

1 **The role of pH on leaching of heavy metals and chlorides from Electric**
2 **Arc Furnace Dust in cement-based mortars.**

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13 **Abstract**

14 Electric arc furnace dust (EAFD) presents a pollution hazard due to its heavy metal leach-
15 ability. Its characterization is essential for recovery or disposal in hazardous waste land-
16 fills. The present work discusses the results of encapsulating two electric arc furnace dust
17 samples (EAFD₁ and EAFD₂) by means of the stabilization/solidification (S/S) technique
18 in cement-based mortars. The pH influence on the leaching levels of Se, Mo, Cd, and Pb
19 was evaluated using a pH dependence test as analytical support. In addition, the compres-
20 sive strength of the S/S mortars was measured to verify its behaviour as a monolithic
21 block. The leaching procedures for granular and monolithic mortars were carried out in
22 order to classify the EAFD samples and the S/S mortars made with EAFD according to
23 the legal limit values imposed by the EU Council Decision 2003/33/EC. Thus, the release

24 levels of Se, Mo, Cd, Pb and chloride in the leachate form of EAFD exceeded the legal
25 limit values, and both EAFDs were classified as hazardous wastes. Concerning the com-
26 pliance test of S/S mortars after crushing, despite lower releases being registered, none of
27 the S/S mortars in the granular state could be classified as non-hazardous waste due to
28 the high Pb level registered. Finally, after analysing the release levels of S/S mortars of
29 EAFD in the monolithic state, the S/S mortars of EAFD₁ were classified as non-hazardous
30 waste. Regarding the S/S mortars of EAFD₂, despite the high reduction in the Pb release
31 level after the S/S treatment, these mortars exceeded the hazardous legal limits.

32 **Keywords:** Electric Arc Furnace Dust; pH dependence test; Encapsulation; Stabiliza-
33 tion/Solidification; Leaching behaviour; Heavy metals.

34 **1. Introduction.**

35 Spain is one of the world's largest steel producers, ranking third in the Europe an Union
36 behind only Germany and Italy. According to Spanish Steel Companies Union
37 (UNESID), over 75% of the steel produced in Spain is made from recycled steel. In fact,
38 in 2011 Spain recycled 12.5 million tons of steel. For every ton of steel that is recycled
39 this industry saves around one and a half tons of iron ore, as well as water usage by 85%,
40 energy consumption by 80% and carbon dioxide output by 95% in addition to related
41 emissions [1].

42 In Spain, there are 22 steel mills, 21 of which use electric arc furnace (EAF) and scrap as
43 input. During the meltdown of the scrap, the electric arc furnace (EAF) can reach tem-
44 peratures of 1600°C, or even higher, and many components of the charge, including Fe,
45 Zn and Pb, are volatilized. These volatiles are carried by the exhaust gases and retained
46 in filters [2, 3]. Consequently, a large quantity of dust is generated when the vapor is

47 cooled and collected. These particles are defined as electric arc furnace dust (EAFD) [4,
48 5], being generated in the Spanish sector around 115000 EAFD tons/year [6].

49 When steel operation is based only on the melting scrap, the heavy metals content in
50 EAFD can reach up to 25% by weight. Due to this heavy metal content EAFD was clas-
51 sified as hazardous waste by European Waste Catalogue [7], code 100207 and a pretreat-
52 ment for disposal in hazardous waste landfills is needed.

53 EAFD chemical composition varies significantly depending on the type of steel produced
54 in the furnace, raw material and additives used in the process. A small change in the
55 furnace can lead to a possible variation in the composition of EAFD. Regarding the min-
56 eralogical composition, EAFD is a mixture of metal oxides, silicates and sulphates [8]
57 composed of magnetite (Fe_3O_4) and zincite (ZnO) as the major oxides [4, 9, 10].

58 The pollutant potential of this type of waste lies in the leachability of heavy metals such
59 as Zn, Cu, Ni, Cd, Cr, Hg and Pb. This implies that the first basic palliative action must
60 be the disposal of EAFD in hazardous waste landfills protected from rain to prevent the
61 generation of contaminated leachates that could pollute surrounding areas. It is known
62 that approximately 70% of EAFD is sent to landfills worldwide and the remaining 30%
63 is processed for Zn recovery and other purposes [4, 11, 12]. However, remedial treatments
64 are increasingly used as alternatives for simply disposing of the waste in landfills.

