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Research article

Evaluation of the influence of mechanical activation on physical and chemical properties of municipal solid waste incineration sludge

V. Caprai*, M.V.A. Florea, H.J.H. Brouwers

Department of Built Environment, Eindhoven University of Technology, Eindhoven, 5600 MB, The Netherlands

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A B S T R A C T

Despite numerous studies concerning the application of by-products in the construction field, municipal solid waste incineration (MSWI) residues are not widely used as secondary building materials. In some European countries, washing treatment to the full bottom ash (BA) fraction (0–32 mm) is applied, isolating more contaminated particles, smaller than 0.063 mm. Therefore, a MWSI sludge is produced, having a high moisture content, and thus a limited presence of soluble species. In order to enhance its performance as building material, here, dry mechanical activation is applied on MSWI sludge. Thereafter, a reactivity comparison between reference BA and untreated and treated MSWI sludge is provided, evaluating their behaviour in the presence of cement and their pozzolanic activity. Moreover, the mechanical performances, as 25% substitution of Portland cement (PC) are assessed, based on the EN 450. Mechanical activation enhances MSWI sludge physically due to the improved particle morphology and packing. Chemically, the hydration degree of PC is enhanced by the MSWI sludge by ≈25%. The milling treatment proved to be beneficial to the residues performances in the presence of PC, providing 32% higher strength than untreated sample. Environmentally, the compliance with the unshaped material legislation is successfully verified, according to the Soil Quality Decree.

1. Introduction

In recent times, the management of municipal solid waste (MSW) has become important to guarantee an economically and environmentally sustainable way of living. After collection, MSW is initially processed in waste-to-energy plants, by incineration. The created municipal solid waste incineration (MSWI) residues consist of various products such as bottom ash (BA) (80% wt.), grate siftings, heat recovery ash, fly ash and air pollution control residues. Despite the risk of releasing harmful substances into the soil (Sabbas et al., 2003; Florea, 2016), in the EU, BA is usually applied for road construction or landfilled (Allegrini et al., 2015; Forteza and Far, 2004; Hjelmar, 1996; Lam et al., 2010; Lin et al., 2012). Due to the increasing environmental concern, since 2011, the Netherlands defined stricter limitations for leaching of contaminants from landfilled materials (Soil Quality Decree, 2015), aiming to reduce the number of landfilling sites and to address the design of secondary raw materials from incineration by-products (Zhen et al., 2013). Nowadays, various applications of bottom ash (BA) in the construction field have been studied (Bertolini et al., 2004; Boesten et al., 2012; Polettini et al., 2009; Revathi et al., 2014). Among them, the application of BA as inert replacement of Ordinary Portland Cement (PC) has found successful, up to 30% of the binder mass (Kula et al., 2002; Tang et al., 2014a; Targan et al., 2003). Before being reused, BA undergoes additional treatments, such as removal of ferrous and nonferrous compounds, in order to achieve a more homogeneous and less contaminated material (Keulen et al., 2012). Among these procedures, a washing separation treatment is applied to the full fraction of BA (below 32 mm), isolating most of the incineration residues smaller than 0.063 mm, generally more contaminated (Biganzoli, 2012; Alam et al., 2016). Therefore, a MWSI sludge is produced characterized by a high moisture content (MC), and consequently limited ions available for dissolution. Due to the restrained application of this washing separation treatment, the use of MSWI sludge as secondary building material is generally not taken into account outside the Netherlands. Consequently, to the authors' knowledge, the investigation of MSWI sludge is not extensively performed, due to the low availability of the by-product in Europe. Furthermore, the storage conditions of MSWI sludge lead to particles agglomeration, increasing the particle size of the supplied material. As mentioned,

* Corresponding author.
E-mail address: v.caprai@tue.nl (V. Caprai).

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due to the source of this by-product, the monitoring of contamination levels and leaching is also a concern. In addition to this, the initial MC of MSWI sludge favors the dissolution of soluble phases, reducing the final reactivity of the by-product.

The suitability of a by-product as cement replacement is linked to its chemical and physical contribution to the PC hydration. In general, the reactivity of ashes is affected by many aspects, from the source of the waste to its collection and disposal. In the case of low reactivity ashes, many studies propose activation treatments to increase the final dissolution of particles (Cheriaf et al., 1999; Onori et al., 2011; Polettini et al., 2009; Qiao et al., 2009, 2008; Tang et al., 2014b). There are three main methodologies applied for this purpose: thermal, chemical and mechanical activation (MA). Thermal activation is not suitable from an environmental point of view, due to the high incineration temperature (between 800 °C and 1500 °C) (Hernández-Montoya et al., 2012; Zhen et al., 2013). Chemical activation of MSWI residues is not always possible, due to the potential high organic content of the by-products (Fernández-Jiménez and Palomo, 2003; Sathonsaowaphak et al., 2009). Therefore, economically and environmentally, the most sustainable method for increasing the reactivity of ashes is MA. Generally, MA improves the bulk and surface area of the solid through physicochemical changes (Kumar and Kumar, 2011), which might lead to the acquisition of additional pozzolanic activity. However, the mineralogical changes are not always remarkable in case of MA of ashes (Temuujin et al., 2009). Its application is beneficial also due to the formation of smaller less reactive particles (external core of coarser particles), which act as a filler among cement grains (Hela and Orsáková, 2013). Increasing the fineness, the prolonged grinding rises the amount of active centres with higher energy state respect to the rest of the structure. The more of those sites, the faster the rate of reactivity in the presence of PC (Onori and Polettini, 2011; Sajedi and Razak, 2011; Temuujin et al., 2009). Until now, the application of MA on MSWI sludge has not been deeply investigated. However, the improvement achieved can lead to performances comparable to BA fines, favouring the application of MSWI sludge in the construction field. Since the behaviour of BA fines as cement replacement has been more extensively studied, a comparison with this residue provides a useful reference for the evaluation of MSWI sludge performances.

