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Electrodialytic removal of heavy metals and chloride from municipal solid waste incineration fly ash and air pollution control residue in suspension – test of a new two compartment experimental cell

Gunvor M. Kirkelund [a,*], Cátia Magro [a,b], Paula Guedes [b], Pernille E. Jensen [a], Alexandra B. Ribeiro [b], Lisbeth M. Ottosen [a]

[a] Department of Civil Engineering, Brovej, Technical University of Denmark, DK-2800 Kgs., Lyngby, Denmark
[b] CENSE, Departamento de Ciências e Engenharia do Ambiente, Faculdade de Ciências e Tecnologia, Universidade Nova de Lisboa, Campus de Caparica, 2829-516 Caparica, Portugal

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A B S T R A C T

Municipal solid waste incineration (MSWI) residues such as fly ash and air pollution control (APC) residues are classified as hazardous waste and disposed of, although they contain potential resources. The most problematic elements in MSWI residues are leachable heavy metals and salts. For reuse of MSWI residues in for instance concrete, the aim of remediation should be reduction of the heavy metal leaching, while at the same time keeping the alkaline pH, so the residue can replace cement. In this study a MSWI residues were subjected to electrodialytic remediation under various experimental conditions. Also a newly developed 2 compartment experimental cell was tested. The results show that the pH development in the MSWI residue suspension depended on the type of MSWI residue and the experimental cell type. The acidification of the suspension occurred earlier when using the 2 compartment setup and the acidification of the fly ash occurred earlier than for the APC residue but the highest removal was seen with the 3 compartment cell. The lowest final pH for the fly ash and APC residue was 6.4 and 10.9, respectively. The results showed that the leaching of Cd, Cu, Pb and Zn was reduced compared to the initial heavy metal leaching except when the pH was reduced to a level below 8 for the fly ash. On the other hand, Cr leaching increased by the electrodialytic treatment. Cl leaching from the MSWI residues was less dependent on experimental conditions and was reduced in all experiments compared to the initial levels.

1. Introduction

Incineration of municipal solid waste (MSW) is commonly used to gain energy and hygenize and minimize the amount of solid waste. The residues from MSWI are flue gas, bottom ash and fly ash or air pollution control (APC) residues depending on the air pollution control system of the incineration plant. In Denmark, all MSWI plants have air pollution control systems to minimize the emissions of acidic gases and dioxins. Fly ash is typically removed in electrofilters or cyclones, while the air pollution control process can be either a dry/semidry (injection of lime in dry form or in a slurry to the flue gas) or wet (flue gas is subjected to a scrubber after removal of fly ash). Either treatment method, the resulting residues are classified as hazardous waste due to the high content of contaminants and salts. The main management system of MSWI residues is disposal in hazardous waste facilities [1].

The electrodialytic remediation (EDR) method has been used to remove mainly heavy metals from particulate waste materials such as soil, harbor sediment and different ashes (biomass, sewage slugde, MSW) [2–6]. Ions electromigrate in the applied electric field during EDR and to maximize their desorption and availability, ionic forms are essential for the process. To reuse the MSWI residues as secondary resources, removal of contaminants and salts is necessary to reduce their toxicity. EDR treated APC residue has indicated potential for substitution of cement in mortar [7], but improvements in reduction of heavy metal leaching and chloride content is still needed.

The removal of heavy metals from MSWI fly ashes using EDR was first applied in a conventional stationary EDR cell with the fly ash saturated with water or desorbing agent [8]. These initial experiments resulted in severe precipitations in the electrolytes, on the ion exchange membranes and in the fly ash. Furthermore,

* Corresponding author. Tel.: +45 45 25 17 30.
E-mail address: gunki@byg.dtu.dk (G.M. Kirkelund).

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Nomenclature:

APC	Air pollution control
EDR	Electrodialytic remediation
MSW	Municipal solid waste
MSWI	Municipal solid waste incineration
I/S REFA	Waste-to-energy incineration plant, semi-dry APC system
SD	APC residue from REFA
I/S Vest for brænding	Waste-to-energy incineration plant, wet APC system
W	Fly ash from Vestforbrænding

