Effects of Chlorides and Sulphates on Heavy Metal Leaching from Mortar with Raw and Electrodialytically Treated MSWI Fly Ash

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Abstract

Municipal solid waste incineration (MSWI) fly ash could be used as supplementary cementitious material in cement-based materials. However, heavy metal leaching, such as Cd, Cr, Cu, Pb and Zn, both from the MSWI fly ash and cement-based materials containing MSWI fly ash, remains a persistent obstacle. Here, an up-scaled electrodialytic treatment was used as a pre-treatment to remove heavy metals from MSWI fly ash before using the fly ash in mortar. Mortar samples with 10 wt% replacement of cement with either raw or electrodialytically treated MSWI fly ash were subjected to monolithic (in-use scenario) and crushed mortar (end-of-life scenario) leaching tests. The environmental conditions (e.g., exposure to chlorides or sulfates) at the surface of cement-based materials can affect leaching. Acidified H₂O, NaCl or Na₂SO₄ solutions were, therefore, used for the leaching tests. Up to 80% heavy metal removal by the up-scaled electrodialytic pre-treatment was feasible. Regulatory limits for disposing of the MSWI fly ash in non-hazardous waste landfills were exceeded, even if the electrodialytic treatment removed heavy metals. However, leaching from monolithic mortar samples complied with the regulatory limits, while Cr leaching exceeded the regulatory limits for all crushed mortar samples when using NaCl or Na₂SO₄. Both NaCl and Na₂SO₄ generally increased the heavy metal leaching yield from fly ash and mortar compared to leaching with acidified H₂O. The results of the study suggest that environmental conditions should be taken into account when assessing leaching from cement-based materials with MSWI fly ash.

Graphical Abstract

Keywords Electrokinetic remediation · Bench-scale · Leaching · Cement-based materials · Environmental factors

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Statement of Novelty

MSWI fly ash is considered hazardous waste due to a high content of leachable metals and salts. We used an electro-dialytic process to remove and reduce the leaching of metals from fly ash before use in mortars. Typically, studies where secondary raw materials replace cement in mortars apply standard leaching tests (TCLP and EN-12457) when evaluating metal leaching from the mortars. By performing leaching tests representing exposure to different external environmental conditions and service life scenarios of mortar, we provide an in-depth look at how metals leach from mortars with raw and electro-dialytically treated MSWI fly ash.

Introduction

Cement is regarded as the second most used material globally after water and the most produced material in terms of mass [1]. The production of ordinary Portland cement clinker represents a significant environmental issue due to high CO₂ emissions. It has been estimated that cement production accounted for 8% of the global anthropogenic CO₂ emissions in 2015 [2]. A well-established strategy to reduce CO₂ emissions is to partly replace cement clinker with supplementary cementitious materials (SCMs) [1]. However, limited supplies of commonly used SCMs necessitates the identification of new SCMs.

Municipal solid waste incineration (MSWI) is a preferred waste management solution in many countries, reducing the non-recyclable waste volume while producing energy. However, MSWI results in hazardous fly ash residues that are disposed of. Disposing MSWI fly ash may have long term environmental consequences due to contaminants leaching, e.g. toxic heavy metals [3], and results in the loss of any valuable metals in the residues.

Therefore, a treatment process that can reduce MSWI fly ash's hazardousness, recover any valuable metals, and enable the treated ash to replace cement clinker partially is of interest. Several treatment methods (e.g. solidification/stabilisation, separation/extraction or thermal treatments) have been investigated previously [4]. Cement solidification has been shown to effectively fix the heavy metals in untreated MSWI fly ash, but has a negative impact on the mechanical strength of the hydrated cement [4]. Thermal treatments can solidify the heavy metals in the MSWI fly ash [4] and if used in cement-based materials, has a positive impact on the mechanical strength [5–7]. However, both methods result in the loss of metals as a valuable resource, as the metals are not extracted by these methods.

It would, therefore, be advantageous to use a treatment for MSWI fly ash that extracts and recovers the metals, and that also has a positive impact on the treated MSWI fly ash if used in cement-based materials. One of the methods that can remove the heavy metals [8] and potentially enable MSWI fly ash for use in cement-based materials [9, 10] is electro-dialytic remediation (EDR). Electro-dialytic remediation uses a direct current to remove metals from a suspension of a contaminated material into electrolyte solutions separated from the suspension by ion-exchange membranes (anion, cation). The process acidifies the suspension due to water splitting ($\text{H}_2\text{O} \rightarrow \text{H}^+ + \text{OH}^-$) at the anion exchange membrane [11], releasing the metals in the material as ions that can migrate in the electric field. During remediation, soluble chlorides can migrate towards the anode as Cl^- and form toxic chlorine gasses if present [12]. A typical laboratory-scale EDR cell (Fig. 1) consists of three compartments. Compartments I and III are filled with a circulating electrolyte solution (0.01 M NaNO_3 adjusted to a $\text{pH} < 2$ with HNO_3) and contain the anode and cathode, respectively. Compartment II contains the suspension, and is kept stirred.

