


Article

Granular Material Development Applied in an Experimental Section for Civil Engineering Purposes

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Abstract: In this study, a granular material (GM) derived from wastes generated in waste-to-energy plants was developed. Weathered bottom ash (WBA) and air pollution control (APC) ashes obtained from municipal solid waste incineration (MSWI) were used as raw materials. A mortar (M) with 50 wt. % of APC and 50 wt. % of Ordinary Portland Cement (OPC) CEM-I was prepared. The GM formulation was 20 wt. % M and 80 wt. % WBA. At the laboratory scale, WBA, APC, M, and crushed GM were evaluated by means of dynamic leaching (EN 12457-4) tests, and WBA, M, and crushed GM by percolation column (CEN/TS 16637) tests. The metal(loid)s analyzed were below the non-hazardous limits, regarding the requirement of the metal(loid)s released for waste revalorization. In order to simulate a road subbase real scenario, the crushed GM was tested in an experimental section (10 × 20 × 0.2 m). During a 600-day period, the leachates generated by the percolation of rainwater were collected. This research shows outstanding results regarding the metal(loid)s released for both the “accumulated” and “punctual” leachates collected. An accomplishment in the immobilization of metal(loid)s from APC residues was achieved because of the encapsulation effect of the cement. The GM formulation from both MSWI wastes can be considered an environmentally safe procedure for revalorizing APC residues.

Keywords: MSWI; bottom ash; APC residues; field scale road; mortar; granular material; leaching evaluation; percolation column test

1. Introduction

Incineration in waste-to-energy plants is currently the main treatment of municipal solid waste (MSW) in the European Union (EU), being the most reasonable alternative to landfilling because it allows both waste volume facilitation and weight reduction [1,2]. In 2017, about 70 Mt of MSW were incinerated in the EU. In fact, in the EU, Norway and Switzerland, a total amount of 463 municipal solid waste incineration (MSWI) plants are operating, and they incinerate around 90 Mt/y [3].

In the specific case of Spain (including Andorra), 11 waste-to-energy plants operated in 2019, treating approximately 2.5 Mt of MSW [3]. Specifically, in Catalonia (Northeast Spain), the separate collection of recyclables materials and bio-waste was introduced in 2000. It was expected they could recycle around 50% of the MSW by 2020. Nevertheless, the data in Figure 1 show that targets were not achieved, and the recycling rate was less than 40% [4]. This recycling rate is still very low, despite the efforts made by the public administration.

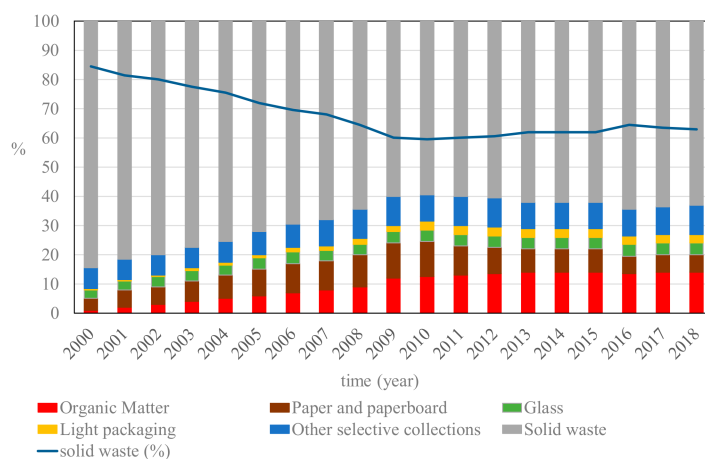


Figure 1. Catalan recycling tendency in the last few years [4].

MSWI presents mainly two types of waste, named bottom ash (BA) and air pollution control (APC). BA represents the major solid output stream residue from MSWI, and it is the 85 wt. % of the solid resulting from combustion [5]. This residue is classified as non-hazardous waste (EWC 190101), and it is mainly composed of Si, Al, Ca, and Na oxides as major elements, with traces of metal(loid)s [6]. Regarding the materials' compositions, BA is heterogeneous and contains glass, ceramic, concrete, brick, debris, sand, metal, stone and fused clinker, among others [1]. The current practice of BA utilization in Europe involves its use in relatively simple civil engineering applications or disposal by landfilling. Some of the applications of BA are as (a) a precursor of alkali-activated materials (AAM), (b) adsorbent materials for the removal of hazardous elements from wastewater and landfill gases, (c) a soil replacement in agricultural activities, (d) a raw materials substitute for ceramic-based products manufacturing, (e) landfill cover, and (f) a biogas upgrading and production enhancer [7–9]. After a combustion temperature of 950 °C [10], BA is further processed in a conditioning plant to recover some valuable materials (ferrous and non-ferrous metals) and to obtain a homogenized granular material. When BA is stabilized through an outdoor maturation (ageing) treatment for 2–3 months, the obtained by-product is called weathered bottom ash (WBA), which is valorized for engineering purposes. This maturation mainly consists of carbonation and a pH stabilization between 8 and 10 [10], which ensures that metal(loid)s are stabilized. There are different applications for WBA as a secondary raw material, such as in civil engineering purposes as a secondary aggregate for road construction [11,12], embankments [13] and pavements [14], and as concrete aggregate [15]. WBA is also used in the building sector as a ceramic material, such as in tiles, bricks and glass-ceramics, as reported elsewhere [16,17]. In the case of Spain (except in Catalonia) and some other EU countries [3], there is no regulation regarding WBA utilization, and disposal via landfill is still the most common MSW management.