poor control of the pH both in the electrolytes and in the fly ash itself, revealed the need to suspend the fly ash matrix in the cell. Pedersen et al. [9] introduced a stirrer in the suspension cell compartment II, Fig. 1a. After the introduction of the stirrer, several studies about EDR of fly ash were made, especially with the use of assisting agents to further enhance the remediation process. Ammonium citrate and gluconate were mostly used as assisting agents, with the purpose of enhancing solubilisation of the heavy metals and form stable charged complexes that could be removed in the electric field and at the same time keeping the alkaline pH in the fly ash. Allowing acidification of the ash during EDR means dissolution of a high quantity of the ash and is thus not an energy efficient separation of the ash and heavy metals. Ammonium citrate as assisting agent was used in EDR of fly ash and these electrodialytic experiments showed higher removals than just by batch extractions and up to 20% Cr, 60% Cd, 6% Pb, 40% Zn and 60% Cu were removed after 2 weeks of remediation [9]. When increasing the remediation time, even higher removals could be obtained, with the lowest still for Cr and Pb [10]. Gluconate was tested as assisting agent for APC residue and high desorption of the metals was seen in batch extraction but much lower removal was seen in EDR experiments, probably due to the size of the metal-gluconate complexes that could not pass the ion exchange membranes [11]. EDR with gluconate was later improved with pre-treatment of the APC residue before EDR, such that 82% Cd, 10% Pb, 63% Zn and 22% Cu were removed after 2 weeks of remediation [6]. One major drawback of using assisting agents is however the significant matrix changes observed both in the mineralogy and the morphology of the fly ash [12] which could hinder reuse. The addition of assisting agents also adds an extra cost to the process.

Suspending the fly ash in distilled water during EDR also results in removal of heavy metals [9]. The removal process in this case is dependent on the solubility and release of the heavy metals from the fly ash particles at high pH. In the 3 compartment cell (Fig. 1a), acidification of the suspension is caused by water splitting at the anion exchange membrane and proton leakage through the anion exchange membrane [13,14,], which leads to enhanced removal of heavy metals during EDR. Some of the few experiment of EDR with fly ash suspended in distilled water in the stirred set-up, showed varying removal results of Cd, Cr, Cu, Pb and Zn and indicate that the removal occurs both towards the anolyte and the catholyte [9,15]. The removal towards the anode can occur due to the complexation between divalent metal ions and chloride to form stable negatively charged complexes ($MeCl_3^-$, $MeCl_4^{2-}$) and for Cr also the presence of negatively charged Cr (VI) ions.

Recently, a new cell design was developed and a patent filed for EDR of suspensions, where the anode is placed directly in the material suspension and thus combines the anolyte and the suspension compartment into compartment I, Fig. 1b [16,17]. This new cell design promotes a more direct acidification of the suspension, where all the produced H^+ ions from the electrolysis at the anode ($2H_2O \rightarrow O_2(g) + 4H^+ + 4e^-$) will be supplied directly in the suspension. Anions are in the 3 compartment cell removed to the anolyte and in the case of Cl^- an oxidation to Cl_2 (g) will occur at the anode. In the 2 compartment cell, it is expected that the oxidation and subsequent removal could be faster, since the electrode is in direct contact with the suspension. Contrarily, if the heavy metals are present as anions in the 2 compartment cell, they will not be removed from the suspension.

The purpose of this study was to investigate 1) the remediation of two types of MSWI residue (fly ash and APC residue) for heavy metals and chloride according to several different experimental variables and 2) to which extent the new 2 compartment cell could improve the remediation of heavy metals and chloride from MSWI residues. The aim was to obtain a stable residue in the sense that it is not leaching heavy metals and chlorides above the Danish limiting values for reuse of waste in constructions after the treatment.

2. Materials and methods

2.1. MSWI residues

Two types of MSWI residues were used in this study from Danish municipal waste incineration plants:

- **SD**: APC residue collected after a semi-dry process after the injection of slaked lime and activated carbon from I/S REFA, where 3,000 tons APC residue is produced annually.
- **W**: A fly ash sample collected prior to the neutralization of acidic components by a wet scrubber process from I/S Vestforbrænding, where 15,000 tons of APC residues, including fly ash is produced annually.

2.2. Analytical methods

The MSWI residues characterization and extraction experiments were carried out using dried residue at 105 °C for 24 h and triplicates were made for the analysis. Total concentration of Cd, Cr,

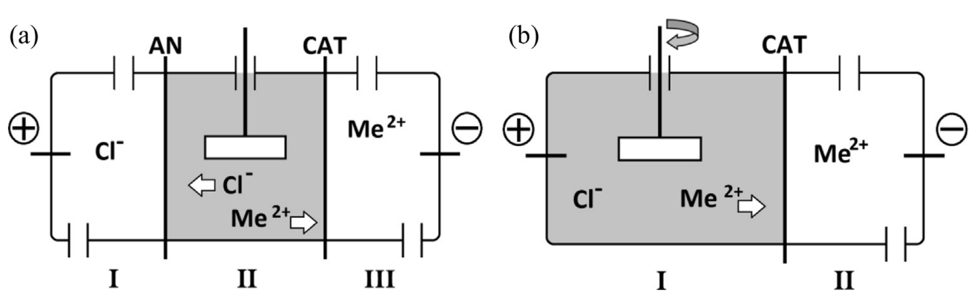

Fig. 1. The experimental set-up of the 3 and 2 compartment electrodialytic cell. AN-anion exchange membrane, CAT-cation exchange membrane.