Electro-dialytic remediation has been used in lab-scale to remove toxic metals from MSWI fly ash [12–15], investigating the method's efficiency and potential improvements. However, for EDR to be a viable treatment process for metal removal and improving the properties of MSWI fly ash residues, the process needs to be up-scalable while retaining its efficiency. Efforts have been made in up-scaling the technology to treat MSWI fly ash at bench-scale [16, 17] and pilot-scale [18, 19] based on the set-up principles used for the electro-dialytic treatment of liquid solutions. However, the purpose of the up-scaled set-ups used in the previous studies was to remove the easily mobile parts of toxic metals from the MSWI fly ash and reduce the potential leaching instead of investigating the metals' total recovery from the fly ash (metal removals were typically below 5% [16]). To further valorise MSWI fly ash and test the recoverable content of

Fig. 1 Principal sketch of the electro-dialytic remediation set-up. AN anion exchange membrane, CAT cation exchange membrane, as presented in [13]

valuable metals, the total content of recoverable metals using an up-scaled EDR process should be investigated.

Although EDR can remove metals from MSWI fly ash, metal leaching may still occur after treatment. Furthermore, if the treated MSWI fly ash partially replaces cement in cement-based material, metals could potentially leach from the cement-based material. Therefore, the content of metals leaching from the EDR treated MSWI fly ash and cement-based materials with EDR treated MSWI fly ash should be investigated. Elements that can potentially leach from MSWI fly ash are typically studied through batch leaching methods with deionised water [20] or the toxic characteristic leaching procedure (TCLP) [21]. Leaching characteristics of cement-based materials containing raw or treated MSWI fly ash have been investigated for both monolithic and crushed samples, e.g. [9, 22–25]. Typically, these leaching tests are performed using the same leaching tests for the raw materials, with distilled/deionised water [26–28] or the TCLP test [21]. However, the typically used tests to assess leaching from cement-based materials do not consider the effect that environmental conditions may have on leaching. Although cement-based materials effectively retain metals through chemical bonding and physical entrapment [23, 24] the environmental conditions at the material's surface can affect leaching [24, 25]. Müllauer et al. [25] investigated the leaching yield from concrete containing coal incineration fly ash. They determined that environmental factors such as chloride (contact with deicing salts or the ocean) and sulphate exposure (ocean water, groundwater, or soil contact) increased Ba, Cr, Sr, and V leaching [25]. Furthermore, they surmised that standard leaching tests do not consider the environmental conditions, potentially leading to a wrong assessment, when concrete contains higher heavy metal content due to secondary fuels in cement production or industrial by-product SCMs [25]. The effects of groundwater, sea water and acid rain on metal leaching from cement products containing hazardous waste, showed highest As, Co, Cr, Ni and V leaching, when leaching simulating sea water was applied [29]. When SO_4^{2-} penetrate cement-based materials, expansive gypsum and ettringite forms, while $\text{Ca}(\text{OH})_2$, AFm (e.g. monosulphate), and the C–S–H phase tends to

dissolve [24]. Leaching with simulated sea water showed removal of ettringite by SEM images, from cement mortars [29]. This exposure to Cl^- may lead to Friedel's salt formation at the expense of the AFt (such as ettringite) and AFm phases. These phase changes are expected to change the solubility of both major and trace elements, as well as leading to a lower mechanical strength [29]. Therefore, before MSWI fly ash is used in cement-based materials, the effect various environmental conditions have on these materials should be investigated. This includes both during the materials use and at end-of-life.

The objectives of the present study were, therefore, (1) to further evaluate the potential of electrodialytically treating MSWI fly ash by up-scaling the process and investigating the total metal recovery percentage and leaching reduction and (2) to determine the effect environmental conditions have on cement-based materials containing MSWI fly ash and EDR treated MSWI fly ash, measuring the Cd, Cr, Cu, Pb and Zn leaching yield during exposure to acidified water, chlorides or sulfates.

Materials and Methods

Experimental Materials

The experimental MSWI fly ash residues used in this study were acquired in 2018 from three different incineration plants. Information about the incineration plants is summarised in Table 1. Six samples were used in this study; three untreated residues (AMA, NUU, RYA) and three EDR treated residues (EDR AMA, EDR NUU, EDR RYA). The AMA and NUU samples were pure fly ash samples (without air pollution control products) whereas the RYA sample also contained air pollution control products.

The residues used for the bench-scale EDR experiments were washed with deionised water in three stages at liquid to solid (L/S) ratio of 5 [31], as part of the treatment process, to remove easily soluble salts detrimental to the electro-dialytic process. The three residues water-soluble mass was

Table 1 Average operating conditions of the incineration plants during sampling

	Amager bakke	Nuuk Incineration	Ryaverket
Denotation	AMA	NUU	RYA
Furnace	Grate fired	Grate fired	Fluidised bed
Waste type	Household/Industrial	Household/Construction	Household/Industrial
Incineration capacity [ton/day]	1680	40	300
Incineration temp. [°C]	1025	1064	900
Flue gas filter type	Electrostatic	Electrostatic	Textile
Fly ash [ton/day]	14.8	0.3	8.9

As shown in [30]

Table 2 Toxic metal concentration (mg/kg) in CEM I cement and quartz

	Pb	Zn	Cd	Cu	Cr
CEM I	17	180	0.4	140	33
Quartz	1.2	6.2	0.0	0.4	0.4

Fig. 2 Principal sketch of the EDR bench-scale set-up. Anode compartment (I), suspension chamber (II) and cathode compartment (III)

determined to be 56, 38 and 28 wt% for AMA, NUU and RYA, respectively [30].