APC is the other waste generated during the MSWI, which mainly contains a mixture of incinerated fly ashes (IFA), products of the neutralization of acid gases (calcium chloride, calcium sulfate or calcium carbonate, among others) and an excess of lime (in semi-dry scrubbers using lime slurry). Some authors evaluated the stabilization of IFA from MSW using hydraulic binders such as Ordinary Portland cement (OPC), lime, coal fly ash or blast furnace slag [18,19]. The main concern regarding the APC residues is the high content of heavy metal(loid)s, dioxins, furans and soluble salts [20,21]. Consequently, APC is currently classified as a hazardous waste [22], and it is handled by landfill disposal after stabilized [23]. Hence, cement-based treatment for formulating mortars [24] is the most employed means of APC stabilization, not only due to economic factors but also because of its ease of handling [15].

This paper aims to present the most relevant findings related to the WBA and APC derived from MSWI in formulating a granular material (GM). The assessment of its potential joint reuse was initiated at laboratory scale, evaluating the chemical performance of both the raw materials (WBA and APC) as

well as the materials formulated from them, i.e., mortar (M) generated for the APC stabilization, and the final GM. The environmental evaluation by means of dynamic leaching assessments (EN-12457-4) [25] for WBA, APC, M and GM, and percolation column tests (CEN/TS 16637) [26] for WBA, M and GM, were also performed. Finally, the optimal formulation of GM was tested in a real experimental section as a granular layer, simulating a road subbase scenario, where two types of leachates were generated by the percolation of rainwater (“accumulated” and “punctual”), and they were evaluated over almost two years by means of inductively coupled plasma mass spectrometry (ICP-MS), analyzing the elements described in the European Landfills Directive.

2. Materials and Methods

2.1. Materials

The WBA and APC samples were collected from a MSWI facility located in Tarragona (Catalonia). This facility is mainly fed with household rubbish with a minor input from commercial sources. Every year, an average of 33,500 t and 3500 t of BA and APC are produced, respectively. Prior to the outdoor process of weathering, a supplementary treatment via a conditioned/revalorization process is performed for the recovery of valuable metals.

For potential reuse, the APC was previously stabilized by a solidification process to obtain a mortar, using OPC as a binder. The mortar (M) was formulated with 50 wt. % of OPC CEM I and 50 wt. % of APC [1]. Considering these percentages, and anticipating their use in a real scenario, 20 t of M was prepared, using the necessary water to provide a workable mixture.

In order to prepare 50 m³ of GM, after 28 days of curing, the hardened M was crushed (Figure 2a) to obtain an aggregate with a particle size lower than 30 mm. The WBA ($\rho = 1.73 \text{ kg}\cdot\text{cm}^{-3}$) was also prepared (Figure 2b). Afterwards, M ($\rho = 1.85 \text{ kg}\cdot\text{cm}^{-3}$) and WBA were stored in stockpiles (Figure 3a). Finally, WBA was mixed to create the GM [1,27], as shown in Figure 3b. Around 18.5 t of crushed M and 69.2 t of WBA were taken for the formulation of GM.



Figure 2. (a) Image of the particle size of crushed M and (b) spillage of WBA in the plant feeding silos.



Figure 3. (a) Crushed M (grey) and WBA (dark) stockpiles (b) GM obtained from the mixture between 80 wt. % WBA and 20 wt. % crushed M.

The real experimental section with the GM developed in this study was implemented, simulating a road subbase layer in Clariana de Cardener (Lleida, Catalonia, Spain). To this end, over a high-density polyethylene (HDPE) geomembrane, different 1.5 mm thick pipes distributed as fish spines were placed to collect the leachates after raining, as Figure 4 shows. On the top of the pipes, a draining natural

gravel cover was put in order to avoid the obstruction of the pipes produced by the fine particles of the GM. Regarding the experimental scenario, the total surface of the experimental section with GM was 200 m² (10 m × 20 m), and it was 0.2 m thick. In addition, a 10 cm thick natural gravel cover was also put over the GM layer to avoid the materials' losses due to adverse weather conditions in the area.

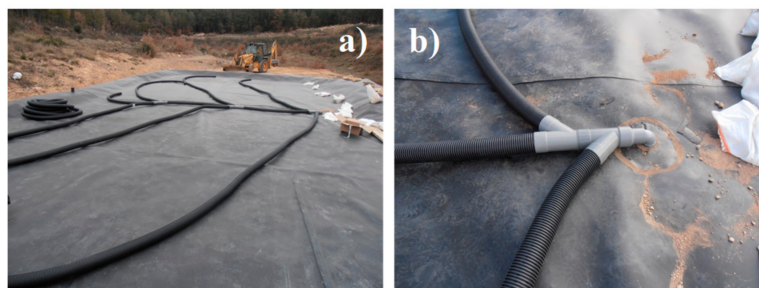


Figure 4. (a) Image of the distribution of the pipes as fish spines. (b) Detail of the pipes unification before collecting the leachates in the collection tank.

2.2. Methods

The WBA, APC, crushed M and GM were homogenized and quartered (1/16), obtaining different subsamples. Representative subsamples of WBA, APC, crushed M and GM were taken for chemical characterization. The semi-quantitative elemental analysis of WBA, APC, crushed M and GM was carried out by a spectrophotometer Panalytical Philips PW 2400 sequential X-ray fluorescence (XRF) equipped with the software UniQuant® V5.0. Besides this, X-ray diffraction (XRD) analysis was used to determine the crystalline phases of the samples by means of a Bragg–Brentano Siemens D-500 powder diffractometer device with CuK α radiation. The humidity (H, %) and dried weight (W_D, %) of the samples were also evaluated.

