



# Electrodialytic remediation of municipal solid waste incineration residues using different membranes



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## HIGHLIGHTS

- Municipal Solid Waste Incineration (MSWI) residues are common in western countries.
- Four membrane brands are compared to treat MSWI residues using electrodialysis.
- Element leaching is generally reduced regardless the membrane used.
- Leaching of some elements in the treated residues varies depending on the membrane.

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## ABSTRACT

In the present work, three different commercial membrane brands were used in an identical electro-dialytic cell setup and operating conditions, in order to reduce the leaching of metals and salt anions of two types of municipal solid waste incineration residues: air pollution control residues of a semi-dry flue-gas cleaning system and fly ashes from a plant with wet flue-gas cleaning system. The results showed a general reduction of the leaching in both residues after ED remediation. For the following elements, the leaching was found to be different after ED treatment depending on the membrane used, with statistical significance:

- Air pollution control residues of the semi-dry flue-gas cleaning system: Cr, Cu, Ni, Pb, Zn;
- Fly ashes from a plant with wet flue-gas cleaning system: Al, Ba, Cu, Ni, Zn, Cl, SO<sub>4</sub>.

Final leaching values for some elements and membranes, but not the majority, were below than those of certified coal fly ash (e.g. Al or Cr), a material which is commonly used in construction materials; at the same time, some of these values were reduced to below the Danish law thresholds on the use of contaminated soil in constructions. These results show the potential of ED as a technology to upgrade municipal solid waste incineration residues.

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## 1. Introduction

Worldwide municipal solid waste generation is expected to double by 2025 (Hoornweg and Bhada-Tata, 2012). Improving solid waste management is therefore an urgent priority. In many developed countries, a common practice is the incineration of solid waste, as it reduces the volume of waste by up to 90% (Hoornweg and Bhada-Tata, 2012) and allows energy recovery. Worldwide, approximately 120 million tonnes of waste are incinerated every year (Hoornweg and Bhada-Tata, 2012). This practice generates fly ashes (FA) and air pollution control (APC) residues, originating from

the treatment of the flue gas coming out of the combustion chamber, and constituting 1–5% of the original waste by mass (Sabbas et al., 2003). FA and APC residues are hazardous and contain several pollutants; among them, salts and heavy metals (Quina et al., 2008a).

Both residues can have different potential applications as substitution of other raw materials in production of construction materials (Ferreira et al., 2003). However, such use is not allowed in some countries like Denmark, where (as in many other western countries) FA and APC residues are normally deposited after treatment (Quina et al., 2008a). At the same time, huge amounts of virgin resources are used in the construction sector. An upgrade to a higher environmental standard (like a reduced metal and salts leaching), could make them suitable for substitution of raw materials in e.g. concrete production.

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Electrodialysis (ED) has been proven a promising technology for this purpose (Kirkelund et al., 2010; Jensen et al., 2010). ED is based on application of an electrical current to a solution or a solid suspension containing ions, forcing cations to move towards the cathode and anions towards the anode. Ion exchange membranes (IEMs), which are able to let chemical species pass depending on their charge, are strategically placed to separate cations and anions from an ion-rich compartment into different compartments. The ED setup used in this work can be seen in Fig. 1.

Previous research has focused on optimising different processing parameters for ED treatment of different materials such as contaminated soils and harbour sediments, municipal solid waste incineration (MSWI) FA/APC residues or sewage sludge ashes; important parameters being the liquid-to-solid ratio (L/S), duration of treatment, current intensity, pH as well as the number of cell compartments or the use of chemicals like  $\text{H}_2\text{SO}_4$  (Jensen et al., 2007a; Nystroem et al., 2005; Kirkelund et al., 2013; Guedes et al., 2016). The performance of different commercial IEMs has been compared in several ED processes like the denitrification of an aqueous solution (Ulises et al., 2011), the removal of organic salts from a fermentation broth (Gong et al., 2006), as well as the production of acid from waste pickle solutions (Akgemci et al., 2005). Their results show that the transport of the same ion through the different IEMs can vary substantially among the different brands: up to a threefold for  $\text{Na}^+$  and  $\text{NO}_3^-$ , up to a 1.3-fold for organic salts, and up to a fourfold for  $\text{Cl}^-$ . Two different ion exchange membrane (IEM) brands have been used in different studies of ED upgrading of MSWI APC residues and FA: Ionics (Kirkelund et al., 2013, 2015; Pedersen et al., 2003) and Neosepta (Kirkelund et al., 2010; Jensen et al., 2010, 2015). The influence of the IEM brand used for ED treatment of MSWI residues has, however, never been investigated.

To better understand the influence of the IEM type on upgrading of APC residues and other particular materials by ED, the present work assesses the performance of four IEM trademarks on the ED treatment of MSWI residues. All IEMs were tested for treatment of two common and different waste materials (MSWI FA and APC residues) in order to evaluate if the IEMs differently affect the MSWI residue characteristics after the ED treatment. The overall aim of the investigation was to compare the leachability changes of these residues, induced by the ED treatment, among the four different IEM brands.

