Waste Management 56 (2016) 216–228

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

Analysis and interpretation of the leaching behaviour of waste thermal treatment bottom ash by batch and column tests

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ARTICLE INFO

Article history:
Received 14 March 2016
Revised 14 July 2016
Accepted 24 July 2016
Available online 29 July 2016

Keywords:
Bottom ash
Leaching behaviour
Column test
Batch test
Modelling

ABSTRACT

This paper investigates the leaching behaviour of specific types of waste thermal treatment bottom ash (BA) as a function of both pH and the liquid-to-solid ratio (L/S). Specifically, column percolation tests and different types of batch tests (including pH-dependence) were applied to BA produced by hospital waste incineration (HW-I), Refuse Derived Fuel (RDF) gasification (RDF-G) and RDF incineration (RDF-I). The results of these tests were interpreted applying an integrated graphical and modelling approach aimed at identifying the main mechanisms (solubility, availability or time-controlled dissolution and diffusion) governing the release of specific constituents from each type of BA. The final aim of this work was in fact to gain insight on the information that can be provided by the leaching tests applied, and hence on which ones may be more suitable to apply for assessing the leaching concentrations expected in the field. The results of the leaching tests showed that the three samples of analysed BA presented differences of orders of magnitude in their leaching behaviour, especially as a function of pH, but also in terms of the L/S. These were mainly related to the differences in mineralogy of the samples. In addition, for the same type of bottom ash, the comparison between the results of batch and percolation column tests, expressed in terms of cumulative release, showed that for some constituents (*e.g.* Mg for HW-I BA and Cu for RDF-G BA) differences of over one order of magnitude were obtained due to variations in pH and DOC release. Similarly, the eluate concentrations observed in the percolation tests, for most of the investigated elements, were not directly comparable with the results of the pH-dependence tests. In particular, in some cases the percolation test results showed eluate concentrations of some constituents (*e.g.* K and Ca in HW-I BA) of up to one order of magnitude higher than the values obtained from the pH-dependence experiments at the same pH value. This was attributed to a rapid washout from the column of the soluble phases present in the BA. In contrast, for other constituents (*e.g.* Mg and Ba for the RDF-G BA), especially at high L/S ratios, the concentrations in the column tests were of up to one order of magnitude lower than the solubility value, indicating release under non-equilibrium conditions. In these cases, batch pH-dependence tests should be preferred, since column tests results could underestimate the concentrations expected in the field.

1. Introduction

In the last decades, several studies have focused on the potential utilisation of granular waste materials such as steel slag, construction and demolition waste and bottom ash from waste thermal treatment, as a replacement of natural aggregates in civil engineering applications (*e.g.* Forteza et al., 2004; Petkovic et al., 2004; Das et al., 2007; Huang et al., 2007). However, compared to natural aggregates, these materials generally contain higher concentrations of harmful compounds (*e.g.* metals and metalloids) that may be released upon contact with water (*i.e.* leached) posing a potential risk for the environment and ultimately human health. The leaching process has been reported to be influenced by both the field conditions to which the material can be subjected to in specific application/disposal scenarios (pH, the amount of water in contact with the waste solid matrix, *i.e.* the liquid-to-solid ratio, redox potential, aging processes and temperature) and the intrinsic characteristics of the material itself (particle size distribution, chemical composition and the solubility of the mineral phases constituting the residues) (van der Sloot, 1996; Sabbas et al., 2003). Over recent years, in order to evaluate the release of contaminants from waste materials as a function of pH and the liquid-to-solid ratio (L/S) different leaching test methods, making use of both

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http://dx.doi.org/10.1016/j.wasman.2016.07.034
0956-053X/

static (batch) and dynamic (column percolation) procedures, were developed and standardized (*e.g.* van der Sloot et al., 1997; Kosson et al., 2002; Hage and Mulder, 2004). Typically, in batch tests, a sub-sample of the studied material is placed in contact with a liquid phase for fixed time (generally 24–48 h) and conditions (*e.g.* L/S or pH), during which chemical equilibrium between the aqueous and solid phase is assumed to be established (López Meza et al., 2008; Garrabrants et al., 2010). Batch tests have the advantage of being of simple and fast application, but do not provide information on release kinetics under dynamic conditions (Kalbe et al., 2008; Grathwohl and Susset, 2009; Grathwohl, 2014). To assess the differences between constituents' release under static and dynamic conditions as a function of L/S, several authors have compared batch and column tests data, finding different results depending on the type of analysed granular waste material. For example, López Meza et al. (2008) conducted a comprehensive experimental study comparing leaching data obtained by column and batch tests as a function of the L/S on five different types of materials (*i.e.* concrete, construction debris, aluminium recycling residue, coal fly ash and bottom ash) concluding that overall the two types of tests provide comparable results. Al-Abed et al. (2008) and Quina et al. (2011) reached the same conclusions by analysing mineral processing waste and air pollution control (APC) residues from municipal solid waste incineration (MSWI), respectively. On the contrary, Kalbe et al. (2008), who focused on contaminated soil, demolition waste and waste incinerator bottom ash, found a good agreement between the two types of tests only with regard to the release of sulphates and chlorides, while for the leaching of metals (*e.g.* Cu and Cr) they observed significant discrepancies. Regarding the evaluation of the pH dependency of leaching, several studies (*e.g.* Astrup et al., 2006; Dijkstra et al., 2006, 2008; Quina et al., 2009; Galvín et al., 2012; Kosson et al., 2014a) have demonstrated that for alkaline waste materials (*e.g.* construction and demolition waste and solid residues from thermal treatment processes) pH plays a particularly crucial role in controlling the release of several constituents. Hence, for a comprehensive evaluation of the leaching behaviour of these types of materials, several types of leaching tests, assessing the release as a function of pH and also the L/S, are necessary (Kosson et al., 2002, 2014b).

