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## **Environmental potential assessment of MSWI bottom ash-based alkali-activated binders**

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25 **Abstract**

26 Alkali-activated binders (AABs) stand out as a sustainable alternative to ordinary Portland  
27 cement (OPC) as they can be formulated using by-products or waste as raw materials. However,  
28 the presence of hazardous compounds in residues can lead to an increase in AABs' toxicity due  
29 to the highly alkaline media. Therefore, it is extremely important to evaluate their  
30 environmental risks to validate their use as building materials. This study environmentally  
31 assessed AABs prepared with two different fractions (0-30-mm and 8-30-mm) of weathered  
32 bottom ash (AA-WBA) from WtE plants. The potential leachate toxicity of AA-WBA was  
33 assessed using granular and monolithic leaching tests that simulated end-of-life and service life  
34 scenarios, respectively. Furthermore, an acute toxicity test with crustacean *Daphnia magna* as  
35 model organisms was conducted to determine the relationship between the leachate metal(loid)  
36 concentrations and the ecotoxicity of AA-WBA. The results showed higher metal(loid)  
37 concentrations in AA-WBA specimens prepared with the 0-30-mm fraction of WBA. The  
38 service life scenario revealed multiple metal(loid)-release mechanisms. The 48-hour EC<sub>50</sub> value  
39 (close to 10%; moderate toxicity) indicated that the use of the coarse fraction of WBA increased  
40 the immobilisation of the metal(loid)s. Finally, the correlation between the concentrations of  
41 some of the metal(loid)s and toxicity was demonstrated.

42 **Keywords:** Alkali-activated binder; Weathered bottom ash; Heavy metal immobilisation;

43 **Ecotoxicity; Daphnia Magna.**

## 44 **1. Introduction**

45 Concerns about municipal solid waste (MSW) management are increasing every year due  
46 to the huge amount of residue generated worldwide [1]. MSW incineration (MSWI) can  
47 mitigate the MSW management issues and also provide some by-products that can be used to  
48 develop new sustainable materials. Many EU countries see MSWI in waste-to-energy (WtE)  
49 plants as an opportunity to recover energy. In 2016, there were 512 WtE plants in the EU with  
50 an incineration capacity of 93 Mt [1]. Incineration reduces the mass (70%) and volume (90%)  
51 of the MSW [2], which can be important in countries with a reduced landfill area. Furthermore,  
52 around 20% of the incinerated MSW becomes a by-product known as incinerator bottom ash  
53 (IBA). IBA is mainly composed of a mineral fraction (85%) and also contains ferrous metals  
54 (10-12%) and non-ferrous metals (2-5%) [3]. It can be valorised as secondary aggregates after  
55 metal recovery and weathering. The resulting material is known as weathered bottom ash  
56 (WBA), which is mainly composed of heterogeneous mineral fractions [4]. However, some  
57 legal, chemical, and technological barriers hamper the valorisation of WBA in many countries,  
58 which leads to its landfilling [5,6]. For this reason, the scientific community is studying the  
59 potential valorisation applications of WBA to increase its added value. One of these potential  
60 applications is the use of WBA as a precursor material in the formulation of alkali-activated  
61 binders (AABs) [7–9].

62 AABs are one of the sustainable alternatives to ordinary Portland cement (OPC), which  
63 remains as the most widely used material in the building and civil engineering field. It is  
64 important to highlight that the massive production of OPC is the cause of the release of around  
65 5 - 8% of the anthropogenic worldwide emissions [10,11] and 2% of the global primary energy  
66 consumption [12]. AABs cements are produced through an alkaline activation process that  
67 involves the reaction between an aluminosilicate-rich powdered precursor and an alkaline  
68 activator solution. Apart from WBA, other aluminosilicate-based by-products or wastes, such  
69 as blast furnace slag (BFS), red mud (RM) or fly ash (FA) from thermal power plants, can be

70 used as precursors in the formulation of AABs [13]. In this regard, the formulation of AABs  
71 has a low carbon footprint and low energy consumption, and also preserves raw materials and  
72 significantly reduces landfilling [14,15], demonstrating that these materials can be sustainable  
73 alternatives to OPC [16]. However, some precursors can potentially increase the toxicity of the  
74 resulting binders due to their complex and dangerous compositions, as well as the aggressive  
75 conditions produced by the highly alkaline media of the alkaline activator solution. Moreover,  
76 toxic elements such as trace metal(loid)s can be released into the environment [17].

77 Both silica-rich composition and substantial calcium and aluminium content [9,18] turn the  
78 WBA into a potential precursor in alkali activation technology, as has been demonstrated in the  
79 literature [19–22]. However, the environmental assessment to validate the commercial use of  
80 alkali-activated WBA (AA-WBA) binders as a building material has been scarcely performed.  
81 Only the potential release of metal(loid)s in deionised water according to granular leaching test  
82 (EN 12457-2) has been studied by the authors in previous works [19,23]. Most of these heavy  
83 metal(loid)s (Cr, Cu, Hg, Mo, Pb, and Zn) remain below the limit established by the EU  
84 legislation for its acceptance at landfills as non-hazardous waste [24]. However, the  
85 concentration of other metal(loid)s such as As and Sb exceeded this limit, leading to the  
86 classification of AA-WBA binders as hazardous waste. Therefore, considering that heavy  
87 metal(loid)s release is the main concern regarding the applications of WBA [25], it is necessary  
88 to thoroughly study AA-WBA binders' toxicity to determine their environmental impact.

89 In this sense, the EU Regulation 305/2011 established harmonised conditions for the  
90 marketing of construction products. The “*Basic requirements for construction works*” are listed  
91 in Annex I, where the environmental analysis of construction products is proposed by assessing  
92 the hazardous substances' release in aqueous systems and soils [26]. This environmental  
93 analysis can be performed through assessment methods such as horizontal harmonised leaching  
94 tests, which have been standardised by the European Committee for Standardisation (CEN).

95 However, according to Bandow et al. (2018), although these leaching tests can be used to  
96 determine the leaching potential of certain compounds, they, unfortunately, do not reproduce  
97 real scenarios due to the complexity of some construction products and some experimental  
98 limitations that affect the experimental conditions [27]. For this reason, the CEN/TC 351  
99 published a technical report (CEN/TR 17105) to develop modular horizontal standardised  
100 ecotoxicity tests for construction products [28]. These tests allow assessing any potential  
101 environmental damage through bioassays. Concretely, when the composition of the leachates  
102 or their interaction with the environment is unknown.

103 Environmental risk assessments of alkali-activated materials (AAMs) are a relatively  
104 unexplored area at the international level. The ecotoxicity of AABs formulated with by-  
105 products and waste as precursors has been scarcely evaluated. The inclusion of this type of  
106 bioassays in the research of AABs is extremely important to validate their use as building  
107 materials, as well as to determine their hazardousness. Only some studies have evaluated the  
108 ecotoxicity of mortars and concrete formulated with by-products and residues which partially  
109 substitute the OPC [29–32]. These assays have confirmed the suitability of materials in which  
110 OPC has been partially replaced with some industrial wastes. However, the highly alkaline  
111 medium of the leachates has a lethal impact on most of the tested organisms, while lowering of  
112 the pH lead to a reduction of the potential toxicity.

113 This study aimed to assess the environmental potential of the AABs produced through the  
114 alkaline activation of WBA (two different particle size fractions) and PAVAL<sup>®</sup> (PV), which is  
115 an aluminium oxide by-product generated in the aluminium recycling process. The novelty  
116 mainly lies in the application of bioassay in these novel cement-based materials to evaluate  
117 their environmental risks. The main goal of this work was based on determining the relationship  
118 between the AA-WBA binders' leachates and their ecotoxicity, as well as the leachate  
119 metal(loid)s depending on the precursors used. The environmental evaluation was carried out

120 through different tests: (i) a compliance leaching test for granular materials (EN 12457-2) to  
121 simulate the material end-of-life scenario, (ii) a horizontal dynamic surface leaching test for  
122 monolithic materials (CEN/TS 16637-2) to simulate the material service life scenario, and (iii)  
123 an acute toxicity test using *Daphnia magna* (*D. magna*) mobility inhibition test [33] to  
124 determine the global toxicity of AA-WBA binders. In addition, the results were compared with  
125 those for two reference materials: an alkali-activated metakaolin (AA-MK) binder and OPC  
126 binder.

## 127 **2. Materials and methods**

### 128 2.1. Materials

129 The IBA was produced by a Waste-to-Energy (WtE) plant located in Tarragona (Spain),  
130 which incinerates around 380 t·day<sup>-1</sup> of MSW. After recovering ferrous and non-ferrous metals  
131 in a treatment process and removing lightweight unburned materials, around 88 t·day<sup>-1</sup> of fresh  
132 IBA is obtained at this WtE plant. The fresh IBA is then stockpiled outdoors for 2-3 months to  
133 stabilise the metal(loid)s and WBA. The collected WBA (50 kg) was quartered to obtain a  
134 representative sub-sample of 10 kg and, which was then dried on a stove at 105°C for 24 h. The  
135 dried sample was sieved to determine the particle size distribution (PSD), as shown in Fig. S1  
136 (Supplementary Material). Afterwards, a metal magnet (Nd; 0.485 T) was passed over the  
137 sample to remove magnetic particles. Finally, the WBA was crushed and milled until obtaining  
138 a powder with a particle size below 80 µm. The formulation of the AA-WBA binders was  
139 carried out by using two samples of WBA with different particle size fractions (see section  
140 2.2.): (i) the 0-30-mm (WBA0/30) fraction, which is the total WBA fraction, and (ii) the 8-30-  
141 mm (WBA8/30) fraction, which is around 30 wt.% of total WBA. The chemical composition  
142 was analysed by X-ray fluorescence (XRF) with a Panalytical Philips PW 2400 sequential X-  
143 ray spectrophotometer (Table 1). X-ray diffraction (XRD) was conducted with a Bragg-  
144 Brentano Siemens D-500 powder diffractometer equipment with CuK $\alpha$  radiation (Fig. S2).

