml. The digestion method was modified from Danish Standard DS 259 for determination of metals in soils and sediments. pH in the ash was measured with a "Radiometer" combined pH-electrode in 1 M KCl at a liquid-to-solid (L/S) ratio of 5. Water content was found as loss of weight after

Table 1
Characteristics of the experimental fly ash

Parameter	Mean ± S.D.
Water content (%)	0.74 ± 0.01[a]
Loss on ignition (%)	1.28 ± 0.02[a]
pH (L/S = 5)	12.16 ± 0.01[a]
Ca (mg/kg DM)	140100 ± 2300[b]
Cd (mg/kg DM)	241 ± 16[b]
Pb (mg/kg DM)	8070 ± 45[b]
Zn (mg/kg DM)	17140 ± 710[b]
Cu (mg/kg DM)	1570 ± 70[b]
Cr (mg/kg DM)	285 ± 3[b]

S.D.: standard deviation.
[a] $n = 3$.
[b] $n = 5$.

drying at 105 °C and content of organic matter was found as loss on ignition (550 °C).

3.2. Chemicals

All chemicals used were of analytical grade. The ammonium citrate solution used as assisting agent was prepared from di-ammonium hydrogen citrate dissolved in 1.25% NH_3, and it was 0.25 M with respect to citrate concentration. The pH of the solution was approximately 9.5.

3.3. The experimental ash

A MSWI electrostatic precipitator ash, obtained from the Danish mass-burn MSWI facility "Vestforbrænding" was used for the experiments. At the point of sampling (the electrostatic precipitator), the gas stream had not been exposed to any flue gas cleaning additives, e.g. lime. Some characteristics of the fly ash are given in Table 1.

3.4. Electrodialytic remediation experiments

The electrodialytic remediation cell used in the remediation experiments was made of cylindrical Plexiglas compartments, designed and developed at the Technical University of Denmark. It consisted of five compartments as illustrated in Fig. 1. The ash compartment (compartment III) was 10 cm in length and with an inner diameter of 8 cm. In all electrodialytic remediation experiments initially 75 g of naturally dry fly ash was placed in the ash compartment, and the compartment was filled up to 500 ml with 0.25 M ammonium citrate solution, corresponding to a liquid-to-solid (L/S) ratio of approximately 6.5.

Two hundred and fifty milliliters of electrolyte solution was circulated in each of compartments II and IV, respectively, and 500 ml of electrolyte was circulated in each of compartments I and V, respectively, using "Pan World" magnetic pumps with a flow rate of 2.6 l/min. The electrolyte solution in compartment I was 2.5% NH_3, in compartment II it was 0.25 M ammonium citrate solution, and in compartments IV and V it was acidified 0.01 M $NaNO_3$ (pH < 2).

A "Heto" motor, with a rotation velocity of 1300 rpm was used to stir the ash suspension during the electrodialytic treatment. The stirrer, which was especially designed for the cell, was made of a flexible plastic piece fastened to an insulated metal wire. The total length of the piece was 5.5 cm and the width was 6 mm. The stirrer as well as the ash slurry was introduced into the compartment III through holes in the top of the cell. Extra liquid could be added through these holes as well if necessary.

The electrodes were made of platinum coated titanium wire (Ø = 3 mm), obtained from "Permascand". The ion exchange membranes used were obtained from Ionics (anion exchange membrane 204SZRA B02094A and cation exchange membrane CR67HMR N10262A). The charged groups were sulfonated copolymers at the cation exchange membrane and quaternary ammonium groups at the anion exchange membrane. With an inner diameter of the cell of 8 cm, the surface area of the membranes corresponded to approximately 50 cm^2 each.

Voltage drop and electric current was read on weekdays during the remediation period, and pH was measured in the electrolytes. pH in electrolyte compartment IV and V was adjusted with 1:1 HNO_3 if pH > 2, and pH in electrolyte compartments I and II was adjusted with NaOH tabs if pH < 9.

At the end of the experiments the ashes were drained on filter paper before drying in order to separate the solids from the aqueous phase. Water content and pH was measured. Ashes, aqueous phases, electrolytes, membranes and electrodes were analysed with respect to heavy metal content (Cd, Pb, Zn, Cu and Cr) using AAS. Membranes and electrodes were soaked in HNO_3 (1 and 5 M, respectively) over-night to release metals prior to AAS analysis. All ash analyses were made in two or three replicates.