Dynamic leaching tests were performed for four samples per duplicate, following the standard UNE-EN 12457-4. The dynamic leaching tests followed the Department of the Environment of the Generalitat de Catalunya's [28] classifying of solid wastes in accordance with the limits established in the Criteria and Procedures for the Waste Acceptance in Landfills [29], described in the European Landfills Directive. Thus, based on their leaching potential, three levels of different wastes were established, as showed in Table 1: inert solid wastes, non-hazardous wastes and hazardous wastes. The dynamic leaching test consisted of placing a solid with a particle size < 10 mm in contact with deionized water in an L/S ratio of 10 L·kg^{−1} for 24 h at room temperature, with continuous rotating agitation. The leachates were filtered with a 0.45 μ m membrane of nitrocellulose. Two aliquots were tested per sample. Two replicates of each replicate were extracted for further analysis by means of the ICP-MS technique by a PerkinElmer ELAN device, evaluating As, Ba, total Cr, Cu, Hg, Mo, Ni, Pb and Zn. Likewise, the pH and conductivity (k) of the leachates were analyzed.

Table 1. Limit values (mg·kg^{−1}) established by the European Landfill Directive [29] to classify solid waste for its admission to landfills.

| Element | Inert | Non-Hazardous | Hazardous |
|---------|-------|---------------|-----------|
| As | 0.5 | 2 | 25 |
| Ba | 20 | 100 | 300 |
| Cr | 0.5 | 10 | 70 |
| Cu | 2 | 50 | 100 |
| Hg | 0.01 | 0.2 | 2 |
| Mo | 0.5 | 10 | 30 |
| Ni | 0.4 | 10 | 40 |
| Pb | 0.5 | 10 | 50 |
| Zn | 4 | 50 | 200 |

In addition, percolation column long-term tests on the WBA, crushed M and GM were performed per duplicate, following the CEN/TS 16,637 standard. These tests consist of placing around 1 kg of dry material (separately WBA, crushed M and GM) in a cylindrical column ($\emptyset = 50$ mm; $h = 310$ mm). The deionized water was passed through from the bottom to the top of the column (bottom-up) with the help of a pump, set at 24 mL·h⁻¹. The L/S ratios evaluated were 0.1, 0.2, 0.5, 1, 2, 5 and 10. Afterwards, for each of the seven eluates (E1, E1, E3, E4, E5, E6 and E7) obtained for each column, the determined parameters were pH, k, and the analysis by ICP-MS of As, Ba, total Cr, Cu, Hg, Mo, Ni, Pb and Zn.

There exist some leaching test methods available that can differ according to aspects such as the material type to be tested, the mass, the particle size, the leachate volume and the duration of the assay. Leaching tests for soils can be classified as batch tests, percolation column tests, and sequential leaching tests or monolithic and bulk tests [30]. These methods aim to determine the concentrations of chemicals in the water that was in contact with the soil for a certain period [31]. Different standardized batch tests and percolation column tests are available for assessing the metal(loid) solubility and associated leaching. It is necessary to homogenize and sieve the soil before leaching [32]. The leaching behavior depends on the following variables: leachate composition (e.g., pH), method and time of contact, and liquid-to-solid (L/S) ratio [33].

Batch tests are widely used for compliance testing because of the low costs, simple design and low duration [31]. The contact time of 24 h is too short for some metals to reach equilibrium, resulting in an underestimation of metal(loid) solubility [32]. The batch test represents the process of mixing a solid mass into groundwater so as to confirm the effect of this process of groundwater [34]. Nevertheless, the main disadvantage of the batch test is that the provided information is limited, as it only offers a single result in terms of L/S ratio, which does not reflect real-world conditions [31]. On the other hand, percolation column tests better simulate the field conditions and are appropriate to assess the long-term release of chemical constituents from the soil into water and groundwater. The percolation column tests resemble the process of the percolation/seepage of water into a soil structure to examine the effect of leachate on the environment [34]. Besides, the column percolation leaching tests simulate the time-dependent percolation behavior of solids in surface waters and groundwater. Even though column percolation tests are laboratory assays, they simulate natural conditions closer than any other test, and provide more robust results than the batch tests [30]. The advantages of the percolation column test over a batch test are related to the high initial concentrations of the percolates at low L/S ratios and the time-dependent release of the chemicals (necessary for the prediction of leaching behavior under field conditions). However, percolation column tests are more expensive, and more labor and effort is needed compared to batch tests [31]. Therefore, while the static batch tests offer information about the cumulative mobilized mass of pollutant, the percolation column tests are more representative of source term behavior [35], but are also time-consuming [31]. Percolation column tests approximate better to leaching processes under field conditions, and thus are more suitable for prediction purposes [33].

As stated elsewhere [36], column and batch leaching tests are of increasing importance for the recycling of waste materials, especially in the context of the recent Waste Framework Directive. They show that the L/S ratio is a convenient normalization parameter to compare different dynamic leaching tests [35]. Besides, it is important to highlight that the L/S ratio is a proper parameter for comparing between both tests, as it is shown that the leaching behavior is independent of the duration and the physical dimensions of the leaching test.

It is usual that when characterizing the leaching behavior of a material using different dynamic leaching tests, the results obtained are different in terms of the metals concentrations released (mg/kg) [37]. Nevertheless, their order of magnitude and their relative significance depends on the type of L/S contact (which in the dynamic leaching tests and the percolation column tests is the same). Usually, the physico-chemical mechanisms governing pollutant release are identical, although they can present different values on the leaching time scale and in the different tests performed [37]. Apart from the laboratory assays, realistic impact simulations for leaching behavior must consider the

physico-chemical characteristics of the metals studied. To sum up, both percolation and batch tests can be used for characterization or compliance [33].

Considering that all the rain percolates, an extrapolation of the time can be predicted for the experimental section, simulating a road subbase layer over real years, considering the four following parameters: (i) 200 m² surface and 0.2 m thick, (ii) an average rainfall in the area over the last 14 years of 584 L·m⁻²·year⁻¹, (iii) a volume of the column of 589 cm³, (iv) and 10 L·kg⁻¹. Therefore, the end of the percolation column test can be approximately extrapolated to 60 years, regardless of the percolation rate. In fact, according to the previous data, the first eluate (E1) corresponds to half a year, the second eluate (E2) corresponds to 1 year, the third eluate (E3) corresponds to 3 years, the fourth eluate (E4) corresponds to 6 years, the fifth eluate (E5) corresponds to 11 years and a half, the sixth eluate (E6) corresponds to 29 years, and the seventh (E7) corresponds to 58 years.