65 According to Pensaert et al. [13], the stabilization/solidification (S/S) technology was in-
66 itially developed as a treatment concept for hazardous waste prior to landfilling. During
67 the last decades, the S/S technique has also been applied as a remediation technology for
68 contaminated soils in developed countries for a wide variety of contaminants such as or-
69 ganics and heavy metals. In this sense, it has been proven that the S/S technique is

70 especially suitable for heavy metals immobilization. However, the fixing mechanisms of
71 some metals such as Pb are not sufficiently known.

72 This technique is being gradually applied in the remediation of contaminated soils due to
73 its cost-effectiveness, in addition to the reduction in the leachability of hazardous constit-
74 uents from the disposed wastes in landfills [14]. Specifically, in Spain, approximately
75 30% of EAFD is managed using the S/S technique and deposited in appropriate hazardous
76 waste landfills after its treatment [15]. This is the starting point of this investigation: the
77 assumption that a remedial technology can reduce the mobility of the contaminants [6].

78 Cement is a material representative of a family having conceptual and practical ad-
79 vantages as S/S matrices. It is a building material that has been extensively tested with a
80 long history of use in different environmental conditions [16]. The advantage of using
81 cement is solidification resulting in a monolithic, water tight final product. In prior studies
82 [17-19], high doses of cement are often required (60-90% by weight or more), resulting
83 in high treatment costs and an increased mass and volume of the treated material.

84 Therefore, the S/S technique aims to avoid the contamination of ecosystems by EAFD.
85 To that end, this technique intends to give monoliths two key parameters: (1) suitable
86 mechanical properties and (2) immobilization of potentially polluting elements, so that
87 they are not released into the medium. Mechanical properties such as compressive
88 strength are necessary for managing the monolith, making it possible for transportation
89 and reducing the risk of accidental pollution and/or fracture. Moreover, pollution of the
90 environment through water (leaching effect) is another major risk to avoid.

91 One of the mechanisms used in the S/S technique is the encapsulation of waste in the
92 matrix. Encapsulation is a process in which the waste is incorporated into a matrix without
93 chemically fixing the toxic elements to the material used. Multiple factors affect the

94 mobility of the elements, in this sense, Van der Sloot and Dijkstra [34] reported that the
95 pH of the material, the pH of its environment and the pH of the leaching liquid are crucial
96 in determining the release of elements in monoliths or granular materials. Although the
97 release curves of elements vs. pH are very similar, each material has its own pH dependent
98 release curve. Previous studies of S/S of EAFD have been carried out by Issa et al. [5]
99 who analysed the compressive strength and leaching behaviour in concrete specimens
100 made with fly ash and EAFD, replacing these different proportions by fine aggregate and
101 cement, respectively. These authors obtained Pb immobilization with the replacement of
102 50% of fine aggregate by EAFD because the pH registered during the Toxicity Charac-
103 teristic Leaching Procedure (TCLP) reached values between 7-8, while for Zn, the release
104 levels increased with increased EAFD content. Another research study on EAFD encap-
105 sulation in concrete was carried out by de Souza et al. [18]. In this case, the leaching
106 results were favourable, remaining below the limits for all metals except for Al, likely
107 due to the dissociation of the calcium aluminate present in the clinker of Portland cement.
108 Navarro et al. [52] experimented mobilization of Pb at high alkaline conditions, when
109 $\text{Pb}(\text{OH})^{4-}$ was the most stable species. These authors affirmed that the optimum pH range
110 to Pb precipitation is about 10.

111 Due to the complex composition of EAFD and the variety of its typology, it is not possible
112 to establish general rules for encapsulation technique of EAFD in cement based mortars.
113 Therefore, an analysis of the pH dependence of its elements is necessary for a better
114 knowledge of the S/S treatment.

