



## Combined use of MSWI bottom ash and fly ash as aggregate in concrete formulation: Environmental and mechanical considerations

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### ARTICLE INFO

#### Article history:

Received 15 January 2009

Received in revised form 30 March 2009

Accepted 30 March 2009

Available online 7 April 2009

#### Keywords:

MSWI bottom ash

MSWI fly ash

Concrete

Waste management

Aggregate material

### ABSTRACT

This paper reports the experimental results obtained after casting concrete formulated with different mix proportions of municipal solid waste incineration (MSWI) by-products, bottom ash (BA) and air pollution control fly ash (APCFA), as aggregates. Several tests were performed to determine the properties of the mixed proportions. Mechanical properties of the formulations, such as compressive strength, were also determined, and two different leaching tests were performed to study their environmental effects. Some suitable concrete formulations were obtained for the 95/5 and 90/10 BA/APCFA mix proportions. These formulations showed the highest compressive strength test results, above 15 MPa, and the lowest amount of released trace metals in reference to the leaching test. The leaching mechanisms involved in the release of trace metals for the best formulations were also studied, revealing that the washing-off process may play an important role. Given the experimental data it can be concluded that these concrete mix proportions are suitable for use as non-structural concrete.

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### 1. Introduction

Waste-to-energy incineration process has been shown worldwide to be a feasible management strategy for treating the increasing amounts of municipal solid waste (MSW) that cannot be recycled [1]. After combustion treatment the waste volume is reduced by 90% and 70% in mass. Consequently, two mainly solid by-products are produced through MSWI. Accounting for nearly 80%, bottom ash (BA) is the most significant by-product, with the remaining amount being ascribed to air pollution control fly ashes (APCFA).

BA can be described as heterogeneous particles consisting of glass, magnetic and paramagnetic metals, minerals, synthetic and natural ceramics, and unburned materials [2]. Freshly quenched bottom ash shows a high reactivity due to its reactive silica and lime. Therefore, the mineralogical metastable phases becomes thermodynamically stable, such that chemical and mechanical properties may change under natural conditions [3]. After this period of natural weathering, which lasts for at least two months, BA is catalogued as non-hazardous waste according to the European Landfill Directive [4], and is thus considered suitable for land filling or reuse.

In 2000 the estimated amount of MSWI residue produced in the USA, Japan and the European Union was about 25 Mtons/year [5]. To face up to the consequences, European governments encourage the reuse of BA as a secondary building material both to prevent the use of non-renewable natural gravels and to avoid excessive land filling. Accordingly, several applications have been found for MSWI bottom ash. The most widespread practice is the reuse of BA as an aggregate substitute for road base [6,7], which is subject to strict requirements that are defined by each European country, for example, the Spanish specifications for road construction [8]. Another important reuse of BA is as an aggregate for concrete. Some studies [9,10] have shown that replacing natural gravel by no more than 50% of BA is possible without affecting the durability or strength of the concrete. Other researchers [11] have studied the reuse of BA as a cement replacement, where up to 15% substitution is considered acceptable.

However, there is a handicap to the reuse of BA as a binder or aggregate in concrete. Freshly quenched BA tends to suffer expansive reactions that could create swelling problems. According to the literature [10–13] freshly quenched BA may fail basically as a result of three chemical reactions that take place during a short-term aging period: (a) formation of gel by oxidation of metallic aluminium, with the generation of hydrogen; (b) ettringite formation when BA is saturated with water; and (c) the hydration of calcium oxide and magnesium oxide, leading to an increase in their volume. The first of these reactions seems to make the most important contribution. However, the use of naturally weathered

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bottom ash (WBA), rather than freshly quenched BA, as secondary building material may avoid these adverse reactions, which take place when the material is disposed of in an open environment [3].

APCFA is considered a hazardous waste which must be properly stabilized before its final disposal in a landfill site. Although its composition may vary due to the technology used in the MSWI incineration process, MSWI fly ash (FA) consists of fine particles, usually with a high content of heavy metals, organic compounds and chlorides [14].

Cement-based treatment is the most extended APCFA stabilization, not only due to economic factors but also because it is easy to apply. Alba et al. [15] evaluated the stability/solidification relationship between FA and Portland cement, and considered the carbonation effect as a way to stabilize final disposal. Other researchers [15,16] have shown that cement can be replaced by up to 30–40% of FA for use as cement paste or cement clinker. However, some studies [17,18] have found that cement only maintains its strength with a maximum of 20% FA, although this depends on the nature and the technology involved in FA production. The reuse of FA as aggregate for concrete is also feasible. Thus, Collivignarelli and Sorlini [19] showed the possibility of casting concrete with a compressive strength higher than 15 MPa when replacing BA aggregate by FA.

However, although there are studies considering both by-products combined in mortar formulations [20] in different percentages and merged with natural aggregates, BA and APCFA have rarely been studied together as an aggregate material used for concrete formulation. In this case, the proper use of both by-products could correct the low content of fine material that characterizes BA [21], thus improving, for example, the compaction of the concrete.

