



## Recycling of air pollution control residues from municipal solid waste incineration into lightweight aggregates



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### ARTICLE INFO

#### Article history:

Received 24 May 2013

Accepted 15 October 2013

Available online 14 November 2013

#### Keywords:

APC residues

MSW incineration

Lightweight aggregates

Recycling

### ABSTRACT

This work focuses on the assessment of technological properties and on the leaching behavior of lightweight aggregates (LWA) produced by incorporating different quantities of air pollution control (APC) residues from municipal solid waste (MSW) incineration. Currently this hazardous waste has been mostly landfilled after stabilization/solidification. The LWA were produced by pelletizing natural clay, APC residues as-received from incineration plant, or after a washing treatment, a small amount of oil and water. The pellets were fired in a laboratory chamber furnace over calcium carbonate. The main technological properties of the LWA were evaluated, mainly concerning morphology, bulk and particle densities, compressive strength, bloating index, water adsorption and porosity. Given that APC residues do not own expansive (bloating) properties, the incorporation into LWA is only possible in moderate quantities, such as 3% as received or 5% after pre-washing treatment.

The leaching behavior of heavy metals from sintered LWA using water or acid solutions was investigated, and despite the low acid neutralization capacity of the synthetic aggregates, the released quantities were low over a wide pH range.

In conclusion, after a washing pre-treatment and if the percentage of incorporation is low, these residues may be incorporated into LWA. However, the recycling of APC residues from MSW incineration into LWA does not revealed any technical advantage.

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### 1. Introduction

The municipal solid waste (MSW) incineration has been increasingly used in most of developed countries, with the advantages of energy recovery and high reduction in mass and volume of the initial wastes. For minimizing the negative impact on environment resulting from emissions to air, the flue gases must be cleaned. Currently, there are scrubbers, fabric filters and/or electrostatic precipitators to take out particulate and gaseous pollutants, before gases being released into the atmosphere. However, this cleaning procedure creates hazardous air pollution control (APC) residues, which require further management. This waste has been mostly treated by a stabilization/solidification technology with hydraulic binders (usually cement) and then landfilled in appropriate disposal sites (Chandler et al., 1997; Sabbas et al., 2003; Quina et al., 2008a). The main characteristics of concern of the APC residues are the high content of heavy metals and soluble salts. The sintering/thermal processes seem to be able to solve these problems (Wang et al., 1998, 2002; Sakai and Hiraoka, 2000;

Mangialardi, 2001; Wainwright and Cresswell, 2001; Dimech et al., 2008; Haiying et al., 2011; del Valle-Zermeño et al., 2013).

Besides, the ceramic industry uses high quantities of silicate-based natural raw materials and thus it is a possible field for ash utilization (Ferreira et al., 2003). In addition, particulate materials can be incorporated into ceramic pastes without pre-treatment, and toxic heavy metal (e.g. Pb, Cd, Cr, Ni, Cu) initially present in the waste may be integrated in the final ceramic products, by choosing carefully the quantity of waste incorporated. In the literature there are references to studies involving this and similar wastes incorporation into ceramic materials, such as lightweight aggregates (LWA). In general, LWA can be obtained from natural resources (e.g. volcanic aggregates or pumice) or produced by thermal processes in kilns. The synthetic aggregates are ceramic products manufactured by thermal treatment of raw materials with expansive properties, such as perlite, vermiculite, clay and shale and industrial by-products (e.g. fly ash and slags) (Chandra and Bertsson, 2002). Lighter LWA may be obtained from coarser grain sizes made from a homogeneous paste of bloating clay, water and a small percentage of mineral oil (Chandra and Bertsson, 2002). The green pellets are sintered at high temperatures to obtain a glassy material characterized by an external hard ceramic shell and very

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porous core with holes of different sizes. The main properties of LWA are lightness, good thermal characteristics, sound insulation and fire resistance, low water absorption, chemically resistant against alkaline and acidic conditions, and high durability. Accordingly, LWA have been used to produce lightweight concrete, insulation materials, geotechnical fill, as well as used for soil engineering, in drainage systems and roofs.

In the literature there are several studies aiming to produce LWA using secondary materials (Ducman et al., 2002; Cheeseman and Virdi, 2005; Cheeseman et al., 2005; Quina et al., 2006; Chiou et al., 2006; Huang et al., 2007; Qiao et al., 2008; Gonzalez-Corrochano et al., 2009, 2012; Chen et al., 2010; Kourti and Cheeseman, 2010; Latosinska and Zygadlo, 2011; Tan et al., 2012; Hwang et al., 2012) such as combustion ashes, waste glass, sewage sludge ash, incinerator bottom ash, mining residues, heavy metal sludge, washing aggregate sludge, lignite coal fly ash, contaminated mine soil. In other cases, natural materials such as zeolitic rocks or volcanoclastites were considered as well (Gennaro et al., 2004, 2005).