145 Both XRF and XRD results showed that both fractions contain silica and alumina-rich mineral  
146 phases. The high SiO<sub>2</sub> content is mainly due to the presence of primary and secondary glass,  
147 whose weight percentage increased with the particle size fraction. The CaO and Al<sub>2</sub>O<sub>3</sub> contents  
148 originated mainly from the cementitious materials containing OPC, as well as fired ceramics  
149 and metallic aluminium [18]. In previous studies, the most abundant trace elements (mainly  
150 metal(loid)s) were more concentrated in the fine fractions [18,34]. Consequently, the content  
151 of metal(loid)s in was greater in the WBA0/30 than in the WBA8/30 fraction, mainly those of  
152 Cu, Zn and Pb (Table 1). Previous studies have reported the SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> availability from  
153 each WBA fractions regardless of their content [34].

154 The WBA8/30 fraction had a substantial lack of aluminium (Table 1) due to the high amount  
155 of non-ferrous metals recovered by an Eddy current device in the fractions above 6 mm [18]. It  
156 is important to highlight the role of aluminium in alkaline activation since it affects the nature  
157 of the reaction products and the strength development of AABs [35,36]. For this reason, an  
158 aluminium oxide by-product named PAVAL<sup>®</sup> (PV) was used in the formulation of AA-WBA  
159 binders as a source of aluminium [37]. This by-product is generated during the recovery of  
160 metallic aluminium from the salt slags in the secondary aluminium refining process. PV was  
161 provided by Befesa Company and collected from its aluminium recycling plant in Valladolid  
162 (Spain). After homogenisation, the PV sample was dried overnight at 105°C to avoid possible  
163 moisture variations in the sample received. The dry sample was then milled and sieved (< 80  
164 µm) to adapt PV powder for its use as a precursor in the AA-WBA formulations. The XRF  
165 analysis of PV (Table 2) showed that it predominantly contained Al<sub>2</sub>O<sub>3</sub>. These results agreed  
166 with the XRD pattern of PV (Fig. S3), revealing the presence of a greater number of aluminium  
167 crystalline phases, with corundum (Al<sub>2</sub>O<sub>3</sub>) being the main mineral phase.

168 AA-MK binders were formulated using commercial metakaolin (MK) powder to compare  
169 to AA-WBA binders. There is a wide range of references and several studies carried out

170 formulating AABs using MK as a precursor [38–40]. The chemical composition of MK powder,  
171 supplied by Bal-Co (Italy), is shown in Table S1. Ordinary Portland cement (OPC) paste was  
172 also used as reference material in the ecotoxicological study along with the AA-MK binders.  
173 In this case, CEM I 52.5 R supplied by Cementos Molins (Spain) was used (Table S1) as raw  
174 material.

175 A mixture of sodium silicate ( $\text{Na}_2\text{SiO}_3$ ) and sodium hydroxide (NaOH) was used as the  
176 alkaline activator solution. The  $\text{Na}_2\text{SiO}_3$  solution with an  $\text{SiO}_2/\text{Na}_2\text{O}$  molar ratio of 3.22  
177 (26.44% of  $\text{SiO}_2$  and 8.21% of  $\text{Na}_2\text{O}$ ;  $\rho = 1.37 \text{ g}\cdot\text{cm}^{-3}$ ) was supplied by Scharlab, S. L. NaOH  
178 solutions (4 M ( $\rho = 1.16 \text{ g}\cdot\text{cm}^{-3}$ ), 6 M ( $\rho = 1.20 \text{ g}\cdot\text{cm}^{-3}$ ), and 8 M ( $\rho = 1.24 \text{ g}\cdot\text{cm}^{-3}$ )) were  
179 prepared by dissolving NaOH pearls (Labbox Labware S.L.; purity > 98%) in deionised water.

## 180 2.2. OPC and alkali-activated binders (AABs) preparation

181 In earlier studies [19,23,41], the AA-WBA binders have been exhaustively characterised  
182 from a physicochemical and mechanical point of view. One formulation of each study was  
183 selected as optimal to formulate again for this environmental assessment. The OPC, AA-MK,  
184 and AA-WBA (AA-WBA0/30, AA-WBA8/30, and AA-WBA/PV) binders were formulated  
185 using the mix proportions shown in Table 3, which specifies the precursors (referred to solid,  
186 S), water or alkaline activator solutions (referred to liquid, L) ratios, and the liquid/solid ratio  
187 (L/S). The preparation of the pastes was started by mechanically stirring the  $\text{Na}_2\text{SiO}_3$  and NaOH  
188 solution in a plastic beaker (except for OPC). The precursors were then gradually added into  
189 the alkaline activator solution for 2 minutes at 500 rpm to favour the dissolution of reactive  
190 phases in the alkaline media. Afterwards, the mixture was mixed for 3 minutes at 750 rpm. The  
191 pastes were poured into 40-mm cubic moulds and vibrated for 5 min. The moulds were then  
192 sealed in plastic bags for 3 days in a climate chamber at  $25^\circ\text{C} \pm 1^\circ\text{C}$  and relative humidity of  
193  $95\% \pm 5\%$ . Finally, the specimens were demoulded after 3 days and kept in a climate chamber



194 under the same conditions until testing (28 days). Nine cubic shape specimens were prepared  
195 for each formulation.

### 196 2.3. Environmental assessment methods

197 Figure 1 shows a diagram flow where the methodology that has been carried out for the  
198 environmental assessment of AABs is specified schematically. It can be observed both  
199 process conditions and testing methods, as well as the main goal for each test.

#### 200 2.3.1. Granular leaching test (EN 12457-2)

201 Leaching tests of the powdered raw materials (OPC, MK, WBA0/30, WBA8/30, and PV)  
202 and AABs were carried out according to a standardised leaching test for granular materials (EN  
203 12457-2). The analysis of the leaching concentration of powdered raw materials was conducted  
204 using deionised water to determine their initial hazardousness. The leaching behaviour of AABs  
205 was evaluated by testing its crushed fragments to represent its end-of-life scenario after a  
206 potential demolition. It was performed in duplicate using two different extraction solutions: (i)  
207 deionised water, according to the standard procedure EN 12457-2, and (ii) freshwater prepared  
208 with different salts concentrations (Table 4), which is standardised in the OECD TG 202 [33].  
209 The deionised water leachates were used as an indicator to estimate the mobility of heavy  
210 metal(loid)s, allows classifying the examined materials. The freshwater leachates were later  
211 used for the ecotoxicity bioassay. The leaching test consisted of applying continuous rotating  
212 agitation ( $10 \text{ min}^{-1}$ ) to the crushed specimen (particle size  $< 4 \text{ mm}$ ) that was in contact with the  
213 extraction solution at an L/S ratio of  $10 \text{ L}\cdot\text{kg}^{-1}$  for 24 h at room temperature. The resulting  
214 leachates were filtered with a  $0.45\text{-}\mu\text{m}$  nitrocellulose membrane. Two replicas per raw material  
215 and AABs formulation were conducted. One aliquot per replica was extracted for further  
216 analysis by ICP-MS (Inductively coupled plasma mass spectrometry) technique with a  
217 PerkinElmer ELAN device, evaluating As, Ba, Cd, Cr, Cu, Hg, Mo, Ni, Pb, Sb, Se, V, and Zn  
218 concentrations.

219 2.3.2. Monolithic tank leaching test (CEN/TS 16637-2)

220 The semi-dynamic leaching test CEN/TS 16637-2 using monolithic specimens was used to  
221 simulate the service life scenario of AABs. Following the standard test procedure, a cured  
222 specimen (28 days) with a defined geometry (40-mm cubic) was immersed in deionised water  
223 at room temperature. In this scenario, only the interaction of the surface of the monolith with  
224 the solvent leads to the leaching. The water volume to surface area (L/A) ratio was  $80 \pm 10$   
225  $\text{L}\cdot\text{m}^{-2}$  ( $\approx 850$  mL). The top, bottom, and lateral surfaces of the tested specimen were  
226 covered/submerged by at least 20 mm. Leaching solutions were exchanged with deionised  
227 water at predetermined cumulative time intervals of 0.25, 1, 2.25, 4, 9, 16, 36, and 64 days to  
228 quantify the long-term diffusive leaching from the binder materials. The eluate was sampled  
229 and changed, with the pH and conductivity determined for each interval. Two replicas per  
230 formulation were conducted. Both filtration procedure and the trace elements analysed in the  
231 eluates were the same of the granular leaching test. The concentrations of metals and metalloids  
232 were determined for each interval, and the cumulative release was calculated following the Eqs.  
233 (1) and (2).

234 
$$r_i = \frac{c_i x V}{A} \quad (1)$$

235 
$$R_n = \sum_{i=1}^n r_i \text{ for } n = 1 \text{ to } 8 \quad (2)$$

236 where

237  $c_i$  is the concentration of the substance in eluate  $i$ , in  $\mu\text{g}\cdot\text{L}^{-1}$ ;  $r_i$  is the area release of the  
238 substance in fraction  $i$ , in  $\text{mg}\cdot\text{m}^{-2}$ ;  $R_n$  is the cumulative area release of the substance for period  
239  $n$  including fraction  $i=1$  to  $n$ , in  $\text{mg}\cdot\text{m}^{-2}$ ;  $A$  is the area of the specimen, in  $\text{m}^2$ , and  $V$  is the volume  
240 of the leachate, in L.

241 The leaching mechanisms were also investigated to determine the leaching behaviour of  
242 metal(loid)s, following the procedures, calculations, and requirements described in the CEN/TS  
243 16637-2 standard. In addition, the cumulative metal(loid)s release ( $\text{mg}\cdot\text{m}^{-2}$ ) in AABs was

244 compared with leaching limits by the Dutch tank leaching test (NEN 7345). This regulation  
245 classifies the building materials in two categories: (i) materials without any environmental  
246 restriction, which are those that do not exceed the  $U_1$  limit (ii) materials with restriction use,  
247 which are those that exceed the  $U_2$  limit. Although materials whose any cumulative metal(loid)  
248 release is comprised  $U_1$  and  $U_2$  limits do not have any environmental restriction, at the end of  
249 their life cycle should remove the pollutant that exceeds the threshold (dismantling) [42].