In this study, the leaching behaviour of three types of bottom ash (BA) generated from different waste thermal treatment plants was analysed experimentally and interpreted applying an integrated graphical and modelling approach. The examined samples were produced by hospital waste incineration (HW-I), Refuse Derived Fuel (RDF) gasification (RDF-G) and RDF incineration (RDF-I). These residues were selected since, to our knowledge, their properties have been investigated only by few studies, mainly focused on the analysis of their physico-chemical and mineralogical characteristics (*e.g.* Gori et al., 2011; Kougemitrou et al., 2011; Rocca et al., 2012, 2013). In addition, the results of the above mentioned studies indicated that, even though these residues present a similar chemical composition, their mineralogy and leaching behaviour may differ considerably, owing in particular to the differences in the type of thermal treatment applied, *i.e.* incineration or gasification (Baciocchi et al., 2010; Gori et al., 2011; Rocca et al., 2012, 2013). Hence, in this work different types of leaching test procedures (both static and dynamic) were applied in order to investigate the influence of pH, the L/S and the type of test on the release of main and trace constituents from each BA sample. The approach employed to evaluate the results attained by the different types of leaching tests performed was based on the combination of a graphical method and a modelling method. The first one, which consists in comparing column and pH-dependence test results, allows to assess if the release of a constituent observed in the column test is governed by the solubility at the specific pH observed in the column. Then, the column test results are compared to the predictions of simplistic models that simulate solubility, availability or time-controlled dissolution and diffusion release. By coupling the information provided by the graphical and modelling methods the main mechanisms possibly controlling the leaching behaviour of a specific constituent may be identified. This may provide useful insight on the information that can be obtained by different types of leaching tests, and hence on which ones may be more suitable to apply for assessing the leaching concentrations of a specific type of BA expected in the field.

2. Materials and methods

2.1. BA physico-chemical and mineralogical composition

Three different types of BA were examined in this study: (i) BA generated at a dedicated hospital waste incineration plant (HW-I BA) equipped with a rotary kiln combustion chamber with operational temperatures of 1000–1200 °C; (ii) BA produced by a fluidised bed gasifier treating RDF (RDF-G BA) and operating at 1200–1400 °C which results in the direct melting of the slag; (iii) BA from a RDF incineration plant (RDF-I BA) employing a water-cooled grate combustion chamber and an operating temperature of 850–1000 °C.

The BA samples collected at each plant were first homogenised through quartering (UNI 10802, 2013) and then the grain size fraction $\geqslant 12$ mm (which for the three types of materials was <5% by weight) was removed and not analysed. Furthermore, coarse metallic, ceramic and glass fragments larger than 5 mm were manually removed from the RDF-I BA sample.

For each type of BA, the particle size distribution (ASTM D422-63, 2007), the total organic carbon (TOC) and carbonate ($CaCO_3$) contents, as well as the chemical and mineralogical composition, were determined. The total carbon (TC) and total inorganic carbon (TIC) were determined with a Shimadzu solid sample module (SSM-5000A) analyser. TC was measured by catalytic combustion of the BA sample (<0.2 mm) at 900 °C, while TIC was determined after H_3PO_4 addition at 200 °C for carbonate ($CaCO_3$) decomposition. The TOC was quantified by subtracting TIC from TC (UNI 13137, 2002). The chemical composition was derived by ICP-OES analysis (Agilent 710-ES) of the solutions obtained after alkali digestion of the samples, carried out at 1050 °C with lithium tetraborate. The mineralogy of the materials was determined by X-ray powder diffraction (XRD) analysis (Philips Expert Pro diffractometer, equipped with a copper tube operated at 40 kV and 40 mA). More details on the characterization methods employed are provided in Rocca et al. (2012).

The three types of BA exhibited a similar grain size distribution, with a median diameter (d_{50}) in the range of 2–2.2 cm (see Fig. A.1 in the Supplementary information). However, while the HW-I BA showed a higher content of fine particles, the opposite was observed for the RDF-I BA. The gasification BA instead, presented a slightly greater homogeneity compared to the other two types of BA, as indicated by the lower gradation of the curve (refer to Fig. A.1). The results obtained for the chemical composition, TOC and calcite content (see Table 1) showed to be quite in line with the ones reported in our previous studies regarding these specific types of BA, *i.e.* Rocca et al. (2012, 2013). In addition, for the HW-I BA all the measured constituents showed to be in the range of values typically observed for this type of BA (*e.g.* Idris and Saed, 2002; Filipponi et al., 2003; Kougemitrou et al., 2011).

Comparing the concentration values reported in Table 1, it may be observed that the three types of BA presented a quite similar content of trace and macro-constituents. The most significant differences are related to Cu, which resulted more abundant in the RDF-G BA, and Pb and Zn that were found at higher concentrations