Research paper

Compatibility analysis of municipal solid waste incineration residues and clay for producing lightweight aggregates

Margarida J. Quina [a,*], Marisa A. Almeida [b], Regina Santos [b], João M. Bordado [c], Rosa M. Quinta-Ferreira [a]

[a] CIEPQPF — Research Centre on Chemical Processes Engineering and Forest Products, Department of Chemical Engineering, University of Coimbra, Rua Sílvio Lima, 3030-790 Coimbra, Portugal
[b] Centro Tecnológico da Cerâmica e do Vidro, Ap. 8052, 3020-053 Coimbra, Portugal
[c] Department of Chemical Engineering, Instituto Superior Técnico, Av. Rovisco Pais, 1049-001 Lisboa, Portugal

ABSTRACT

This work provides insight into the most important properties of air pollution control (APC) residues from municipal solid waste incineration (MSWI) and clay used at industrial level aiming at producing a ceramic material known as lightweight aggregates (LWA). Both clay and APC residues were characterized to assess the most important chemical, physical, thermal and mineralogical properties to evaluate their compatibility. The maximum percentage of incorporation tested was 10% of APC residues. The results showed that the waste does not own bloating properties and it is not expectable other improvements into the technological properties of LWA, since the fluxing components contribution is high and the amount of silica is low. Although during the thermal analysis of the APC residues the loss on weight is high (about 32% until 1200 °C), they seem not to be suitable as a gas-release additive, once for 10% of incorporation the external shell appears to be significantly broken. The environmental impact due to leaching of pollutants from the LWA incorporating less than or equal to 5% of waste appears not to be relevant. In conclusion, the results showed that synthetic LWA may be considered as a possible approach to recycling of APC residues in case of low amount of waste is used or if a pre-treatment is performed.

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1. Introduction

The municipal solid waste (MSW) incineration has been widely used in most of developed countries, as part of integrated management systems for their destruction and energy recovery. However, one of the important shortcomings of this process is the generation of hazardous air pollution control (APC) residues, which comprise particles captured in the flue gas cleaning system prior to discharge into the atmosphere (Chandler et al., 1997; Sabbas et al., 2003; Quina et al., 2008a). APC residues may represent 2–3% of the MSW incinerated, and thus a huge amount of these wastes have been landfilled in the world. These residues are produced at high-temperature, and therefore they are thermodynamically unstable and highly reactive under ambient conditions (Sabbas et al., 2003). In the literature there are various studies aimed at APC residues characterization, where properties such as moisture, loss on ignition (LOI), size distribution, density, porosity, specific surface area, morphology, total elemental content (TC), total availability (TA) and the leaching behavior, mineralogy and thermal behavior can be found (Chandler et al., 1997; Quina and Quinta-Ferreira, 2002; Quina et al., 2008b, 2009, 2011a, 2011b). The principal characteristics of concern are the high heavy metals content, soluble salts that leach in quantities that do not comply with the regulatory constraints, as well as the trace organic pollutants. Other studies analyze in particular the thermal behavior of such incinerator residues (Tettamanti et al., 1998; Wang et al., 1998; Sakai and Hiraoka, 2000; Mangialardi, 2001; Wang et al., 2002). A pre-treatment stage involving washing with water can be applied to APC residues in order to remove soluble salts prior to further utilization or chemical stabilization processes (Mangialardi, 2001; Nzihou and Sharrock, 2002; Sabbas et al., 2003). Salts may account for up to 10 to 20% of the material and are responsible for many of the negative effects of such residues. Some studies have indicated that a liquid-to-solid ratio of 10 allows for extraction of more than 90% of the soluble salts (mainly chlorides) (Nzihou and Sharrock, 2002). However, for evaluating the overall benefits of a washing pre-treatment, the shift of the pollutants from solid to liquid streams must be considered (Sabbas et al., 2003). Indeed, to assess the suitability for a specific application, besides the environmental impact, the ability for processing and the technical performance must be also considered (Ferreira et al., 2003).

In this framework, some European countries have devoted efforts to find feasible applications for APC residues rather than just disposed of in environmental acceptable landfills (Sabbas et al., 2003). Although these residues generally have limited applications, thermal treatments can produce a material that is environmentally stable, and may be further used for producing ceramics (Quina et al., 2008a). In this scope,

* Corresponding author. Tel.: +351 239 798 700; fax: +351 239 798 703.
 E-mail address: guida@eq.uc.pt (M.J. Quina).

the production of lightweight aggregates (LWA) has been considered for many secondary materials (Wainwright and Cresswell, 2001; Ducman et al., 2002; Cheeseman and Virdi, 2005; Cheeseman et al., 2005; Chiou et al., 2006; Quina 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) such as combustion ashes, waste glass, sewage sludge ash, incinerator bottom ash, mining residues, heavy metal sludge, washing aggregate sludge, lignite coal fly ash, and contaminated mine soil. Indeed, LWA can be defined as materials lighter than water and more porous than sand, gravel or ground rock (commonly referred to as "dense" aggregates) and can be produced from natural resources (e.g. volcanic aggregates or pumice), requiring only mechanical treatment, or obtained by thermal processes in kilns (Chandra and Berntsson, 2002; de' Gennaro et al., 2004, 2005). In a broad sense, synthetic aggregates are ceramic materials obtained by thermal treatment of materials with expansive properties, such as (i) perlite, vermiculite, clay and shale and (ii) industrial by-products (e.g. fly ash, slags, and sludge) (Chandra and Berntsson, 2002). One of the most widespread types of LWA is produced from expanded clay and commercially known as Leca® (Lightweight Expanded Clay Aggregates), or from coal fly ash and referred as Lytag®. Expansion (bloating) occurs by heating materials to fusion temperature, where pyroplasticity and formation of gas must take place simultaneously. The technological applications of LWA are mostly linked to the low density and high compressive strength (de' Gennaro et al., 2005).

