



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



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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-Corcochano 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 (CO₂ and water), the decomposition of carbonates (CO₂), the reduction of ferric iron (O₂ and CO₂), the sulfides oxidation (SO_x) 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 CaCO₃.

2.3. Technological properties

The bulk density, ρ_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, ρ_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 ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	Na ₂ O	K ₂ 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 _{residues}	14.1	7.4	2.3	41.9	1.0	3.7	3.2	1,924	232	456	121	57	4,308	
APC _{washed}	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 (%)	ρ_r (g/cm ³)	pH	EC (mS/cm)	TDS (g/kg)	Pb	Cr	Cu	Ni	Cd	Zn	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	ND	
APC _{residues}	0.85	8.3 [*]	2.52	12.5	37.5	222	324	5.97	1.38	2.54	0.15	50.5	98,400	
APC _{washed}	0.93	nd	nd	12.1	nd	24.5	130	9.0	<0.9	<1.45	<0.07	18.0	ND	

^{*} Loss on ignition measured at 750 °C; nd – not determined; ND – not detected by titration with AgNO₃.

Table 2
Formulations tested in the laboratory experiments.

Reference	APC residues as-received							APC residues washed	
	W0	W1	W2	W3	W4	W5	W10	W5_w	W10_w
Clay (%)	100	99	98	97	96	95	90	95	90
Residues (%)	0	1	2	3	4	5	10	5	10

Table 3
Mean mass and median diameters of unfired granules.

Reference	APC residues as-received							APC residues washed	
	W0	W1	W2	W3	W4	W5	W10	W5_w	W10_w
Mean mass (g)	1.47	1.41	1.47	1.44	1.40	1.43	1.48	1.29	1.14
Median diameter (mm)	11.4	11.2	11.2	11.3	11.3	11.4	11.6	11.2	11.1
Number of granules analyzed	105	162	197	162	134	184	159	217	164

$$\rho_{a,3} = \frac{M_A}{V_f - V_{sand}} \quad (3)$$

where M_1 is the mass of the pycnometer filled with water (g), M_2 mass of the pycnometer filled with water and aggregates (g), M_A mass of the aggregates (g), ρ_{H_2O} specific gravity of water (g/cm^3), D the mean diameter of the aggregates (cm).

The bloating index (BI) was assessed in terms of the diameter increase after firing, Eq. (4)

$$BI = \frac{D - d}{d} \times 100 \quad (4)$$

where D is the mean diameter of sintered aggregates (cm) and d the mean diameter of green pellets (cm). The particle size change of aggregates before and after sintering was measured by using a pachymeter. BI parameter can also be measured in terms of volume ratio, representing in this case the number of times that the volume increases during the firing process (Gennaro et al., 2004).

The compressive strength was determined according to the industrial procedure (DIN 4226 – Part 3), by using a press Tecnilab LST. The LWA are placed in a vessel with a capacity of 1 L, and then the force required to cause a lowering of 20 cm for 100 s was recorded.

The water absorption capacity (WAC) was determined following the standard EN 1097-6, by measuring the increase in the mass of the oven dry sample through immersion in water using a pycnometer at 22 ± 1 °C, according to Eq. (5):

$$WAC = \frac{M_w - M_A}{M_A} \times 100 \quad (5)$$

where M_w is the mass of the sample after a specific period of immersion (d), M_A the mass of the dry aggregates. The method allows the assessment of the WAC at specific times, and in our study, measurements were made at 5, 15, 30 and 45 min and 1, 2, 4, 6, 9 and 24 h.

The porosity of LWA was evaluated only for formulation W0, at three levels: whole aggregate crushed, external shell and core part. The techniques considered were mercury porosimetry, Micromeritics – PoreSizer 9320, and helium pycnometry, Micromeritics AccuPyc 1330. In the mercury porosimetry, the pressure was increased from 1.5 psia to about 30,000 psia, and due to the size of Hg atoms macro and mesopores were characterized.