A total of six electrodialytic remediation experiments (experiments 1–6) and two reference experiments (experiments 7 and 8) were made. The six electrodialytic remediation experiments were all run at 40 mA, corresponding to 0.8 mA/cm^2, for varying periods of time (5–70 days). The reference experiments (experiments 7 and 8) were different kinds of batch extraction experiments both with an extraction time of 14 days (same duration as experiment 3). Experiment 7 was a 0-current experiment (no electric current was applied to the cell, otherwise alike experiment 3). Experiment 8 was a "simple" batch extraction experiment; in that 75 g fly ash was agitated with 500 ml ammonium citrate solution for 14 days in a 1 l plastic bottle.

The experimental parameters for the different experiments are shown in Table 2.

From Table 2, it is seen that the cell voltages vary in all electrodialytic remediation experiments, ranging from 2.6 to 28.7 V with a tendency of increasing upper values with increasing remediation time. The cell voltage increases when the electrical resistance increases since the current is kept constant. Various factors may contribute to the overall cell voltage. Cathodic and anodic overpotentials, the electric resistance in the electrolyte solutions and over the ash com-

Table 2
Experimental parameters for the remediation experiments carried out

Experiment no.	Duration (days)	Current (mA)	Current density (mA/cm^2)	Voltage drop (V)	Comments
1	5	40	0.8	3.2–10.5	Electrodialytic remediation
2	10	40	0.8	3.6–6.4	Electrodialytic remediation
3	14	40	0.8	2.6–8.2	Electrodialytic remediation
4	21	40	0.8	3.5–6.6	Electrodialytic remediation
5	35	40	0.8	3.4–18.3	Electrodialytic remediation
6	70	40	0.8	3.5–28.7	Electrodialytic remediation
7	14	0	0	–	0-current experiment
8	14	0	0	–	Batch extraction experiment

partment and ion exchange membranes. The electrical resistance in the electrolyte compartments is considered low due to high ionic strength of the electrolytes and low polarisation potential at the electrodes, since the electrolytes are circulated. On the other hand, increasing resistance over the ash compartment and ion exchange membranes is expected as the ash compartment is being depleted in ions [7]. Fouling of the membranes during the electrodialytic remediation process may also lead to increased resistance. Studies of ion exchange membranes used in electrodialytic soil remediation experiments found, however, that the resistance only increased slightly for membranes that had been used for a 3 months remediation experiment [14].

4. Results and discussion

4.1. Electrodialytic remediation experiments

The distribution of Cd, Pb, Zn, Cu and Cr in the different experiments after remediation is shown in Fig. 2 together with the initial amount (mg) of each metal.

From Fig. 2 it is seen that the metal distribution is changing with changing remediation time. By comparing experiments 1–6 it is seen that for all metals, the amount of metals in solution (aqueous phase + removed to the electrolytes) generally is increasing with increasing remediation time, and consequently the residual amount of metals bound in the ash is decreasing. This indicates that the metals are continuously being released from the solid phase during the remediation process. However, especially for Zn, there are diverging results. These may be explained by imperfect mass balances (% recovery) for Zn. The Zn recovery in the different experiments varies from 1266 to 2191 mg (*y*-axis values), corresponding to 99–170% recovery (% recovery = mg final/mg initial × 100%) with no general tendency. The imperfect mass balances may be explained by inhomogeneous distribution of Zn in the ash, although incomplete digestion of the ash samples or precipitation of metals in, e.g. tubes or elsewhere in the system may also be part of the reason. Large variations in recovery are also seen for Cr, varying between 61 and 142% recovery. For Cd and Cu the mass balances are generally good (close to 100%), except for Cd in experiment 4 (138%). The recovery of Pb is a little low (74 and 71%) in the two remediation experiments with the longest remediation time (experiments 5 and 6), possibly due to precipitation of Pb in tubes, etc. As a consequence of these variations in metal recovery, the presented remediation results are to be regarded as general tendencies rather than implicit results, especially for Zn and Cr.

When an electric dc current is applied to the ash system, the heavy metals in solution and in exchangeable forms are removed continuously as charge-bearing ions in the electric field. Therefore, it is expected that the chemical equilibrium reactions are shifted toward enhanced dissolution. As a result, the application of an electric dc current should enhance metal desorption compared to traditional batch extraction experiments. Changes in chemical associations of Cu in soil during electrodialytic remediation toward increased solubility have been observed [15]. By comparing the results from Experiment 3 with the two reference experiments (experiments 7 and 8), this theory is supported. With the exception of Pb, a larger fraction of the metals are released in the electrodialytic remediation experiment (experiment 3), compared with the two reference experiments. Incomplete recovery of Pb in experiment 3 may explain why less Pb was found in solution in this experiment compared with experiment 7. When looking at, e.g. experiment 2 (which had shorter duration than experiment 3), more Pb was found in solution.