The rainfall area was considered to properly interpret the results obtained. The rainwater samples were collected on the first not-rainy day after one (or some) rainy day(s) with the help of two different pluviometers. From the total extension of 200 m², 199 m² were intended for the rainwater leachates' evaluation. The leachates must pass through the 0.2 m of the surface's thickness. This area was called "punctual" Figure 5a). A 1000 L "punctual" tank collected the percolated rainwater samples. The "punctual" leachates were collected each time it rained, and afterwards the "punctual" 1000 L tank was emptied. On the other hand, a 1 m² area was called "accumulated", and we evaluated the leaching behavior of that specific surface (Figure 5b). The tank for the "accumulated" leachates was not emptied in any case. The collection of the "accumulated" percolated rainwater was made through two connected volume-measured deposits, one of 30 L in order to be more precise when collecting the first leachates, and one of 1000 L to collect the "accumulated" leachates of the 1 m² area (Figure 6). Therefore, both kinds of leachates were analyzed each time it rained for almost two years. The L/S ratio value in the experimental section at the end of the study was 2, considering both "accumulated" and "punctual" leachates.



Figure 5. Experimental section image (a) "punctual" area, 199 m², (b) "accumulated" area, 1 m².



Figure 6. The two "accumulated" rainwater tanks (front) and the "punctual" rainwater tank (behind).

After every rainy day (or after a period of rainy days), around 500 mL of "accumulated" and "punctual" rainwater leachates was collected. After 600 days, 54 leachates from the percolated rainwater tanks were compiled; 27 from the rainwater "punctual" tank and 27 from the "accumulated" tank. It is important to highlight that, in order to make a proper comparison between the "accumulated"

and “punctual” leachates, the “punctual” concentration for each element analyzed was calculated considering the previously determined results, making an integration by adding each previous punctual concentration; this was the punctual–accumulated. From the “accumulated” and “punctual” tanks, the pH, conductivity ($\mu\text{S}\cdot\text{cm}^{-1}$) and As, Ba, total Cr, Cu, Hg, Mo, Ni, Pb and Zn metal(loid)s ($\text{mg}\cdot\text{kg}^{-1}$), by means ICP-MS, were analyzed for all the 54 collected leachates.

3. Results

3.1. Chemical Composition: XRF and XRD

The chemical composition results in terms of the most stable oxides determined by XRF for WBA, APC, crushed M and GM are presented in Table 2. The major compounds for WBA sample are SiO_2 and CaO , due to the packaging glass and ceramics content of MSW [38]. The Fe_2O_3 and Al_2O_3 contents are likewise noteworthy, wherein the former is related to the existence of unrecovered ferrous metals while the latter is the consequence of non-ferrous metals and ceramics such as clays. Nevertheless, as expected, the SiO_2 content is low in APC residues, which are mainly composed of lime (CaO), the main compound used as an alkaline neutralizer during flue gas scrubbing from combustion. The Cl amount is significant in APC because of the polyvinyl chloride (PVC) combustion, and its leaching causes concern in terms of the APC residues’ landfill treatment [39]. Besides, the LOI value is significant due to the use of $\text{Ca}(\text{OH})_2$ for gas neutralization in the APC system, whereby part of the excess of $\text{Ca}(\text{OH})_2$ leads to a formation of CaCO_3 . Meanwhile, the SO_3 amount is related to the presence of CaSO_4 . After the stabilization of APC with OPC to obtain the crushed M, its chemical composition is close to that of APC residues, with the supplementary contribution of OPC affecting SiO_2 content. Similarly, the GM composition was comparable to that of WBA, with the additional amount of CaO from APC residues.

Table 2. Chemical composition determined by means XRF for the WBA, APC, crushed M and the GM formulated.

| Compound | wt. % | | | |
|-------------------------|-------|-------|-----------|-------|
| | WBA | APC | Crushed M | GM |
| SiO_2 | 43.30 | 6.64 | 21.70 | 39.15 |
| CaO | 16.90 | 48.35 | 45.30 | 26.59 |
| Cl | 0.14 | 8.86 | 9.08 | 0.89 |
| Fe_2O_3 | 14.10 | 0.79 | 1.65 | 16.89 |
| Na_2O | 7.58 | 4.28 | 4.44 | 3.71 |
| Al_2O_3 | 5.80 | 4.02 | 3.82 | 4.87 |
| MgO | 2.22 | 1.73 | 1.87 | 2.02 |
| K_2O | 1.11 | 4.33 | 2.10 | 1.67 |
| CuO | 0.23 | - | - | 0.13 |
| SO_3 | 0.65 | 6.24 | 4.53 | 1.53 |
| TiO_2 | 0.35 | 0.86 | 0.68 | 0.30 |
| P_2O_5 | 2.97 | 1.36 | 0.51 | 1.10 |
| ZnO | 0.18 | 0.72 | 0.32 | 0.19 |
| PbO | - | 0.12 | 0.13 | 0.03 |
| LOI | 4.47 | 11.7 | 3.87 | 0.93 |

Considering the mineral phases, the XRD patterns obtained for WBA and GM show as main components the amorphous SiO_2 and CaCO_3 . For the crushed M sample, the predominant phases are halite (NaCl) and sylvite (KCl) from APC, due to the fact that the hydrated phases of OPC are less crystalline than the determined ones. If more information about the present mineral phases is required, the following references [1,40] should be reviewed.