The aim of the present study was to consider the viability of a combination of WBA and APCFA as aggregate materials for concrete formulation without using any other kind of aggregate. The research takes into account both mechanical aspects, which determine whether the concrete can be used as a building material, and environmental aspects that may limit its use. In accordance with these goals the main objective of the study was to determine the proper formulation of a concrete that allows APCFA to be stabilized, by means of a stabilization-solidification (S/S) process, and also to produce suitable material for building.

## 2. Methods and materials

### 2.1. Characterization of materials

Both WBA and APCFA were collected from a single municipal solid waste incinerator in Tarragona, Spain. This facility produces 35,000 tons/year of freshly quenched BA, which is fully reused as secondary building material. An additional 3000 tons/year of APCFA is also produced, which is currently stabilized with Portland cement for final disposal in a landfill site. Prior to this, to allow the short-term natural weathering process to stabilize the freshly quenched BA, it is homogenized in a conditioned plant where the quality of this material is improved and recovery of the revalorized metals also becomes possible.

Around 600–700 kg of WBA was taken from various stockpiles of bottom ash that had been naturally weathered for three to four months, while about 150 kg of APCFA was taken from the fly ash collection system. The samples were previously homogenized and representative sub-samples of about 1 kg were taken for physical and chemical characterization.

The material characterization of WBA was carried out for particle sizes ranging from 3.2 to 25 mm, according to the classification

**Table 1**

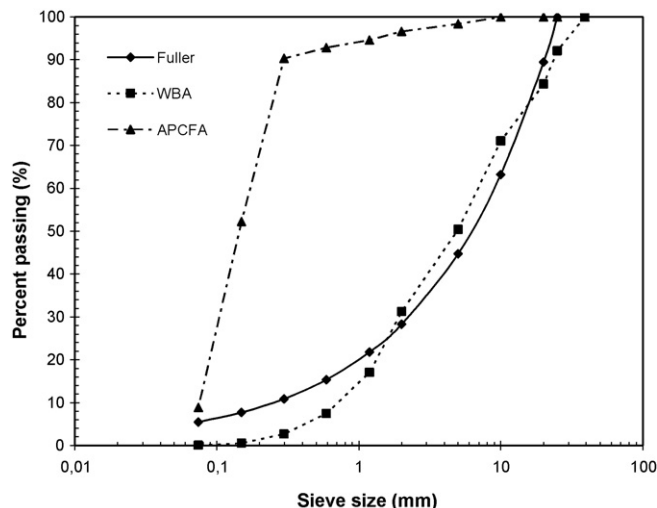
Average chemical composition of MSWI natural weathered bottom ash (WBA) and air pollution control fly ashes (APCFA).

Oxides	WBA (%)	APCFA (%)
SiO <sub>2</sub>	49.38	6.35
CaO	14.68	43.05
Cl	n.d.	8.38
Fe <sub>2</sub> O <sub>3</sub>	8.38	0.63
Na <sub>2</sub> O	7.78	5.80
Al <sub>2</sub> O <sub>3</sub>	6.58	3.5
MgO	2.32	1.38
K <sub>2</sub> O	1.41	4.59
CuO	1.26	–
SO <sub>3</sub>	0.57	4.64
ZnO	0.38	1.41

described in previous studies [2]. It showed that nearly 50% of WBA consisted of glassy material (mostly from domestic sources), a further 37% was composed of ceramic materials (natural or synthetic), while the rest was classified as metals and almost 2% of unburned material.

The chemical composition of the major elements in the bulk WBA and APCFA was determined in duplicate by X-Ray Fluorescence Spectroscopy (XRF) and the results are shown in Table 1. As can be observed, the XRF results agree with the material characterization of WBA, which is mainly composed by lime-soda-glass, the most prevalent type of glass used in bottles. The X-ray powder diffraction (XRD) pattern of APCFA revealed the presence of Halite, Sylvite, Calcite, Anhydrite and/or Bassanite and the excess of Portlandite that results from its use for gas neutralization in the air pollution control system.

Particle size distribution was also determined by sieving the samples as indicated by the standard EN 933-2 [22]. Fig. 1 shows the grading curve of WBA and APCFA and compares both of them with a Fuller reference particle size distribution. Fuller curves are grading curves which give the minimum void space and closest packing for sands and other mineral aggregates containing particles of varying sizes. The shape of the Fuller curve will depend on the maximum particle size, but will be a single curve for any given maximum particle size, this being a suitable curve that allows the best mechanical properties of a concrete to be obtained [23]. Table 2 also shows the determination of dried-oven density, surface dried density and apparent particle density of WBA according to the standard EN 1097-6 [24].



**Fig. 1.** Particle size distribution for BA and APCFA compared with Fuller curve.

**Table 2**

Dried-oven density ( $\rho_{od}$ ), surface dried density ( $\rho_{ssd}$ ), apparent particle density ( $\rho_a$ ) and water absorption (WA) for WBA determined by the standard EN 1097-6.