Another advantage of using electrodialytic remediation compared with batch extraction is that the dissolved metals becomes physically separated from the ash when they are electro-migrating from the ash compartment, through the ion exchange membranes and into the electrolyte compartments. Thus, a subsequent filtration step, which may be both difficult and expensive, can be avoided. When comparing experiment 3 specifically with experiment 7 (which was the 0-current experiment) it is observed that the application of current increases the amount of metals removed from the ash suspension to the electrolytes by a factor of at least 50% for all metals (see Fig. 2). There is, however, still a significant amount of metals removed to the electrolytes in the 0-current experiment (experiment 7). This transport can be explained by interdiffusion phenomena. Interdiffusion is defined as an exchange of counter ions across an ion exchange membrane separating two electrolytes, as is the case in the electrodialytic remediation cell [16]. For example, if the ion exchange membrane is a cation exchange membrane separating elec-
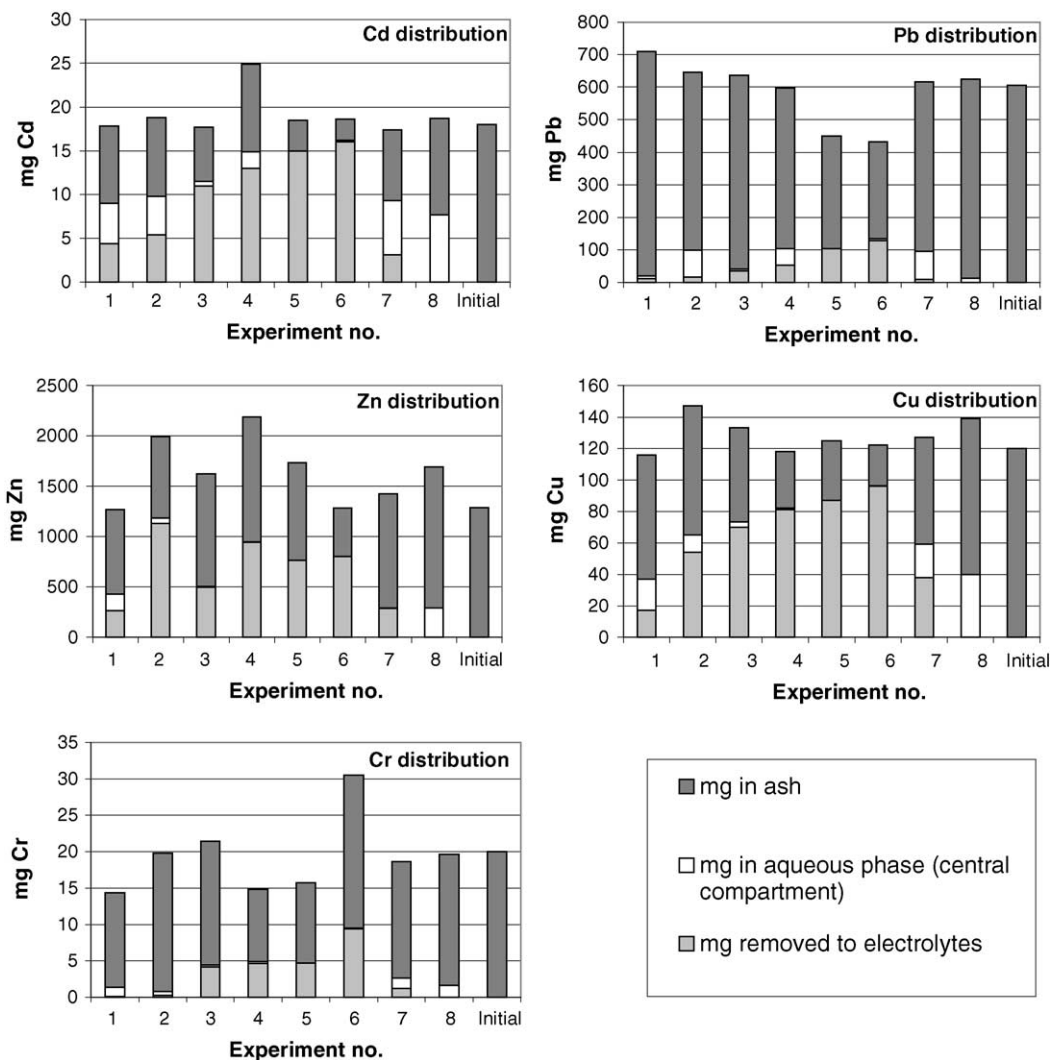
Fig. 2. Distribution of Cd, Pb, Zn, Cu and Cr in the different experiments. "mg in aqueous phase" represents the amount of metals found in the aqueous phase of the ash suspension after draining. "mg removed to electrolytes" includes the metals found in the electrolyte solutions, on the electrodes and in the ion exchange membranes after remediation. "mg in ash" is the amount of metals found in the ash residue at the end of remediation (mean value of two or three replicates). "Initial" is the initial amount of each metal present in the ash (mean value of five replicates).