3.2. Dynamic Leaching Test (EN-12457-4)

In Table 3 is shown the WBA, crushed M and GM dynamic leaching tests results. Most of the metal(loid)s concentrations in the leachates from the samples under study are below the limits established for inert and/or non-hazardous waste. From the APC residues results, the Pb and Zn concentrations are noticeable, as the leaching is noteworthy. Nevertheless, the encapsulation effect of OPC was very effective, as could be observed in the crushed M results. With these results, the GM developed could be revalorized as a secondary source, or as secondary arid.

Table 3. Results of humidity (H, %), dried weight (W_D, %), pH, k (mS·cm⁻¹) and heavy metal(loid)s concentrations (mg·kg⁻¹) for the WBA, APC, crushed M and GM.

| | H (%) | W _D (%) | pH | k (mS·cm ⁻¹) | As | Ba | Total Cr | Cu | Hg | Mo | Ni | Pb | Zn |
|--------------------------|---------------------|--------------------|-------|-----------------------------|--------|------|----------------|-------|------|-----|------|--------|--------|
| WBA | 7.34 | 93.00 | 12.09 | 2.97 | 0.010 | 2.5 | 1.780 | 9.215 | 0.01 | 2.0 | <0.2 | 0.395 | 1.535 |
| APC | 2.54 | 97.46 | 12.50 | 5.84 | <0.520 | 39.3 | <0.040 | 0.790 | 0.01 | 2.5 | <1.3 | 47.350 | 19.830 |
| Crushed M | 15.19 | 84.81 | 12.66 | 16.33 | 0.010 | 97.4 | 1.595 | 0.600 | 0.01 | 2.8 | <6.1 | 5.600 | 1.805 |
| GM | 10.66 | 98.5 | 11.31 | 1.61 | 0.010 | 9.7 | 1.140 | 5.600 | 0.01 | 1.8 | <1.1 | 0.415 | 4.310 |
| Inert limit | - | - | - | - | 0.5 | 20 | 0.5 | 2 | 0.01 | 0.5 | 0.4 | 0.5 | 4 |
| Landfill ^a | Non-hazardous limit | - | - | - | 2 | 100 | 10 | 50 | 0.2 | 10 | 10 | 10 | 50 |
| | Hazardous limit | - | - | - | 25 | 300 | 70 | 100 | 2 | 30 | 40 | 50 | 200 |
| Utilization ^b | | | | | 1 | - | 5 ^c | 20 | 0.2 | - | 5 | 5 | 20 |

^a Catalan Order Number 5370/30.4.2009 [41], ^b Catalan Order Number 2181/13.3.1996 [42], ^c Max. Cr(VI): 1 mg·kg⁻¹.

3.3. Percolation Column Test (CEN/TS 16637) for the WBA, Crushed M and GM

The total amount analyzed for each material and the amount of stabilized soil (m₀) were evaluated per duplicate for the WBA, crushed M and GM. The results are shown in Table 4.

Table 4. Data required in the percolation column tests.

| Material | Total Mass (kg) | | m ₀ (kg) | |
|----------|-----------------|-------|---------------------|-------|
| WBA | 0.825 | 0.784 | 0.767 | 0.729 |
| M | 0.710 | 0.578 | 0.600 | 0.490 |
| GM | 0.830 | 0.829 | 0.741 | 0.741 |

As was previously detailed, seven L/S ratios (eluates) were evaluated for each material (WBA, crushed M, GM), up to a L/S ratio of 10 L/kg, as specified in the standard. Table 5 presents the mean values of the metal(loid)s for the two replicas of the percolation column tests performed for each material. It is senseless to discuss the results from the eluates E1–E6 since nowadays there is no legislation. In this regard, it would be inappropriate to describe the tendency between E1 and E6 and to compare the values with the limits in the EN-12457 standard, as E1–E6 only represent trends in the metal(loid)s release. As expected, when the L/S ratio increases, the eluate concentration values decrease. Although it is important to observe the behavior when L/S = 10, E5 (L/S = 2) is the value used to compare between both standards, CEN/TS 16,637 and EN-12457-4. The results in Table 5 show that in E7 the leaching in the WBA is below the inert limits. When evaluating the crushed M's results in E7, the As, Cu, Mo and Zn are below the inert limits; meanwhile, Ba, total Cr, Hg, Ni and Pb are above the inert limits. This is because of the APC's composition. Despite all these results, it is very important to highlight that in E7 for GM the leaching behavior is below the inert limits. As such, by comparing the E7 values for GM and for crushed M, it can be concluded that the APC fly ash residue is stable when it is used for preparing the GM evaluated in this study.

Table 5. Percolation column tests (CEN/TS 16,637) results for the WBA, crushed M and GM. The metal(loid)s units are $\text{mg}\cdot\text{kg}^{-1}$. (Red color, orange color and yellow color for values above hazardous, non-hazardous and inert limits, respectively, according to Catalan Order Number 5370/30.4.2009).