Mg/m <sup>3</sup>	Fraction <5 mm	Fraction >5 mm
$\rho_{od}$	1.64	2.31
$\rho_{ssd}$	1.88	2.34
$\rho_a$	2.22	2.48
WA	15.75	2.72

**Table 3**

Results of the leaching test EN 12457-2 for WBA and APCFA in comparison with limits set by European Landfill Directive. Results expressed in mg kg<sup>-1</sup>.

Element	WBA	APCFA	Limit values		
			Inert	Non-hazardous	Hazardous
As	0.01–0.05	0.10–0.64	0.5	2	25
Ba	0.24–0.36	18.6–32.2	20	100	300
Cd	0.01	0.05	0.04	1	5
Cr	0.08–0.31	1.60–3.63	0.5	10	70
Cu	1.02–2.61	1.60–2.71	2	50	100
Hg	<0.01	0.10–0.17	0.01	0.2	2
Mo	0.19–0.25	0.26–3.25	0.5	10	30
Ni	0.06–0.10	0.57–1.84	0.4	10	40
Pb	0.05–0.10	66.1–211	0.5	10	50
Sb	0.13–0.35	0.01–0.07	0.06	0.7	5
Se	0.02–0.25	0.25–3.45	0.1	0.5	7
Zn	0.04–0.51	7.90–100	4	50	200

To evaluate the potential release of pollutants from raw materials and how they are catalogued according to the European Landfill Directive [25], the corresponding EN-12457-2 [26] leaching test was carried out in triplicate. In this test, samples of WBA and APCFA are reduced, when necessary, to a particle size below 4 mm and brought into contact with ten times the weight of water under continuous stirring for 24 h. Concentrations of different trace metals in eluate, after being filtered through a 45  $\mu$ m pore size polypropylene filtration membrane and preserved as appropriate, were determined by inductive coupled argon plasma mass spectrometry (ICP-MS). Results are given in Table 3. According to these results and the limits established for the acceptance of waste at landfills [27], WBA can be catalogued as non-hazardous residues, while APCFA shows considerably more toxicity, it being catalogued as hazardous residue.

## 2.2. Methods

The cement used for concrete cast was Portland CEM I 52.5 R, and the ratio of cement:aggregate material (C/A) was constant at 1:4 in all formulated specimens. According to the literature [20], additions of fly ash in concrete, used as a replacement for cement, must not exceed more than 25% in weight. Therefore, different formulations were cast based on variations in the percentage of both MSWI by-products (Table 4). Previously, the particle size distribution of WBA and APCFA used as aggregate for all studied formulations was also considered to predict their mechanical properties comparing them with the Fuller curve (see Fig. 2) and their similitude in terms of fineness module.

**Table 4**

Mixture proportions of WBA and APCFA used as aggregate for concrete casting.

Concrete	WBA (kg)	APCFA (kg)	Cement (kg)	(C/A)	Water (kg)	Water/cement
75/25	18.75	6.25	6.25	1:4	7.00	1.12
80/20	20.00	5.00	6.25	1:4	6.41	1.02
83/17	20.75	4.25	6.25	1:4	6.00	0.96
85/15	21.50	3.56	6.25	1:4	5.63	0.90
90/10	22.50	2.50	6.25	1:4	4.91	0.78
95/5	23.75	1.25	6.25	1:4	4.63	0.74
100/0	25.00	0.00	6.25	1:4	4.22	0.67

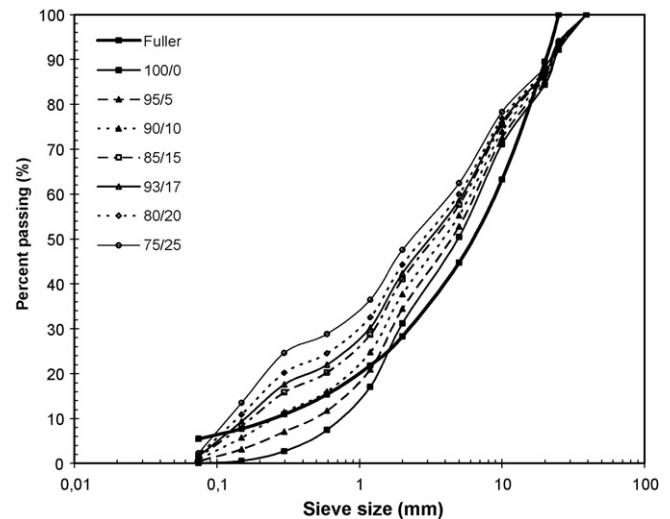


Fig. 2. Comparison for the different mixtures of concrete, cast with BA and APCFA, particle size distribution with Fuller curve.