trolyte AX from electrolyte BX, there will be a flux of A cations to the BX electrolyte and a similar flux of B cations to the AX electrolyte to obtain electroneutrality [16].

The "electrodialytic" remediation efficiency, defined as the percentage of metals removed from the ash suspension to the electrolytes as a function of remediation time, is illustrated in Fig. 3 for experiments 1–6. In Table 3 the metal removal rates are given as millimoles removed per hour per square meter of membrane.

From Fig. 3, it is seen that the major part (>70%) of both Cu and Cd is removed during the first 3 weeks of remediation, and further remediation does not improve the removal efficiency for these two metals considerably. This indicates a quite fast (instant) mobilisation of Cu and Cd. Table 3 reveals that the metal removal rates generally increase during the first 2 weeks of remediation, then decrease. This means that at the given experimental conditions the most efficient remedi-
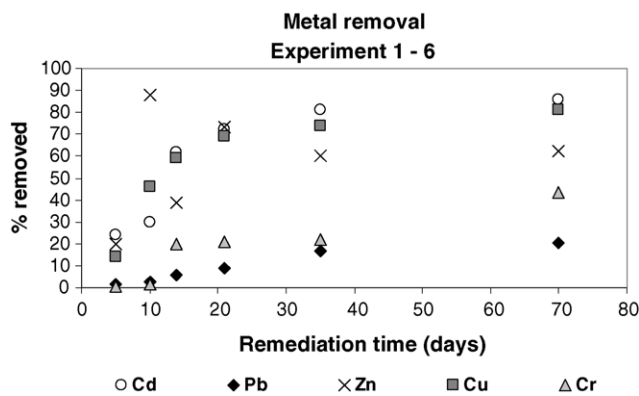

Fig. 3. Percentage of metals removed (calculated on the basis of the initial amount) from the ash suspension to the electrolytes in experiments 1–6.

Table 3
Metal removal rates in the different electrodialytic remediation experiments given as millimoles removed per hour per square meter of membrane

Experiment no.	Metal removal rate (mmol h^{-1} m^{-2})				
	Cd	Pb	Zn	Cu	Cr
1	0.065	0.088	6.64	0.44	0.003
2	0.040	0.064	14.33	0.70	0.005
3	0.058	0.100	4.50	0.65	0.048
4	0.046	0.101	5.70	0.50	0.035
5	0.032	0.119	2.78	0.32	0.021
6	0.017	0.074	1.45	0.18	0.021
7	0.016	0.026	2.60	0.35	0.014

ation is achieved when the remediation time is set to approximately 2 weeks. The highest removal rate is seen for Zn, reaching 14.3 mmol Zn removed h^{-1} m^{-2} in experiment 2 (10 days). With an initial concentration of 17.1 g Zn kg^{-1} ash (see Table 1), which corresponds to approximately 260 mmol Zn kg^{-1}, it would take approximately 18.5 h to clean 1 kg ash in a plant containing 1 kg ash per square meter of membrane at this removal rate. The lowest removal rate is seen for Cr, never exceeding 0.05 mmol h^{-1} m^{-2}, meaning that it would take at least 110 h to clean 1 kg of ash with respect to Cr.

The removal of Pb, on the other hand, is an almost linear function of the remediation time, indicating a continuous but rather slow mobilisation of Pb. After 70 days, only 20% Pb is removed.