The valorization of fly ash and/or APC residues from MSW incineration in LWA has been analyzed only in few studies (Quina et al., 2006; Huang et al., 2007; Hwang et al., 2012; Tan et al., 2012). However, other sintered or melting materials and ceramic bricks have been considered (Wang et al., 1998; Sakai and Hiraoka, 2000; Mangialardi, 2001; Wang et al., 2002; Dimech et al., 2008; Haiying et al., 2011; Valle-Zermeño et al., 2013). Some studies point out that a washing treatment of APC residues with water to separate soluble salts may be important in the scope of their recycling (Mangialardi, 2001; Sabbas et al., 2003; Quina et al., 2011b), but attention should be given to the mobilization of toxic heavy metals (Nzihou and Sharrock, 2002). The stabilization of most heavy metals can be accomplished with chemical additives (Nzihou and Sharrock, 2002; Quina et al., 2010). It should be noted that an important benefit of recycling polluted wastes into synthetic aggregates is the low leachability usually observed (van der Sloot et al., 2001).

This work focuses on the characterization of APC residues from MSW incineration and natural clay currently used in industry in order to assess the possibility of producing lightweight aggregates.

2. Materials and methods

2.1. Characterization of APC residues and natural clay

In our study, APC residues samples were collected in an incineration plant in the region of Lisbon, comprising gas cleaning residues from economizer, semi-dry scrubbers (with lime and activated carbon) and fabric filters; the sample of natural clay was taken in an industry that produces LWA in the center region of Portugal. APC residues were extensively characterized in our previous study (Quina et al., 2008b), where moisture content, loss on ignition (LOI), particle size distribution, density, porosity, specific surface area, morphology, total elemental content (TC), total availability (TA), leaching behavior with common compliance tests, and the acid neutralization capacity (ANC) can be found. The main mineralogical crystalline phases were identified, and the thermal behavior of the APC residues is also shown. Those parameters were determined for 5 different samples, collected over two years. The specific features of the leaching behavior of these residues may be found in other publications (Quina et al., 2009, 2010, 2011a, 2011b). In the present study, the analysis of the thermal behavior was deepened for one sample (referred as A2), aiming at considering the recycling of

the waste by its incorporation in LWA, and includes simultaneous thermal analysis (STA), X-ray diffraction (XRD) and heating microscopy.

The natural clay was characterized regarding moisture content through weight loss over night in an oven at 105 °C; loss on ignition (LOI) was determined as the weight loss after exposing the clay at 1000 °C using a muffle furnace during 1 h; the real specific density was determined by using helium pycnometry, Micromeritics-AccuPyc 1330, based on the amount of displaced gas (helium); the total dissolved solids (TDS), pH and electrical conductivity were determined in the liquid obtained at liquid-to-solid ratio (L/S) of 10 mL/g, under agitation for 24 h with demineralised water; the particle size distribution was determined by sedimentation, Micromeritics-SediGraph 5100. In addition, the elemental composition of clay was assessed using X-ray fluorescence (XRF), Philips-PW 1480, equipped with an Rh X-ray tube. The samples were dried, finely grinded and mixed with $Li_2B_4O_7$ (66.5%) and $LiBO_2$ (33.5%). The major elements were reported as wt.% oxide. Powder XRD, Philips-PW 1710, with Cu Kα radiation (40 kV and 30 mA) was also employed and the minerals were recognized according to the data base ICDD (International Centre for Diffraction Data).

The thermal behavior of both clay and APC residues was obtained by using STA, Netzsch — STA 449C, with a heating rate of 10 °C/min in air. This test allowed us to obtain simultaneously the thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) of samples previously milled to less than 75 μm, using about 20–30 mg of powder. Heating microscopy, Leitz-2A, enabled us to observe with video recording 3 mm sided cubic samples of clay and APC residues by heating them to 1500 °C at a rate of 10 °C/min.

The total content (TC) of previously calcined APC residues at 1100 °C for 1 h was determined by acid digestion with aqua regia, using 3 mL of HNO_3 (65%) and 10 mL of HCl (38%) in a digestion bomb (Parr 4744). The solid sample (0.4 g) and the acid mixture (13 mL) within the bomb were heated in a conventional oven during 6 h at 150 °C. These analyses were carried out in triplicate.

2.2. Washing of APC residues

The washing treatment of APC residues involved extractions with water in one or two stages, at L/S of 10 mL/g, using an open beaker under agitation through a magnetic stirrer at 120 rpm. The separation of both phases was conducted by vacuum filtration through a 0.45 μm membrane filter. The solid and liquid phases were collected and preserved for further chemical analysis. The times of extractions were 0.5, 1, 5, 10, 30, 60, 480 min and 24 h. In the washing solutions pH, electrical conductivity (EC), total dissolved solids (TDS), concentration of the soluble ions (Cl^-, Na^+, K^+) and toxic heavy metals (Pb, Zn, Cr) were measured. The flame atomic absorption spectrometry (FAAS), Perkin Elmer-3300, and ion chromatography, Waters-Action Analyzer, were used as the main analytical techniques.

Moreover, some experiments were conducted by adding a small amount of phosphoric acid, in order to reduce the mobilization of toxic heavy metals (mainly Pb which is the most critical element). The experimental conditions involved 248.5 mL of water plus 1.5 mL of H_3PO_4 (85%) and 25 g of APC residues, which means a L/S of 10 L/kg for 10 min.

2.3. Preparation and characterization of LWA

The LWA were produced not only using natural raw materials but also incorporating APC residues, according to the formulations shown in Table 1. The LWA were prepared and sintered based on the method used for quality control in the laboratory of the industrial plant that produces commercial LWA. The experimental procedure to obtain LWA followed different phases: homogenisation of clay and APC residues during 30 min; blend the mixture by adding about 20% of water and 1% of oil (as expanding agent) to achieve sound pelletization

Table 1
Formulations tested and firing conditions used to produce lightweight aggregates.

Formulation	APC residues (%)	Clay (%)	Firing conditions
LA0	0	100	8 min at 1100 °C
LA1	1	99	8 min at 1100 °C
LA5	5	95	8 min at 1100 °C
LA10	10	90	8 min at 1070 °C

in a mechanical device; the pellets were first dried at 110 and 200 °C for 2 h, and then fired in a muffle furnace over $CaCO_3$ at 1100 or 1070 °C for 8 min to obtain a glassy material. The formulation LA10 requires lower temperature (1070 °C) otherwise it disintegrates during the firing process.