For all concrete mixtures the consistence was fixed in the range of 5–8 cm according to the Abrams cone [28] and different water:cement (W/C) ratios were used. Three cylindrical specimens with dimensions of 15 cm diameter and 30 cm height were cast for each formulation. Immediately after the concrete was cast and compacted the specimens were kept in the climatic chamber for 24 h. They were then demolded and stored in the curing room at a constant temperature of 20 °C and a relative humidity of 95% for 28 days, after which the compressive strength was tested. Density and porosity of the hardened concrete were also determined according to the standard EN 12390-7 [29]. A control mixture using natural gravel as aggregate material, with the same particle size distribution as WBA, was also performed under the same conditions to compare the mechanical properties.

To determine the potential release of toxic and pollutant elements from the studied mixtures, leaching behaviour was characterized in two different ways as a criterion to define the final disposal of these concretes as possible residues, e.g. at the end of use. Firstly, from the specimens broken during the compression tests, the leaching test procedure EN 12457-2 [26] was performed for granular materials. Secondly, and in order to determine the set criteria for monolithic materials, the semi-dynamic test NEN 7375 [30] was carried out. In accordance with this test a moulded and cured specimen (28 days) of defined geometry (>40 mm in any direction) was immersed in deionised water. The water volume must be approximately 5 times greater than that of the specimen. Leaching solutions are exchanged with fresh deionised water at predetermined cumulative time intervals of 0.25, 1, 2.25, 4, 9, 16, 36 and 64 days. The 64-day tank test quantifies long-term diffusive leaching from the stabilized waste. For each extraction, water pH and conductivity are determined. Concentrations of different trace metals in eluate, after being filtered through a 45  $\mu$ m pore size polypropylene filtration membrane and preserved as appropriate, were determined by ICP-MS, whereas chloride and sulphate concentrations were analysed by ion chromatography. Accumulated heavy metal and metalloid concentrations in water were also calculated after all the extractions.

Finally, to compare both the mechanical and leaching behaviours of all the specimens formulated with WBA and APCFA with those obtained from natural gravel commonly used as aggregate, a number of control specimens (Control) were cast and cured under the same conditions as those described previously.

### 3. Results and discussion

#### 3.1. Mechanical and physical properties

Although WBA shows a good match to the Fuller curve, it could be improved by the addition of APCFA (Fig. 2). Indeed, using a small quantity of APCFA in order to correct the lack of fine particles in WBA would appear to be necessary. However, considering the variation in fineness module for each of the mixtures it would seem advisable not to exceed 15% of APCFA as aggregate material. These measures, in addition to hardened concrete densities and porosities, are given in Table 5. The apparent density ( $\rho_a$ ) and porosity increase as the use of APCFA in the concrete formulation decreases. This may be explained by the low bulk density and high specific surface area of APCFA ( $0.77 \text{ g cm}^{-3}$  and  $6.04 \text{ m}^2 \text{ g}^{-1}$ , respectively), as well as by the W/C ratio (see Table 4). However, the values of the 100/0 and control specimens indicate that the porosity and density (see Table 2) of WBA are lower than those of natural gravel commonly used as aggregate. These results indicate that mixtures of 95/5 and 90/10 are the most suitable from a physical point of view. The fact that they show a particle size distribution that is more similar to the Fuller reference contributes in having less porosity and, in consequence, higher density.

Compressive strength for all the mixtures is shown in Fig. 3. The mixture corresponding to 95% WBA and 5% APCFA (95/5) shows the highest value, reaching nearly 18 MPa. This result is only 4 MPa lower than the mean compressive strength obtained from control specimens (22.15 MPa). However, compressive strength showed a linear decay when the proportions of APCFA used were increased (see Fig. 3). Compressive strength for the 100% WBA (100/0) formulation is lower than expected when considering this trend, and there could be two reasons for this: (i) the 100/0 formulation does not show as good a particle size distribution as the 95/5 and 90/10 formulations; and (ii) not take advantage from the effects of pozzolanic reactions occurring between portlandite in the APCFA and silicates from WBA, which accounts for the increase in compressive strength due to the formation of hydrated pastes. In this regard, some authors [15,16,20] have replaced cement by FA due to its binding properties. However, the use of a high percentage of APCFA as aggregate material in a concrete formulation implies a high W/C ratio, which reduces compressive strength (see Fig. 4).

Given the characteristics of both by-products, e.g. water absorption, abrasion resistance and particle morphology [6,15], the concrete formulated with a combination of WBA and APCFA as aggregate materials should mainly be used as mud slab concrete. However, the 95/5 and 90/10 cast mixtures have a compressive strength higher than the minimum needed for non-structural concrete (15 MPa) according to Spanish regulation EHE-8 [31], which denotes a good mechanical behaviour that enables these mixes to be used in, for example, non-structural precast concrete pavers.