250 Finally, it is important to highlight that the feasibility of this horizontal harmonised test was  
251 validated by different European laboratories [43]. However, it is also required more realistic  
252 scenarios to determine environmental risks. For this reason, apart from leachate metal(loid)s  
253 evaluation, a bioassay test was performed to determine a global parameter of toxicity which  
254 validates the use of AABs as construction material.

#### 255 2.4. *D. magna* acute toxicity test

256 The *D. magna* mobility inhibition bioassay (ISO 6341) is used to evaluate the ecotoxicity  
257 of binder materials formulated with (or containing) secondary resources in freshwater aquatic  
258 ecosystems [29–32]. This test has been chosen because it allows assessing the global toxicity  
259 of the formulated AABs through the eluates obtained in the granular leaching test. Besides, it  
260 is important to highlight that *D. magna* is highly sensitive to a wide range of chemicals and is  
261 relatively easy to culture and maintain in the laboratory.

262 Seven different dilutions were prepared with each of the binders' eluates (EN 12457-2 using  
263 standard freshwater) prior to the toxicity test. The test solutions obtained were the following  
264 (volume of eluate:volume of standard freshwater): 0:1 (0%, the negative control group), 1:16  
265 (6.25%), 1:8 (12.5%), 1:4 (25%), 1:2 (50%), 3:4 (75%), and 1:0 (100%). The negative control  
266 group was used to ensure the acceptability of the ecotoxicity test (> 90% survival and normal  
267 mobility). Eluates obtained from the EN 12457-2 leaching test represented the worst-case  
268 scenario, involving a small particle size and dynamic leaching conditions that favoured leaching

269 and increased the potential release of metal(loid)s from the binders. Consequently, this can  
270 result in higher leaching and ecotoxicity compared to normal service conditions.

271 The OECD TG 202 “*Daphnia* sp. Acute Immobilisation Test” standard [33] was followed  
272 to assess putative aquatic toxicity and obtain the EC<sub>50</sub> value (the concentration that is estimated  
273 to immobilise 50% of the daphnids within a stated exposure period). This acute toxicity test  
274 assesses the inhibition of the mobility of *D. magna* after 24 and 48 hours of exposure to the  
275 eluate being tested. A total of 600 *D. magna* neonates (< 24 hours in age) were kept at 20 ± 2°C  
276 and under a light/dark cycle of 16/8 hours prior to the assay. Three hours after spirulina feeding,  
277 groups of 5 neonates were randomly exposed to 10 mL of each test solution for 48 hours under  
278 darkness. Immobilisation measurements (when daphnids were not able to swim within 15  
279 seconds) were recorded at 24 and 48 hours and compared with the values obtained in the  
280 negative control group. For each test solution, four replicates were tested. The results were  
281 analysed to calculate the EC<sub>50</sub> of each test item at 24 and 48 hours. Temperature (20 ± 2°C),  
282 light/dark cycles (16/8 hours), pH (6.9-7.5) and the oxygen concentration (8.3-8.8 mg·L<sup>-1</sup>) were  
283 controlled, while any adverse events and/or abnormal daphnid behaviour were monitored  
284 during the experimental period.

#### 285 2.5. Statistical analysis

286 Percentage of immobilization by treatment group and material was analysed by using the  
287 EPA probit analysis program version 1.5, which allowed the calculation of EC<sub>50</sub> values as well  
288 as the 95% confidence limits of the slope, intercept of the probit-concentration curve and any  
289 EC value from 1 to 99%.

290 Spearman's rank correlation was used to determine significant relationships ( $\alpha = 0.05$ )  
291 between the presence of trace elements in the eluate and changes in the mobility response of *D.*  
292 *magna*. Statistical analysis was performed with SPSS Statistics 24 for Mac.

### 293 3. Results and Discussion

### 294 3.1. Granular leaching test (EN 12457-2)

#### 295 3.1.1. Raw materials hazardousness

296 The initial hazardousness of powdered raw materials was determined using the EN 12457-  
297 2 procedure. Table 5 summarises the metal(loid) leaching concentrations and the limits  
298 established by EU for the acceptance of waste at landfills [24]. Some differences can be found  
299 in the release of metal(loid)s between the two fractions of the WBA assessed (WBA0/30 and  
300 WBA8/30 fractions). The metal(loid) leaching concentration was higher in the WBA0/30  
301 fraction in agreement with previous studies [44–46]. This fraction contains a large amount of  
302 particles size below 4 mm (Fig. S1), in which mainly the metal(loid)s and soluble salts are  
303 accumulated [18,45]. These metal(loid)s could be released during WBA0/30 fraction milling  
304 and re-dissolved at this pH (11.33). Concerning the pH of WBA samples, it can be observed a  
305 substantial decrease in the WBA8/30 compared to the WBA0/30 fraction. This is due to the  
306 initial content of portlandite is much higher in the fine fractions, and their pH is controlled by  
307 the ettringite formed during the weathering process of IBA. By contrast, as the initial content  
308 of lime is lower in the coarse fractions, it is quickly carbonated, lowering the pH controlled by  
309 the calcite [46]. Both samples were below the threshold established by the landfill legislation  
310 to classify wastes as non-hazardous materials. Moreover, it is remarkable that the  
311 concentrations of most of the elements were found below the limits for inert materials. Only  
312 the leaching concentration of Cu, Mo, Sb, and Se in the WBA0/30 fraction, and the leaching  
313 concentration of Sb in the WBA8/30 fraction, exceeded the limits for inert materials.

314 Leachates from PV had Sb concentration that was above the established non-hazardous  
315 limit. The presence of antimony is due to its use in some aluminium-bearing alloys, which may  
316 contain up to 4 to 6 wt.% of Sb [47]. This metalloid is probably removed during the refining  
317 secondary aluminium process and concentrated into the saline slags. The leaching

318 concentrations of the remaining metal(loid)s were below the threshold of inert waste or non-  
319 hazardous waste.

320 Among the reference materials OPC and MK, only the concentrations of Ba, Cr, and Mo in  
321 the leachates from OPC stood out. Barium is added to raw meal in carbonate or sulphate form  
322 before the clinkerisation process in OPC manufacturing [48]. Chromium is primarily  
323 incorporated in the silicate phases during the clinker burning process [49]. Its stabilisation in  
324 OPC has been extensively studied [50–52]. Finally, molybdenum is incorporated into the  
325 procurement and milling processes of the clinker [49].

326 On the basis of the results obtained in the raw materials, it is proposed how the highly  
327 alkaline medium of the activator can affect the leaching conditions of AA-WBA binders. The  
328 different and complex composition of the precursors could lead to potential increases in  
329 ecotoxicity due to the release of some toxic elements that would otherwise be stabilised. In this  
330 regard, we investigated whether the synthesis of AA-WBA binders increased the leaching of  
331 the metal(loid)s or if the formation of a cementitious matrix encapsulated these elements,  
332 thereby preventing their leaching.

### 333 3.1.2. AA-WBA binders (End-of-life scenario)

334 Table 6 shows the concentrations of the metal(loid)s and the pH of the leachates from AA-  
335 WBA binders determined with the EN 12457-2 standard procedure. The leachate results of OPC  
336 and AA-MK specimens were used as references. This test was conducted in the AA-WBA  
337 binders to simulate the leaching behaviour at the end of life, after a potential demolition. The  
338 metal(loid) concentrations of the leachates from the AA-WBA binders (deionised water) were  
339 below the limits set for classification as non-hazardous wastes, except for those of As (for AA-  
340 WBA0/30) and Sb (for all AA-WBA binders). These two metalloids showed a considerable  
341 increase in their concentration in the leachates as a result of the strong alkaline reaction of the  
342 activators. The rest of the metal(loid)s only showed a slight increase in concentration due to the

343 alkaline reaction and the pH of the aqueous leaching solution. As and Sb originated mainly  
344 from the high content of glass cullet in the MSW [18]. Their oxides are used as clarifying or  
345 (de)colouring agents in glass manufacturing [53]. Moreover,  $\text{Sb}_2\text{O}_3$  is used as a pigment in dyes  
346 and paints as well as in the textile industry [54]. Several studies link the release of both As and  
347 Sb to calcium-bearing minerals [55–58] and Fe, Al, Mn-(hydr)oxides [59], which are  
348 neoformed during the natural weathering process of IBA. When the pH increases in the WBA  
349 as a result of the addition of a strongly alkaline solution (i.e.,  $\text{pH} > 12.5$ ), calcium precipitates  
350 as portlandite and the metalloids are released into the solution. In addition, the neoformed  
351 secondary glass during the combustion of MSW can encapsulate metal(loid)s in its structure,  
352 preventing their release [18]. This amorphous structure, however, can dissolve in a strongly  
353 alkaline medium, releasing some of these metals.

354 As expected, most of the metal(loid)s showed a higher concentration in the leachates  
355 obtained from the AA-WBA0/30 binder due to the high content of fine fractions in the  
356 WBA0/30 fraction (Fig. S1) [45]. The leachates from the AA-WBA/PV specimens had the  
357 lowest concentrations of metal(loid)s among those that had been formulated with only WBA  
358 (WBA0/30 and WBA8/30 fractions). This fact is probably due to the use of PV as a precursor,  
359 which increased the formation of N-A-S-H and C-A-S-H gels and the number of cementitious  
360 phases [60]. Thus, the increase in the encapsulating effect of the binder decreased the release  
361 of metals into the leaching solutions [61].

362 The concentration of metal(loid)s in the leachates obtained from the two reference binders  
363 (OPC and AA-MK) was generally lower than those from the AA-WBA binders. On one hand,  
364 the leachates from OPC had substantial concentrations of Ba, Cr, and Mo, coming from the raw  
365 materials and/or the clinker milling process [48,49,52], as mentioned previously. On the other  
366 hand, the leachates from the AA-MK specimens showed high concentrations of As, which was  
367 above the limits established for the classification as non-hazardous waste ( $> 2 \text{ mg/kg}^{-1}$ ). The

368 high leaching concentration of As is probably due to the desorption of arsenate from the natural  
369 kaolinite [62].