115 The present work studies the pH influence on the leaching of Se, Mo, Cd, Pb and chloride
116 from EAFD encapsulated in cement based mortars. To reduce the EAFD polluting poten-
117 tial, an S/S treatment with PC clinker was applied by means of incorporating EAFD in
118 mortars. To observe the influence of EAFD typology on encapsulation treatment

119 effectiveness, two different EAFDs were used in the mixtures. In order to provide
120 complete characterization of EAFD and a better understanding of the mobility of the
121 EAFD elements, the pH dependence test according to UNE-EN 14429:2015 [20] was
122 carried out. The compressive strength and leaching behaviour were examined to deter-
123 mine the encapsulation treatment effectiveness and release of hazardous elements. Re-
124 garding the leaching behaviour, it was analysed by performing the compliance test ac-
125 cording to the standard UNE-EN 12457-4:2003 [21] for the EAFD after its S/S treatment.
126 In addition, the tank leaching test according to XP X31-211:2012 [22] was conducted for
127 S/S monolithic mortars of EAFD. The main contribution of this work is to justify the
128 encapsulation treatment effectiveness of EAFD with the role of the pH on the release
129 level of different heavy metals (Pb, Mo, Cd and Se).

130 **2. Materials and Methods.**

131 **2.1. Experimental programme.**

132 Two EAFDs from different sources were used in the present study to observe the influ-
133 ence of EAFD typology on the encapsulation treatment. EAFD₁ and EAFD₂ came from
134 two steel industries located in Olaberría and Zumárraga (Guipúzcoa, Spain), where sam-
135 pling were carried out according to UNE-EN 14899:2007 [23]. Both facilities used EAF
136 in the production of steel. Zumárraga produces up to 320000 tons of bars and 730000
137 wire rods per year, whereas Olaberría manufactured 900.000 tons of structural profiles in
138 2017 [24].

139 When EAFD is introduced into a cementitious matrix, the compressive strength and
140 leaching behaviour are affected. To observe the influence in both aspects, two control
141 mortars (named CM₁ and CM₂) were manufactured without EAFD content. The control
142 mortars were acquired from Valsec-Valderribas Company, which were manufactured

143 with Portland cement CEM I 52.5 R-SR (C), limestone filler (LF) and siliceous natural
144 sand (NS). Their dosage in weight percentage is show in Table 1.

145 Six mixtures were created to evaluate the influence of two EAFD on mortars, giving pri-
146 ority to the analysis of the compressive strengths registered and the leaching behaviours
147 of all samples. The design of these mixtures consisted of incorporating an amount of
148 EAFD in the control mortars (CM₁ and CM₂), using a ratio of 2:1 by weight (mor-
149 tar:EAFD). These ratios were used by authors such as Pereira et al. [25] and Ledesma et
150 al. [26]. Mortars with EAFD content were referred to as S/S mortars. The nomenclature
151 and dosage used for each mix are shown in Tables 2 and 3.

152 The amount of water added was the equivalent to obtain a consistency of 230 ± 10 mm in
153 accordance with UNE-EN 1015-3:2000 [27]. This high fluidity was achieved by increas-
154 ing the water/cement ratio (Table 3). For each mixture, 9 cylindrical samples 80 mm high
155 and 40 mm in diameter were made, according to XP X31-212:2011 [28]. These samples
156 were cured in a climatic chamber at $20^{\circ} \text{C} \pm 2^{\circ} \text{C}$ and relative humidity of $95\% \pm 5\%$ for
157 28 days. Regarding the replication of the analysis, 3 repetitions were carried out for each
158 laboratory test performed.

159 Thus, the experimental programme was as follows: 3 specimens were used to determine
160 the mechanical strength of mortars, and 6 specimens were used to develop the environ-
161 mental assessment of the EAFD by means of testing. Three specimens were used accord-
162 ing to the compliance test UNE-EN 12457-4:2003 [21] for analysing the leaching behav-
163 iour of S/S mortars of EAFD in the granular state, and 3 specimens were used according
164 to the tank leaching test XP X31-211:2012 [22] for evaluating the leaching release of
165 monoliths after the S/S treatment of waste.