The cement used was a CEM I 52.5 N (MS/LA/ ≤ 2) with a bulk and absolute density of 1.1 and 3.2 g/cm³, respectively. Portland cement contains toxic metals in varying concentrations due to the raw materials used during cement production. The metals are either bound during cement hydration or dissolved in the pore solution [32]. M8 grade inert quartz with 99.4% SiO₂ was used as the inert material for cement replacement. CEN standard sand was used for the mortars. Table 2 contains the toxic metal concentration of the cement and quartz used. The cement was treated with Fe(II) to reduce Cr(VI) to Cr(III) at the cement production plant.

Bench-Scale Electrodialytic Remediation

Three electrochemical treatment experiments, one with each residue, were performed using the bench-scale experimental set-up shown in Fig. 2. The bench-scale set-up was built in a PVC plastic container (60 × 40 × 32 cm³). The suspension was a mixture of residue and distilled water at L/S 10: first, 27 L deionised water was added to the suspension chamber (II) and then a slurry of 3 L deionised water and 3 kg residue was gradually added. The residue was kept suspended

to avoid sedimentation in the container by three propellers connected to Vos 14 overhead stirrers from VWR. The anode and cathode were placed in electrode units (I and III), plastic boxes with circulating electrolytes. The electrolytes were circulated using Plastomec Magnetic Pumps. The unit's side facing the suspension contained a 27 × 37 cm² anion exchange membrane (20 SZRA B02249) or cation exchange membrane (CR67 HUY N12116B) from Ionics. Each electrode unit had a separate electrolyte circulating system (5 L of 0.01 M NaNO₃ adjusted to a pH < 2 with HNO₃) and one electrode. The electrodes were platinum-coated titanium meshes (4 × 20 cm²) from durAnode®. A power supply (Blanko- Model Q J-3003C III) maintained a constant current of 1 A. The applied current and membrane area of ~ 1000 cm² corresponded to a current density of 1 mA/cm². The voltage varied between 5 and 18 V during the experiments depending on the residue. Intermittently the voltage peaked at 32.1 V.

The suspension chamber's pH was measured daily by Radiometer electrodes. The pH in the catholyte was adjusted daily during the experiments to pH ~ 2 with 7 M HNO₃. The experiments lasted 28 days, and the current applied was equivalent to 800 C/g ash.

The suspension was filtered through 45 µm filters at the end of each experiment. The treated ash was then dried at 50 °C and crushed. Afterwards, the toxic metal content was measured with X-ray fluorescence (XRF) analysis. The membranes and stirrer were submerged in 1 M HNO₃ and the electrodes in 5 M HNO₃ to release any accumulated metals. The toxic metal accumulations were then analysed with inductively coupled plasma—optical emission spectrometry (ICP-OES).

The wt% distribution of metals removed from the residue, e.g. in the suspension, catholyte or anolyte, was calculated based on the residues initial metal concentration before the EDR treatment, measured with XRF.

During the EDR experiments, liquid flowed between the electrolyte and suspension chambers, resulting in an unbalanced system. Additional 0.01 M NaNO₃ adjusted to a pH < 2 with HNO₃ was added to the electrolytes, while additional deionised water was added to the suspension chamber during the experiments. However, this did not influence the pH of the fly ash suspensions. In the experiment with the AMA residue, one of the pumps failed after eight days and was replaced.

Treated Ash Characterisation

The elements in the EDR treated MSWI fly ash were analysed with XRF analysis. An external laboratory performed the XRF analysis, using samples ground to a particle size below 200 µm and a Pd-tube equipped SPECTRO GmbH X-LAB 2000.

The content of Al, Ca, Fe, K, Mg, Na, P, S and Si were back-calculated as oxides, while Cl and the studied metals Cd, Cr, Cu, Pb and Zn are presented as-is. Furthermore, the mineralogy and phase composition were analysed using X-ray diffraction (XRD) and thermogravimetric (TG) analysis. The XRD analysis was performed on back-loaded samples using a Pan-Alytical X'pert PRO Θ – Θ System (2 wt% detection limit). The ICDD PDF4 database was used for peak and phase identification. The TG analysis was performed using a NETZSCH STA 449 F3 Jupiter, aluminium oxide sample holders and 50 ml/min of nitrogen as the purge gas. The samples were heated from 29 to 900 °C at 10 °C/min. The TG analysis results are included as the 1st derivative of the thermogravimetric graph (DTG).

Leaching Experiments

Leaching experiments were performed on the raw and treated residues and mortar cubes made with 10 wt% residue substituting cement. In addition to the mortar cubes with residue, two reference cubes were made, one without cement replacement and one with 10 wt% inert M8 quartz filler substituting the cement.

The leaching experiments were performed using three different solutions, based on Müllauer et al. [25] and the commonly used Dutch diffusion test NEN 7345 [28, 33–36].

1. NaCl dissolved in distilled water at a concentration of 30 g/L. Simulates deicing salt for concrete highways in moderate and cold climates [25].
2. Na₂SO₄ dissolved in distilled water at a concentration of 30 g/L. It corresponds to the upper limit of exposure class XA2 according to EN 206 and is the most often concentration used to test resistance to sulphate attack [25].
3. Distilled (DI) water acidified to pH 4 using 1 M HNO₃ [28, 33–36].

Figure 3 shows an overview flow chart of the leaching experiments performed.

