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Effect of accelerated carbonation and zero valent iron on metal leaching from bottom ash

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ABSTRACT

About 85% of the ashes produced in Sweden originated from the incineration of municipal solid waste and biofuel. The rest comes from the thermal treatment of recycled wood, peat, charcoal and others. About 68% of all ashes annually produced in Sweden are used for constructions on landfills, mainly slopes, roads and embankments, and only 3% for construction of roads and working surfaces outside the landfills (SCB, 2013). Since waste bottom ash (BA) often has similar properties to crushed bedrock or gravel, it could be used for road constructions to a larger extent. However, the leaching of e.g. Cr, Cu, Mo, Pb and Zn can cause a threat to the surrounding environment if the material is used as it is. Carbonation is a commonly used pre-treatment method, yet it is not always sufficient.

As leaching from aged ash is often controlled by adsorption to iron oxides, increasing the number of Fe oxide sorption sites can be a way to control the leaching of several critical elements. The importance of iron oxides as sorption sites for metals is known from both mineralogical studies of bottom ash and from the remediation of contaminated soil, where iron is used as an amendment.

In this study, zero valent iron (Fe(0)) was added prior to accelerated carbonation in order to increase the number of adsorption sites for metals and thereby reduce leaching. Batch, column and pH_{stat} leaching tests were performed and the leaching behaviour was evaluated with multivariate data analysis. It showed that leaching changed distinctly after the tested treatments, in particular after the combined treatment.

Especially, the leaching of Cr and Cu clearly decreased as a result of accelerated carbonation. The combination of accelerated carbonation with Fe(0) addition reduced the leaching of Cr and Cu even further and reduced also the leaching of Mo, Zn, Pb and Cd compared to untreated BA. Compared with only accelerated carbonation, the Fe(0) addition significantly reduced the leaching of Cr, Cu and Mo. The effects of Fe(0) addition can be related to binding of the studied elements to newly formed iron oxides. The effects of Fe(0) addition were often more distinct at pH values between 7 and 9, which indicates that a single treatment with only Fe addition would be less effective and a combined treatment is recommended. The pH_{stat} results showed that accelerated carbonation in combination with Fe(0)⁰ addition widens the pH range for low solubility of about one unit for several of the studied elements. This indicates that pre-treating the bottom ash with a combination of accelerated carbonation and Fe(0) addition makes the leaching properties of the ash less sensitive to pH changes that may occur during reuse. All in all, the addition of Fe⁰ in combination with carbonation could be an effective pre-treatment method for decreasing the mobility of potentially harmful components in bottom ash.

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1. Introduction

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Each year about 1 million tons of bottom ashes are produced in Sweden as a result of waste incineration. Bottom ash (BA) has similar properties to virgin ballast material like crushed bedrock and gravel and can therefore replace some of the 40 million tons of virgin material used for road constructions each year. However using wastes such as ash must not harm the environment and must not exceed the guidance levels for emissions from waste that is used as construction material set up by the Swedish EPA (SNV, 2010). Today, the most common areas of application for bottom ash in Sweden are in landfill constructions such as slopes, roads and embankments, since for this type of application the emission limits are less strict than for uses outside of landfills.

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Nevertheless, pre-treatment prior to use on the landfill is anyhow needed in most cases as ashes from waste incineration contain considerable amounts of soluble salts and partly soluble constituents, such as metals. A common method is carbonation that will reduce the pH and thereby decrease the leaching of elements like Cd, Pb, Cu, Zn and Mo (Meima and Comans, 1999). This treatment is, however, not always sufficient and alternative methods are needed. One possibility is to increase the number of adsorption sites for trace elements. The importance of iron oxides as sorption sites for metals is known from both mineralogical studies of bottom ash (Saffarzadeh et al., 2011; Cornelis et al., 2008; Cornell and Schwertmann, 2003) as well as from remediation of contaminated soil, where iron is used as an amendment in order to immobilize metals (Kumpiene et al., 2008).

Fettihydrite is the first iron oxide phase formed during iron oxidation. Ferrihydrite is poorly crystalline and therefore has more sorption sites than more crystalline iron oxides. However, it is thermodynamically unstable, and will transform into other more crystalline iron oxides, most commonly goethite and hematite. A more crystalline iron oxide will have less sorption sites, and can thus retain less metal. However, the rate of transformation can be reduced if foreign species, like metals, are retained within the structure (Cornell and Schwertmann, 2003). Formation of iron oxides in an environment that contains metal ions would possibly enhance the chance of metals to be sorbed to ferrihydrite and thereby decreasing the chance of transformation. Iron oxides can bind ions like Cu, Cr(III) and Zn (Sabbas et al. 2003), oxyanions like Mo and Cr as well as fulvic acids that in turn can bind e.g. Cu (Dijkstra et al., 2006).