370 The metal(loid) concentrations in the leaching test conducted using standard freshwater  
371 presented substantial differences to that obtained using deionised water (Table 6). These  
372 differences can be mainly attributed to the decreasing in binders' pH due to the standard  
373 composition of the freshwater (Table 4), as well as the difference in the solubility of  
374 metal(loid)s depending on the pH of the medium [58]. This decrease in pH led to a decrease in  
375 the concentrations of the trace elements with high oxidation states that can form oxyanions (As,  
376 Cr, Mo, Sb, and Se) and an increase in the concentrations of divalent pH-dependent metals (Ni,  
377 Cu, and Zn). As a result of the decrease in some metal(loid)s concentration, the leachates of  
378 AA-WBA binders formulated with WBA8/30 (AA-WBA8/30 and AA-WBA/PV) were below  
379 the limits for its classification as inert waste, excepting for Sb, which exceeded the threshold  
380 established for non-hazardous waste ( $> 0.7 \text{ mg}\cdot\text{kg}^{-1}$ ).

### 381 3.2. Monolithic leaching test (Service life scenario)

382 The generic horizontal dynamic surface leaching test (DSLTL) was conducted mainly to  
383 determine the surface-dependent release of metal(loid)s from monolithic specimens of AABs,  
384 as well as to identify the mechanisms controlling the leaching processes. It was also performed  
385 to simulate the service life scenario of AABs and compare their leachability. The DSLTL of the  
386 AA-MK was also included as a reference. Table S2 (Supplementary material) shows the  
387 concentrations of the metal(loid)s and the pH of the leachates from AA-MK and AA-WBA  
388 binders, using the CEN/TS 16637-2 test procedure. Overall, the results from the CEN/TS  
389 16637-2 leaching test agreed to those from the EN 12457-2 leaching test. The metal(loid)s with  
390 the highest leaching concentrations in the granular test also showed the highest surface-  
391 dependent release in the leaching test using monolithic samples.



392 Figures 2 to 4 show the cumulative release of metal(loid)s from the AA-MK and AA-WBA  
393 binders. Figure 2 groups the elements (As, Se, and V) that had a larger cumulative release from  
394 the AA-MK leachate compared to those from AA-WBA specimens. These metal(loid)s  
395 probably come from natural kaolinitic soils [62–65]. Moreover, Se and V can also be released  
396 in soils via anthropogenic activities such as fuel combustion [66–68], which is necessary for  
397 the thermal dehydroxylation process of kaolinite. Although our results for Se were similar to  
398 those of other studies [69], the cumulative release of As and V was much higher, probably due  
399 to the initial content of these metals(loid)s in MK.

400 Figure 3 shows the cumulative release of the metal(loid)s (Cu, Mo, Ni, Sb, and Zn) that  
401 presented a higher leaching concentration for the AA-WBA0/30 specimen compared to the AA-  
402 WBA formulated with WBA8/30 (AA-WBA8/30 and AA-WBA/PV). As expected, the use of  
403 WBA0/30 fraction led to a greater release since their finer fractions have a higher content of  
404 metal(loid)s and soluble salts [45]. It should be noted that the difference in the cumulative  
405 release of these metals at 64 days ( $R_8$ ) was much lower when the WBA8/30 fraction was used.  
406 In the specific cases of Cu and Zn, these differences were 4 and 5 times lower, respectively. In  
407 all the cases, the metal(loid) leaching concentrations were similar or slightly lower for the AA-  
408 WBA/PV specimen compared to the AA-WBA8/30 specimen. Additionally, Figure 3 depicts  
409 the cumulative release of the metals (Ba, Cr, and Pb) that had higher leaching concentrations  
410 for AA-WBA8/30 than for AA-WBA0/30. This is probably due to the glassy and ceramic nature  
411 of the WBA8/30 fraction, which contains a large amount of fired ceramics coming from small  
412 domestic works, as well as primary and secondary glass coming from beverage containers [18].  
413 Barium carbonate and lead oxide are widely used as additives in ceramic glazes [70–72], while  
414 trivalent chromium is extensively used as a green colouring agent in glass manufacturing [73].  
415 In addition, the WBA8/30 fraction also contains construction wastes such as OPC-cement based  
416 materials that can be generated in the household sector and municipal cleaning services. As

417 mentioned above, OPC has a high content of barium and chromium [49] that show long-term  
418 release [58,69].

419 The release of metal(loid)s from cementitious materials that are in contact with water can  
420 be controlled and affected by various physical and chemical retention mechanisms [58]. In this  
421 regard, the leaching mechanisms were determined using the data analysis procedure introduced  
422 in CEN/TS 16637-2 (see Fig. S4 in Supplementary Material). Table 7 summarises the release  
423 mechanisms identified for each metal(loid) with the DSLT using AA-MK binder as the  
424 reference. Some of the metals that leached from the AA-MK specimen showed a low  
425 concentration or surface wash-off followed by a low concentration (Ba, Cd, Mo, Ni, and Sb).  
426 These same metals had shown low leaching in the EN 12457-2 batch test. A surface release  
427 mechanism controlled by dissolution was identified for the rest of the metals (As, Cr, Cu, Pb,  
428 Se, and V), except for Zn, which showed diffusive control. By contrast, for the AA-WBA0/30  
429 specimen, the surface release of all the metal(loid)s under consideration was controlled by a  
430 dissolution mechanism, except for As and Cd, which showed a low concentration and an  
431 unidentified mechanism, respectively. In the AA-WBA8/30 and AA-WBA/PV specimens, the  
432 dissolution-controlled release was also the main mechanism for surface release (i.e., As, Cu,  
433 Mo, Ni, Sb, and V). The release mechanism determined for Ba, Pb, and Zn was a surface wash-  
434 off preceding a diffusion-controlled or unidentified mechanism (see Table 7). Once again, a  
435 concentration below the detection limits was determined for Cd in both samples. However,  
436 earlier studies determining the surface release mechanisms of metal(loid)s from cementitious  
437 matrix materials have reported no clear differences between diffusion-controlled and  
438 dissolution-controlled mechanisms [58,69,74].

439 Figure 5 shows the cumulative surface release ( $R_n$ ) of some of the metal(loid)s from AA-  
440 WBA binders vs. leaching time plotted on a log-log scale. According to van der Sloot et al.  
441 [58], the slope for leaching times controlled by a diffusive surface release mechanism should

442 be 0.5. As can be seen in Figure 5, the leaching from some of the AA-WBA specimens showed  
443 a different trend in the first aliquots compared to the last ones, with a significant change in the  
444 slope. Likewise, the slope of the line of best fit was close to 0.5, indicating a diffusion-  
445 controlled leaching process. This trend was more noticeable for the AA-WBA binders  
446 formulated with the WBA8/30 fraction (AA-WBA8/30 and AA-WBA/PV), corroborating a  
447 decrease in open porosity and an increase in the tortuosity of the binder matrix.

448 The cumulative metal(loid)s release (expressed in  $\text{mg}\cdot\text{m}^{-2}$ ) was compared to the limits set  
449 in NEN 7345 standard (Table 8), aiming to determine the feasibility of AABs from an  
450 environmental point of view. Although most metal(loid)s release are below  $U_1$  threshold, the  
451 results revealed that AA-WBA binders should be used with environmental restrictions. This is  
452 due to Sb exceeded the  $U_2$  limit. In the case of AA-MK, the results evidenced the possibility of  
453 using this binder without any environmental restriction. However, according to NEN 7345,  
454 some metal(loid)s (As, Se, and V) should be removed at the end of its life cycle. It is important  
455 to highlight that the most problematic metal(loid)s release (in granular and monolithic test) are  
456 those that can form oxyanions (As, Sb, and V) as reported elsewhere [75]. In this sense, it was  
457 demonstrated that longer curing periods increase the immobilisation efficiency of these  
458 metal(loid)s [76]. Finally, the authors want to emphasise that in this study they have been tested  
459 AABs instead of alkali-activated mortars or concretes. Thereby, the preparation of AABs with  
460 sand and/or gravel could lead to a dilution of raw materials and consequently decrease in  
461 leachate metal(loid)s concentration.

### 462 3.3. *D. magna* acute toxicity test

463 The standard mobility test with the freshwater crustacean *D. magna* is considered the most  
464 sensitive when analysing the ecotoxicity of building materials [29]. In this study, all binders  
465 (OPC paste, AA-MK, and AA-WBA binders) were assessed at concentrations ranging from  
466 6.25 to 100% of the eluates (EN 12457-2 using standard freshwater). The immobilisation of *D.*

467 *magna* individuals was recorded at 24 and 48 hours. The validity criteria were met since no  
468 control daphnids were immobilised after 48 hours of exposure and the daphnids from the  
469 negative control group did not show any abnormal behaviour or adverse events. The influence  
470 of the leachate pH was minimised by using standard freshwater that contained pH-buffering  
471 chemicals [77]. The number of *D. magna* individuals immobilised after 24 and 48 hours of  
472 exposure is summarised in Tables S3 to S5 and Figure S5. The addition of the leachates from  
473 all the specimens tested induced a sequential (at 24 and 48 h) and concentration-dependent  
474 immobilisation of the daphnids. The percentages of immobilised *D. magna* individuals in each  
475 eluate after 24 and 48 hours of exposure are plotted in Figure 5. The percentage of immobilised  
476 daphnids was used to determine the EC<sub>50</sub> by probit analysis, which is summarised in Table 8.  
477 Due to the distribution results, there was no option to estimate the 95% confidence limits by  
478 probit analysis for three of the binders (AA-MK, AA-WBA0/30 and AA-WBA/PV at 24 hours  
479 and AA-WBA0/30 at 48 hours), which were extrapolated. Lower EC<sub>50</sub> values indicate greater  
480 toxicity because the material is toxic even at low concentrations (high dilutions). Accordingly,  
481 the toxicity of the AA-WBA0/30 sample was higher than that of the rest of the binders studied,  
482 which showed similar toxicity (AA-MK  $\cong$  AA-WBA8/30  $\cong$  AA-WBA/PV). The toxicity of the  
483 formulated AABs was significantly higher than that determined for OPC. The EC<sub>50</sub> of OPC  
484 obtained in this study was lower than that reported previously [78], probably due to the greater  
485 release of Mo and Ba as a consequence of a more acidic pH. It should also be noted that the  
486 toxicity of the binders formulated with WBA8/30 fraction (AA-WBA8/30 and AA-WBA/PV)  
487 was similar to that of the AA-MK binder. Furthermore, the EC<sub>50</sub> results demonstrated that the  
488 addition of PV as a precursor had practically no effect on toxicity (AA-WBA/PV).