2.4. Leaching properties

For analyzing the leaching behavior of LWA, several pellets of each formulation were crushed to less than 1 mm. Then, the compliance tank leaching test based on DIN 38414-S4 (superseded by

EN 12457-2) and the pH dependence test (prEN 14429) were performed. For each of those tests, at least two replicates were conducted. The compliance test involved one stage batch using distilled water at a liquid to solid ratio (L/S) of 10 L/kg for 24 h, conducted at natural pH. On the contrary, in the pH dependence test, a series of subsamples were mixed with solutions of varying acidity with L/S 10 L/kg. In practice, ten solutions with different final pH values were prepared, covering the minimum range of 4–12 (Quina et al., 2009). The suspensions were shaken on a rotary agitator (Heidolph Reax 20) for 48 h in closed vessels. The leachate was then separated by filtering through a 0.45 μm membrane filter and the pH was recorded before acidification prior to metal analysis. The pH dependence test allows the assessment of the buffering capacity of the material or acid neutralization capacity (ANC), which affords perception in the sensitivity of the materials under analysis to externally imposed conditions. Metals were quantified by flame atomic absorption spectroscopy, Perkin Elmer – 3300, chlorides and sulfates were quantified by ion chromatography, Waters Action Analyzer, with a 100 μL loop.

3. Results and discussion

3.1. Morphology

The outer surface of LWA may be observed in Fig. 1 for formulations with only clay (W0), incorporating increasing amounts of APC residues as-received from 1% to 10% (W1, W2, W3, W4, W5, W10) and also using 5% and 10% of washing APC residues (W5_w and W10_w). By visual observation of the outer surface of LWA, a dense glassy outer shell is noted. W0 is the reference case, and it is perceptible that the increasing incorporation of waste leads to irregular surfaces, mainly for formulations W4, W5 and W10. By comparing W5 with W5_w it is clear that the washing treatment has a positive contribution to the smoothness of the surface of the particles. The replacement of 10% of clay was very detrimental for the external appearance of LWA. In addition, as predicted through the Riley diagram (Quina et al., 2006) both incorporations of 10% with and without washing significantly reduce the bloating process.

3.2. Technological properties of sintered LWA

Fig. 2a shows that although the initial median diameter, d_{50} , were in the range of 11.1–11.6 mm, after the firing process the LWA exhibits d_{50} in the range of 16.9–21.2 mm. Fig. 2b illustrates for each formulation tested the median diameter before and after firing plus the associated standard deviation. Though we tried to

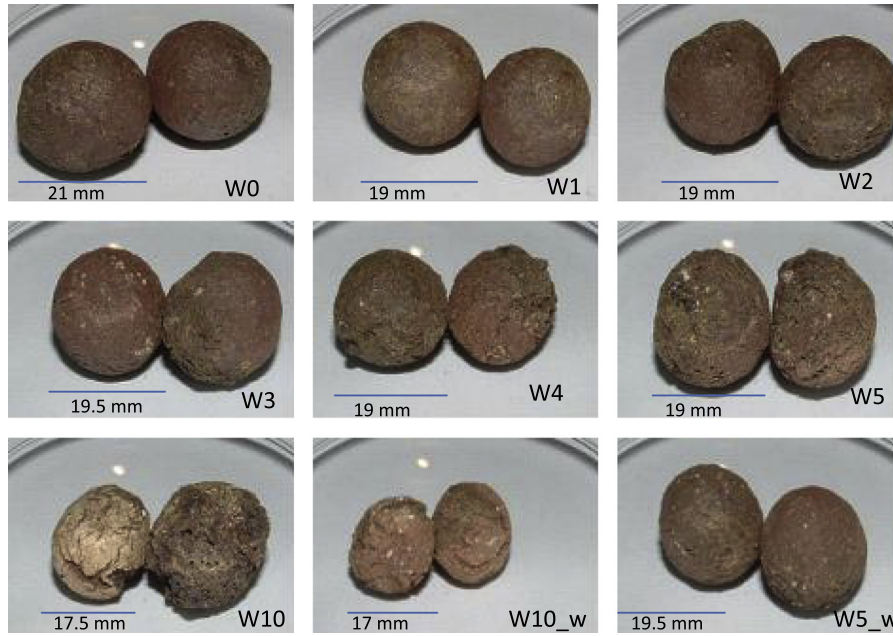


Fig. 1. External surfaces appearance of LWA pellets without APC residues (W0), by incorporating 1–10% of waste as-received (W1–W10) and 5% and 10% of waste pre-washed (W5_w and W10_w).