| | Eluates | L/S | pH | k (μS/cm) | As | Ba | Total Cr | Cu | Hg | Mo | Ni | Pb | Zn |
|---------------------|---------|------|-------|-----------|------|-------|----------|-------|------|-------|------|-------|------|
| WBA | E1 | 0.1 | 11.90 | 26.15 | 0.28 | 1.06 | 28.77 | 28.18 | 0.01 | 10.26 | 0.27 | 1.29 | 1.34 |
| | E2 | 0.2 | 12.08 | 28.65 | 0.30 | 1.25 | 19.88 | 29.77 | 0.01 | 10.77 | 0.20 | 0.76 | 1.28 |
| | E3 | 0.5 | 11.76 | 18.95 | 0.26 | 0.75 | 12.59 | 21.34 | 0.01 | 7.68 | 0.13 | 3.61 | 1.36 |
| | E4 | 1.0 | 11.97 | 9.23 | 0.10 | 0.16 | 6.07 | 10.33 | 0.01 | 4.00 | 0.04 | 2.82 | 1.35 |
| | E5 | 2.0 | 12.20 | 3.85 | 0.03 | 0.10 | 3.28 | 4.77 | 0.01 | 2.06 | 0.03 | 7.41 | 0.30 |
| | E6 | 5.0 | 10.87 | 1.38 | 0.02 | 0.05 | 0.86 | 1.59 | 0.01 | 0.73 | 0.01 | 0.07 | 0.28 |
| | E7 | 10.0 | 10.11 | 0.81 | 0.01 | 0.04 | 0.30 | 0.70 | 0.01 | 0.27 | 0.03 | 0.05 | 0.28 |
| Crushed M | E1 | 0.1 | 13.33 | 90.05 | 1.26 | 17.00 | 5.54 | 3.25 | 0.22 | 4.84 | 1.66 | 35.37 | 2.60 |
| | E2 | 0.2 | 13.42 | 90.40 | 1.71 | 20.21 | 7.06 | 5.28 | 0.15 | 7.31 | 1.32 | 19.31 | 2.60 |
| | E3 | 0.5 | 13.29 | 68.75 | 1.05 | 17.74 | 6.39 | 2.44 | 0.15 | 3.61 | 0.89 | 2.70 | 2.60 |
| | E4 | 1.0 | 13.57 | 45.05 | 0.67 | 24.00 | 3.39 | 1.40 | 0.15 | 2.05 | 0.88 | 4.26 | 2.60 |
| | E5 | 2.0 | 13.46 | 24.90 | 0.32 | 31.53 | 3.00 | 0.69 | 0.11 | 0.92 | 0.91 | 5.78 | 1.41 |
| | E6 | 5.0 | 13.33 | 12.15 | 0.17 | 31.99 | 2.53 | 0.48 | 0.11 | 0.40 | 0.69 | 5.23 | 1.60 |
| | E7 | 10.0 | 12.94 | 4.37 | 0.05 | 27.99 | 2.32 | 0.13 | 0.02 | 0.19 | 0.44 | 5.78 | 0.40 |
| GM | E1 | 0.1 | 13.11 | 49.40 | 0.83 | 6.12 | 3.96 | 36.63 | 0.01 | 6.08 | 0.93 | 3.42 | 4.94 |
| | E2 | 0.2 | 13.14 | 47.50 | 1.08 | 3.66 | 4.18 | 47.51 | 0.01 | 8.09 | 0.98 | 4.86 | 8.04 |
| | E3 | 0.5 | 13.37 | 30.70 | 0.48 | 2.10 | 2.70 | 19.68 | 0.01 | 3.23 | 0.38 | 1.92 | 3.37 |
| | E4 | 1.0 | 13.54 | 18.23 | 0.18 | 2.73 | 2.70 | 7.64 | 0.01 | 1.20 | 0.43 | 1.63 | 2.70 |
| | E5 | 2.0 | 12.81 | 8.04 | 0.04 | 2.81 | 0.03 | 2.31 | 0.01 | 0.34 | 0.20 | 1.00 | 0.69 |
| | E6 | 5.0 | 12.89 | 4.00 | 0.02 | 2.71 | 0.03 | 0.90 | 0.01 | 0.14 | 0.15 | 0.76 | 0.39 |
| | E7 | 10.0 | 11.82 | 2.82 | 0.02 | 1.97 | 0.03 | 0.51 | 0.01 | 0.10 | 0.13 | 0.46 | 0.15 |
| Inert limit | | - | - | - | 0.5 | 20 | 0.5 | 2 | 0.01 | 0.5 | 0.4 | 0.5 | 4 |
| Non-hazardous limit | | - | - | - | 2 | 100 | 10 | 50 | 0.2 | 10 | 10 | 10 | 50 |
| Hazardous limit | | - | - | - | 25 | 300 | 70 | 100 | 100 | 30 | 40 | 50 | 200 |

3.4. Experimental Section: “Accumulated” and “Punctual” Rainwater Leachates

As previously mentioned, the rainwater leachates were collected each time it rained in the area of the experimental section. The collection of the leachates was done on the first not-rainy day. This means that if, for example, on three consecutive days it rained, the leachates were assembled on the fourth day. The rainwater samples were assembled 27 times, after almost 600 days. The following results (Figures 7 and 8a–i) compare the “accumulate” and the “punctual” leachates, expressing the “punctual” as an addition of the concentration of each previous metal(loid) concentration analyzed. This means the “punctual” leachates are expressed as an integration of each previous concentration result, in order to compare them with the “accumulated” results.

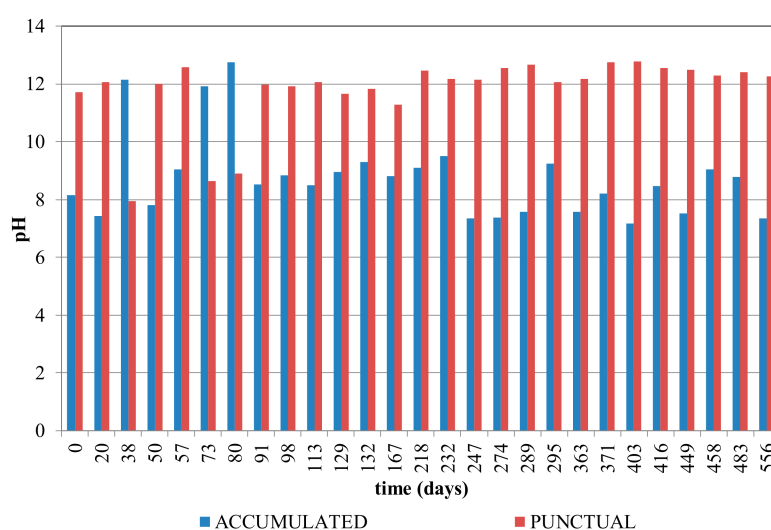


Figure 7. pH results obtained from the leachates of the “accumulated” (blue) and “punctual” (red) rainwater tanks.