166 Fig. 1 summarizes the experimental procedure developed in the present research.

167 **2.2. EAFD characterization.**

168 EAFD₁ and EAFD₂ were previously characterized by the following procedures: X-rays
169 fluorescence (XRF), specific surface area and real particle density. XRF was carried out
170 by the following equipment: Power: 4kW; Model: S4PIONEER; Brand: BRUKER. The
171 specific surface areas of EAFD₁ and EAFD₂ were determined by the BET method, using
172 a Micromeritics ASAP 2010 equipment and the real particle density was estimated ac-
173 cording to UNE 80103:2013 [29].

174 To evaluate the leaching behaviours of EAFD₁ and EAFD₂ and analyse their pollutant
175 potentials before being treated by S/S, the compliance test according to UNE-EN 12457-
176 4:2003 [21] was performed.

177 The compliance test analysed the leaching behaviour of granular wastes (grain size less
178 than 10 mm). This laboratory procedure established a single leaching state with a liquid
179 to solid ratio (L/S) of 10 L/kg. In the experimental procedure 0.90 kg of dry waste are
180 tested. First, the powder sample was introduced into a vessel and deionized water was
181 added to establish an L/S ratio = 10 L/kg. Immediately, the mixture was shaken in a tum-
182 bler for 24 hours. Then, a sample of eluate was filtered with a membrane (0.45 µm) and
183 analysed by inductively coupled plasma mass spectrometry (ICP-MS) using a Perkin
184 Elmer ELAN DRC-e spectrometer.

185 The EU Council Decision [30] of the acceptance of wastes in landfills has established
186 three limits for granular waste: "inert," "non-hazardous," and "hazardous," according to
187 the pollutant behaviour and depending on the release levels of heavy metals on leachates.
188 Thus, in the following the obtained data were compared with the legal limits established
189 by the Landfill Directive [30], which are illustrated in Table 4.

190 Both EAFDs were also tested by the pH dependence procedure in accordance with the
191 standard UNE-EN 14429:2015 [20] in order to analyse the influence of pH value on the
192 release levels of the parameters listed in the Landfill Directive. The pH dependence test
193 requires at least 8 samples of EAFD at different pH values. It is necessary to cover a pH
194 range between 4 and 12. Thus, 15 g of EAFD and deionized water were introduced in 8
195 vessels, establishing an L/S ratio = 9 L/kg. Starting from the natural pH of EAFD, differ-
196 ent amounts of acid/base were added until the desired pH was reached in each case (7,
197 7.75, 8.5, 9.25, 10, 10.75, 11.5 and 12.25). The acid and base used were HNO₃ and NaOH,
198 respectively (4 mol/l). Then, deionized water was added, and an L/S ratio = 10 L/kg was
199 re-established. The samples were shaken with a magnetic agitator for 48 hours. After this
200 contact time, eluates were extracted from the suspensions with the different pH values,
201 filtered with a membrane (0.45 µm) and analysed by ICP-MS.

202 **2.3. Mortar characterization.**

203 **2.3.1. Compressive strength.**

204 The mechanical properties of S/S mortars of EAFD, such as compressive strength, are
205 necessary for managing the monolith, its manipulation and transport and for reducing the
206 risk of accidental fracture. For that reason, according to the standard XP X31-212:2011
207 [28], the compressive strengths of control mortars (CM₁ and CM₂) and S/S mortars of
208 EAFD (CM₁₋₁, CM₁₋₂, CM₂₋₁, and CM₂₋₂) were tested at 28 days of age.

209 These results were compared with the minimum requirement of acceptance (compressive
210 strength of 1 MPa) imposed by the English Environmental Agency (EEA) [31] for depos-
211 iting waste in landfills. This criterion has been previously applied by authors such as Fer-
212 nández et al. [32], Lampris et al. [33] and Ledesma et al. [26].

213 S/S hardened mortars of EAFD were analysed with an X-rays diffraction (XRD) powder
214 method, using a Siemens D5000 with monochromatic radiation of Cu K α ($\lambda= 1.5405 \text{ \AA}$;
215 40 kV; 30 mA). A scanning speed of 2°/min with a step of 0.02° every 0.6 s and scanning
216 angles between 3 and 80° in units of 2 θ were used. This technique aims to observe the
217 formation of different phases after the setting of S/S mortars.