## 2. Materials and methods

### 2.1. MSWI residues

Two different Danish MSWI residues were studied:

- I APC residues collected from a semi-dry (SD) flue-gas cleaning system from the MSWI plant REFA I/S. REFA I/S incinerates approximately 120,000 tonnes of household and industrial waste annually. SD were collected on 28th of June 2011 from line 3 after APC additives (slaked lime and activated carbon).
- II FA collected from the electrostatic precipitator in a wet flue-gas cleaning system from the MSWI plant Vestforbrænding I/S, which is the largest plant in Denmark, incinerating approximately 520,000 tonnes of household and industrial waste annually. FA was collected in week 50 in 2011 from the ash silo 5 prior to mixing with the wet scrubber sludge.

After sampling, both residues were dried at 40 °C. They were subsequently dry sieved through a 1 mm sieve, as performed in previous pilot experiments (Kirkelund et al., 2010; Jensen et al., 2010, 2015), to prevent clogging in the flow spacers when scaling

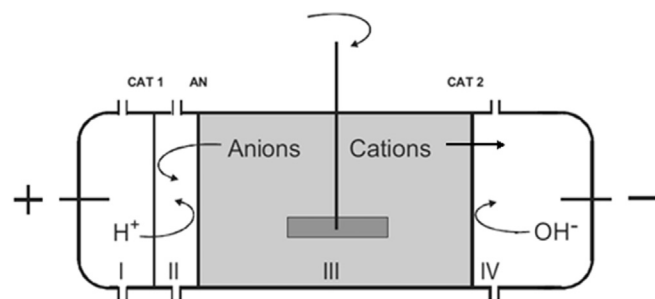


Fig. 1. Schematic view of a cell used for the ED treatment of both APC residues. AN: anion-exchange membrane; CAT1/CAT2: cation-exchange membranes.

up. The removed fraction represented a 1% of the total initial mass (Jensen et al., 2015).

### 2.2. Analytical methods

Mineralogy, the metal and water soluble salt concentrations and leaching were investigated for both treated and raw MSWI residues after drying them at 105 °C. Water solubility was measured only for the untreated residues. The targeted metals were: Al, As, Ba, Ca, Cd, Cu, Cr (total Cr and Cr (VI)), Mn, Mo, Na, Ni, Pb, Zn, V, Zn; whereas the target anions were:  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ .

The mineralogy of each residue sample was examined using X-ray powder diffraction (XRPD) with a PANalytical X'Pert PRO. Water solubility was calculated by measuring the weight loss after washing the untreated residues: 100 g of each residue was shaken with 500 mL of distilled water for 5 min, and thereafter the supernatant was separated and filtered at atmospheric pressure. This procedure was repeated two more times for the same residue before all the suspension was filtered, and dried overnight at 105 °C. The metal content was measured by ICP-OES (Varian 720-ES ICP-OES) in triplicates after pre-treatment by Danish Standard DS259 (Danish Standard, 2003): 1 g of each residue and 20 mL 7.3 M  $\text{HNO}_3$  were heated at 200 kPa (120 °C) for 30 min. The liquid was thereafter separated by vacuum filtration through a 45  $\mu\text{m}$  filter and diluted to 100 mL by de-ionised (DI) water.

The water soluble content of  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$  was determined on IC (DIONEX DX-120 IC) in triplicates according to DS/EN ISO 10304-2 (Danish Standard, 2009) after water extraction. This was performed as follows: 2.5 g of each residue and 25 mL distilled water were mixed, shaken for 16 h on a horizontal shaker operating at 150 rpm, and the extractants were vacuum filtered through 45  $\mu\text{m}$  filters.

Leaching experiments were made according to DS/EN 12457-1 (Danish Standard, 2002) in triplicates. The L/S was 2, mixing 40 g of each residue with 80 mL DI water. The resulting suspension was shaken at 150 rpm for 24 h before vacuum filtration through a 45  $\mu\text{m}$  filter and the filtrate was divided into two subsamples after measuring the pH by a Radiometer Analytical pH electrode. One subsample was used to measure sulphate ( $\text{SO}_4^{2-}$ ) and chloride ( $\text{Cl}^-$ ) concentrations on ionic chromatograph (IC). The other was diluted at a ratio of 1.25 with concentrated  $\text{HNO}_3$ , and heated at 200 kPa (120 °C) for 30 min, before the metal content analysis on ICP-OES (Varian 720-ES ICP-OES).

### 2.3. Experimental set-up

The Electrodialytic (ED) cell was similar to the ones used in previous experiments (Pedersen et al., 2003; Ferreira et al., 2005). It consisted of four (I, II, III, IV) compartments (Fig. 1) made of cylindrical Plexiglas® with an internal diameter of 8 cm. Compartment III was 10 cm long and contained in each experiment 35 g ash and

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