#### 3.2. Leaching behaviour

It is recognized that the environmental performance of a material should be based on release rather than on total content of

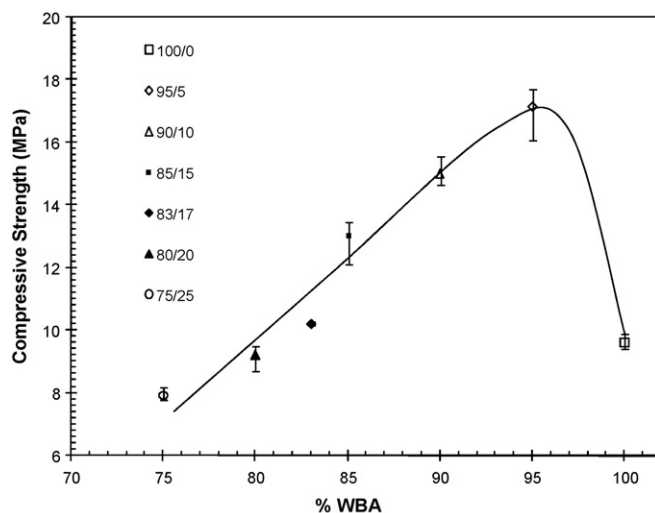


Fig. 3. Variation in compressive strength with percentage of WBA.

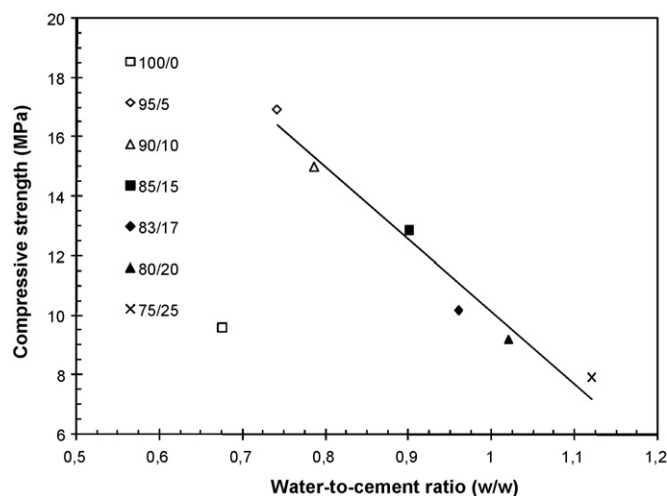


Fig. 4. Variation in compressive strength with water-to-cement ratio.

potentially dangerous constituents [32]. Currently, leaching tests are widely used to estimate the potential release of contaminants from waste materials over a range of possible waste management activities, including during recycling or reuse [33]. However, leaching tests designed to simulate release under specific environmental scenarios, typically worst-case management, often do not provide information about scenarios other than the one being considered. Therefore, taking into account all possible scenarios, the acceptance criteria and regulations based on leaching test procedures have not been provided for the reuse of stabilized waste materials, e.g. precast concrete (or structural concrete) formulated with by-products or stabilized waste. Thus, in order to evaluate the potential release, the leaching behaviour of the materials studied in this work was

Table 5

Fineness modules ( $\sigma$ ), apparent density ( $\rho_a$ ), relative density ( $\rho_r$ ) and porosity for hardened concrete formulated with a combined use of WBA and APCFA as aggregate materials.

	75/25	80/20	83/17	85/15	90/10	95/5	100/0	Control	Fuller
$\sigma$	6.24	6.48	6.62	6.71	6.95	7.19	7.49	7.49	7.13
$\rho_a$ (Mg/m <sup>3</sup> )	1.41	1.53	1.50	1.58	1.61	1.64	1.63	2.12	
$\rho_r$ (Mg/m <sup>3</sup> )	2.86	2.72	2.65	2.64	2.59	2.58	2.53	2.62	
Porosity (%)	50.43	43.75	43.26	40.1	37.4	36.8	35.6	18.96	



**Table 6**

Results for the leaching test EN 12457-2 determined for concrete specimens formulated with bottom ash (WBA) and air pollution control fly ash (APCFA) as aggregate materials.

	Mixture WBA/APCFA (mg kg <sup>-1</sup> )							Limit values (mg kg <sup>-1</sup> )			
	100/0	95/5	90/10	85/15	83/17	80/20	75/25	Control	Inert	Non-hazardous	Hazardous
As	<LOD	<LOD	<LOD	<LOD	0.06	0.06	0.08	<LOD	0.5	2	25
Ba	14.5	15.5	17.5	19.2	21.2	20.2	29.5	6.65	20	100	300
Cd	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.04	1	5
Cr	0.26	0.25	0.05	0.07	0.06	0.08	0.06	0.23	0.5	10	70
Cu	0.72	0.30	0.49	0.34	0.23	0.29	0.40	0.06	2	50	100
Hg	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.01	0.2	2
Mo	0.36	0.30	0.30	0.32	0.27	0.29	0.29	0.06	0.5	10	30
Ni	0.15	0.26	0.85	1.44	0.31	2.11	0.36	<LOD	0.4	10	40
Pb	0.52	1.06	0.16	0.19	1.37	0.16	0.97	0.21	0.5	10	50
Sb	0.02	0.02	0.03	0.03	0.03	0.02	0.02	<LOD	0.06	0.7	5
Se	<LOD	<LOD	0.01	<LOD	<LOD	<LOD	<LOD	<LOD	0.1	0.5	7
Zn	0.31	0.29	0.53	0.95	0.45	0.33	0.51	0.39	4	50	200

LOD: Limit of detection

compared with the considered acceptance criteria for landfills and waste classification [25,34]. In addition, analytical data from monolithic leaching tests enable the predominant release mechanism to be determined for each trace metal of interest.