Fly Ash

Leaching experiments with the untreated and treated residues were performed according to the batch leaching test described in DS/EN 12,457–2 [37], although modified to study the potential influence of environmental factors on leaching using the solvents 1–3. The experiments were performed by mixing 5 g of ash with 50 mL of solvent (L/S ratio of 10). The mixture was shaken for 24 h at 195 rpm, then filtered using 0.45 µm syringe filters before pH and metal analysis with a Radiometer electrode and ICP-OES, respectively. The experiments were performed in duplicate.

Mortar Cubes

The mortar cube specimens were prepared according to EN 196–1 [38]. A total of sixteen cubes were prepared, eight for measuring leaching from crushed mortar and eight for measuring monolithic (in-use scenario) leaching. Mortar cubes with 10 wt% cement replaced by either fly ash or inert quartz (admixtures) were a mix of 45 g admixture, 405 g cement, 1350 g CEN standard sand and 225 g distilled water. The 100 wt% cement cubes contained 450 g cement, 1350 g CEN standard sand, and 225 g distilled water. The mixtures were vibrated and moulded in 5 × 5 × 5 cm³ cubical fibre wood moulds at 50 Hz for 2 min after mixing the ingredients. The cubes were demolded after 20–24 h of curing at > 90% relative humidity and 20 °C and stored to hydrate further for 28 days in desiccators with 90% relative humidity at 20 °C.

The monolithic leaching experiments were performed using a modified version of the Dutch leaching test NEN 7345 described in [35] by submerging the cubes in one of the solvents 1–3 at an L/S ratio of 4. The submerged cubes were stored in 1.4 L sealed containers for 70 days at ambient temperatures. The cubes were placed flat on the containers' bottom, exposing five of the cubes' sides. After 6 h, 24 h, 54 h, 4 days, 9 days, 16 days, 36 days and 70 days, 20 mL of the effluent was extracted from the containers and replaced with 20 mL of the fresh solution. The extracted effluent's pH

Fig. 3 Flow chart of the performed leaching experiments

and metal content were then measured with a Radiometer electrode and ICP-OES, respectively. The measured concentration of metals was corrected to account for the initial concentration of metals in the solvents. One experiment with crushed and monolithic mortar was performed for each of the raw and EDR treated residues.

Leaching experiments on crushed (end-of-life scenario) mortar cube specimens were performed according to DS/EN 12,457–1 [39]. The mortar cubes were crushed to a particle size below 4 mm with a hammer after 28 days of curing. The experiments were performed by mixing 175 g of the crushed specimen with solutions 1–3 at an L/S ratio of 2. The volume of solution used was adjusted for each specimen to account for each crushed specimen's water content.

Results and Discussion

Bench-Scale Electrodialytic Remediation

Experimental Conditions

The pH development in the suspension chamber during the bench-scale electrodialytic remediation experiments is shown in Fig. 4. In the electrodialytic remediation experiments, the suspensions' final pH was 1.5 for AMA and 5.5 for NUU and RYA.

The differences in final suspension pH between the residues may be attributed to their initial pH differences (pH 6 for AMA and pH 12 for NUU and RYA) and their buffer

capacities. Therefore, a longer remediation time would be required for NUU and RYA to reach an acidic pH similar to AMA. However, after 15 days, AMA's pH rate of decrease changed, stabilising at a pH of 1.5. The acidification occurred slower during the bench-scale experiments than what has been observed at laboratory scale. This may be due to the L/S ratio differences between the bench-scale and lab-scale experiments or the difference between the ion exchange membrane area and current compared to the amount of residue remediated.

Metal Removal

The distribution of Cd, Cu, Cr, Pb and Zn after the three electrodialytic experiments is shown in Fig. 5. The metal removal order was $\text{Cu} > \text{Zn} > \text{Cd} > \text{Cr} > \text{Pb}$ for AMA, while, $\text{Cd} > \text{Zn} > \text{Cr} > (\text{Cu}, \text{Pb})$ for NUU and RYA. For AMA, the highest removal was 80 wt% Cu, while the highest removal for NUU and RYA were 20 and 10 wt% Cd, respectively. The release of metals as ions from MSWI fly ash has been shown to depend on pH [15, 40]. HNO_3 batch leaching experiments with MSWI fly ash [15, 40] indicate that Cd leaches below pH 8, Zn below pH 7, Cu below pH 5, while Pb and Cr leach below pH 4. Therefore, a high metal removal could be expected in the experiment with AMA. The differences in metal removal between the experiments correlate with the differences in pH. A final pH of 5.5 was insufficient to remove high concentrations of metals from NUU and RYA. Due to the higher pH of NUU and RYA and their buffer capacities, more than 28 days of remediation would be required to treat more alkaline MSWI fly ashes under the current bench-scale experimental conditions. The initial pH differences suggest that adjusting the residues' pH before the EDR treatment may shorten the time needed for the treatment. However, Jensen et al. [17] found that Pb and Zn leaching from EDR treated air pollution control residue was higher when the residue was pre-washed with acid compared with water before the EDR experiments.

At the acidic pH reached in the AMA experiment, the valency of metals in a pure acidic and aerated solution could be expected to be Cd^{2+} , Zn^{2+} , Pb^{2+} and Cu^{2+} and Cr^{3+} [41], suggesting a release to the suspension and subsequent removal to the catholyte, confirming the release of positively charged metals. However, most metals released from AMA remained in the suspension instead of migrating to the electrodes. The experiment with AMA was not hydraulically stable. As previously mentioned, the pump failed and a significant amount of liquid flowed from the electrolytes to the suspension compartment. This experiment's metal distribution indicates that the metals were transported with the liquid instead of retained in the electrolytes.