489 Comparison with the results from the EN 12457-2 leaching test (Table 6) indicated good  
490 correspondence between toxicity and the concentrations of trace elements in the eluates (see  
491 Fig. S6 in Supplementary Material). Spearman's correlation was used to link the concentration

492 of trace elements in the eluates with the immobilisation response of *D. magna* (Table 10). This  
493 identified significant correlations ( $p < 0.05$ ) between the presence of Cu and Sb in the eluates  
494 and the immobilisation of *D. magna* (i.e., EC<sub>50</sub> values). A previous study showed that Cu and  
495 Sb concentrations in elutriates from sewage sludge have a strong impact on the mobility of  
496 *Daphnia* [79]. Although As, Pb, and Zn concentrations in the leachates correlated with acute  
497 toxicity in *Daphnia*, these correlations were not statically significant ( $p = 0.188$ ,  $p = 0.054$  and  
498  $p = 0.104$  for As, Pb, and Zn, respectively). The toxicity of metals on *D. magna* has been  
499 reported previously by several studies [80–82]. However, other factors rather than just the  
500 released metals mentioned (e.g., chlorides, sulphates, sodium, etc.) may affect the biological  
501 responses of the crustacean [29].

502 Comparing the 48-h EC<sub>50</sub> values for the AABs obtained in this study (Table 9) with those  
503 reported by other studies using ecotoxicity tests with *D. magna* for WBA (EC<sub>50</sub> 0.5-17.0) [83],  
504 it can be concluded that the decrease in particle size (< 80 µm) and the alkaline activation did  
505 not significantly affect the toxicity of the binder materials. Nevertheless, taking into account  
506 the ecotoxicity limit values established in the French proposal of the Criterion and Evaluation  
507 Methods of Waste Ecotoxicity (CEMWE) document [84], the alkali-activated binders  
508 formulated with WBA (and MK) showed evidence of acute toxicity (48-h EC<sub>50</sub> < 10%). Since  
509 the EC<sub>50</sub> values for AA-WBA8/30 (8.5%) and AA-WBA/PV (7.8%) were less than 10% but  
510 considerably greater than 1%, it can be said that they showed moderate-low acute ecotoxicity.  
511 By contrast, the EC<sub>50</sub> for AA-WBA0/30 was closer to 1%, indicating moderate-high  
512 ecotoxicity.

#### 513 **4. Conclusions**

514 The results obtained in this study provide a clear picture of the environmental and  
515 ecotoxicological potential of AA-WBA binders. The synergistic combination of the leaching

516 tests (EN 12457-2 and CEN/TS 16637-2) and the acute toxicity test could become an interesting  
517 tool to determine the environmental and ecotoxicological risks of the binder materials.

518 Granular leaching test (EN 12457-2) was used to assess the initial hazardousness of  
519 powdered raw materials, as well as to simulate the leaching behaviour of AABs binders after  
520 their end-of-life. This test demonstrated the different hazardousness of raw materials, which  
521 could be classified as inert (OPC and MK), non-hazardous (WBA/0/30 and WBA8/30), and  
522 hazardous (PV) waste according to the EU landfill legislation. Moreover, the metal(loid)s  
523 concentrations in the leachates from AA-WBA binders revealed severe activation of As and Sb,  
524 both for deionised water and freshwater. These two metalloids are commonly used as additives  
525 during the manufacturing of container glass and were affected by alkaline activation, showing  
526 higher concentrations than in the raw materials leachates. It is also important to highlight the  
527 substantial decrease in some metal(loid)s concentration (As and Sb) when freshwater was used  
528 as a leaching solution. This is due to the standardised composition of freshwater led to the  
529 decreasing in pH of AA-WBA binders, which in turn contributed to a decrease in the  
530 concentrations of the trace elements with high oxidation states that can form oxyanions.

531 Monolithic leaching test (CEN/TS 16637-2) was used to simulate the leaching behaviour of  
532 AABs during their service life. The surface-dependent release mechanism in some metal(loid)s  
533 was unpredictable, except for those that showed low concentrations or a depletion mechanism.  
534 A mixed release mechanism, diffusion-dissolution, probably controlled the surface release of  
535 most of the metal(loid)s. In the AA-WBA binders, given that the pH was very similar in all the  
536 aliquots, the differences in the release mechanism determined for the same metal(loid) could be  
537 attributed to the use of different size fractions or the use of PV (i.e., Ba, Pb, and Zn). In this  
538 case, higher SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> availability led to a greater formation of reaction products, which  
539 decreased open porosity and increased the tortuosity of the cementitious matrix.

540 The present study demonstrated the suitability of the *D. magna* acute toxicity test for the  
541 ecotoxicological assessment of AA-WBA binders. To the authors' knowledge, there are no  
542 previous studies that have included an ecotoxicological analysis in the environmental  
543 assessment of AABs. In this study, only the eluates obtained from the EN 12457-2 leaching test  
544 (with freshwater) were selected for the ecotoxicological assessment. These eluates represented  
545 the worst-case scenario, involving a small particle size and leaching conditions that favoured  
546 leaching and increased the potential release of metal(loid)s from the binder materials.  
547 Consequently, this can result in much higher leaching and ecotoxicity compared to normal  
548 service conditions. The specimens formulated with WBA8/30 fraction showed less toxicity,  
549 presenting similar toxicity as AA-MK binder. The AA-WBA8/30 and AA-WBA/PV  
550 formulations showed moderate-low acute ecotoxicity, according to the ecotoxicity limit values  
551 established in the French proposal of the CEMWE document, with EC<sub>50</sub> values close to 10%.  
552 Hence, the potential ecotoxicity of materials studied could be ordered as follows: AA-  
553 WBA0/30 > AA-MK ≈ AA-WBA8/30 ≈ AA-WBA/PV >> OPC.

554 Therefore, the environmental and ecotoxicological behaviour of the AA-WBA binders  
555 formulated from the WBA8/30 fractions was significantly better than those formulated with the  
556 WBA0/30 fraction due to the higher content of metal(loid)s in the fine fractions. The increase  
557 in the availability of reactive aluminium by using PV as a precursor that partial substituted  
558 WBA (8-30 mm) also slightly decreased the mobility of the metal(loid)s, probably due to the  
559 greater formation of cementitious phases.

#### 560 **CRedit authorship contribution statement**

561 **A. Maldonado-Alameda:** Investigation, Writing – review & editing; **J. Giro-Paloma:**  
562 Validation, Writing – review & editing; **A. Rodríguez-Romero,** Validation, Writing – review  
563 & editing; **J. Serret:** Investigation; **A. Menargues:** Validation, Writing – review & editing; **A.**

564 **Andrés:** Conceptualization; Writing – review & editing; **J.M. Chimenos** Conceptualization,  
565 Supervision, Writing - Original Draft, Funding acquisition.

### 566 **Declaration of competing interest**

567 The authors declare that they have no known competing financial interests or personal  
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**Table 1.** Major, minor, and trace elements composition of WBA depending on the particle size fraction (wt. %).

<i>Major and minor elements</i>			<i>Trace elements</i>		
	<b>WBA0/30</b>	<b>WBA8/30</b>		<b>WBA0/30</b>	<b>WBA8/30</b>
<b>SiO<sub>2</sub></b>	45.44	52.08	<b>Cu</b>	0.13	0.04
<b>CaO</b>	17.55	20.72	<b>Zn</b>	0.51	0.12
<b>Al<sub>2</sub>O<sub>3</sub></b>	10.38	6.35	<b>Br</b>	<0.01	<0.01
<b>Na<sub>2</sub>O</b>	5.04	3.38	<b>Rb</b>	<0.01	0.01
<b>K<sub>2</sub>O</b>	1.54	2.09	<b>Sr</b>	0.07	0.05
<b>Fe<sub>2</sub>O<sub>3</sub></b>	6.08	4.12	<b>Y</b>	<0.01	<0.01
<b>MgO</b>	2.66	2.43	<b>Zr</b>	0.03	0.10
<b>TiO<sub>2</sub></b>	0.65	0.65	<b>Nb</b>	<0.01	<0.01
<b>Cl<sup>-</sup></b>	1.42	0.54	<b>Sn</b>	0.02	0.01
<b>SO<sub>3</sub></b>	2.57	1.07	<b>Sb</b>	0.01	0.01
<b>Mn</b>	0.03	0.02	<b>Ba</b>	0.07	0.04
<b>LOI*</b>	5.78	6.10	<b>Pb</b>	0.11	0.04

LOI\*: Loss on ignition at 1100 °C



**Table 2.** Major, minor, and trace elements composition of PAVAL<sup>®</sup> (wt. %).

*Major and minor elements*

<b>SiO<sub>2</sub></b>	<b>CaO</b>	<b>Al<sub>2</sub>O<sub>3</sub></b>	<b>Na<sub>2</sub>O</b>	<b>K<sub>2</sub>O</b>	<b>Fe<sub>2</sub>O<sub>3</sub></b>	<b>P<sub>2</sub>O<sub>5</sub></b>	<b>MgO</b>	<b>TiO<sub>2</sub></b>	<b>MnO</b>	<b>LOI*</b>
8.21	1.99	61.24	2.70	0.63	1.57	0.06	6.21	0.80	0.18	15.71

*Trace elements*

<b>Ba</b>	<b>Cr</b>	<b>Cu</b>	<b>Mn</b>	<b>Ni</b>	<b>Pb</b>	<b>Sr</b>	<b>V</b>	<b>Zn</b>	<b>Zr</b>
0.16	0.05	0.29	0.16	0.02	0.02	0.02	0.01	0.12	0.01