### 3.2.1. Granular test

Results for the leaching test EN 12457-2 are presented in Table 6. The leachate pH of all granular samples studied was between 12.3 and 12.5. No difference in pH was observed while increasing the amount of APCFA added to the specimen. Therefore, it can be argued that the pH is controlled by the solubility of the portlandite  $\text{Ca}(\text{OH})_2$  content in the Portland cement binder and/or in the WBA and APCFA added as aggregates. In this pH range, many pH-dependent metals such as Pb or Zn exhibit relatively high solubility. However, none of the cast formulations exceed the established limits for a non-hazardous landfill disposal (Table 6). Despite the high amount of MSWI waste added as aggregate the amounts metals released were of the same order of magnitude as for the control specimen.

It can also be seen that the concentration of Ba increases along with the quantity of APCFA used in the concrete mixture. Moreover, the 100/0 formulation exhibits a higher Ba release than was obtained from the WBA and APCFA used in the concrete formulation (see Table 3). The Ba leaching mechanism in both WBA and APCFA waste seems to be controlled by the solubility of barite  $\text{BaSO}_4$  or  $\text{Ba}(\text{S,Cr})\text{O}_4$  solid solutions [35]. However,

during the hydration process of Portland cement, sulphates (and chromates) are strongly involved in the formation of ettringite  $\text{Ca}_6\text{Al}_2(\text{SO}_4)_3(\text{OH})_{12}\cdot 26\text{H}_2\text{O}$  [36], thus displacing the solubility equilibrium of barite. The relationship between Ba release from the cement-stabilized MSWI waste and the sulphate available to neoform the ettringite mineral phase has been argued [37] to explain the leachability of this element.

Other elements such as Cu, which are typically prevalent in WBA (see Table 3), show a concentration decrease when using lower amounts of WBA as aggregate. The release of Cu is mainly related to the complexation of this metal with dissolved organic carbon from the organic matter contents of the WBA [38]. However, the release of metals such as Pb and Zn, whose content is notably higher in APCFA, is not significantly different in the specimens formulated with a greater amount of APCFA. This fact is due to the solidification/stabilization process that takes place when using cement binders. This also explains why the concentration of As, Cd and Se in the leachate was lower than the limit of detection.

### 3.2.2. Monolithic test

In order to assess the leaching behaviour of the concrete and the mechanism of metal release, the semi-dynamic 64-day NEN 7375 tank test for monolithic waste [30] was performed. Table 7 shows the results obtained with this test, which are given as the accumulative concentration for the different heavy metals and metalloids that could represent a possible environmental hazard, calculated

**Table 7**

Results for the semi-dynamic leaching test for monolithic waste (NEN 7375) determined for concrete specimens formulated with bottom ash (WBA) and air pollution control fly ash (APCFA) as aggregate materials.

Element	$\varepsilon_n^*$ (mg/m <sup>2</sup> )							Limit values		
	Mixture WBA/APCFA							Control	Stable non-reactive hazardous waste in non-hazardous landfill	Hazardous waste landfill
	100/0	95/5	90/10	85/15	83/17	80/20	75/25			
Zn	1.19	4.27	4.91	2.69	3.73	6.87	13.9	2.90	30	100
Pb	0.49	0.39	0.78	0.85	0.80	1.10	6.66	1.81	6	20
Ni	1.02	0.92	0.93	0.76	0.76	0.76	0.86	<1.91*	6	15
Cr	4.61	1.22	1.15	1.00	1.15	0.96	0.89	<1.91*	5	25
Cu	8.40	2.22	4.37	3.69	2.19	2.87	3.88	5.17	45	60
Ba	0.51	23.4	133	412	366	445	390	4.67	45	150
Se	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.4	5
As	1.76	1.36	1.19	0.60	4.95	2.42	2.80	<LOD	1.3	20
Cd	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.2	1
Sb	5.01	4.98	2.42	1.40	1.74	2.57	4.22	<1.00*	0.3	2.5
Mo	1.12	0.91	1.13	1.04	0.98	1.22	1.04	<0.90*	7	20

LOD: Limit of detection.

\* At least one  $E_i$  is lower than LOD.