218 **2.3.2. Leaching tests for environmental assessment.**

219 To evaluate the leaching behaviour of the mortars and analyse their pollution potential,
220 the compliance test UNE-EN 12457-4:2003 [21] was conducted for the control (CM₁ and
221 CM₂) and S/S mortars of EAFD (CM₁₋₁, CM₁₋₂, CM₂₋₁, and CM₂₋₂) after crushing and siev-
222 ing by a 10 mm sieve for correct performance of the leaching procedure.

223 In addition, for the monolithic samples, the tank leaching test was carried out according
224 to the standard XP X31-211:2012 [22]. This test evaluates the leaching behaviour due to
225 the phenomenon of binding and diffusion that controls the release of contaminant ele-
226 ments in monolithic materials [34]. It estimates the leaching of contaminants from the
227 solid waste initially or generated by a solidification process (monolithic waste). This tank
228 leaching test allows to evaluate the elements release in the first contact with the leaching
229 liquid in 24 hours, however, the development of the long term leaching is not evaluated.

230 In previous works, Cyr et al. [35] used this test to evaluate the leaching behaviour of
231 sewage sludge ash in cement-based monoliths. Subsequently, Cyr et al. [36] and Quina et
232 al. [37] analysed the leaching of fly ash from the incineration of municipal solid waste
233 introduced into cement-based matrices.

234 The tank leaching test performed according to the mentioned laboratory procedure is il-
235 lustrated in Fig. 2. The tested mortars were manufactured as cylindrical shapes with a
236 height of 80 mm and diameter of 40 mm according to the standard XP X31-212:2011

237 [28]. After curing for 28 days, the control mortars and S/S mortars of EAFD were weighed
238 and introduced into a polypropylene tank for the leaching assessment. According to the
239 standard, the tank should have a volume of between 2 l and 6 l, an inside diameter of 140
240 mm and an opening diameter of at least 80 mm. The samples should be placed on a raised
241 surface 20 mm from the bottom. This surface must be a 5 x 5 mm mesh to ensure good
242 circulation of the reagent (deionized water). The reagent was introduced along with the
243 samples into the tank with L/S = 10 L/kg \pm 0.2 L/kg, leaving these samples fully im-
244 mersed, and the volume of liquid added ranged between 1 l and 6 l. The samples remained
245 immersed in reagent for 24 hours at a temperature of 20° C \pm 5° C and were moved
246 through a magnetic agitator at 120 rpm \pm 20 rpm. After this contact time, the eluate was
247 filtered with a membrane (0.45 μ m) and analysed by ICP-MS.

248 Due to the absence of monolithic limits, the EEA [31] proposes that the release levels
249 obtained on leachates are compared with the limits of Landfill Directive [30] to classify
250 the pollutant behaviour of the mortars (Table 4).

251 **3. Results and discussion.**

252 **3.1. EAFD characterization.**

253 Table 5 shows the chemical composition of EAFDs (XRF-analysis) and a comparison
254 with previous values from literature. The major species for each EAFD were oxides of
255 Zn and Fe, with similar values for both: 36.28% and 28.32% for EAFD₁ and 32.77% and
256 30.48% for EAFD₂. These values were in agreement with the literature [4, 9, 10].

257 The specific surface results of EAFD₁ and EAFD₂ were 3.7 m²/g and 4.6 m²/g, respec-
258 tively. These results were in agreement with those provided by de Paula et al. [38] and
259 Vargas et al. [39]. The real particle density was 3.85 g/cm³ for EAFD₁ and 3.81 g/cm³ for
260 EAFD₂. Studies carried out by de Souza et al. [18], Salihoglu and Pinarli [40] and Vargas

261 et al. [39] reflected real particle density results similar to those of the present study (4.08
262 g/cm³, 4.30 g/cm³ and 4.23 g/cm³, respectively).