The bulk density was determined by weighing the mass of 1 L of aggregates, and the density of particles was assessed by displacement of mercury. The mechanical strength was measured according to DIN 4226-Part 3, using a specific press (Tecnilab LST) for applying a pressure to a container with 1 L of LWA during 100 s.

In addition, to evaluate the potential environmental impact of LWA to soil and water, the classic standards DIN 38414-S4 (leaching for 24 h in agitation, at L/S equal 10) and TCLP (leaching test with acetic acid solution pH 2.88 and L/S 20, over 18 h) were used. In both leaching tests, the LWA were milled up to 1 mm, with a hammer mill. The metals Pb, Cd, Cr, Ni and Cu were quantified through graphite furnace atomic absorption spectrometry (GFAAS), Perkin Elmer-AAnalyst 600, Zn by flame atomic absorption spectrometry (FAAS), Perkin Elmer-3300. The anions Cl^- and SO_4^{2-} were measured by ion chromatography, Waters-Action Analyzer, and F^- by potentiometry.

3. Results and discussion

3.1. Characterization of natural clay

The natural clay considered in our study has been used in industry for producing LWA of expanded clay. Table 2 summarizes some of the properties of the clay, such as moisture content (MC), loss on ignition (LOI) and real specific density (ρ_r). In addition, certain properties were measured in eluates obtained by leaching clay at L/S of 10:1 and under agitation for 24 h. As expected, the values determined in the eluate for EC, TDS and metal concentrations are low, since the clay was stored next to the industrial plant, in a stockpile for aging for 2 to 3 months under weather conditions (rain, wind, sun, etc.). At industrial level, it is well known that this aging period improves the technological properties when compared with freshly-mined clay.

The particle size distribution determined by sedimentation showed that the clay comprises 36.3% of clay fraction (<2 μm), 56.7% of silt (2–63 μm) and 7.1% of sand fraction (>63 μm).

The analysis of major elements of natural clay using XRF revealed the chemical composition indicated in Table 3. Thus, the requirement for bloating materials indicated in the literature (Riley, 1951; de' Gennaro et al., 2004; Chiou et al., 2006; Gonzalez-Corrochano et al., 2009; Latosinska and Zygadlo, 2011) is observed for clay since SiO_2: 48–70%, Al_2O_3: 8–25% and fluxing oxides (Fe_2O_3 + CaO + MgO + Na_2O + K_2O) 4.5–31%. Indeed, Fig. 1 shows that by plotting the normalized composition

of the clay in the ternary diagram SiO_2–Al_2O_3–fluxing, it is within the bloating area established by Riley (1951). Therefore, enough viscous phases are expected during the sintering of LWA for trapping gas released. For comparison purposes, Table 3 includes also data from the literature (Chiou et al., 2006; Latosinska and Zygadlo, 2011, termed as [C1] and [C2], respectively) and in general close values were observed mainly for SiO_2 and Al_2O_3. The clay characterized by Latosinska and Zygadlo (2011) comprises lower content of Fe_2O_3 and fluxing, but it is still recommended for bloating materials.

Moreover, major (Si, Al, Fe and K) and minor (Mg, P, S, Ba, Sr, Rb, Zn, Cu, Ni, Ti, Ca, Cd and Mn) elements were detected by XRF in clay. It must be noticed that sodium and chloride were not identified in this analysis.

Crystalline phases present in the clay were determined by XRD and the spectrum is shown in Fig. 2. The data point out that quartz (SiO_2) and illite ($(K,H_3O)(Al,Mg,Fe)_2(Si,Al)_4O_{10}$) are the major crystalline phases present together with some hematite (Fe_2O_3), smectite ($(Na, Ca_{0.5})_{0.3}(Al,Fe,Mg)_2(Si,Al)_4O_{10}(OH)_2 \cdot nH_2O$), kaolinite ($Al_4Si_4O_{10}(OH)_8$) and traces of feldspar ($(Na,K)AlSi_3O_8$) and calcite ($CaCO_3$). Although the XRD analysis was carried out qualitatively, by taking into account the intensity of peaks at a five star rating, quartz (****) and illite (***) are the most significant crystalline phases, and kaolinite, smectite and hematite (*) were detected in minor amounts. In fact, according to the literature (Konta, 1995; de' Gennaro et al., 2004; Latosinska and Zygadlo, 2011) the main minerals of expanding clays are variable amounts of quartz, illite, smectite, kaolinite and numerous accessory minerals. Increasing amounts of kaolinite and mica do not favor the production of LWA (Konta, 1995).

The thermogravimetric behavior of the clay, Fig. 3, was analyzed by STA between 20 and 1200 °C yielding a total weight loss of 8.6%, which is the result of two main contributions: 3.1% till 330 °C and 4.9% up to about 850 °C. The initial reactions are predominantly endothermic (DSC curve) due to the loss of free water and the exothermic reactions were observed probably due to the oxidation of organic matter. The mass loss observed to around 850 °C probably resulted from carbonate minerals decomposition. In the last phase of the thermogram (higher than 850 °C) a low weight loss of 0.6% and a significant heat absorption by the sample are observed. In the literature, few studies were found concerning TGA analysis of such clays. Chen et al. (2012) analyzed sediments with a similar composition to the clay and the thermogravimetric behavior revealed a global weight loss of about 7%. In Fig. 3 the results observed for the residue and for a mixture of both are also indicated, but these data will be discussed in the next section.

Some heating microscope images of a cubic clay sample, during sintering at a heating rate of 10 °C/min are shown in Fig. 4, where the dimensional changes of the sample can be detected by comparing the silhouette captured from 40 to 1362 °C. Until 927 °C the reduction of the sample dimensions was almost undetectable, and thus this may be considered the initial sintering temperature. This marks the beginning of changes to the phase state in the structure of the sintered material (Latosinska and Zygadlo, 2011). The final sintering temperature is about 1200 °C, and at 1224 °C the softening phenomenon is perceptible. The maximum expanded temperature occurs at 1300 °C, where the image of the sample achieved the largest area in the projection on the field. By increasing the temperature further, the sample becomes softer and viscosity significantly decreases until 1362 °C, where the sample is

Table 2
Properties of the clay and APC residues (metal and Cl^- concentrations in mg/kg).