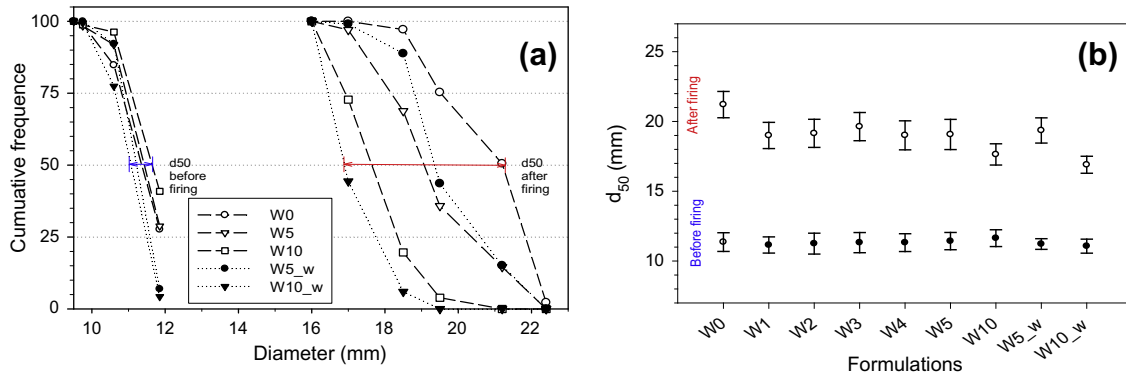


Fig. 2. (a) Granules size distribution and (b) comparison of median diameter before and after firing.

form spherical pellets of similar diameter, after firing the distribution became larger for all formulations (higher standard deviations), and this created some difficulties for comparison purposes.

The bloating index (BI) measured as a percentage of expansion of the material during the firing process, Fig. 3, decreased with increasing percentage of incorporation. For the base case W0, the

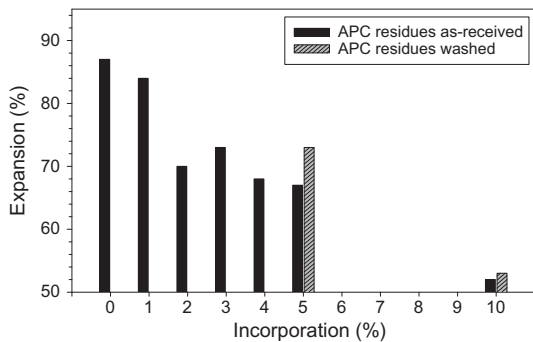


Fig. 3. Percentage of expansion of LWA as a function of the formulation.

observed BI was 87%, whereas for W10 that index decreased for 52%. This means that the waste hampers the expanding process. In fact, when the composition of the APC residues was represented in the Riley ternary diagram (Quina et al., 2006), by plotting the amount of SiO₂, Al₂O₃, and the fluxing (Fe₂O₃ + CaO + MgO + Na₂O + K₂O), it was observed that it falls outside the bloating area. Only replacements lower than 5% led to formulations inside the bloating region. By using washed APC residues, the decrease in the expansion is not so significant, being in this case only 16%.

The variation of density of fired LWA with the percentage of incorporation is shown in Fig. 4. The bulk density, ρ_b , rises as the amount of APC residues increase, meaning that by replacing a certain amount of clay by waste the LWA become heavier. In fact, the value observed for W0 was about 152 g/L and for W10 239 g/L. Considering as example the case of 5% of APC residues (W5) the increase in the bulk density was about 19% (181 g/L), whereas for pre-washed APC residues, the replacement of 5% led to an increase of only 13%. Regarding the apparent specific gravity, ρ_a , the values obtained with Eqs. (1)–(3) were also represented in Fig. 4. The method used in industry for quality control, $\rho_{a,3}$, resulted in lower estimates, ranging from 0.27 to 0.45 g/cm³. According to the experimental procedures involved, the method based on the