Fig. 4 Suspension chamber pH during bench-scale EDR experiments

Fig. 5 Wt% of toxic metals after bench-scale electrochemical experiments in the treated ash (ash), electrolyte solutions (catholyte/anolyte), chamber II suspension and on the suspension stirrers (suspension + stirrer) and on the anode or cathode

The bench-scale experiments' experimental performance showed that further developments and adjustments of the experimental set-up and conditions are needed (1) to obtain hydraulically stable experiments and (2) to achieve faster acidification and higher metal removal. This development is essential to properly assess technical concerns that can increase the technological readiness level (TRL) of electrochemical remediation for MSWI fly ash treatment and enable full-scale treatment [42].

Characteristics of Treated Fly Ash

The element concentrations of the raw [30] and EDR treated residues are included in Table 3. The EDR treatment reduced

the residues' K, Na, and Cl concentrations due to the dissolution of easily soluble salts, decreasing the mass of the residues after the experiments. Even if metals were removed from the residues (Fig. 5), the EDR treated AMA was the only sample with reduced toxic metal concentrations (Cd, Cu, Zn). The Cr and Pb concentration in AMA were concentrated. The EDR treatment also concentrated the metals in NUU and RYA, except Pb, while reducing the residues' Ca content. The Ca concentration in AMA increased after the treatment, which may be attributed to an insoluble Ca content. Furthermore, the EDR treatment concentrated the Si, Al and Fe in the residues, which would be advantageous when used as an SCM in cement-based materials.

Table 3 Calculated oxides composition of the raw residue, as presented in [30], and EDR treated residues, with trace elements, measured with XRF analysis and pH

		AMA	EDR AMA	NUU	EDR NUU	RYA	EDR RYA
pH	[–]	6.5	4.1	12	7.7	12	7.2
Al ₂ O ₃	[Wt%]	1.4	2.3	4.6	9.6	4.7	5.3
CaO	[Wt%]	13	28	43	29	43	31
Fe ₂ O ₃	[Wt%]	1.0	2.4	0.9	1.4	1.8	2.7
K ₂ O	[Wt%]	14	1.0	12	0.4	2.7	0.7
MgO	[Wt%]	0.6	0.4	1.4	1.3	1.8	2.2
Na ₂ O	[Wt%]	7.4	0.7	22	–	7.0	–
P ₂ O ₅	[Wt%]	2.6	2.1	1.8	2.7	2.2	3.7
SO ₃	[Wt%]	34	30	8.9	9.7	8.9	8.7
SiO ₂	[Wt%]	4.6	14	6.1	14	6.4	12
Cl	[Wt%]	2.8	0.1	24	0.1	13	0.2
Cd	[mg/kg]	480	60	420	460	80	100
Cr	[mg/kg]	300	700	800	1100	400	600
Cu	[mg/kg]	1400	520	1300	2000	7900	10,200
Pb	[mg/kg]	12,000	31,000	3,200	3200	3400	3400
Zn	[mg/kg]	55,000	26,000	33,000	41,000	6600	9100

(–) below the detection limit

Fig. 6 XRD diffractograms of the raw and EDR treated residues

Figure 6 depicts the XRD diffractograms of the raw and EDR treated residues. The XRD diffractograms showed that the mineral phases of AMA were CaSO_4 , and CaAl_2O_4 , NUU contained NaCl , KCl , CaCO_3 , CaSO_4 and CaO , while RYA contained CaClOH , CaSO_4 , NaCl , KCl and CaCO_3 . After the EDR treatment, Cl containing mineral phases could no longer be identified. Instead, EDR AMA contained $\text{CaSO}_4 \cdot 0.5\text{H}_2\text{O}$, CaSO_4 and PbSO_4 , while EDR NUU and RYA contained $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ and CaCO_3 . EDR AMA might not contain CaSO_4 at all as the peaks for CaSO_4 overlap with

$\text{CaSO}_4 \cdot 0.5\text{H}_2\text{O}$. The EDR AMA contained PbSO_4 , which would account for the increased Pb concentration due to precipitation.

Figure 7 depicts the DTG graphs of the raw and EDR treated residues. In a previous study by Ebert et al. [30] it was determined that raw NUU and RYA contained CaCO_3 (mass loss at 650–700 °C). The three samples potentially also contained sulphate phases that started decomposing above 700 °C. The mass losses observed between 0 and 500 °C for NUU and RYA indicate water loss from several

Fig. 7 DTG patterns of the raw and EDR treated residues

Table 4 Resulting pH in the raw and treated residue batch leaching experiments

	DI _{pH 4}	NaCl	Na ₂ SO ₄
AMA	6.6	6.4	6.6
EDR AMA	5.1	4.9	5.3
RYA	11.5	11.6	11.5
EDR RYA	8.3	8.3	8.8
NUU	11.9	11.8	12.0
EDR NUU	9.1	9.1	9.9

different phases not present in AMA. The DTG curves of the EDR treated residues suggest that the samples still contain CaCO_3 and various sulphate phases, as supported by the XRD graphs. However, EDR NUU did not show the same mass loss after 700 °C as raw NUU. Furthermore, the mass losses between 0 and 500 °C are no longer observable. Instead, the three samples show a mass loss between 100 and 150 °C, corresponding to $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ [43]. The mass loss size suggests that NUU and RYA contained a higher concentration of $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ than AMA, despite containing less sulphur. However, The XRD analysis showed that AMA contained $\text{CaSO}_4 \cdot 0.5\text{H}_2\text{O}$ instead of $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$, suggesting that the smaller mass loss could be due to less bound H_2O per CaSO_4 . EDR NUU and EDR RYA also had a mass loss at 70 °C and 300 °C, which was not as easily identified that could be due to monosulphate [43]. After the EDR treatment, the irremovable Ca then remained as CaCO_3 or as either unhydrated or hydrated CaSO_4 .