A local waste-to-energy plant in Northern Sweden incinerates waste and the BA can possibly be used as construction material. A previous study (Oja, 2012), showed that a number of trace elements (Cr, Cu, Mo, Pb and Zn) exceeded the guideline values of the Swedish EPA for the use of waste as construction material (SNV, 2010), even though the BA had been stored outdoors for more than six months. Additional treatment is therefore necessary before the BA can be used as construction material.

The aim of this study was to reduce the leaching of Cr, Cu, Mo, Pb and Zn by pre-treating the bottom ash using two different methods: accelerated carbonation in a 30 °C humid CO_2 atmosphere and equal accelerated carbonation but with the addition of zero valent iron (Fe(0)), a by-product from the steel industry, in order to increase the number of sorption sites.

The objectives of the study were to investigate (1) how pretreatment including accelerated aging and accelerated aging with addition of Fe(0) affects the leaching of Cr, Cu, Mo, Pb and Zn and (2) which processes are behind these effects.

2. Material and methods

2.1. Material

BA from a combined heat and power plant in the municipality of Boden in Northern Sweden was tested. The ash was derived from two fired grate stoker boilers burning a mixture of municipal, industrial and wood waste. The ash is collected in one big heap during a whole year and once per year, typically in July and August, the material is treated in a mobile metal separation plant. During this process, the material is also classified into three grain size fractions: 0-10 mm, 10-50 mm and >50 mm. The samples were taken from the 0-10 mm fraction resulting from the metal separation treatment. At the time of sampling the ash in that heap was between 1 and 10 months old. In order to take a representative samples of the whole heap subjected to the separation treatment, one sample was taken each day during the metal separation process (that had a duration of 32 days).

In a pre-study Nilsson et al. (2013) screened the samples for systematic inhomogeneity with regard to TDS, LOI, electrical conductivity and pH and came to the conclusion that the heterogeneity of the material was evenly distributed. The 32 samples were therefore combined before being employed for the tests described in this paper.

2.2. Pre-treatment

The 32 samples were homogenized using a rotary splitter. After homogenization sub-samples of the BA were pre-treated with accelerated carbonation (BA_A) and accelerated carbonation with addition of 2% zero valent iron (BA_AFe). 15 kg of the 0–10 mm fraction were pre-treated. Non-treated bottom ash (BA) was used as control (note: the ash had been stored outside for 1–10 months prior to sampling, *i.e.* it cannot be considered as "fresh" BA). For accelerated carbonation (with/without Fe(0)) the ash was moistened and placed in a CO₂-filled barrel at 30 °C for two weeks. CO₂ was refilled and the ash was agitated by occasionally rolling the barrel.

The Fe(0) used in this experiment is a by-product from a steel factory in Sweden called SSAB Merox, where it is used as a blasting material. The Fe(0) powder presented the following particle size distribution: 2.5% $d \ge 1$ mm; 20.5% $0.5 \le d < 1$ mm; 68% $0.25 \le d < 0.5$ mm and 9% d < 0.25 mm. The chemical composition was determined by a laboratory specialised in ore and metal analyses (Nilab AB): 96.50% Fe_{tor}, 0.91% Mn, 0.81% C, 0.76% Si, 0.24% Cr, 0.11% Ni, 0.04 Mo and 0.02% S.

2.3. Chemical composition

Samples of 1.5 kg from fractions of BA, BA_A and BA_AFe were dried at 50 °C for 7 days, comminuted in a jaw crusher to <3 mm and subdivided into samples of 30 g each before milling (disc mill). The total content was analysed in triplicates at ALS Scandinavia AB using ICP–AES (Inductively Coupled Plasma – Atom Emission Spectroscopy) and ICP–SFMS (– Sector Field Mass Spectroscopy). The carbon content was analysed with a TOC analyser (Shimadzu, TOC-V CSH).

2.4. Leaching behaviour

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The leaching of bottom ash was evaluated with (1) column tests, CEN/TS 14405, (SIS, 2004), (2) batch test, SS EN 12457-2, (SIS, 2003) and (3) pH-stat test, CEN/TS 14497, (SIS, 2007). The tests all reach a liquid to solid ratio (L/S) of 10, but differ with regard to a number of factors, see Table 1. The pH_{stat} tests were performed at pH 2, 4, 7, 8, 9 and 10 and also with only water in order to uncover possible impacts of the test conditions in comparison to the standard batch test.

Table I
Characteristics of the leaching tests used.

	Column test ^a	Batch test	pH _{stat} test
Duration	About 30 days	24 h	48 h
L/S ratio	0.1, 0.2, 0.5, 1, 2, 5, 10	10	10
Particle size	<10 mm	Sieved < 4 mm	Crushed < 1 mm
Sample mass (DM)	2.7 kg	90 g	15 g
Separation leachate – solids	0.45 µm filter	0.45 µm filter	0.45 μm filter
pH adjustment	None	None	HNO ₃

^a Up-flow column tests in 40 cm long columns with a diameter of 10 cm. Flow rate: 12 cm/day.

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