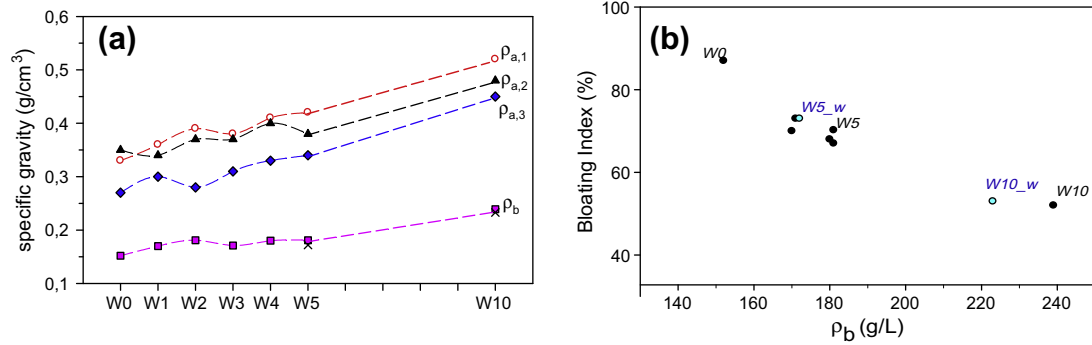


Fig. 4. (a) Apparent ($\rho_{a,1}$; $\rho_{a,2}$; $\rho_{a,3}$) and bulk specific gravity (ρ_b) of LWA as a function of the formulation; (b) Bloating index as a function of bulk density.

Archimedes' principle using water, $\rho_{a,1}$, is probably the most accurate one, but also the most time consuming. Thus, the apparent specific gravity is in the range of 0.33 (for W0) to 0.52 g/cm³ (for W10). This property was not measured for the cases W5_w and W10_w. Fig. 4b shows that, as expected, there is a strong correlation between the bloating index and the bulk density of the material. It should be noted that the expansion of the aggregates during the process of firing is driven by the release and entrapment of gas in the solid matrix. Therefore, LWA with lower bulk densities correspond to higher bloating indexes.

The compressive strength is represented in Fig. 5a as a function of the % of APC residues incorporation, and also in this case an increase is observed as the amount of APC residues rise, with the exception of formulations W1 and W2. The behavior observed for W1 and W2 may be due to the fact that the d_{50} was slightly lower than it should have been (Fig. 2b). Taking as example the 5% of replacement, an increase of 36% in the compressive strength is observed when compared to the reference case (W0). The washing of APC residues led to slightly higher compressive strengths. In Fig. 5b it was represented the compressive strengths of commercial LWA and the ones produced in laboratory conditions as function of the median diameter. In general, as expected, the strength of commercial LWA is significantly higher than the ones produced in laboratory conditions, but in any case as the diameter increases the compressive strength drops. It should be noted that the best commercial LWA must be resistant with dense continuous surface shell but with low density (porous ceramic cores) (Cheeseman and Viridi, 2005). According to our results, by replacing a percentage of clay with APC residues, the compressive strength becomes higher but due to an increase in the bulk density, and thus this is considered a disadvantage.

Regarding the water absorption capacity (WAC) of LWA, this is assumed as one of the most important properties to be determined, especially aiming at producing lightweight concrete. In the

literature, there are references indicating that WAC should be as low as possible (Cheeseman and Viridi, 2005); but others refer that the idea of conducting a pre-saturation of LWA may have a beneficial effect of supplying internally curing water (Castro et al., 2011). However, if the LWA were used at dry conditions, they could interfere to a great extent with the hydration reactions of cement by removing water. Fig. 6a represents the rate at which dried LWA absorb water over 24 h, and Fig. 6b the normalized absorption (calculated as the ratio between the actual value and the final total absorption after 24 h) during the first six hours, where it was also included the formulations with washed APC residues (W5_w and W10_w). The water absorption increased over time, but in the first two hours the rate of absorption was higher. Indeed, the normalized data showed that within the first two hours, the absorption reached more than 50% of the final WAC. It is interesting to note that the lowest rates are observed for W0 (base case) and for W5_w, and the highest initial rate occurred for W10. Similar trends can be found in literature for commercially available LWA (Castro et al., 2011), where values of 6% and 30% were attained at 24 h of immersion. Fig. 7a shows WAC at 24 h as a function of the % of the incorporation of APC residues and an increase is detected up to 5% of replacement. In this case the WAC is near 21%, against 11.5% for aggregates produced by using only clay. The commercial LWA (Leca®) showed absorption of about 30.5%. When the APC residues were previously washed, the WAC decreases until 14.8% (for W5_w) which may represent an advantage for the final product. For the incorporation of 10%, the WAC decreases, and this is a consequence of the fact that the expansion in this case is reduced (only 50%) and then the pores structure is not well developed. In Fig. 7b the water absorption is represented against the bloating index (expansion) for the cases that the residues were pre-washed (W5_w and W10_w) and used as-received (W1, W3, W4, W5 and W10). By comparing, for example, the results obtained for the formulations W5 and W5_w, it is clear that the washing process is

