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Electrodialytic extraction of Cr from water-washed MSWI fly ash by changing pH and redox conditions

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ABSTRACT

Electrodialytic process offers a range of possibilities to waste management by electrodialytic separation (EDS) of heavy metals, depending on how the process is designed. Using three EDS cell setups (two two-compartment and one three-compartment) and their combinations, the extraction of Cr from municipal solid waste incineration fly ash by changing pH and redox conditions was investigated in the present work. The experiments were designed into single, two and three steps, based on the number of setups (by changing EDS cells) or effective setups (by shifting working electrode pairs) used. Prior to EDS the ash studied went through pretreatments such as water-washing and dry-sieving with a 50 μm sieve. The results showed that Cr was strongly bound in the ash, and the major fraction remained bound after the different treatments. Two/three-step treatment, which obtained the maximum Cr extraction rate of 27.5%, is an improvement on the single-step that extracted maximum 3.1%. The highest extraction was obtained due to the combined extraction of Cr(III) under low pH (accompanied with high redox) conditions and Cr(VI) under high pH (low redox) conditions subsequently. The Cr leaching from the treated ashes with acidic pH was lower than from those with alkaline pH; after the three-step treatment, Cr leaching was much lower from the coarse fraction (> 50 μm), as compared to the fine (≤ 50 μm) or the unsieved ash. As for the coarse fraction, two/three-step treatment reduced the leaching of Cr compared to the single-step in the same pH range (either acidic or alkaline).

1. Introduction

Recycling of valuable metals from municipal solid waste incineration (MSWI) fly ash (e.g. full-scale Zn recovery in Switzerland (Purgar et al., 2016)) and/or recycling of the ash itself as secondary material resources (e.g. for cement production (Pan et al., 2008)) have become the focus of research in the management of this kind of residual waste. However, as one type of ash residues from solid waste management process, MSWI fly ash is commonly considered as hazardous, which means it can cause environmental concerns if recycled or utilized. Leaching of toxic substances (e.g. As, Cr, Cu, Sb and Zn) is one of the possible aspects concerning environmental issues; for instance, the leaching of Cr, Cr(VI) in particular, contributes to carcinogenic human impact (Allegrini et al., 2015). The high health risk is seemingly linked to the fine particles in the ash due to the enrichment of unstable heavy metals (Zhou et al., 2015). Size-based separation e.g. by sieving could be applied to separate out the fine particles for metal recovery, and meanwhile the coarse fraction could be recycled or used (De Boom and Degrez, 2015).

With respect to metal extraction and MSWI fly ash treatment, electrodialytic separation (EDS) is one option (Chen et al., 2017). Metals are extracted from ash when suspended in e.g. water, attributed mainly to the electromigration of mobile free ions in the suspension through ion exchange membranes in current field: cations towards the cathode end and anions to the anode. Fig. 1 illustrates three EDS cell setups. Setup-a (Fig. 1a), a two-compartment (2C) cell, has an anode and a cathode compartment separated by a cation exchange membrane. The distilled water-ash suspension to be treated is placed in the anode compartment (as anolyte) (Chen et al., 2015). The two compartments in a 2C cell can also be separated by an anion exchange membrane for anion removal, i.e. setup-b (Fig. 1b). However, setup-b is not commonly seen in MSWI fly ash treatment, and with good reason: most heavy metals are mobilized to the highest extent at low pH values in mainly cationic form. In setup-c (Fig. 1c), a three-compartment (3C) cell, there are anode and cathode compartment for electrolyte solutions, plus a center compartment for ash suspension. These three compartments are separated by a pair of cation and anion exchange membranes.

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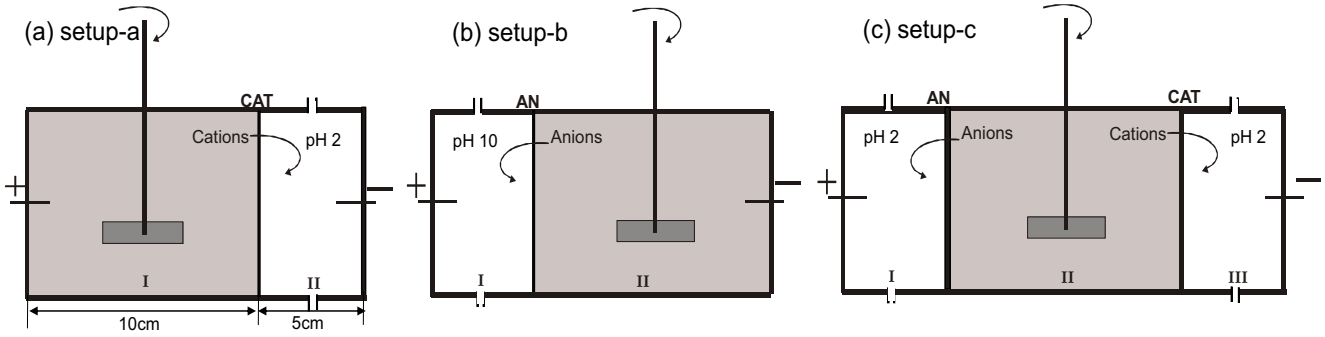

Fig. 1. Cylindrical electrodialytic cell setups with an inner diameter of 8 cm (AN – anion exchange membrane; CAT – cation exchange membrane; number of compartment – I, II and III).