	MC (%)	LOI (%)	ρ_r (g/cm³)	Measured in the eluate (L/S = 10)											
				pH	EC (ms/cm)	TDS (g/kg)	Pb	Cr	Cu	Ni	Cd	Zn	Ca	K	Cl^-
Clay	15	6.6	2.66	8.49	0.346	2.65	<0.9	<1.17	<0.9	<1.45	<0.07	0.23	749	104	nd
APC$_{residues}$	0.85	8.3[a]	2.52	12.5	37.5	222	324	5.97	1.38	2.54	0.15	50.5	40,900	26,600	98,400

nd — not determined.

[a] Determined at 750 °C.

Table 3
Chemical composition of clay (through XRF) and APC residues (% w/w).

Ref.	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	ΣF[a]	Ti_2O	MnO	P_2O_5	LOI
Clay_our study	55.4	18.2	8.0	1.9	3.9	<0.5	4.3	18.1–18.6	0.9	0.11	<0.5	6.8
[C1]	62.8	16.8	7.0	1.0	2.3	1.7	3.1	15.1	ni	ni	ni	ni
[C2]	68.1	15.2	4.3	0.74	1.05	0.4	1.9	8.4	0.77	0.015	ni	7.1
APC residues												
A2	14.1	7.4	2.3	41.9	1.0	3.7	3.2	52.1	nd	nd	nd	8.3[b]
A2_1st_extr.	18.7	9.8	3.1	36.2	2.2	1.3	0.8	43.5	nd	nd	nd	nd
A2_2nd_extr.	26.1	10.3	4.3	33.3	2.3	0.3	<DL	40.2	nd	nd	nd	nd

[C1] — Chiou et al., 2006; [C2] — Latosinska and Zygadlo, 2011; ni — not indicated; nd — not determined; DL — detection limit.

[a] ΣF = Fluxing oxides = Fe_2O_3 + CaO + MgO + Na_2O + K_2O.
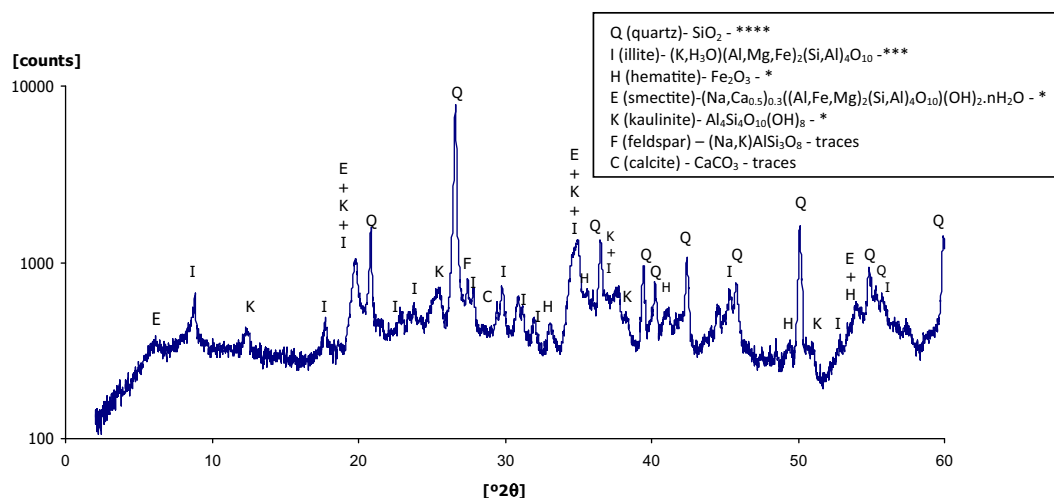
[b] Determined at 750 °C.
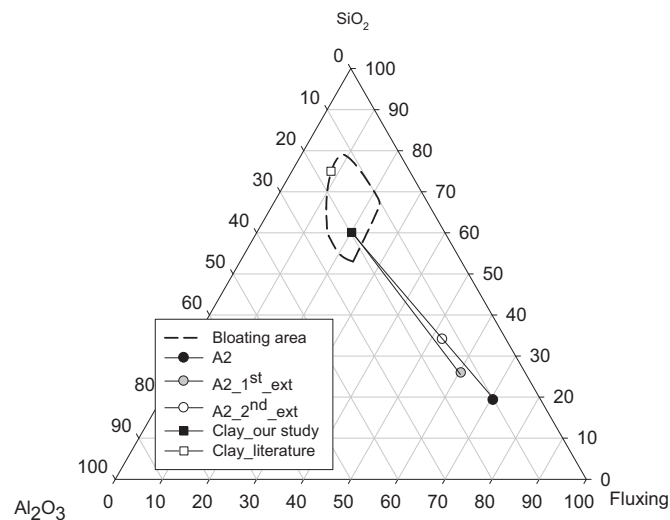
Fig. 1. Composition of clay of our study and from the literature (Latosinska and Zygadlo, 2011), as well as APC residues as received (A2) after one extraction (A2_1st_ex.) and after two extractions (A2_2nd_ext).

completely melted, and this may be considered the fluxing temperature. Similar measurements were reported in the literature (de' Gennaro et al., 2005; Latosinska and Zygadlo, 2011) for clay and Neapolitan yellow tuff, respectively, aiming at producing lightweight aggregates. It is important to notice that these results were obtained at a heating rate of 10 °C/min, and thus the sintering and bloating processes which occur in the industrial kiln will be different. Indeed, the industry that uses the same clay produces LWA at a maximum kiln temperature of 1170 °C.