Leaching Experiments

Raw Materials

The pH of the batch leaching experiments with the raw and EDR treated residues are included in Table 4. The pH of the leachate varied between 6.4 and 12.0, depending on the sample. The batch leaching results with the raw and EDR treated residues are shown in Fig. 8. The figure includes the criteria for granular hazardous waste acceptable at landfills for non-hazardous waste [44]. The leaching yield of the raw residues exceeded the regulatory limit for landfilling as non-hazardous waste for some of the metals. Raw AMA leached more Cd, Cr, Pb and Zn, raw NUU leached more Cr and Pb, while raw RYA leached more Cu and Pb than the regulatory limits. Treating the residues with electrodiolytic remediation generally reduced the Cd, Cu, Pb and Zn leaching yield. However, it increased the Cr leaching yield, although it depended on the residue and the leaching solution. The leaching yield from EDR AMA was still above the regulatory limits, except for Cu leaching with distilled water adjusted to pH 4. However, the EDR treatment did increase the Cu leaching yield from AMA. For EDR NUU and EDR RYA, Cr was the only metal above the regulatory limit. The observed differences in leaching concentrations may be due to the decreased pH after the EDR treatment (Table 4).

In pure acidic or alkaline solutions within the water stability range, Zn and Cu are present as Zn^{2+} and Cu^{2+} below a pH of 7, while Cr is present as Cr^{3+} below a pH of 4 [41]. Pb is present as Pb^{2+} in the entire pH range [41]. In carbon dioxide containing solutions, Pb can be passivated as PbO_2 at a pH between 5 and 10 and as PbSO_4 at a pH between 0

Fig. 8 Residues leaching using distilled water adjusted to pH 4 with HNO₃, a 30 g/L NaCl solution and a 30 g/L NaCl solution. Included are the limit values from [44]

and 10 in sulphate containing solutions [41]. The residues' characterisation showed the presence of CaCO_3 and phases containing SO_4^{2-} , with PbSO_4 being identified in AMA. In a Cd–S–C–O–H system, Cd is present as Cd^{2+} below a pH of 8 [45]. Furthermore, Zn and Pb can be present as oxyanions at a pH above 11 [41]. This may explain the higher leaching yield from the raw residues than the reduced Pb and Zn yield from the treated residues.

The NaCl and Na_2SO_4 solutions effect on leaching is not easily discerned, as the leaching yield varied depending on the metal and sample. Generally, the NaCl and Na_2SO_4 solutions leached more Cd and Cr than the pH 4 adjusted solution. Adding extra Cl^- and SO_4^{2-} could result in additional soluble (e.g. metal-chloride complexes) or insoluble complexes (e.g., PbSO_4), increasing or decreasing the leaching yield. The increased leaching due to added NaCl and Na_2SO_4 appears to be higher for the EDR treated samples than the untreated, suggesting they could be more susceptible to

leaching during varying environmental conditions. NaCl or Na_2SO_4 did not generally affect whether the leaching yield was within the regulatory limits. However, they did increase the Cd yield from EDR NUU, Cr yield from RYA and Cu yield from EDR AMA to above the limit, compared to the pH 4 adjusted water.

Monolithic Leaching Test (in-Use Scenario)

Monolithic leaching tests represent an in-use scenario for the mortars and can be compared to leaching limits for construction materials. Table 5 shows the monolithic leaching experiments' results as the maximum leaching measured between 0–4 days, 4–16 days and 16–70 days. The leaching results are compared to the regulatory limit for total leachability specified in the Dutch Building Decree [46]. After 6 h to 70 days, the pH was between pH 11.4 and pH 12.5 for all experiments performed. Carbonation of cement-based

Table 5 Maximum monolithic leaching (mg/m^2 surface) between 0–4 days, 4–16 days and 16–70 days