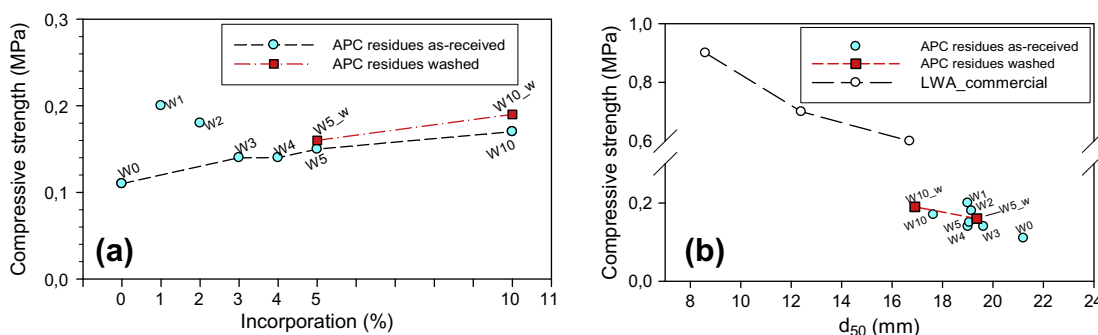


Fig. 5. Compressive strength of LWA as a function of (a) the formulation; (b) median diameter.

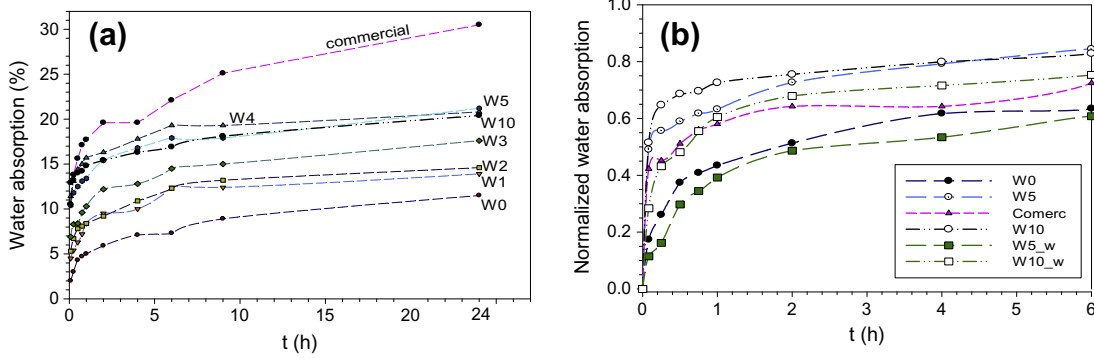


Fig. 6. (a) Water absorption of LWA over 24 h; (b) Normalized water absorption (referred to the total WAC after 24 h) for the initial 6 h.

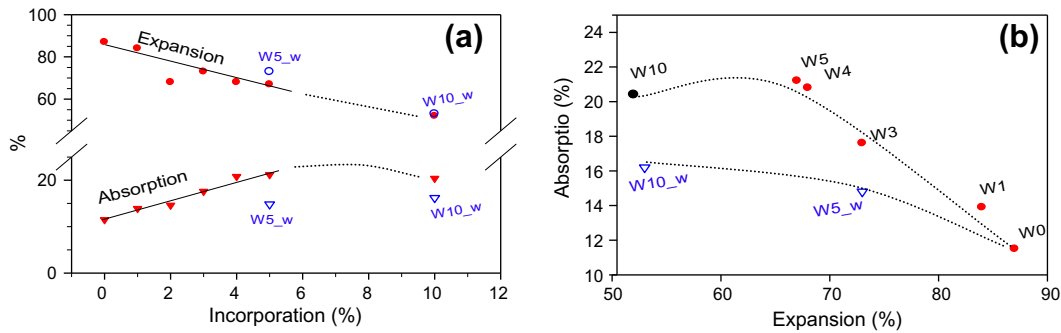


Fig. 7. (a) Water absorption capacity and expansion of the LWA as a function of the incorporation of APC residues; (b) as a function of expansion.