3.2. Characterization of APC residues before and after washing

As mentioned previously, APC residues were extensively characterized in our former studies (Quina and Quinta-Ferreira, 2002; Quina et al., 2008b, 2009, 2010, 2011a,b). Nevertheless, Table 2 summarizes the same properties as the ones reported for clay for comparison purposes, and the differences are rather clear. Indeed, contrarily to the results observed to clay, the values of EC, TDS and metals determined in leaching solutions with water were high. In particular, the leaching of Pb exceeds the environmental criteria even for disposal of this material in hazardous landfills. Therefore, one important point of this research will be the evaluation of the possible immobilization of heavy metals into the ceramic matrix. The particle size distribution of APC residues diameters was between 0.001 and 1 mm being characterized by a mean diameter of 124 μm and a median of 86 μm (Quina et al., 2008b). This waste was also analyzed through XRF, which allowed the identification of about 20 different elements: Ca, Cl, K and S as major elements; Fe, Cr, Ti, P, Si, Al, Mg, Na, Cu, Zn, Ba, Sn, Sb, Sr, Br and Pb as minor. However, given that XRF analysis was only qualitative, the oxide composition was calculated based on the total elemental content (of sample with reference A2), determined by acid digestion (Quina et al., 2008b), and the values obtained are indicated in Table 3. These records were also represented in the Riley diagram in Fig. 1, after normalization to 100%, where it is observed that the composition is far from the bloating area. Therefore, for recycling APC residues into LWA, the use of a bloating material is essential, otherwise the material expansion will not occur. Also other studies (Chen et al., 2010) showed that MSW incineration APC residues were not within the limits of the bloating region of the ternary diagram (Riley diagram), due to the low content of SiO_2. Hence, the sintering at high temperature does not afford a liquid phase with enough viscosity. On the other hand, these residues had high CaO content (>30%), while the preferred content

**Fig. 2.** XRD diagram for natural clay.

Fig. 3. Simultaneous thermal analysis (STA) of the clay (—), APC residues (—·—) and a mixture of both (– –).

Fig. 4. Heating microscopy images of a cubic clay sample (3 × 3 × 3 mm) at different characteristic temperatures during sintering up to 1362 °C with heating rate 10 °C/min.

$(CaO + MgO)$ is normally not more than 7%. Similarly, by normalizing for 100% the data of the chemical composition determined by XRF for similar residues, Dimech et al. (2008) found a fluxing, SiO_2 and Al_2O_3 of 82.6, 12.5 and 4.9%, respectively; after washing the APC residues those values were changed for 77.3, 15.0 and 7.7%. By contrast, in our previous study (Quina et al., 2006) it was shown that there are wastes, such as coal fly ashes, with bloating properties which allow in some cases their direct use for producing LWA.

Fig. 5. XRD-diagram for APC residues as received and after calcined at 700, 900 and 1100 °C: P = $Ca(OH)_2$; Ha = NaCl; S = KCl; C = $CaCO_3$; A = $CaSO_4$; Q = SiO_2; G = $Ca_2Al(AlSiO_7)$; Cl = $Ca(OH)Cl$; H = Fe_2O_3; An = $Ca_3Fe_2(SiO_4)_3$).

Table 4
Total elemental content and leaching behavior of APC residues (mg/kg), before and after calcination.

Sample	Total content						Leaching with water, at L/S = 10 for 24 h					
	Pb	Cd	Zn	Cr	Ni	Cu	Pb	Cd	Zn	Cr	Ni	Cu
A2	2020	64	4308	232	121	456	353	<0.2	28	6.9	<1.5	<0.9
A2 calc_1100 °C	212	1.9	3042	348	117	428	<0.9	<0.2	<0.2	66	<1.45	<0.9
RL[a]							20	5	100	5	20	100

[a] Regulatory limit for hazardous waste landfills.
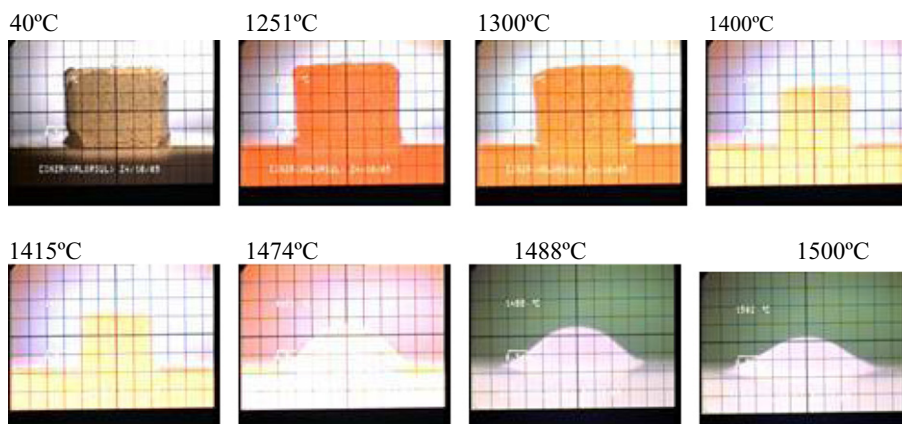
Fig. 6. Heating microscopy images of a cubic APC residues sample (3 × 3 × 3 mm) at different temperatures up to 1500 °C with heating rate 10 °C/min.

Moreover, crystalline phases present in the APC residues were determined by XRD, for the sample as received and after calcined at 700, 900 and 1100 °C, Fig. 5. Initially the main phases identified were halite (NaCl), sylvite (KCl), calcite (CaCO₃), anhydrite (CaSO₄), quartz (SiO₂), gehlenite (Ca₂Al(AlSiO₇)), hematite (Fe₂O₃), portlandite (Ca(OH)₂) and calcium hydroxychloride (Ca(OH)Cl). Most of these phases were identified in other studies as well (Dimech et al., 2008). The interpretation of the spectra after calcination is not easy, since solid phases may suffer only slight rearrangements which can originate distinct peaks. Nevertheless, it can be concluded that SiO₂, KCl, NaCl, CaSO₄ and Ca₂Al(AlSiO₇) are stable for all temperatures; CaCO₃ was identified at 700 °C but not at 900 and 1100 °C; Ca(OH)₂ and Ca(OH)Cl were not identified at any of the calcination temperatures; Fe₂O₃ was not observed at any of the calcination temperatures but probably a new phase andradite (Ca₃Fe₂(SiO₂)₃) emerged, which is characterized by a greenish-gray color and thus it may be responsible for the appearance of the residue after calcination. Although the CaO is a product of thermal decomposition of the phases of calcium, this mineral was not identified in the XRD spectra, probably due to its high reactivity, and thus it tends to react with the phases of silicon, aluminium and iron.