	Cr			Cu			Pb			Zn		
	0–4	4–16	16–70	0–4	4–16	16–70	0–4	4–16	16–70	0–4	4–16	16–70
DI_{pH4}												
CEM I	–	–	–	–	–	–	–	–	–	–	–	–
QUA	–	–	–	–	–	–	–	–	–	–	–	–
AMA	0.76	0.83	1.1	–	0.68	0.81	–	1.8	3.4	1.4	2.2	4.0
EDR AMA	0.78	0.84	0.78	–	–	–	1.8	8.0	14	–	–	–
RYA	0.66	0.75	0.72	–	0.87	–	–	–	–	–	–	–
EDR RYA	–	–	–	–	–	–	–	–	–	–	–	–
NUU	–	–	1.7	–	–	–	–	–	–	1.4	5.4	1.5
EDR NUU	–	–	–	–	–	–	–	–	–	–	–	1.0
NaCl												
CEM I	2.0	5.8	9.2	–	–	–	–	–	–	–	–	–
QUA	2.3	5.2	10	–	–	–	–	–	–	–	–	–
AMA	2.8	6.0	9.8	–	–	–	1.9	3.7	5.4	1.4	3.2	5.4
EDR AMA	1.1	3.4	6.4	–	0.62	–	4.9	12	19	–	1.6	–
RYA	2.3	7.1	13	–	–	–	–	0.83	–	12	–	–
EDR RYA	1.0	4.0	7.6	0.36	0.43	0.59	–	–	1.4	–	0.84	–
NUU	3.3	7.4	12	0.94	1.2	1.2	–	–	–	0.8	2.2	2.3
EDR NUU	2.4	5.6	10	0.86	1.1	1.7	–	–	–	1.3	1.9	2.3
Na_2SO_4												
CEM I	1.9	3.2	4.7	–	–	–	–	–	–	–	–	–
QUA	1.6	3.8	4.3	–	–	–	–	–	–	–	–	–
AMA	3.9	5.7	7.6	–	–	–	1.5	2.4	3.1	3.4	9.1	6.7
EDR AMA	1.2	3.2	5.4	–	0.40	0.40	4.2	7.3	12	2.6	0.93	0.91
RYA	1.3	3.9	7.4	–	0.68	0.46	–	–	0.80	0.73	2.1	–
EDR RYA	1.2	3.1	3.9	1.1	2.1	1.6	0.75	0.53	1.4	0.87	1.3	0.25
NUU	3.4	5.8	8.3	0.97	0.95	1.1	–	–	–	1.8	5.3	4.7
EDR NUU	2.2	4.2	6.3	0.63	0.91	1.1	–	–	–	–	–	1.1
Limit		1500			540	1275	2100					

(–) below detection limit or no higher than the initial solution concentration. The regulation limit values are from the Dutch building material degree, as presented in [46]

materials can affect leaching by reducing the pore solution pH, changing the porosity and pore size distribution and causing the dissolution of the hydration products [32]. Uncontrolled uptake of CO_2 has been shown to have a negligible effect on the leaching conditions in the NEN 7345 test [28]. Therefore, any observed increase in leaching yield is attributed to the effect of the solutions used. However, using MSWI fly ash may result in swelling and cracking in a mortar bar [47] and may make the mortar more susceptible to Cl^- and SO_4^{2-} penetration, resulting in an increased leaching yield. Cd leaching was below the measurement limit of the ICP-OES. This may be attributed to Cd's strong incorporation into the cement matrix due to its similar ionic radius to Ca^{2+} [29, 47–49]. It may also have been encapsulated as $\text{Cd}(\text{OH})_2$ by C–S–H and $\text{Ca}(\text{OH})_2$ [50].

No measurable Cu, Pb and Zn leached from the reference mortar cubes. Only Cr leached from the reference mortar cubes, when using the NaCl or Na_2SO_4 solutions and more Cr leached in the NaCl solution than the Na_2SO_4 solution (9.2 mg/m^2 and 10 mg/m^2 compared to 4.7 mg/m^2 and 4.3 mg/m^2 after 70 days). This suggests that the diffusion of Cl^- into the cement matrix has a more significant effect on Cr leaching than SO_4^{2-} . It has been well documented that Cr may be incorporated into the AFt and AFm through Al^{3+} and SO_4^{2-} substitution [22, 32, 48, 51, 52]. However, it may also be chemically incorporated into all other cement hydration products [47]. The higher Cr leaching yield with NaCl may be attributed to a higher Cr concentration in the AFt phase, which would fit with Cr(III) substituting Al^{3+} in ettringite. The Cr is subsequently released due to AFt dissolution.

Leaching with NaCl or Na_2SO_4 resulted in a higher Cr yield from the cubes with MSWI fly ash than the reference cubes, potentially due to the higher Cr content in the residues. This was especially prevalent for NUU and RYA. Despite the higher Cr content in the cubes with MSWI fly ash, the leaching behaviour was similar to the 100 wt% CEM I and quartz cubes increasing over time. The Cu leaching results showed the highest dependence on residue and leaching solution; as NaCl yielded a higher Cu leaching from NUU while Na_2SO_4 yielded a higher Cu leaching from RYA. For AMA, using either NaCl or Na_2SO_4 reduced Cu leaching compared to the acidified water. NaCl had a similar effect on the mortar cube with RYA, suggesting which phase Cu is predominantly bound in may depend on the residue used. Leaching with NaCl increased the Pb yield more than Na_2SO_4 , while Na_2SO_4 increased the Zn yield more than NaCl. Pb and Zn can replace Ca^{2+} in the AFt or C–S–H phase or be adsorbed to the C–S–H phase [24, 32, 48, 51, 53]. The higher Pb yield with NaCl may be attributed to a higher concentration in the C–S–H phase, while the Zn could have been bound in the AFt phase, similarly to Cr.

The efficiency of EDR treating MSWI fly ash in reducing the leaching yield depended on the metal studied. The EDR

treatment generally reduced the Cr and Zn leaching yields from the mortar cubes, while the Cu and Pb leaching yields increased. This was contrary to the residue batch leaching experiments, where an increase in Cr and a reduction in Pb leaching yield were observed. The Pb leaching yield from mortars with AMA significantly increased after the EDR treatment, even with the pH 4 adjusted solution and represented an issue with the EDR treatment that should be investigated further.