The results for Zn are not consistent. More Zn is removed in the 10 and 21 days experiments compared to all other experiments. These varying results may be caused by small variations in the chemical environment (pH, redox conditions) between the different experiments, or it may be a consequence of the previously described imperfect mass balances. The 10 and 21 days experiments both have a very high percentage recovery for Zn (155 and 170%, respectively).

For Cr, the percentage removal seems to stabilise around 20% after 14 days of remediation, indicating that the residual 80% of the Cr is very tightly bonded in the ash. But, as for Zn, imperfect mass balances for Cr rather than real remediation slow-down may have caused this tendency. The Cr removal seems to increase again between 35 and 70 days.

After 70 days of electrodialytic remediation, totally 86% Cd, 20% Pb, 62% Zn, 81% Cu and 44% Cr is removed to the electrolytes.

The final ash weight (g dry matter), the volume of the aqueous phase collected at the end of the experiments after draining of the ash suspension and the final pH in the ash is listed in Table 4.

As seen from Table 4, the final pH in the ash is lowered to between 6.9 and 10.8. Initially, the ash had a pH of 12.2 (Table 1), and the ammonium citrate solution had a pH of approximately 9.5. The pH lowering, which is more pronounced with longer remediation time, may be explained by both the previously described water-splitting phenomena at the anion exchange membrane during electrodialytic remediation, and by ageing processes (e.g. carbonation) occurring in the ash

Table 4
Volume of aqueous phase collected in the central compartment, final pH in ash suspension and final ash weight in remediation experiments

Exp. no.	Volume of aqueous phase collected (ml)	Final pH in the ash suspension	Final ash weight (g DM)
1	184	9.5	74.8
2	140	9.4	69.6
3	185	8.9	71.6
4	130	9.6	60.4
5	0	8.2	50.6
6	25	6.9	46.8
7	261	8.7	75.1
8	200	10.8	83.9

The final ash weight includes the weight of residual citrate in the ashes. The initial ash weight was 75 g in all experiments, before addition of ammonium citrate solution.

as a result of contact with an aqueous solution. Such ageing processes usually result in a decrease of pH toward neutral [17].

It is also seen from Table 4 that in experiments 7 and 8, the final ash weight (g DM) is higher than the initial value. This is due to a large amount of residual citrate in the ash, increasing the total weight. For all other experiments, however, an increasing weight loss in the ashes is observed with increasing remediation time. Such a weight loss, which is due to dissolution of mineral salts as, e.g. alkali chlorides [18,19], may result in an increased concentration (mg/kg DM) of the residual heavy metals in the ashes, even if a considerable amount (in mg) has actually been removed from the ashes during the remediation process. In Fig. 4 the normalised metal concentration in the ash ($C_{final}/C_{initial}$) in experiments 1–6 is plotted as a function of remediation time, illustrating this effect. After 70 days, for instance, Cr is concentrated by a factor of approximately 1.6 compared to the initial concentration.

Since it is expected that the electrodialytic treatment has removed the most mobile fractions of the heavy metals, an increased concentration of certain heavy metals in the treated ash residue is however not expected to pose a higher risk of leaching if the ash residual is finally deposed of. And the total leaching potential is anyhow reduced due to the reduced quantities of solid residue to be deposed of.
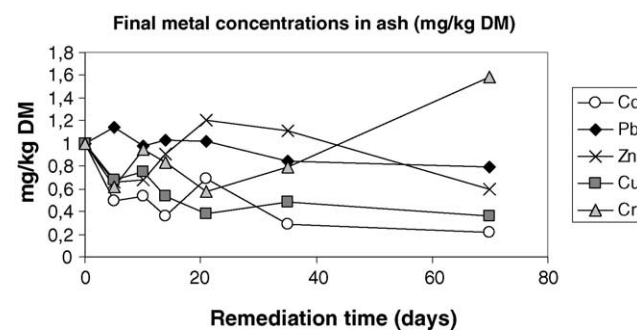

Fig. 4. Normalised metal concentrations ($C_{final}/C_{initial}$) in the ash residues as a function of remediation time (experiments 1–6).

5. Conclusion

Electrodialytic remediation using ammonium citrate as an assisting agent has shown good potential as a method for removal of heavy metals from MSWI fly ashes. The removal efficiency was improved significantly when using electrodialytic remediation compared to simple batch extraction experiments.

After 70 days of electrodialytic remediation, 86% Cd, 20% Pb, 62% Zn, 81% Cu and 44% Cr was removed from 75 g MSWI fly ash using 0.25 M ammonium citrate solution as an assisting agent.

Acknowledgements

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