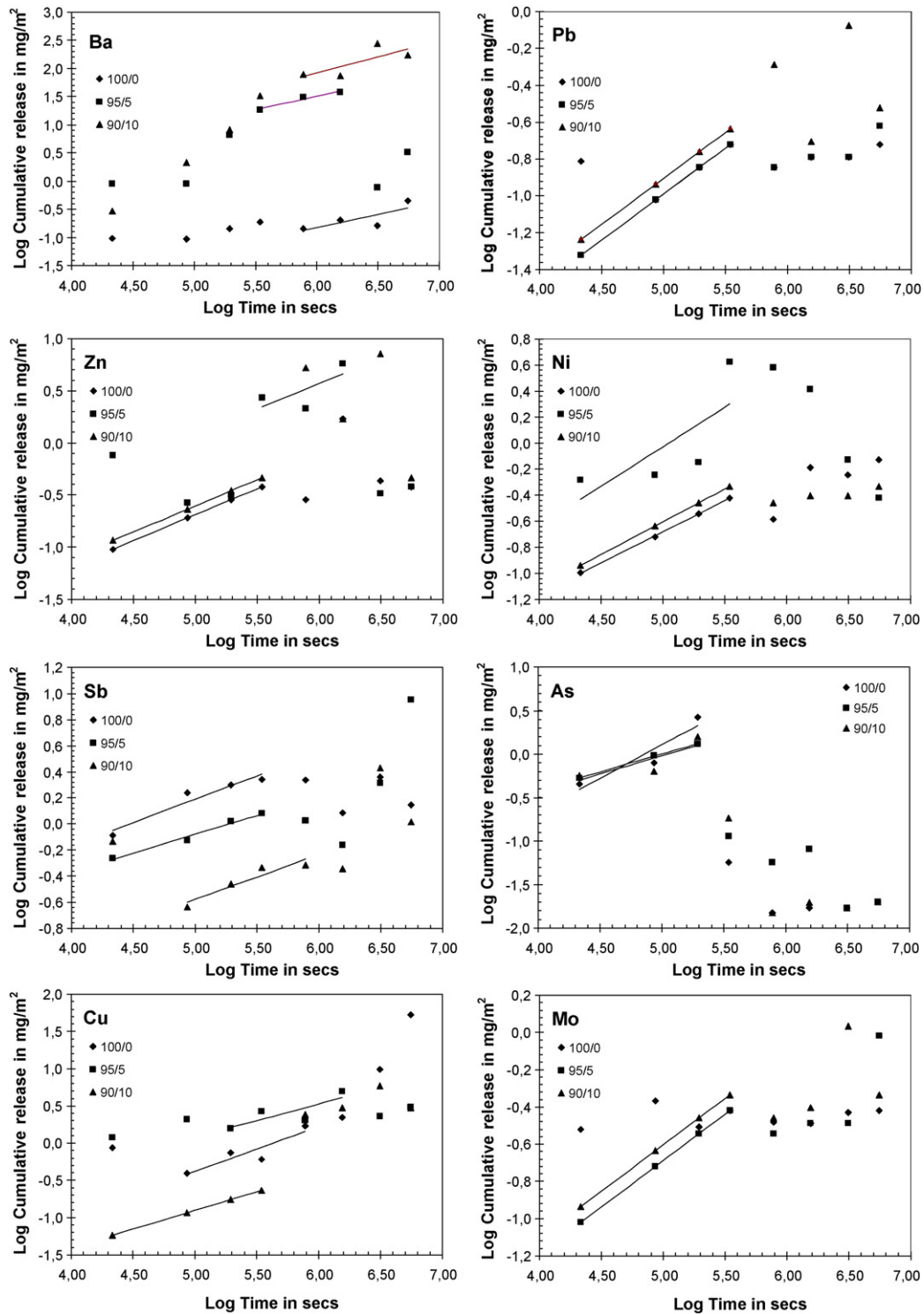


Fig. 5. . Log–log plots of the cumulative derived leaching vs. time.

according to the following formula:

$$\varepsilon_n^* = \sum_{i=1}^n E_i \quad \text{for } n = 1 \text{ to } N \quad (1)$$

where  $\varepsilon_n^*$  is the measured cumulative leaching of a metal for a period  $n$  ( $\text{mg m}^{-2}$ ),  $E_i$  is the measured leaching of the trace metal in the fraction  $i$  ( $\text{mg m}^{-2}$ ) and  $N$  is the total number of leachant renewal periods.

The same table also sets out the proposed leaching limit values for monolithic waste [34]. These values are for both stable non-reactive hazardous wastes in separate cells in non-hazardous landfills, and hazardous waste in hazardous landfills. Firstly, comparing the results of the different formulations with the control specimen showed that most of the trace metals released came from the residues used as aggregate. Only the release of Ba, whose occurrence may be related to the presence of portlandite in both the cement and the two forms of incineration waste, seems to be sig-

nificantly higher in the specimen formulated with natural gravel than in those formulated with WBA (100/0). In addition, it can also be observed in Table 7 that most of the trace metals are released to an extent that is below the established limits for stable non-reactive hazardous waste. However, some concrete formulations did show a release of Ba, Sb and/or As that was higher than the established limits for stable non-reactive hazardous waste, and some values were also higher than the limits for hazardous waste. The release of Ba exhibited the same behaviour as in the granular leaching test (see Table 6). The leachability of Ba increased in line with the total amount of APCFA used as aggregated material. As mentioned above, Ba solubility is strongly related to neoformed sulphate-basis phases.