Similarly to the observations made by Müllauer et al. [25], metal leaching from monolithic mortar cubes containing MSWI fly ash is affected by the surface environmental conditions. Despite the increase in leaching yields observed in this study, the leaching yields were as much as a factor 1000 lower than the regulatory limits [46] from all mortar cubes. This shows that partially substituting cement with the EDR treated residues is feasible in terms of leaching, during the cement-based materials lifetime, despite the environmental conditions and may be attributed to how effectively heavy metals are retained in cement-based materials [23].

Crushed Mortar (End-of-Life Scenario)

The concentration of Cr, Pb and Zn that leached from crushed mortar is shown in Fig. 9, representing an end-of-life scenario. Leached concentrations can be compared to the leaching limits for reusing inert construction and civil engineering waste in Denmark, where Category 3 covers more restricted use than Category 2 [54]. The pH of leachate in the experiments was 12.6–12.9. Similarly to the monolithic tests, the Cd leaching yield was below the detection limit of the ICP-OES measurement. Cu leaching did occur from the various crushed mortars but was always below the Category 2 limit. Furthermore, the Cu yield from the crushed mortars with MSWI fly ash showed little to no variation compared to the reference mortars.

The crushed reference mortars exceeded the Category 2 limit for Cr leaching when using the pH 4 adjusted solution but remained within Category 3. However, the Category 3 limit was exceeded when using the NaCl and Na_2SO_4 solutions. The crushed 100 wt% CEM I mortar also leached more Pb than the Category 3 limit when using the pH 4 adjusted solution, while NaCl or Na_2SO_4 did not affect the yield. However, no measurable Pb leaching was observed from the quartz mortar, and the reference mortars did not leach Zn.

The results show that Cr, Pb and Zn leaching from the crushed mortars containing MSWI fly ash were generally below the Category 2 or 3 limits when using the pH 4 adjusted solution, except for mortar with AMA. Crushed mortar with AMA exceeded the Category 3 limit for Pb leaching and the Category 2 limit for Zn leaching, which may be attributed to this residues' higher concentration of Pb

Fig. 9 Crushed mortar leaching using distilled water adjusted to pH 4 with HNO₃, a 30 g/L NaCl solution and a 30 g/L Na₂SO₄ solution. Included are the upper limit for Category 3 (blue) and Category 2 (teal) for use of residues [54]

and Zn. Like the previous observations, leaching with NaCl or Na₂SO₄ increased the yield, especially for Cr and Zn. For Pb, the results varied from residue to residue. Contrary to the monolithic leaching results, Na₂SO₄ resulted in the highest Cr leaching.

Treating the residues with EDR reduced the Cr leaching yield from the crushed mortars for all three residues. EDR treatment especially reduced the Zn leaching yield for AMA. Contrarily, the EDR treatment increased the Pb yield, similarly to the monolithic leaching.

In general, NaCl and Na₂SO₄ resulted in a higher leaching yield than the pH 4 adjusted solution, further suggesting that when evaluating metal leaching from mortar containing

MSWI fly ash, varying environmental conditions should be considered. However, the effect of the environmental conditions appears to depend on the metal and the MSWI fly ash. A previous study with crushed mortar containing an MSWI fly ash treated with phosphation and calcination showed an increase in Cr and Pb leaching [55], suggesting that these metals might not be as efficiently bound in cement-based materials as the other metals. The effect of environmental factors on leaching was very apparent for Cr leaching from the crushed mortar. Cr leaching was within Category 3 for all samples when using the pH 4 adjusted solution. However, with NaCl and especially Na₂SO₄, the Cr yield was above the Category 3 limit for all mortar samples.

Conclusions

Treating MSWI fly ash with electrodialytic remediation at bench-scale to remove heavy metals was feasible if a low pH is reached, e.g. pH 1.5. The removal order was $\text{Cu} > \text{Zn} > \text{Cd} > \text{Cr} > \text{Pb}$, with close to 80 wt% Cu removal. The effect of simulated environmental factors by NaCl and Na_2SO_4 on leaching behaviour was tested on both fly ash and monolithic and crushed mortar with fly ash. The electrodialytic treatment generally reduced Zn, Pb, Cd and Cu's leaching while increasing Cr. However, an increase in the Cu leaching yield for treated AMA was observed.

The heavy metal leaching yield from monolithic mortar with MSWI fly ash, representing the in-use phase of the mortar, was higher for Cu, Pb, and Zn than references made with 100 wt% CEM I cement and 10% quartz. For Cr, leaching from the reference mortars was observed, attributed to the Cr content in cement. Higher leaching of Cr, Pb, and Zn by NaCl and Na_2SO_4 was also observed in the monolithic time-dependent leaching test. It may be attributed to the accelerated breakdown of the cement hydration phases over time. However, the monolithic leaching test showed that the leaching was within regulatory limits during the mortars service life when the raw and electrodialytically treated residue was used.

The leaching test on the crushed mortar, representing an end-of-life scenario, showed that Pb leaching from the AMA and EDR AMA sample exceeded the regulatory limit for reuse with all leaching solutions. In contrast, Cr leaching only exceeded the limits when the NaCl and Na_2SO_4 solutions were used. Heavy metal leaching from the other samples was below the limit values for reuse. For future studies, it is suggested to supplement the commonly used leaching tests using distilled water with other liquids that can simulate environmental factors to investigate the environmental performance further when using secondary resources such as MSWI fly ash for construction materials.

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Data Availability Data is available upon request to the corresponding author. Data is stored by the Technical University of Denmark.

Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Ethical Approval Not applicable for the manuscript.

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