advantageous since it allows higher bloating index and lower water absorption.

For characterizing the porous structure of LWA, some aggregates W0 were disintegrated and divided into the dense continuous surface layer (external shell) and the low density internal core. Table 4 summarizes the results obtained through mercury porosimetry and helium pycnometry (ρ_r). The former technique allowed to estimate the open porosity of each fraction at about 81% for the internal core and 55% for the external shell. The total porosity (closed plus open), ε , indicated in Table 4 (89.3% and 61.9% for the internal and external parts) was determined by Eq. (6)

$$\varepsilon = \left(1 - \frac{\rho_a}{\rho_r}\right) \times 100 \quad (6)$$

where ρ_a is the apparent density (g/mL) and ρ_r the real density (g/mL). The closed porosity was calculated by difference between total and open porosity, and, according to Table 4, 7.9% for the internal part and 7.2% for the external shell were the values found. Similar figures were indicated in literature (Latosinska and Zygadlo, 2011) for lightweight aggregates incorporating sewage sludge as expanding

agent. It is important to note that it was not possible to analyze adequately a whole aggregate through mercury porosimetry due to logistical limitations of the sample holder, whereas helium pycnometry indicated a real density of 2.44 g/cm³.

3.3. Leaching properties

Based on holistic approaches, our study aims to reduce the environmental impact of APC residues, but without the shift of the pollutants from solid to liquid streams. In this scope, some studies stressed that the leaching behavior of synthetic aggregates should be taken into consideration to evaluate the environmental effect on the various stages of its use (van der Sloot et al., 2001). Table 5 shows the results measured in leaching solutions of milled LWA extracted with water. Thus, it can be concluded that the toxic heavy metals Pb, Cd, Zn, Cr, Ni and Cu were below to the detection limits (DL) of the flame atomic absorption spectroscopy. It is important to note that the APC residues release high concentration of these elements in similar leaching conditions (for instance, about 324 mg/kg of Pb were observed) (Quina et al., 2008b). Therefore, for the formulation W10, a minimum release of 32 mg/kg would be expected if immobilization during the sintering process had not occurred. Regarding the major species, such as K, Na and Ca similar results were obtained for the different formulations. However, the concentration of chlorides rises as the percentage of APC residues increase from 0% to 10% (W0 to W10). The results obtained for W5_w and W10_w indicated, as expected, that after washing almost all chlorides were removed.

In addition, the pH dependence test has been carried out for assessing the release of heavy metals and the buffering capacity of the material. Indeed, one of the most important properties affecting the pH of the leachate is the acid or base neutralization capacity (ANC/BNC) (Sabbas et al., 2003). These parameters measure the capacity of a system to neutralize the influence of acids

Table 4
Results from mercury intrusion porosimetry and helium pycnometry for LWA with formulation W0.

	Internal core	External shell
Pore median diameter, $d_{pore,50}$, (μm)	1.280	0.227
Mean pore ($4V/A$), \bar{d}_{pore} (μm)	0.362	0.059
Bulk density, ρ_b (g/ml) = ρ_a	0.265	0.922
Skeletal density, ρ_{sk} (g/ml)	1.421	2.059
Real density, ρ_r (g/ml)	2.470	2.395
Open porosity (%)	81.4	55.2
Total porosity (%)	89.3	61.9
Closed porosity (%)	7.9	7.2

Table 5
Leaching quantities for LWA tested with water for 24 h (mg/kg).