The total elemental content of the calcined residue at 1100 °C concerning the toxic heavy metals are reported in Table 4, and during this thermal treatment about 14.6% of the global mass was lost due to decomposition of mineral phases (e.g. Ca(OH)Cl). These results show that besides the changes described above for the major mineral phases, also the total content and the leaching behavior of toxic heavy metals were changed. The total content losses of Pb, Cd, Zn, Ni and Cu were about 91, 97, 40, 18 and 20%, respectively, and the leaching release of all these metals were well below the regulatory thresholds for hazardous waste landfills. Regarding Cr the observed behavior is different. Indeed, whereas it was not volatilized during calcination at 1100 °C, the amount released by leaching increased strongly, becoming a potential problematic element. This behavior has been reported in the literature, and the most likely reason is the formation of CaCrO₄ at high temperatures, whose phase is leachable in water (Kirk et al., 2002; Wang et al., 2002).

It is important to notice that the APC residues when analyzed through STA, Fig. 3, by increasing the temperature from 20 to 1200 °C, in a rate of 10 °C/min, a loss of 32.2% was detected. The significant loss observed when temperature is in the range of 600–750 °C is mainly due to the decomposition of Ca(OH)Cl(s) to CaO(s) and HCl(g) and CaCO3(s) to CaO(s) and CO₂(g) (Quina et al., 2008b). For higher temperatures than 750 °C the weight loss is most likely related with the partial decomposition of the NaCl and KCl phases, with the release of HCl gas. The endothermic behavior of the clay and the mixture (clay +5%APC) for a temperature higher than 1000 °C correspond to the sintering of the body.

Some heating microscope images of cubic APC residues samples, during sintering at a heating rate of 10 °C/min are shown in Fig. 6. The reduction of the sample dimensions during the sintering process can be noticed, by comparing the images from 40 to 1415 °C. Indeed, during this heating process the sample kept the cubic shape up to 1415 °C. At 1474 °C the sample melted, and by increasing the temperature further, the glassy phases become softer and the viscosity significantly decreases.

Table 5
Total content (TC), and extracted amounts in the elutes by washing process.

Test	Pb	Cd	Zn	Cr	Ni	Cu	K	Na	Ca	Cl	SO₄	Si	Al	Fe
	(mg/kg)						(g/kg)							
TC	2020	64	4308	232	121	456	26.7	27.6	299	nd	nd	65.9	39.1	16.1
A2W_10 min	408	<DL	51	3.5	0.09	4.2	21.9	20.1	104	101.1	12.4	nd	nd	nd
A2W_24 h_1ext	355	<DL	28	6.9	0.09	1.4	25.4	21.8	95.2	108.5	13.4	<DL	<DL	<DL
A2W_24 h_2ext	130	<DL	18	9.0	0.1	0.05	6.2	5.6	25.5	nd	nd	nd	nd	nd
A2W_PO₄	18	<DL	1.5	5.1	<DL	0.23	25.6	20.9	66.6	nd	nd	nd	nd	nd

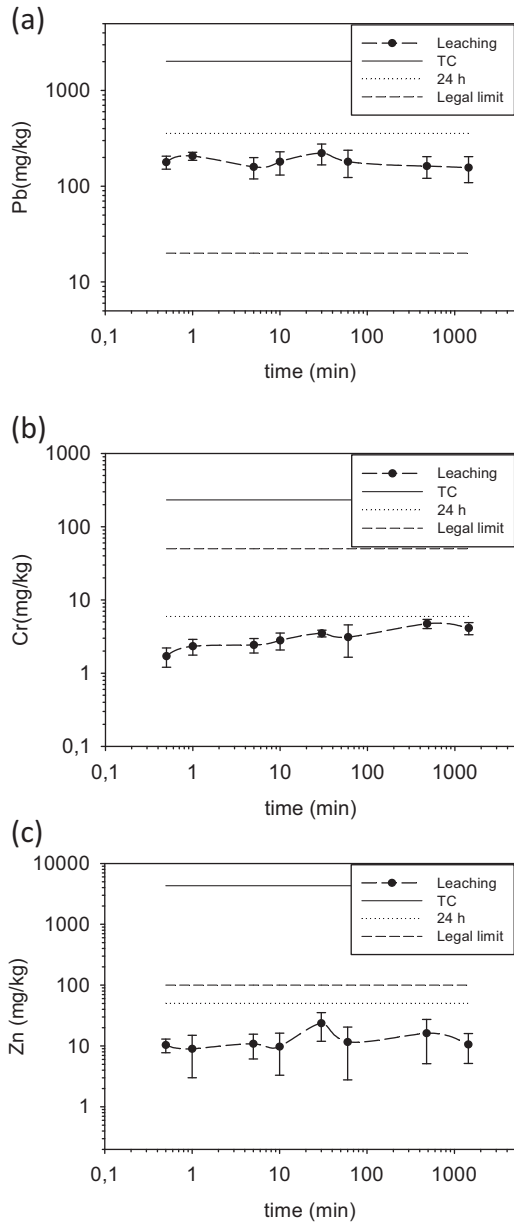DL — detection limit of FAAS; nd — not determined.

Fig. 7. Released amounts of (a) Pb, (b) Cr, and (c) Zn as a function of the time of the extraction from 0.5 min to 24 h.

As previously referred by taking into account the Riley diagram, Fig. 1, no evidence of the bloating process was observed in this case.

As aforementioned, the washing of APC residues can be regarded as a first step of other possible treatments or recycling of the waste as a valuable material (Mangialardi, 2001; Nzihou and Sharrock, 2002;

Sabbas et al., 2003). This process entails the extraction of solid particles with a liquid phase aiming to improve the technological properties of the residue. The results obtained in these washing tests showed that some elements may be released in water in high quantities. Table 5 summarizes some data measured in the eluates obtained after 10 min (A2W_10 min) and 24 h (A2W_24 h_1ext) of washing, after a second extraction with 24 h (A2W_24 h_2ext), and after washing the waste with phosphoric acid (A2W_PO$_4$).