263 Table 6 shows the element release levels by the compliance test (expressed as mg of
264 element per kg of dry matter) and provides parameters of conductivity (μS/cm), temper-
265 ature (°C) and pH. These results reveal that for EAFD₁, three parameters exceeded the
266 hazardous limit: Se, Cd and chloride. However, more elements were detected as hazard-
267 ous for EAFD₂: Se, Mo, Pb, and chloride. By analysing the levels released by EAFD₁ and
268 EAFD₂, both wastes were classified as hazardous according to the Landfill Directive [30]
269 (Table 4).

270 Despite both EAFD presenting a similar chemical composition, which can be observed in
271 Table 5, EAFD₂ exhibited higher pH values (13.28) than EAFD₁ (9.60). Leclerc et al. [41]
272 measured the following pH values in five different types of EAFDs: 8.2, 9.3, 10.4, 11.0
273 and 11.4. In the case of the present study, the variation of pH values in the two EAFDs
274 may be due to the different natures of the wastes. As previously stated, the EAFDs came
275 from different steelworks.

276 The large difference in the release levels between the two EAFDs (Table 6) was attributed
277 to the different pH values registered. Comparing both wastes, the greatest difference be-
278 tween the concentration values was observed in Pb (6.14 mg/kg and 5483.87 mg/kg). The
279 pH dependence test (Fig. 3a) confirmed the lower release of Pb in EAFD₁ (6.14 mg/kg)
280 due to a minimum in the Pb curve between pH values of 9 - 11. However, when the pH
281 was increased to values higher than 12, the Pb concentration in the leachate increased,
282 justifying that the higher concentration of Pb in EAFD₂ (5483.87 mg/kg) was due to the
283 pH value (13.28) recorded. It was previously confirmed by authors such as Mitrakas et
284 al. [42] that the high leachability of Pb at high pH values of the leachate was due to the

285 high hydroxyl concentration. Sebag et al. [43] also reported that the leachability of all
286 other metals in leaching tests with alkaline pH decreased with the increase of pH, except
287 for Pb and Cr. These authors stated that the high concentration of Pb in the leachates
288 should exclude EAFD from landfill disposal suggesting a specific treatment for safe dis-
289 posal in hazardous waste landfills.

290 The Mo concentrations in the leachate were also different between both the two EAFDs
291 (1.87 mg/kg for EAFD₁ and 20.49 mg/kg for EAFD₂). This phenomenon was again at-
292 tributed to the pH values recorded in the two wastes (9.60 and 13.28, respectively). In
293 Fig. 3b, Mo was shown to have a pH dependence similar to Pb (Fig. 3a), increasing its
294 concentration in the medium when the pH value increased. Dung et al. [44] concluded
295 that the release of potentially toxic elements decreased with increasing pH value in their
296 studied samples (except Mo).

297 Differences in the Cd concentration between the two EAFDs (2.71 mg/kg and 0.14
298 mg/kg) were also observed in the leachate. Cd (Fig. 3c) showed pH dependence opposite
299 to that presented for Pb and Mo (Fig. 3a and 3b); with a higher release to the medium at
300 pH values lower than 10. The results of the pH dependence test for Cd (Fig. 3c) justified
301 the highest concentration of this element in EAFD₁ (2.71 mg/kg) due to its lower pH
302 (9.60) compared to EAFD₂ (13.28). These results agreed with the results of Izquierdo and
303 Querol [45] who showed that Cd solubility decreased near neutral conditions.

304 The difference in the release of Se to the medium in the two EAFDs was not relevant
305 (4.56 mg/kg and 2.76 mg/kg). This element showed low pH dependence (Fig. 3d), with a
306 constant release pattern for each EAFD in the pH range studied.

307 According to data shown in Table 6, it can be observed that chloride ion exceeded the
308 hazardous legal limit in both EAFDs. The reason for the high levels registered was

309 explained by Mitrakas et al. [42], who affirmed that the high concentration of chloride
310 ion was due to the composition of the scrap material used during the steel manufacturing.
311 Additionally, differences were found between leaching values of other metals (Cr, Cu and
312 Zn) in the EAFDs (Table 6). However, the pH dependence test was not studied due to
313 they did not exceed the legal limit that the Landfill Directive establishes (Table 4).