\*LOI: Loss on ignition at 1100 °C

**Table 3.** OPC paste and alkali-activated binders (AABs) mix proportion.

Binder	S					L			L/S ratio		
	<sup>1</sup> OPC	<sup>1</sup> MK	<sup>1</sup> WBA	<sup>1</sup> WBA8/30	<sup>1</sup> PV	<sup>2</sup> NaOH (wt. %)				<sup>2</sup> Na <sub>2</sub> SiO <sub>3</sub> (wt. %)	<sup>2</sup> H <sub>2</sub> O (wt. %)
						4M	6M	8M			
OPC paste	100	-	-	-	-	-	-	-	-	100	0.5
AA-MK	-	100	-	-	-	-	-	68	66	-	1.3
AA-WBA0/30	-	-	100	-	-	-	20	-	80	-	1.0
AA-WBA8/30	-	-	-	100	-	-	20	-	80	-	0.8
AA-WBA/PV	-	-	-	98	2	20	-	-	80	-	0.6

<sup>1</sup>wt. % respect to the total solid

<sup>2</sup>wt. % respect to the total liquid

**Table 4.** Reagent grade chemicals used to prepare 1 L of standard freshwater stock solution (OECD, 2004).

	Formula	Amount (g)
Calcium chloride	$\text{CaCl}_2 \cdot 2 \text{H}_2\text{O}$	11.76
Magnesium sulphate	$\text{MgSO}_4 \cdot 7 \text{H}_2\text{O}$	4.93
Sodium bicarbonate	$\text{NaHCO}_3$	2.59
Potassium chloride	$\text{KCl}$	0.23

\* *Distilled water with conductivity < 10  $\mu\text{S}\cdot\text{cm}^{-1}$  was used*

**Table 5.** Leaching concentration values ( $\text{mg}\cdot\text{kg}^{-1}$ ) of powdered raw materials according to EN 12457-2.

Sample	As	Ba	Cd	Cr	Cu	Hg	Mo	Ni	Pb	Sb	Se	Zn	pH
OPC	<0.01	4.80	<0.01	3.23	0.09	<0.01	8.79	0.29	0.03	<0.01	0.2	0.22	12.88
MK	0.01	0.84	<0.01	<0.01	0.02	<0.01	0.01	0.53	<0.01	<0.01	0.2	0.18	7.89
WBA0/30	0.04	0.37	<0.01	0.45	3.33	<0.01	1.26	0.03	<0.03	0.35	0.21	0.44	11.33
WBA8/30	0.02	0.25	<0.01	0.17	0.69	<0.01	0.33	0.11	0.01	0.27	<0.10	0.12	9.54
PV	0.10	0.06	<0.01	0.01	0.15	<0.01	2.90	<0.20	0.01	1.38	<0.10	0.10	10.03
<sup>1</sup> Inert waste ( $\text{mg}\cdot\text{kg}^{-1}$ )	0.5	20	0.04	0.5	2	0.01	0.5	0.4	0.5	0.06	0.1	4	
<sup>1</sup> Non-hazardous waste ( $\text{mg}\cdot\text{kg}^{-1}$ )	2	100	1	10	50	0.2	10	10	10	0.7	0.5	50	
<sup>1</sup> Hazardous waste ( $\text{mg}\cdot\text{kg}^{-1}$ )	25	300	5	70	100	2	30	40	50	5	7	200	

<sup>1</sup>limit for acceptance at landfills. EU landfill legislation [24]

**Table 6.** Leaching concentration values ( $\text{mg}\cdot\text{kg}^{-1}$ ) of OPC paste and AABs according to EN 12457-2.

<i>Using deionised water</i>														
	<b>As</b>	<b>Ba</b>	<b>Cd</b>	<b>Cr</b>	<b>Cu</b>	<b>Hg</b>	<b>Mo</b>	<b>Ni</b>	<b>Pb</b>	<b>Sb</b>	<b>Se</b>	<b>V</b>	<b>Zn</b>	<b>pH</b>
<b>OPC</b>	0.00	4.46	<0.01	0.87	0.07	<0.01	1.19	0.20	0.03	0.01	0.01	0.01	0.18	12.72
<b>AA-MK</b>	3.57	0.04	<0.01	0.54	0.17	<0.01	0.06	0.02	0.08	0.02	0.07	26.75	0.19	10.72
<b>AA-WBA0/30</b>	3.56	0.05	<0.01	0.81	3.15	0.01	1.45	0.15	0.30	3.48	0.21	2.68	1.68	11.29
<b>AA-WBA8/30</b>	1.25	0.23	<0.01	0.61	0.78	<0.01	0.18	0.04	0.44	1.74	0.05	0.26	0.49	11.64
<b>AA-WBA/PV</b>	0.83	0.15	0.01	0.37	0.46	<0.01	0.12	0.03	0.17	1.17	0.06	0.35	0.33	11.19
<i>Using freshwater (OECD, 2004)</i>														
	<b>As</b>	<b>Ba</b>	<b>Cd</b>	<b>Cr</b>	<b>Cu</b>	<b>Hg</b>	<b>Mo</b>	<b>Ni</b>	<b>Pb</b>	<b>Sb</b>	<b>Se</b>	<b>V</b>	<b>Zn</b>	<b>pH</b>
<b>OPC</b>	0.00	5.27	<0.01	1.29	0.12	<0.01	2.09	0.21	0.02	0.02	0.04	0.01	0.37	7.13
<b>AA-MK</b>	1.46	0.06	<0.01	0.22	0.24	<0.01	0.14	0.02	0.02	0.03	0.11	7.25	0.16	6.98
<b>AA-WBA0/30</b>	2.39	0.07	0.01	0.52	3.60	<0.01	1.23	0.18	0.24	1.69	0.15	2.56	2.07	7.24
<b>AA-WBA8/30</b>	0.34	0.14	<0.01	0.24	1.22	<0.01	0.08	0.05	0.11	0.98	0.07	0.15	0.69	7.14
<b>AA-WBA/PV</b>	0.38	0.31	<0.01	0.15	0.74	<0.01	0.08	0.05	0.09	0.72	0.05	0.21	0.44	7.12
<sup>1</sup> Inert waste ( $\text{mg}\cdot\text{kg}^{-1}$ )	0.5	20	0.04	0.5	2	0.01	0.5	0.4	0.5	0.06	0.1	4	0.5	
<sup>1</sup> Non-hazardous waste ( $\text{mg}\cdot\text{kg}^{-1}$ )	2	100	1	10	50	0.2	10	10	10	0.7	0.5	50	2	
<sup>1</sup> Hazardous waste ( $\text{mg}\cdot\text{kg}^{-1}$ )	25	300	5	70	100	2	30	40	50	5	7	200	25	

<sup>1</sup>limit for acceptance at landfills. EU landfill legislation [24]

**Table 7.** Identification of metal(loid)s surface release mechanisms according to CEN/TS 16637-2 standard.

release mechanisms	As				Ba				Cd				Cr				Cu				Mo			
	P2	P3	P4	P5	P2	P3	P4	P5	P2	P3	P4	P5	P2	P3	P4	P5	P2	P3	P4	P5	P2	P3	P4	P5
<b>M1</b>	×	×	×	×	×	×	×	×	✓	✓	✓	✓	×	×	×	×	×	×	×	×	×	×	×	×
<b>M2</b>	×	×	×	×	✓	×	×	×	-	-	-	-	×	×	×	×	×	×	×	×	✓	×	×	×
<b>M3</b>	×	×	×	×	-	×	✓	×	-	-	-	-	×	×	×	×	×	×	×	×	-	×	×	×
M3.1	-	-	-	-	-	-	✓	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
M3.2	-	-	-	-	-	-	×	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
M3.3	-	-	-	-	-	-	×	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
<b>M4</b>	✓	×	✓	✓	-	✓	-	×	-	-	-	-	✓	✓	×	✓	✓	✓	✓	✓	-	✓	✓	✓
<b>M5</b>	-	✓	-	-	-	-	-	✓	-	-	-	-	-	-	✓	-	-	-	-	-	-	-	-	-
M5.1	-	-	-	-	-	-	-	✓	-	-	-	-	-	-	✓	-	-	-	-	-	-	-	-	-
M5.2	-	✓	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-

release mechanisms	Ni				Pb				Sb				Se				V				Zn			
	P2	P3	P4	P5	P2	P3	P4	P5	P2	P3	P4	P5	P2	P3	P4	P5	P2	P3	P4	P5	P2	P3	P4	P5
<b>M1</b>	✓	×	×	×	×	×	×	×	✓	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×
<b>M2</b>	-	×	×	×	×	×	×	×	-	×	×	×	×	×	×	×	×	×	×	×	×	×	×	×
<b>M3</b>	-	×	×	×	×	×	✓	✓	-	×	×	×	×	×	✓	×	×	×	×	×	✓	×	✓	×
M3.1	-	-	-	-	-	-	✓	✓	-	-	-	-	-	-	×	-	-	-	-	-	✓	-	✓	-
M3.2	-	-	-	-	-	-	×	×	-	-	-	-	-	-	✓	-	-	-	-	-	×	-	×	-
M3.3	-	-	-	-	-	-	×	×	-	-	-	-	-	-	×	-	-	-	-	-	×	-	×	-
<b>M4</b>	-	✓	✓	✓	✓	✓	-	-	-	✓	✓	✓	✓	✓	-	✓	✓	✓	✓	✓	-	✓	-	×
<b>M5</b>	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	✓
M5.1	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	✓
M5.2	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-

**M1:** Overall low concentration

**M2:** Surface wash-off followed by low concentration

**M3:** Diffusion controlled release of a substance

  M3.1: Surface wash-off preceding diffusion-controlled release

  M3.2: Diffusion-controlled release followed by depletion

  M3.3: Surface wash-off preceding diffusion-controlled release followed by depletion

**M4:** Dissolution controlled release of a substance

**M5:** Unidentified mechanism

  M5.1: Surface wash-off of a substance preceding the unidentified release

  M5.2: Unidentified mechanism followed by depletion

**P2:** AA-MK

**P3:** AA-WBA0/30

**P4:** AA-WBA8/30

**P5:** AA-WBA/PV

**Table 8.** Cumulative release ( $\text{mg}\cdot\text{m}^{-2}$ ) of AABs after 8 extractions (64d) following CEN/TS 16637-2.