	Pb	Cd	Zn	Cr	Ni	Cu	K	Na	Ca	Cl ⁻	SO ₄ ²⁻
W0	<DL	<DL	<DL	<DL	<DL	<DL	41	30	201	43	36
W1	<DL	<DL	<DL	<DL	<DL	<DL	38	18	424	37	90
W2	<DL	<DL	<DL	<DL	<DL	<DL	56	20	588	42	204
W3	<DL	<DL	<DL	<DL	<DL	<DL	54	11	779	44	354
W4	<DL	<DL	<DL	<DL	<DL	<DL	40	9	403	57	209
W5	<DL	<DL	<DL	<DL	<DL	<DL	36	12	441	54	115
W10	<DL	<DL	<DL	<DL	<DL	<DL	54	13	469	112	180
W5_w	<DL	<DL	<DL	<DL	<DL	<DL	32	7	626	16	161
W10_w	<DL	<DL	<DL	<DL	<DL	<DL	44	27	531	47	198
DL	0.9	0.2	0.1	1.0	1.4	0.9	-	-	-	-	-

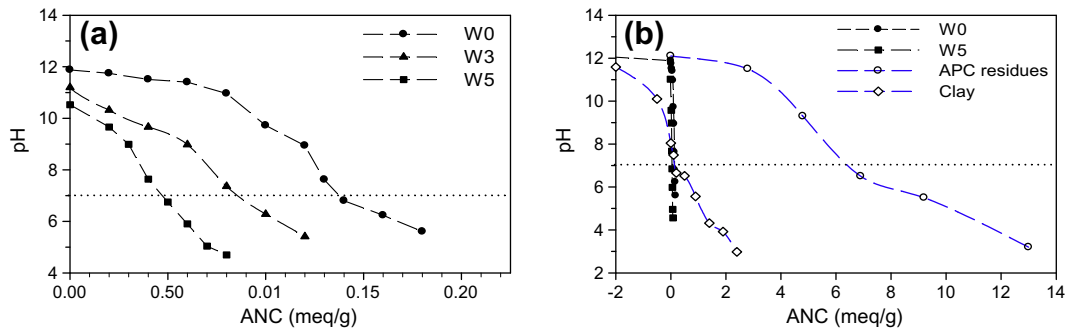


Fig. 8. Acid neutralization capacity of (a) LWA of type W0, W3 and W5 and (b) comparison with APC residues and clay.

or bases, and may reveal the sensitivity of a material itself to external influences and/or internal stresses (e.g. mineralization, organic matter degradation). Besides, the buffering capacity determines the evolution of the pH in the leachate over time. Fig. 8a depicts the ANC for formulations W0, W3 and W5 having as reference the pH 7, and the most important conclusion is that even if the LWA were tested after milling, they exhibited a very low

neutralization capacity (between 0.05 and 0.15 meq/g). Indeed, Fig. 8b shows that ANC for LWA is significantly lower than the observed for the starting materials (clay and APC residues). This is a result of the sintering process, which provokes the degradation of calcite and the incorporation of Ca in the silicate matrix (van der Sloot et al., 2001). For this reason, it is very important to evaluate the sensitivity of LWA to external pH, namely regarding the release