When electricity is supplied to an electrodialytic cell, water is electrolyzed to hydrogen ions and oxygen at the anode, and to hydroxyl ions and hydrogen at the cathode. In ash suspension, current is carried by ions that are either in the suspension originally or released along the treatment process because of ash dissolution caused by the changing properties of the suspension such as pH and redox potential.

The development of pH and redox potential differs between the EDS setups. Setup-a (Fig. 1a) and setup-c (Fig. 1c) acidify ash suspension due to water electrolysis and water splitting, respectively; whereas, setup-b alkalizes ash suspension due to the production of hydroxyl ions. The redox potential of ash suspension is affected by the generation of oxygen and hydrogen from electrode reactions if they enter into the suspension. For instance, setup-a directly introduces oxygen into suspension, whereas setup-b hydrogen.

The pH plays an important role in the mobilization of heavy metals and has great impact on the species of charged mobile ions, therefore influencing the electrodialytic extraction rate. Most metals form soluble cations at acidic pH (e.g. Zn^{2+}), and amphoteric metals (e.g. Pb and Zn) also exist as soluble hydroxides at alkaline pH (e.g. $Zn(OH)_3^-$ and $Zn(OH)_4^{2-}$). Some metals, especially oxyanion forming elements such as Cr, develop different mobile species not only depending on pH but also on redox potential (Cornelis et al., 2008). This suggests that using different EDS setups might lead to different extraction rates of Cr from MSWI fly ash. However, relevant information has not been reported yet. In addition, a deep understanding of the migration patterns of Cr found in MSWI fly ash during EDS treatment under different conditions might be of importance considering that, in most cases without using assisting agents such as ammonium citrate (Pedersen, 2002), the extraction of Cr was low compared to that of other heavy metals such as Cd, Cu and Zn. For instance, compared to 3.3–15% of Cu, 1.1–61% of Cd and 1.5–53% of Zn, the extraction of 1.1–4.5% of Cr was obtained by Kirkelund et al. (2015) through treating a MSWI fly ash using either 2C EDS cell (Fig. 1a) or 3C (Fig. 1c) under the conditions: water-to-ash ratio (L:S) of the suspension of 3.5; current density of 0.1/1 mA/cm^2; and 14 days treatment. Chen et al. (2017) treated the same washed ash which was used in the present study through single-step 2C (Fig. 1a) and two-step 2C + 3C (Fig. 1a+c) EDS. A slight increase in Cr extraction from the washed ash was observed by introducing the second treatment step (effective 3C), but the extraction rate was still less than 1.5% under the conditions: L:S of 3.5; current density of 1 mA/cm^2; and 3/7/12 days treatment.

In view of the above-mentioned facts, the present study investigated the possibility for chromium extraction from water prewashed MSWI fly ash under different pH and redox potential conditions, which were created by using different combinations of the 2C and 3C electrodialytic setups, without using any assisting agents. Setup-b (Fig. 1b) was especially used to investigate the feasibility of extracting Cr under high alkaline and low redox conditions. The Cr extraction was compared between the different EDS combinations: one-step treatment with either acidification or alkalization of ash suspension; two-step using the two 2C setups to combine acidification and alkalization; and three-step involving all the three setups illustrated in Fig. 1. As for ash pretreatment prior to EDS, by prewashing the consumption of current on moving soluble salt ions can be reduced, and the generation of toxic chlorine gases during EDS treatment can be suppressed; and by one-size sieving of the washed ash the differences in Cr extraction between the coarse and the fine fractions were studied. Moreover, the Cr leaching from the variously treated ashes was discussed.

2. Materials and methods

2.1. MSWI fly ash and pretreatment

The MSWI fly ash studied was collected after electrostatic precipitator but before the neutralization of acidic components by a wet scrubber process, from the Vestforbrænding MSW incineration plant located in Glostrup, Denmark. The incinerator has a capacity of 600 000 t/year and generates about 15 000 tons of air pollution control (APC) residue annually, including fly ash. The as-received sample was sieved in the laboratory to remove the few particles exceeding 1 mm.

The ash passing through the sieve was washed with distilled water at a L:S ratio of 5 for 5 min. The suspension was filtered (12–15 μm filter) and the ash retained on the filter paper was dried at 105 °C for 24 h. One part of the washed ash was directly used in the EDS experiment, and another part was fractionated by dry-sieving using a 50 μm sieve prior to EDS treatment.

2.2. EDS treatment

The three setups illustrated in Fig. 1 were used in the present lab-scale study. The ash suspension was placed in the compartments (with a length of 10 cm) highlighted in grey – setup-a: compartment I; setup-b/c: compartment II. The overall experimental design is outlined in Table 1. Each experiment was named based on the ash sample treated (C = coarse fraction; F = fine fraction; W = the whole washed ash unsieved) together with the setup(s) arranged in order of appearance (e.g. b – single-step treatment using setup-b; a-b/b-a – two-step treatment in the order of setup-a and setup-b or in reverse order; and so forth). The experiments were made in three phases:

Phase I (C-a, C-b and F-a): Single-step treatments comparing setups -a and -b, as well as comparing the treatment of the two ash fractions using setup-a.

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