Turning now to the MSWI bottom ash leaching studies, these demonstrated that the dissolved Sb-concentration was controlled by sorption of amorphous Fe/Al-(hydr)oxides [39] and the solubility of calcium bearing minerals, mainly portlandite and ettringite [40], as a function of natural pH of WBA. However, all these compounds are actively involved in cement hydration, and their solubility equilibria are highly influenced by the neoformed phases during Portland cement setting. Accordingly, the Sb incorporated in Fe/Al-(hydr)oxides and/or calcium-bearing minerals may be desorbed and released during the solidification process. Likewise, As is mainly trapped by amorphous Fe-(hydr)oxides [41] and could interact as arsenate with calcium-bearing minerals such as portlandite, calcite or gypsum [40], which are newly affected by the hydration process of Portland cement.

From the environmental point of view, according to the results summarized in Table 7, only those specimens of concrete formulated with a low APCFA percentage (95/5 and 90/10) seem to be suitable. This assertion is consistent with the mechanical results, where the specimens with the lowest percentages of APCFA were also those that showed the best compressive strength behaviour (see Fig. 3). Therefore, in order to determine which leaching mechanisms are involved in the release of different trace metals, log–log plots of cumulative derived leaching vs. time (Fig. 5) were derived for the 100/0, 95/5 and 90/10 formulations. In accordance with the standard NEN 7375 [30], derived cumulative leaching was calculated via the following equation:

$$\varepsilon_n = E_i \cdot \frac{\sqrt{t_i}}{\sqrt{t_i} - \sqrt{t_{i-1}}} \quad (2)$$

where  $\varepsilon_n$  is the derived cumulative leaching for a trace metal for period  $n$  ( $\text{mg m}^{-2}$ ),  $E_i$  is the measured leaching of the trace metal in the fraction  $i$  ( $\text{mg m}^{-2}$ ),  $t_i$  is the replenishment time of fraction  $i$  (days) and  $t_{i-1}$  is the replenishment time of fraction  $i - 1$ . Gradients below 0.35 indicate either surface wash-off or depletion. Gradients between 0.35 and 0.65 indicate diffusion controlled release, whereas gradients greater than 0.65 indicate dissolution.

As can be observed in the Fig. 5, and according to the criteria described above, the formulations including APCFA residue (95/5 and 90/10) yielded slopes for early leaching intervals indicate dissolution control for most trace metals studied, with depletion in the later leaching interval. Only the Ba release in both the 95/5 and 90/10 formulations showed diffusion control in the later periods. However, no trace metal studied showed entire diffusion control over the total increment period (i.e. fractions 2–7), and depletion or dissolution appear as leaching mechanisms in some fractions of the test. Likewise, the initial surface wash-off may play an important role in the final leaching concentration of trace metals. Thus, the initial concentration of components may not be dependent on the aggregates added in the concrete formulation, or the percentage of them, and could, in fact, be directly related to the surface wash-off process.

#### 4. Conclusions

The experimental results confirm the possibility of casting concrete with a combined use of MSWI bottom ash and fly ash as aggregates.

As regards an appropriate mechanical resistance, formulations with no more than 10% of APCFA as aggregate showed values higher than 15 MPa. This could be explained by pozzolanic reactions between BA and APCFA, in addition to an improvement in the particle size distribution. The results of two different leaching tests also show that formulations with less than 10% of APCFA are also the most environmentally suitable. For these formulations the release of possible pollutants appeared to be the lowest. In addition, the leaching mechanism determined for the best formulations showed that the surface wash-off process plays an important role in the final leaching concentration of trace metals.

Given the properties shown by 95/5 and 90/10 BA/APCFA concrete formulations we conclude that these may be appropriate for non-structural precast concrete, especially for pavers, mud slab concrete or New Jersey concrete barriers.

In summary, the combined use of WBA and APCFA as aggregate in concrete formulations has shown very promising results and several potential applications. However, this is a preliminary study and the conclusions are based solely on compressive strength results and the release of heavy metals in leaching tests. In addition to compressive strength a number of other mechanical parameters play a critical role (e.g. abrasion resistance of the aggregate, skid resistance, expansion–retraction, etc.) and should also be taken into consideration. Furthermore, the release of chlorides, sulphates and other anions must also be studied to determine the limits of any potential applications.

#### Acknowledgements

The authors wish to thank the SIRUSA and VECSA companies for their financial support and for providing access to sampling sites. Thanks are also given to Mrs. M. Vizcaino and J. Gómez for their assistance with chemical analysis and data management.

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