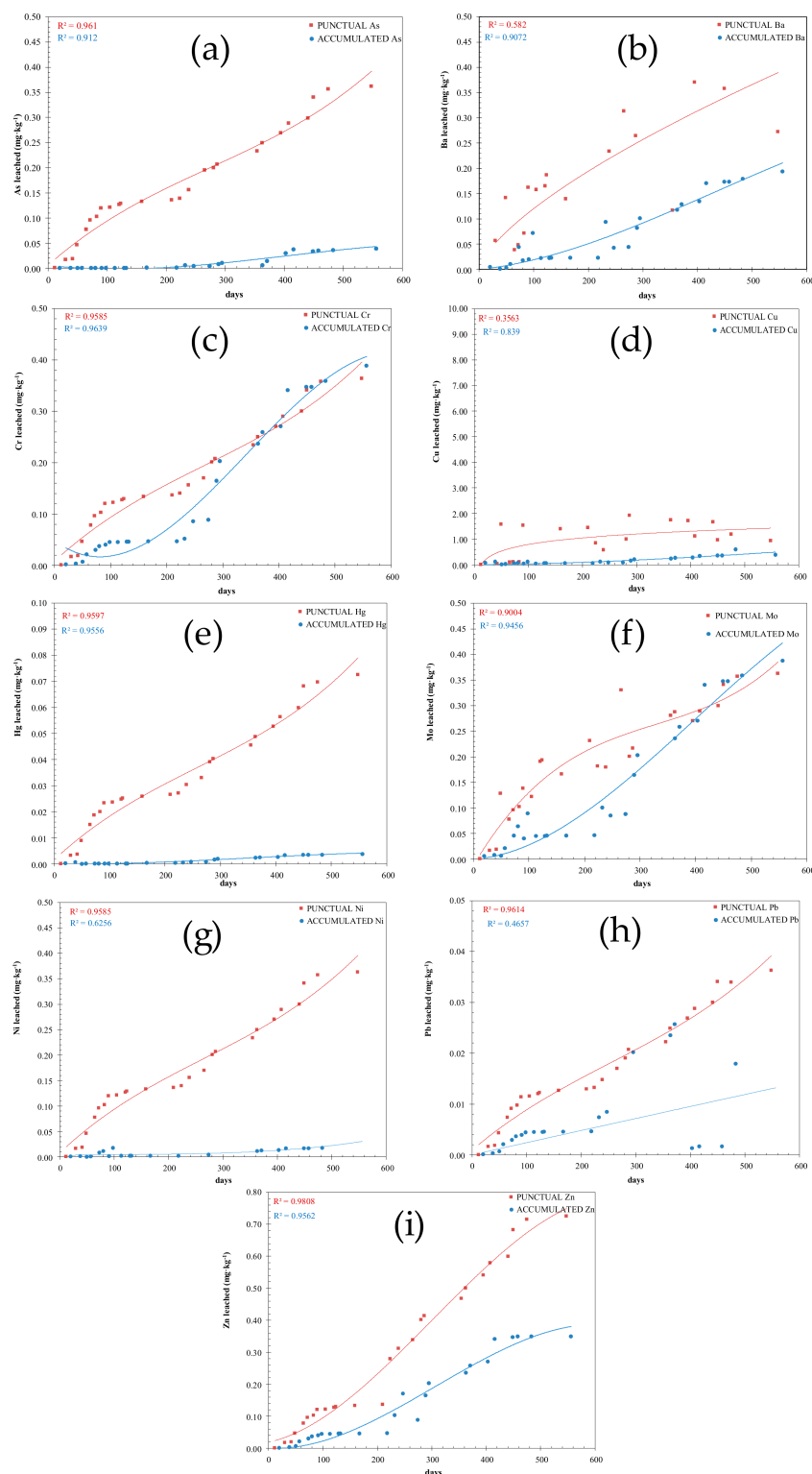


Figure 8. Concentration for both “accumulated” and “punctual” samples (a) As, (b) Ba, (c) Total Cr, (d) Cu, (e) Hg, (f) Mo, (g) Ni, (h) Pb and (i) Zn.

Figure 7 shows the pH results from the “accumulated” and “punctual” rainwater leachates. As the figure shows, in general, the pH in the “punctual” leachates is higher than in the “accumulated” ones, due to the saturation of the solution because of the high lime (CaO) content. The pH in the “accumulated” leachates is close to the pH controlled by the Ca(OH)₂. Besides, although both types of

leachates have the same L/S ratio (a value of 2), the “punctual” pH is higher because, as the surface is higher, the ability to collect rainwater is also higher. Therefore, as the “accumulated” area receives more leaching rainwater, it may not have achieved the saturation, or the lime concentration in this surface is lower. It would also be important to consider the evaporation phenomenon as another parameter to consider in the L/S ratio (it is not contemplated in this study). Regarding the “accumulated” ones, the pH values were between 7 and 9, although two samples at the beginning of the experimental section were $\text{pH} > 12$. The pH of “punctual” samples was more constant, and in general, more alkali (around $\text{pH} = 12$), compared to the “accumulated” leachates. This behavior is usual for the M sample, as it is controlled by the CaO solubility of both the APC and OPC binder at a pH around 12. This pH factor is important to consider regarding the release of some metal(loid)s, because some of them are pH-dependent (such as As (III), Cr (III), Cu, Ni, Pb and Zn).

The results by means of ICP-MS analysis of each of the analyzed metal(loid)s are presented in Figure 8, considering the concentration of the metal(loid)s before the percolation days. A comparison between the “accumulated” and the “punctual” leachates is performed. As previously mentioned, the L/S ratio in the experimental section was 2 in both “accumulated” and “punctual” leachates. Therefore, all the metal(loid)s concentration results are compared with those named E5 in the percolation column tests, as the L/S is 10 in the dynamic leaching tests. Then, it can be expected that, in the real experimental section, when achieving an L/S ratio of 10, the leachates’ concentrations will decrease even more. Besides, as the pH in the “punctual” leachates is much higher than in the “accumulated” ones, the leaching is also higher for the “punctual” ones in most of the cases.