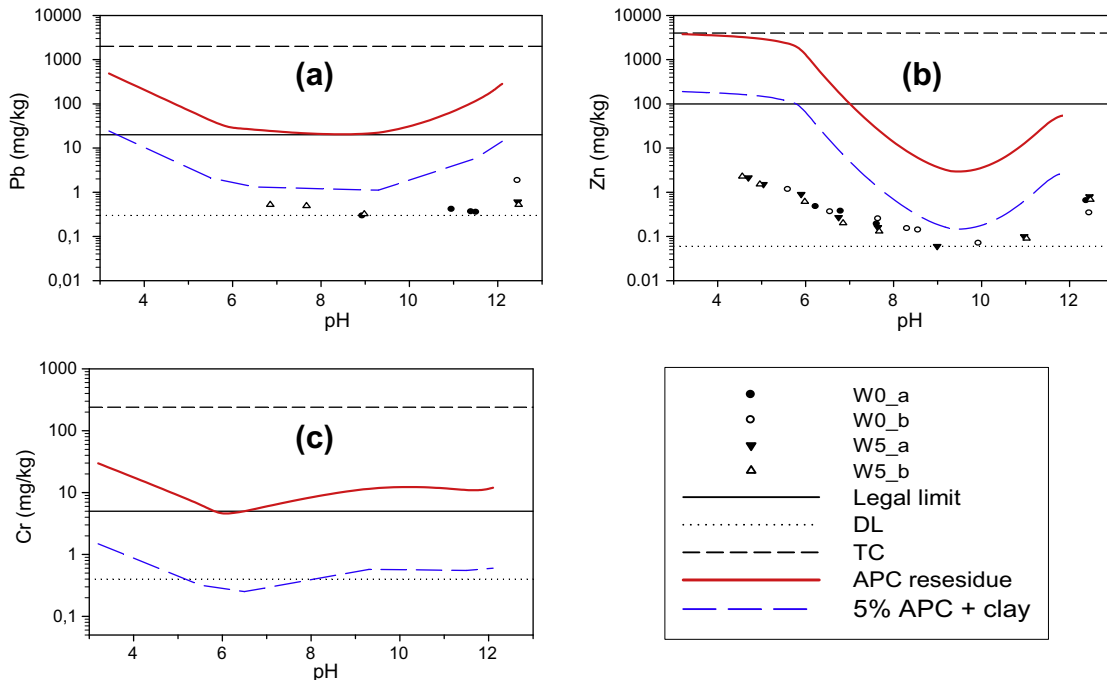


Fig. 9. Leaching release of (a) Pb; (b) Zn and (c) Cr for milled LWA (two replicates for formulation W0 (a and b) and W5 (a and b) in comparison with APC residues, its total content (TC), the limit for deposition in hazardous landfill, and the detection limit (DL) of FAAS.

of Pb, Zn and Cr, Fig. 9a–c. In each of these figures it was represented the leaching observed for two replicates of milled LWA with formulations W0 (without APC residues) and W5 (with 5% of APC residues), the leaching observed for APC residues (Quina et al., 2009), the total content (TC), the limit for disposed of waste in hazardous landfill, and the detection limit of flame atomic absorption spectroscopy (FAAS). In the case of Pb, Fig. 9a, it was observed that the legal limit of 20 mg/kg is exceeded in the all range of pH for APC residues. However, after the incorporation of 5% of APC residues in LWA, the real leaching quantities are below the detection limit for all values of pH. Even so, the curve referred as “5% APC + clay” shows the situation that would be observed if immobilization did not occur. For the case of Zn, Fig. 9b indicates that for pH lower than 7 the released quantities from LWA were above the detection limit, whilst far from the case referred to as “5% APC + Clay”, where it was assumed that immobilization did not occur. For the case of Cr, Fig. 9c, all the released quantities were below the detection limit. Globally, these results show that if LWA were used in concrete for natural gravel replacement, where the predominant pH is above 12, it is unlikely that leaching will have a significant environmental impact. In open storage, due to the low ANC, the leachate pH may decrease to below 7, and a slightly higher undesired leaching might occur, but not exceeding environmental acceptable levels.

4. Conclusions

In this work, several formulations were tested in order to produce synthetic lightweight aggregates (LWA) in a laboratory oven, aiming at recycling air pollution control (APC) residues from MSW incineration. The morphology of the external glassy shell showed that more than 4% of APC residues as-received were very detrimental, leading to irregular external surfaces. However, for the case of a washing pre-treatment, the 5% of incorporation led to good results. In general, the bulk density and the compressive strength rose as APC residues amount increased, whereas the bloating index decreases. The water absorption capacity (WAC) increased as the amount of APC residues increase until 5%, whereas when 10% of waste is used a reduction was measured. This parameter gives an indication about the open porosity, and thus depending on the application of the LWA, a high WAC may (or may not) be advantageous.

The leaching behavior was investigated using water and acidic solutions, and it was concluded that the environmental impact of heavy metals will not be significant. In spite of the low acid neutralization capacity of the LWA, their sensitivity to external pH variations was not relevant, since the pollutants were immobilized into the glassy ceramic matrix.

The main outcome of the work is that none beneficial effect was really identified by incorporating APC residues (with or without washing) into LWA. Accordingly, more research work is still required in order to divert this hazardous waste from landfills.

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