Sample	As	Ba	Cd	Cr	Cu	Hg	Mo	Ni	Pb	Sb	Se	V	Zn
AA-MK	53.8	0.6	0.1	2.9	4.1	< 0.1	1.7	0.4	0.4	0.8	3.4	450	4.5
AA-WBA0/30	25.3	1.3	6.8	10.6	75.6	0.2	10.8	3.6	13.2	55.8	2.2	27.2	115.9
AA-WBA8/30	33.1	13.5	1.6	19.9	19.4	0.1	4.1	1.9	20.1	56.3	1.8	8.7	27.5
AA-WBA/PV	30.1	7.2	1.3	16.8	19.9	0.1	3.9	1.5	15.3	54.9	2.6	20.5	27.9
<sup>1</sup> U <sub>1</sub> limit	40	600	1	150	50	0.4	15	100	50	3.5	1.5	250	200
<sup>1</sup> U <sub>2</sub> limit	300	4500	7.5	950	350	3	95	800	350	25	9.5	1500	1500

<sup>1</sup>Leaching limits set by the Netherlands tank leaching test (NEN 7345)

**Table 9.** EC<sub>50</sub> values and 95% confidence limits by probit analysis in the *Daphnia magna* immobilisation test.

	24 hours			48 hours		
	EC <sub>50</sub> (%)	95% LCL	95% UCL	EC <sub>50</sub> (%)	95% LCL	95% UCL
<b>OPC</b>	29.7	24.1	36.7	24.1	19.4	29.9
<b>AA-MK</b>	12.5	-	-	8.8	6.3	11.1
<b>AA-WBA0/30</b>	6.0	-	-	5.0	-	-
<b>AA-WBA8/30</b>	10.8	8.9	13.0	8.5	7.0	10.2
<b>AA-WBA/PV</b>	13.2	-	-	7.8	5.3	9.9

EC<sub>50</sub> for P2, P3 and P5 at 24 h and P4 at 48 h were extrapolated.

LCL: lower confidence limit to the EC<sub>50</sub>.

UCL: upper confidence limit to the EC<sub>50</sub>.



**Table 10.** Spearman's correlation coefficient for metal content in leachates obtained according to EN 12457-2 (using freshwater) and EC<sub>50</sub> values calculated from *Daphnia magna* immobilisation test.

	<b>As</b>	<b>Ba</b>	<b>Cd</b>	<b>Cu</b>	<b>Mo</b>	<b>Ni</b>	<b>Pb</b>	<b>Sb</b>	<b>Se</b>	<b>V</b>	<b>Zn</b>
<b>EC<sub>50</sub></b>	-0.7	0.3	0.3	-0.9*	0.359	0.051	-0.072	-0.9*	-0.6	-0.4	-0.8

<sup>a</sup> Significant correlations at  $p < 0.05$  are marked with one asterisk. <sup>b</sup> Chromium is not included in the analysis because their values determined in the leachates were below detection limit.

## Figure Caption

**Fig. 1.** Scheme of the methodology and goals of the study.

**Fig. 2.** Cumulative area release of As, Se, and V from AABs specimens according to CEN/TS 16637-2 standard.

**Fig. 3.** Cumulative area release of Cd, Cu, Mo, Ni, Sb, and Zn from AABs specimens according to CEN/TS 16637-2 standard.

**Fig. 4.** Cumulative area release of Ba, Cr, and Pb from AABs specimens according to CEN/TS 16637-2 standard.

**Fig. 5.** Log-log plot of cumulative area release ( $R_n$ ) of some metal(loid)s leached from AABs specimens formulated using WBA as the precursor.

**Fig. 6.** Immobilization rate of *Daphnia magna* according to the concentration of the leachate resulting from the EN-12457-2 standard.

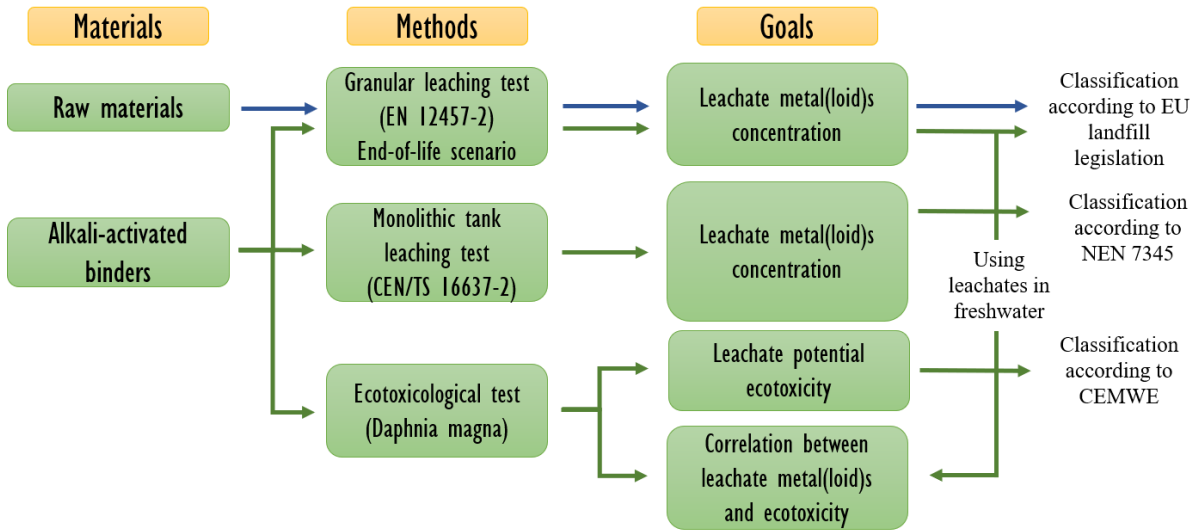


Fig. 1

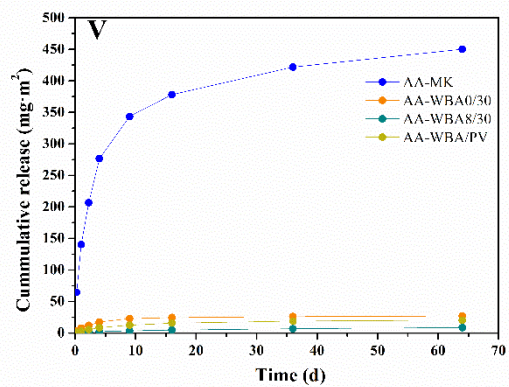
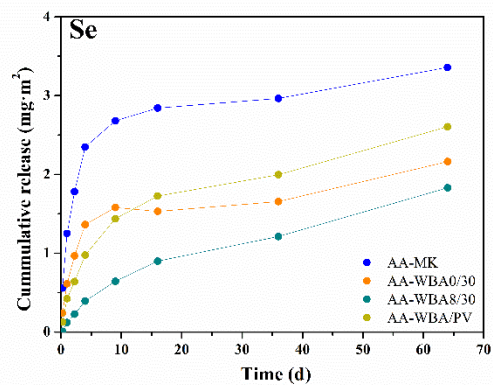
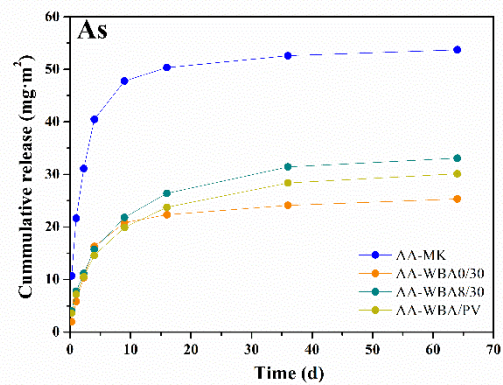