In general, the kinetic of washing is fast. After only 10 min of extraction, some elements were released in water in high quantities (for example K$^+$, Na$^+$, Cl$^-$ and SO$_4^{2-}$), whereas for elements such as Pb, Zn, Cr, Ni and Cu much lower quantities were extracted when compared to TC, and others (Cd, Si, Al and Fe) were below the detection limit (DL).

By using phosphoric acid in the washing procedure, a positive effect in heavy metals (Pb, Zn, Ni and Cu) was observed, while for other elements (e.g. K, Na) a minor effect was detected.

In order to follow the kinetic of releasing of Pb and Cr, dynamic leaching processes were conducted, and the results plotted in Fig. 7, where the TC for each element, the regulatory limit for hazardous landfills and the values obtained by performing a washing process within 24 h were also included. From Fig. 7a it can be observed that the legal limit for Pb is always largely exceeded; the first sample was taken at 0.5 min and also in this case the Pb concentration goes beyond the limit. The amount of Cr shows a slightly increase over time, and the regulatory limit can be surpassed after 60 min, Fig. 7b. The leaching behavior of Zn is shown in Fig. 7c, and in this case higher variability is detected between replicates. However, the legal limit was never exceeded.

According to the overall data, the washing process should be performed at L/S 10, over 10 min, at room temperature, using a small amount of phosphoric acid (adding at least 0.06 mL/g of APC residues). By analyzing a sample of washed APC residues by XRD, the disappearance of the peaks corresponding to NaCl, KCl and CaClOH was observed (Quina et al., 2008b). It is significant to note that for sample A2 during the washing process the mass loss was about 22% due to solubilization. Moreover, by heating the washed residue until 1200 °C an additional loss of 22% was further observed (Quina et al., 2008b), leading to a significant global loss of mass.

3.3. Lightweight aggregates

In the previous sections, it was shown that the clay has characteristics of bloating, whereas the APC residues has high fluxing components (near to 52%) and low silicon oxide content. Thus, aiming to produce LWA, it is expectable that low percentages of incorporation will be feasible, and in this section this aspect will be explored.

In general, some conditions were required for producing bloating materials (de' Gennaro et al., 2004). Namely, the material must yield a high temperature glassy phase with appropriate viscosity to trap the gases; some chemical reactions must occur, releasing gas at the same time that the glassy phase was formed; and in the cooling phase an external glassy shell must be formed. The gases produced may comprise CO$_2$, CO, H$_2$O, O$_2$, SO$_2$, HF and HCl, among others, and can be generated

Table 6
Probable chemical reactions releasing gases during the firing bloating process of LWA.

	Reaction	Eq.	T (°C)	Ref.
Organic matter	$C_{org} + O_2(g) \rightarrow CO_2(g) + CO(g)$	(1)		Büchner et al. (1989)
Calcium sulfate	$2CaSO_4(s) \rightarrow 2CaO(s) + 2SO_2(g) + O_2(g)$	(2)	1200–1350	Garea et al. (2003)
Calcite	$CaCO_3(s) \rightarrow CaO(s) + CO_2(g)$	(3)	600–850	Garea et al. (2003)
Iron oxides	$6Fe_2O_3(s) \rightarrow 4Fe_3O_4(s) + O_2(g)$	(4)	>1150	Büchner et al. (1989)
	$Fe_2O_3(s) + C(s) \rightarrow 2FeO(s) + CO(g)$	(5)	1100	
	$Fe_2O_3(s) + CO(g) \rightarrow 2FeO(s) + CO_2(g)$	(6)		
	$4FeO(s) + 4Al_2O_3(s) \rightarrow 4FeAl_2O_4(s)$	(7)		
	$2Fe_2O_3(s) + C + 4Al_2O_3 \rightarrow 4FeAl_2O_4 + CO_2(g)$	(8)		
Portlandite	$Ca(OH)_2(s) \rightarrow CaO(s) + H_2O(g)$	(9)	350–450	Garea et al. (2003)
Calcium hydroxychloride	$Ca(OH)Cl(s) \rightarrow CaO(s) + HCl(g)$	(10)	465–600	Bodénan and Deniard (2003)

Fig. 8. Lightweight aggregates for 0, 1, 5 and 10% of incorporation.
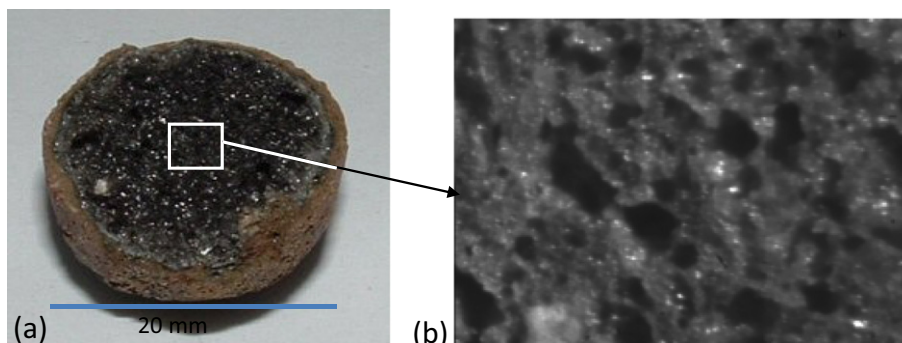

Fig. 9. Broken lightweight aggregates of type LA0 (a) and a zoom of the internal core (b).

from the clay minerals phases, from some additives used for this purpose (e.g. oil) and also from thermal unstable phases from APC residues. Table 6 summarizes some of the many reactions that may be involved.