This study provides an evaluation of the influence of MA on MSWI sludge, compared with reference fines of two streams of BA. The enhancement of MSWI sludge properties aims to achieve comparable results with BA in mechanical and environmental performances, in order to provide an alternative application for this municipal residue. Firstly, the by-products provided by Heros Sluiskil (NL) are characterized physically and chemically. Additionally, the MSWI treated sludge is compared to the untreated material and reference BA, regarding the impact of the MA on physical properties. Thereafter, this paper will address the reactivity of the two reference streams of BA and MSWI sludge (before and after MA), based on the pozzolanic activity index and by isothermal calorimetry analysis. Finally, the application of the by-products as secondary building materials is evaluated by the strength activity index and by the one stage batch leaching test, for the compliance to the Dutch SQD legislation for the unshaped (granular) material ("Soil Quality Decree," 2015).

2. Experimental

2.1. Materials

Three different streams of MSWI residues are considered in this study: two types of bottom ash fines (BA1 and BA2) and MSWI sludge (Ss), all coming from Heros Sluiskil (NL). All the MSWI residues are characterized by a particle size below 0.250 mm. After dividing the BA full fraction in a coarse and a fine particle size streams (Bac and BAf, respectively), a wet separation treatment is initially applied to produce sludge, by removal of fine residues from the BAc. The MSWI sludge is further processed by using a centrifuge to limit the water content of the final product. In parallel, the fine size stream (BAf) is fractioned and particles below 0.250 were isolated (BA1). Part of the BA1 is further clean, by air separation removal, limiting the presence of the ultrafine ash particles, forming BA2. Due to the agglomeration of particles caused by the storage conditions, the application of Ss as untreated material is not possible. Hence, in this study, an implementation by MA was attempted for this residue, and compared with the two BA streams, for evaluation. The pozzolanic activity of BA1, BA2 and MSWI sludge was evaluated in the presence of calcium hydroxide (CH) (CH $\geq$ 96% pure, Fluka Analytical), CEM I 42.5 N (PC) and inert standard sand (98% SiO_2, Norm sand, ISO 679, EN 196-1). The analysis of reactivity was performed without any additional laboratory treatment, since the washing processing might affect the final dissolution rate of the by-products in water.

2.2. Methods

2.2.1. Characterization of the raw materials

Firstly, all materials were dried at 50 °C for 72 h, to constant mass. The PSD was measured by laser diffraction (Mastersizer 2000, Malvern). The specific gravity of the dry powders was tested by Helium pycnometer (AccuPyc II 1340), while the bulk density by a conventional pycnometer. The chemical composition of the by-products was determined by X-ray Fluorescence spectrometer (PANalytical Epsilon 3 range, standardless OMNIAN method), on pressed powder. The specific surface area and total pore volume were measured using nitrogen adsorption measurements (Micromeritics, Tristar II 3020 V1.03). The water demand of the powders was measured by the Puntke test (Hunger and Brouwers, 2009). Information about the morphology of the particles was provided by Scanning Electron Microscopy (SEM, Quanta 650 FEG, FE). The analysis was performed in high vacuum, at chamber pressure of 0.6 mbar, spot size 3.0, voltage 20.0 KV and in low vacuum by coupling large field detector, GSED detection (LFD) and BSE- detector, at chamber pressure 0.6 mbar, spot size 4.0, voltage10 KV. The quantification of $CaCO_3$ was computed from the CO_2 mass loss between 640 and 740 °C (Gabrovšek et al., 2006), with thermogravimetric analysis (TGA), using Jupiter STA 449 F1, Netzsch, heating rate 5 °C/min, purged with N_2 and synthetic air. The evaluation of the metallic Al content was performed by treatment in alkaline environment (3 M NaOH solution) for 24 h (Porciúncula et al., 2012). The loss of ignition was evaluated by heating the samples to 1100 °C for 4 h and measuring the mass loss at constant RH. The evaluation of the Ca^{2+} ions dissolution in the presence of water was measured by ion chromatography (IC) (Thermoscientific Dionex ICS-1100), after 1 h, at 20 °C.

2.2.2. Mechanical activation treatment of MSWI sludge

The MA of Ss was carried out using a Fritsch Pulverisette 5 planetary ball mill, at a constant speed of 200 rpm. Bottom ash particles are characterized by a rigid inner core, mainly composed of stony or glassy material, and a surrounding loose coating layer in the order of few hundred microns, composed by ultrafine ash particles. Due to its structure, composed by light materials as organic matters, fine melt glass and remnants of metallic matter, this layer is defined as "fragile zone" and it contributes to the increase of the final porosity in BA particles (Saffarzadeh et al., 2011). Due to the presence of these dust-like residues on the Ss particles surface and its fine PSD, zirconia grinding balls size 10 mm were

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