To obtain lightweight aggregates it is necessary to heat raw materials at high temperatures and to entrap the released gases within a highly viscous liquid phase; during the cooling period an external glassy film with very low porosity, homogeneous and resistant is formed (Gennaro et al., 2004). The main sources of gases are the combustion reactions of organic compounds (CO<sub>2</sub> and water), the decomposition of carbonates (CO<sub>2</sub>), the reduction of ferric iron (O<sub>2</sub> and CO<sub>2</sub>), the sulfides oxidation (SO<sub>x</sub>) and other compounds that can be released from raw material (F, Cl). The commercial LWA are in general made from appropriate clays, by adding small amounts of oil to improve the gas release.

This study focuses on the analysis of the effect of the APC residues (with and without washing pre-treatment) on the main technological properties of LWA as well as on the environmental impact due to leaching processes with water and acidic solutions.

## 2. Materials and experimental procedures

### 2.1. Materials

In our work, specific natural clay supplied by an industrial plant from the central region of Portugal, that processes about 24 t/h of clay, was considered. The APC residues were collected from a mass burning incinerator that has a nominal capacity of 28 t/h, comprising gas cleaning residues from economizer, semi-dry scrubbers (with lime and activated carbon) and fabric filters. In addition, about 1% of oil (also supplied by the industry) and 20–25% of water were used in the formulations tested. Both APC residues and natural clay were characterized in our previous works (Quina and Quinta-Ferreira, 2002; Quina et al., 2006, 2008b, 2009, 2010, 2011a, 2011b). However, Table 1 summarizes the most important properties of the materials used in this study.

The APC residues were tested as-received from the industrial plant and after a washing pre-treatment with water. The washing treatment consisted of mixing the residue with 10 times of distilled water (L/S 10), during 10 min. The suspension was then filtered and the solids were dried over night at 105 °C. The washing treatment aimed at removing water soluble compounds, in particular the chlorides.

### 2.2. Production of LWA

The industrial process for producing LWA involves a tubular kiln with an angle of tilt less than 5° and with two different zones: 40 m long for drying the particles with a slow rotation (residence time of about 1.8 h); 20 m long where firing takes place at about 1170 °C (residence time of 8–10 min). The granules are fed at the higher end of the kiln in a counter flow process, and as the material moves along the tube the expansion takes place. However, in our laboratory it was not possible to simulate this process, but the industrial methodology used for quality control was followed. The raw materials (waste, clay, water and oil) were thoroughly mixed according to the different formulations summarized in Table 2, including clay, APC residues, about 20% of water and 1% of oil. For each case, spherical pellets were formed by hand, with mean mass values and median diameters reported in Table 3. Indeed, an initial attempt was made to use a rotary drum pelletiser, but since the size and weight of the granules were very different, it was difficult to replicate the tests in the laboratory. Following the conditions used for quality control at the industrial plant, the LWA were obtained by drying at 200 °C (at least 2 h) in a first step and then by firing the spherical pellets in a chamber furnace oven at 1170 °C (during 8 min) over solid CaCO<sub>3</sub>.

### 2.3. Technological properties

The bulk density,  $\rho_b$ , was determined in triplicate by weighing the mass of one litter of fired aggregates, which is also the method used at industry and recommended in the standard EN 1097-3. The apparent specific density,  $\rho_a$ , of LWA was determined through three methods: Met1 – using Archimedes' principle with water (in triplicate), considering 10 aggregates Eq. (1); Met2 – assuming a spherical geometry, Eq. (2); Met3 – based on the displacement method by immersing 6 aggregates in a certain amount of sand,  $V_f$ , and measuring the initial volume of sand in a graduated cylinder,  $V_{sand}$ , Eq. (3).

$$\rho_{a,1} = \frac{M_A}{M_1 - M_2} \rho_{H_2O} \quad (1)$$

$$\rho_{a,2} = \frac{6M_A}{\pi D^3} \quad (2)$$

**Table 1**

Properties of the clay, APC residues as received and after washing.

	Main oxides (%)							Total elemental content (mg/kg)					
	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	Na <sub>2</sub> O	K <sub>2</sub> O	Pb	Cr	Cu	Ni	Cd	Zn
Clay	55.4	18.2	8.0	1.9	3.9	<0.5	4.3	nd	nd	nd	nd	nd	nd
APC <sub>residues</sub>	14.1	7.4	2.3	41.9	1.0	3.7	3.2	1,924	232	456	121	57	4,308
APC <sub>washed</sub>	18.7	9.8	3.1	36.2	2.2	1.3	0.8	nd	nd	nd	nd	nd	nd
	Measured in the eluate (L/S = 10; concentration in mg/kg)												
	Moisture (%)	LOI (%)	$\rho_r$ (g/cm <sup>3</sup> )	pH	EC (mS/cm)	TDS (g/kg)	Pb	Cr	Cu	Ni	Cd	Zn	Cl <sup>-</sup>
Clay	15	6.6	2.66	8.49	0.346	2.65	<0.9	<1.17	<0.9	<1.45	<0.07	0.23	ND
APC <sub>residues</sub>	0.85	8.3 <sup>*</sup>	2.52	12.5	37.5	222	324	5.97	1.38	2.54	0.15	50.5	98,400
APC <sub>washed</sub>	0.93	nd	nd	12.1	nd	24.5	130	9.0	<0.9	<1.45	<0.07	18.0	ND

<sup>\*</sup> Loss on ignition measured at 750 °C; nd – not determined; ND – not detected by titration with AgNO<sub>3</sub>.

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