In Figure 8a is presented the As concentration results. Although the concentration in the “punctual” leachates is higher than in the “accumulated” samples after 556 percolation days, the concentrations of As in the “accumulated” leachates are similar to those in the E5 in the percolation column tests ($\text{L/S} = 2$). Besides, all the concentration results are below the inert limits established for $\text{L/S} = 10$ ($0.5 \text{ mg}\cdot\text{kg}^{-1}$ for inert waste, $2 \text{ mg}\cdot\text{kg}^{-1}$ for non-hazardous waste, and $25 \text{ mg}\cdot\text{kg}^{-1}$ for hazardous waste). Figure 8b shows the Ba concentration, wherein the “accumulated” and the “punctual” samples are comparable, as the obtained concentrations demonstrated the same magnitude values. Nevertheless, the results in the experimental section after almost 2 years of percolation are lower than those obtained in the percolation column tests. Actually, the concentrations in the experimental section and in the percolation column tests are below the inert and non-hazardous limits established by the European Landfill Directive [29]. Figure 8c shows the results of the concentrations of the total Cr for the “accumulated” samples after 556 days of percolation, and the “punctual” samples after 548 days. Both results for the “accumulated” and “punctual” samples in terms of the total Cr analysis are equivalent. The results obtained in the experimental section, compared to E5 in the percolation column tests, show that the former results are higher. Nevertheless, in both cases, the concentration is 25 times lower than the non-hazardous limit in all the eluates studied, where the established threshold value is 10 ppm ($10 \text{ mg}\cdot\text{kg}^{-1}$). The Cu concentrations as determined by ICP-MS in both “accumulated” and “punctual” tanks are shown in Figure 8d. As can be observed, both the “accumulated” and “punctual” results are similar. All the obtained values are much lower than those obtained in E5 in the percolation column tests. Besides, all the values are below the non-hazardous limit established by the European Landfill Directive, of 50 ppm. Following the same procedure, in Figure 8e is presented the Hg concentration results for the “accumulated” and “punctual” leachates after 556 and 548 days of percolation, respectively. The “punctual” leachates’ values are higher than the “accumulated” ones, and the “accumulated” and E5 results are comparable. Besides, all the values obtained are below the non-hazardous limit (0.2 ppm), although the behaviors of the leachates in both tanks are different. When studying the Mo element by means ICP-MS for the “accumulated” and “punctual” leachates (Figure 8f), it can be observed that the results obtained are comparable. Besides, all the values are very similar to those obtained in E5 in the percolation column tests, and all of them are below the non-hazardous limit (10 ppm). Besides, the Ni concentration in Figure 8g elucidates that “accumulated” leachates followed an irregular tendency, as did some other metal(loid)s. Therefore, the tendency between both types of samples is different.

Despite the differences obtained for the “accumulated” and “punctual” leachates, the “accumulated” leachates are lower than E5, and the “punctual” ones are comparable with E5. Furthermore, all the samples are below the inert level. Although Ni concentrations would increase, exceeding the inert limit (0.4 ppm), they would still probably achieve values below the non-hazardous limit (10 ppm). Figure 8h shows the Pb concentration results, and all the analyzed leachates are quite similar, although the “accumulated” samples are more irregular than the “punctual” ones and the concentrations are below that of E5 in the percolation column test. Besides, the “accumulated” and “punctual” samples’ results are below the non-hazardous limit (10 ppm). Finally, in Figure 8i is presented the Zn concentration values for the “accumulated” and “punctual” samples, which are comparable. In Figure 8 the R^2 values are shown, some of which are low. These are the cases of “punctual samples for Cu” and “accumulated tank for Pb”. In the case of Cu, the main reason is because there is a very low concentration in the first 100 days of percolation. In the case of the Pb, the dispersion of values between 300 and 400 days is high. Therefore, it is difficult to fit a trend line for both cases, and the R^2 value becomes small. The R^2 in the rest of the scenarios is around 0.9. The results obtained in the experimental section and in E5 for the percolation column tests are also similar. It can be noticed that all the results obtained are far below the non-hazardous limit (50 ppm). In conclusion, a general observation is that after almost two years, the GM leaching is stabilized for most of the metal(loid)s evaluated, and all the concentrations obtained are lower than the inert limit specified in the European Landfills Directive. A such, the cement-based treatment succeeded in the formulation of hybrid cements with APC. This achievement can be considered in the context of both economic and ease factors, in terms of GM development

4. Conclusions

This paper presents the results obtained from an experimental section simulating a road subbase layer, using a developed granular material (GM) formulated with 80 wt. % of WBA and 20 wt. % of crushed M (50 wt. % OPC CEM I and 50 wt. % APC). Dynamic leaching tests, through ICP-MS, were assessed for the WBA, M and crushed GM samples. The results showed that the main metal(loid)s concentrations were below the limits established for classification as non-hazardous waste. Considering the crushed GM, the results were excellent, as total Cr, Cu, Mo, Ni and Zn were between the inert and the non-hazardous limits. On the other hand, percolation column tests were also performed for the WBA, M and crushed GM samples. Considering the crushed GM, the results indicated that, in the seventh eluate (with L/S = 10, emulating 60 years), all the metal(loid)s analyzed were below the inert threshold. Hence, the GM developed succeeded in stabilizing the APC hazardous waste.

Lastly, an experimental section of 40 m³, utilizing the crushed GM formulated in this study, was constructed and evaluated over almost two years. The most important outcome from this study was that the elements analyzed from the 27 rainwater leachates (“accumulated” and “punctual”) collected after rainy days were much below the inert limit established by the European Landfills Directive.

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