The recipes tested in these preliminary tests follows the industrial one, where clay, water and oil were mixed into a paste (blank: LA0), granulated into particles round in shape. After firing, the LWA obtained are indicated in Fig. 8 for 0, 1, 5 and 10% of incorporation; Fig. 9a–b show a broken aggregate and a zoom of the interior region, respectively. The external appearance of LWA produced in a laboratorial furnace with 0% (LA0), 1% (LA1), 5% (LA5), and 10% (LA10) of APC residues shows that when 5% and 10% of APC residues are used, the external glassy shell loses the smooth surface. These irregularities suggest a possible extra gas release during the firing process, due to the presence of the APC residues, which have the tendency to lose weight within the temperature range used, as previously shown in Fig. 3. Based on the morphology observed in Fig. 8, it is clear that incorporating 10% of APC residues is excessively detrimental to produce LWA. Therefore, the technological properties in this preliminary assessment were only determined for formulations LA0, LA1 and LA5, as indicate in Table 7. These results point out that if 5% of APC residues were incorporated, significant differences in bulk density and mechanical strength were observed. Thus, further studies should be conducted in order to maximize the percentage of incorporation and minimize the consequence in the technological properties. It is important to notice that since the washing treatment revealed that a reduction in the fluxing content is possible to achieve (see Fig. 1), additional studies should consider the possibility of incorporating pre-washed APC residues instead of as received from the incineration plant.

Finally, it is essential to evaluate the environmental impact; in particular, the risks imposed on the surroundings by a recycling strategy must be weighed against creating new pollution sources elsewhere. In this scope, it is important to take into account the specific application under consideration. In the case of LWA the most probable uses are in cement bound products, but its direct contact with water is also possible for example for agricultural utilizations. Nonetheless, in the literature it is referred that the "end of life" condition, in particular when the material becomes debris, should be taken into account (van der Sloot et al., 2001). Therefore, the leaching behavior of LWA was evaluated after size reduction (<1 mm) of the LWA to simulate the worst scenario. Table 8 shows the leaching results obtained for formulations LA0, LA1 and LA5, according to the standards DIN 38414-S4 and TCLP. The main conclusion that can be drawn from these results is that all the measured quantities are well below the regulatory limits for considering the material as hazardous. However, when 5% of APC residues are used (LA5), the release of some species increases slightly (Zn, Cu, Cl^- and SO_4^{2-}). By comparing the results obtained through TCLP and DIN tests it can be seen that in general the releases are higher for the former case, which is due to the fact that the leaching solution has lower pH, and then is more aggressive for the extraction processes. It is very important to note that low leaching values of the product do not necessarily indicate chemical immobilization of the pollutants in the ceramic matrix; indeed, this may indicate that pollutants of concern volatilize during sintering. Therefore, in future work the gaseous emissions should be analyzed in what concerns the most problematic elements (chlorides, volatile and semi-volatile metals).

4. Conclusions

This study aimed to characterize the natural industrial clay and APC residues from MSW incineration in order to evaluate the possibility of producing LWA with the commercial technological properties and simultaneously recycling the waste.

The results obtained for both materials showed that the bloating capacity, based on the chemical composition of silica, alumina and fluxing contents (Riley diagram), is significant for clay but null for APC

Table 7
Bulk and particle densities and mechanical strength of the lightweight aggregates.

Formulation	Bulk density (g/L)	Particle density (g/mL)	Mechanical strength (MPa)
LA0	264	0.6	0.40
LA1	279	0.6	0.46
LA5	178	0.5	0.19

Table 8
Results from leaching tests DIN 38414-S4 and TCLP for formulations LA0, LA1 and LA5.

	Pb (µg/L)		Cr (µg/L)		Cd (µg/L)		Zn (mg/L)		Ni (mg/L)		Cu (mg/L)		Cl⁻ (mg/L)		SO$_4^{2-}$ (mg/L)		F⁻ (mg/L)	
	DIN	TCLP	DIN	TCLP	DIN	TCLP	DIN	TCLP	DIN	TCLP	DIN	TCLP	DIN	TCLP	DIN	TCLP	DIN	TCLP
LA0	4.25	7.52	0.82	8.62	0.028	0.256	<DL	0.037	<DL	0.024	<DL	0.085	0.14	nq	5.11	nq	0.76	1.89
LA1	0.72	6.67	0.78	7.87	0.003	0.397	<DL	0.027	<DL	0.042	<DL	0.065	0.84	nq	4.73	nq	0.67	1.77
LA5	3.42	13.3	1.44	7.83	0.051	0.446	<DL	0.067	<DL	0.044	<DL	0.170	3.24	nq	14.8	nq	0.57	0.77
RL[a]	2000	5000	500	nd	1000	1000	10	nd	2	nd	10	nd	10000	nd	5000	nd	50	nd

nd — not defined; nq — not quantified.

[a] RL — regulatory limit.

residues. The heating microscope images corroborate completely this behavior. The main crystalline phases of clay are quartz (SiO_2) and illite ($(K,H_3O)(Al,Mg,Fe)_2(Si,Al)_4O_{10}$), hematite ($Fe_2O_3$), smectite ($(Na,Ca_{0.5})_{0.3}((Al,Fe,Mg)_2(Si,Al)_4O_{10})(OH)_2 \cdot nH_2O$), kaolinite ($Al_4Si_4O_{10}(OH)_8$) and traces of feldspar ($(Na,K)AlSi_3O_8$) and calcite ($CaCO_3$). Whereas the main crystalline phases of APC residues are halite ($NaCl$), sylvite (KCl), calcite ($CaCO_3$), anhydrite ($CaSO_4$), quartz (SiO_2), gehlenite ($Ca_2Al(AlSiO_7)$), hematite (Fe_2O_3), portlandite ($Ca(OH)_2$) and calcium hydroxychloride ($Ca(OH)Cl$). The thermogravimetry revealed that a total weight loss of 8.6% was observed for clay, while APC residues lost 32% when temperature rose to 1200 °C.

The washing treatment for removing soluble salts has a fast kinetic (only 10 min is enough to solubilize 22%), and a weight loss of 22% was observed by increasing temperature to 1200 °C. Globally, the washing treatment could be beneficial in the scope of producing LWA and thus it will be further explored.

The preliminary tests involving LWA showed that incorporating 10% is excessively detrimental (the external glassy shell lose the smooth surface) and even 5% led to significant differences in bulk density and mechanical strength when compared with the reference case. The leaching experiments conducted to very low releasing amounts for both standards tested.

In summary, the main findings pointed out that the integration of the APC residues for producing LWA does not improve technological properties, but incorporations lower than 5% will be further tested.

Acknowledgments

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