Fig. 2

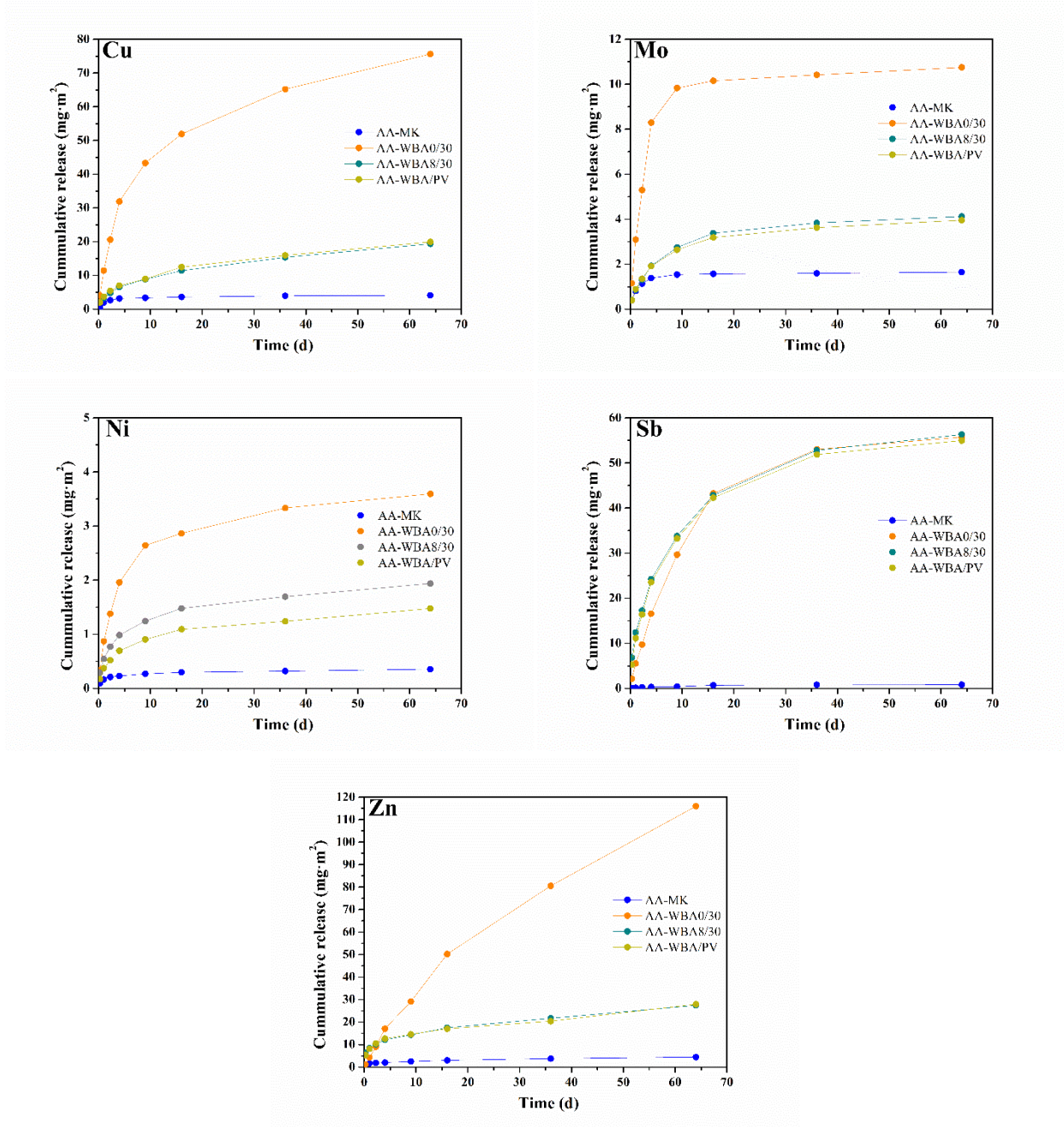


Fig. 3

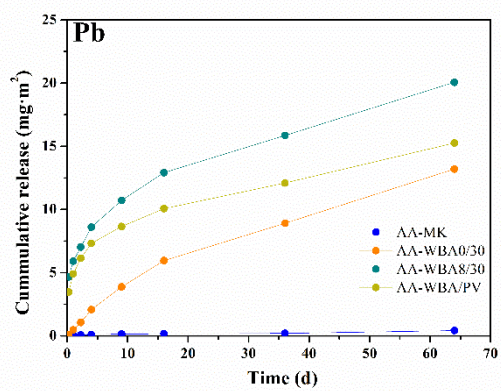
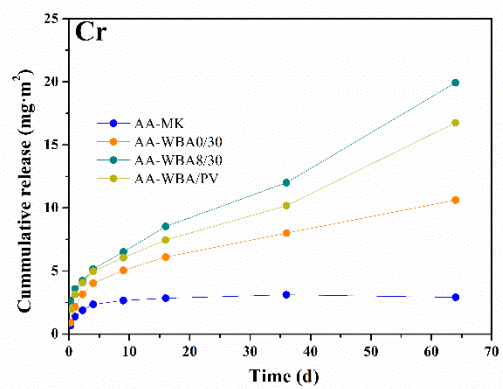
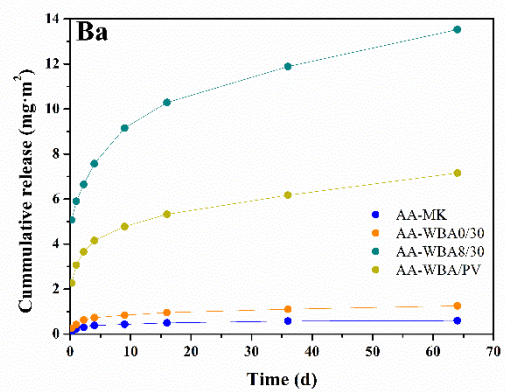


Fig. 4



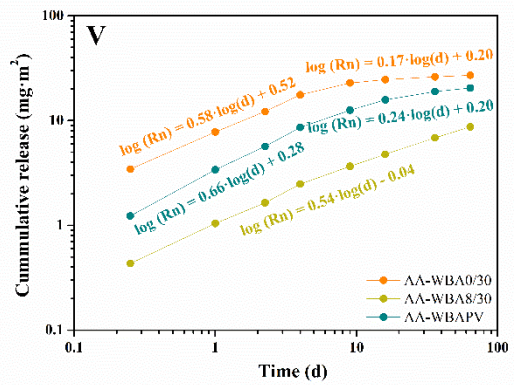
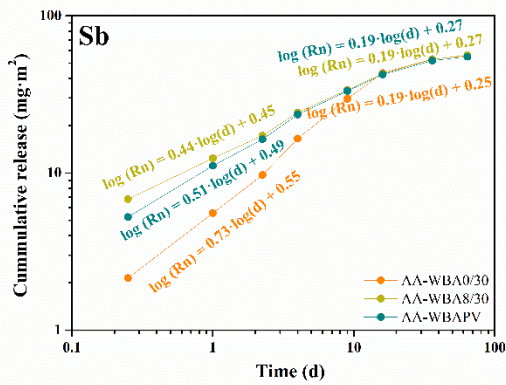
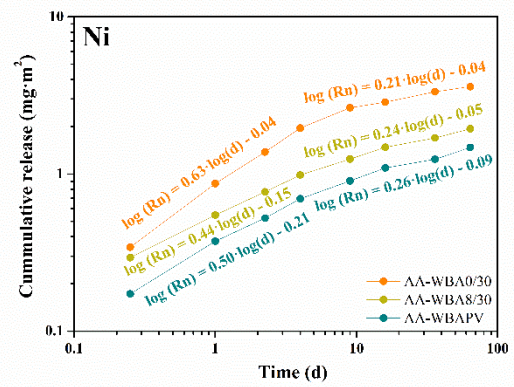
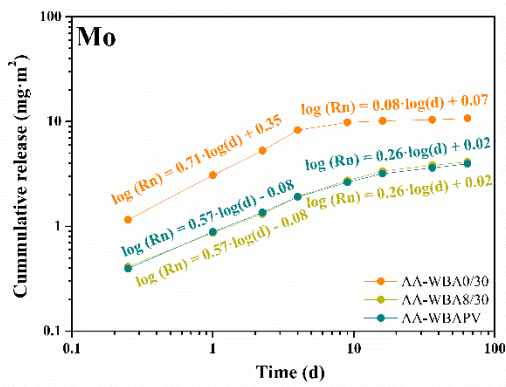
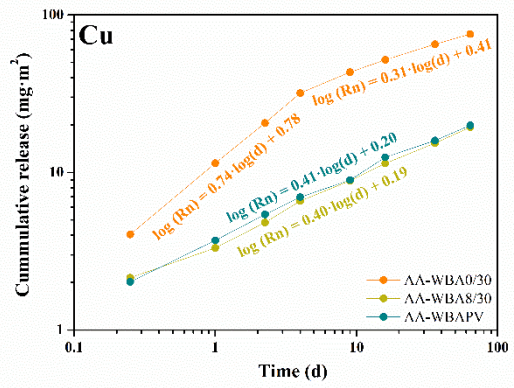
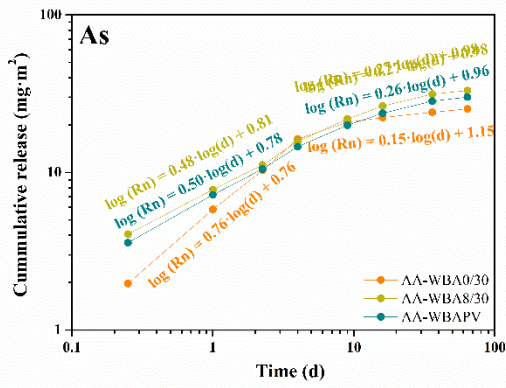


Fig. 5

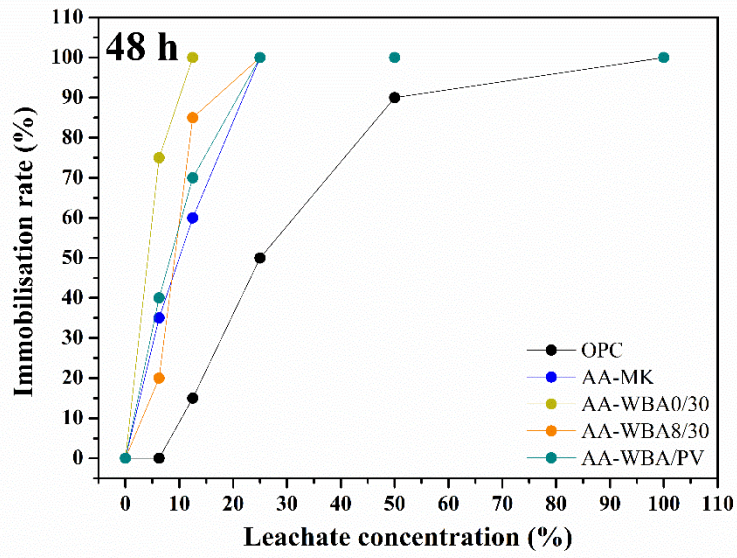
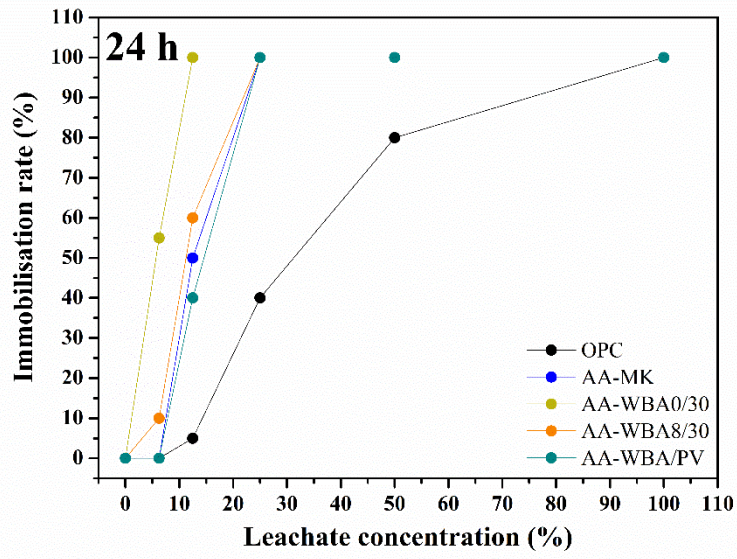


Fig. 6





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**Supplementary Material**

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