314 **3.2. Mortar characterization.**

315 **3.2.1. Compressive strength of control and S/S mortars.**

316 The compressive strength development was affected when EAFD was introduced into a
317 cementitious matrix. Thus, it is important to analyse the compressive strengths of control
318 mortars, and the results are reported in Fig. 4. CM₁ and CM₂ showed the highest com-
319 pressive strength values (17.66 MPa and 18.52 MPa, respectively). The higher compres-
320 sive strength of CM₂ was justified by the mineral skeleton (NS) presented by the mortar
321 [26].

322 Previous research works [46] observed that the typical compressive strength of S/S mor-
323 tars of EAFD was approximately 0.5 MPa - 5 MPa. In accordance with Fig. 4, none of
324 the S/S mortars of EAFD₁ met the compressive strength requirement (1 MPa) imposed
325 by EEA [31]. Instead, the S/S mortars of EAFD₂ met the acceptance criterion for all tested
326 samples. The results of the compressive strengths for S/S mortars of EAFD₂ were appro-
327 priated (7.93 MPa and 8.47 MPa) to be considered monolithic blocks. According to the
328 above, the range expressed by Laskonen [46] as typical for S/S mortars of EAFD is 0.5
329 MPa - 5 MPa, and all S/S mortars of EAFD₂ were in this range.

330 It is known that the presence of Zn in mortars produces a delay of the hydration process.
331 This could be due to the formation of different forms of Zn (as ZnO and ZnSO₄) which

332 act as retardants of cement hydration. Although ZnO seriously retards the cement hydra-
333 tion during an initial period, it has been found that it increases the mechanical strength
334 after ageing [47]. This phenomenon would justify the higher strengths of the S/S mortars
335 of EAFD₂. Balderas et al. [47] noted that during the delay period, a protective amorphous
336 Zn hydroxide cover, Zn (OH)₂, is formed on the surfaces of the grains. Next, high con-
337 centrations of Ca²⁺ and OH⁻ transform the Zn (OH)₂ to crystalline hydrated hydroxide of
338 Zn and Ca, i.e., Ca (Zn₂(OH)₆·2H₂O).

339 Quian et al. [48, 49] proposed three main fixation mechanisms for Zn in alkali-activated
340 slag binder: (i) the formation of insoluble calcium zincate Ca (Zn₂ (OH)₆·2H₂O) precipi-
341 tate; (ii) the formation of insoluble zinc silicate gel (Z-S-H); and (iii) the incorporation of
342 zinc within the lattice of calcium silicate hydrates (C-S-H), i.e., the main hydration prod-
343 uct of PC. They also stated that the latter two mechanisms are preferable at low Zn con-
344 centrations (<0.5% by mass of slag), while excess Zn (2% by mass of slag) will precipitate
345 as calcium zincate.

346 In the present study, during the first stage of characterization, the S/S hardened mortars
347 of EAFD were subjected to XRD analysis (Fig. 5). Portlandite (Ca (OH)₂) did not appear
348 in the S/S mortars. However, a hydrated hydroxide of Ca and Zn (CaZn₂ (OH)₆·2H₂O)
349 was observed in the composition of S/S mortars of EAFD₂. This newly formed compound
350 did not appear in the S/S mortars of EAFD₁. The formation of CaZn₂ (OH)₆·2H₂O could
351 contribute to the acquisition of compressive strength demanded in the S/S mortars of
352 EAFD₂. Ledesma et al. [26] also observed the absence of Portlandite (Ca (OH)₂) and the
353 appearance of CaZn₂(OH)₆·2H₂O in S/S mortars of EAFD, the latter being attributed to
354 the compressive strength obtained in S/S mortars.

355 The formation of $\text{CaZn}_2(\text{OH})_6 \cdot 2\text{H}_2\text{O}$ in S/S mortars of EAFD₂ was justified by the pH of
356 the waste (13.28). Authors such as Zielger and Johnson [50] demonstrated that this was
357 stable at pH values approximately 12. Due to the natural pH of EAFD₂ (13.28) and since
358 the setting process is generated in an alkaline environment, the formation of CaZn_2
359 $(\text{OH})_6 \cdot 2\text{H}_2\text{O}$ is favourable in EAFD₂. However, in S/S mortars of EAFD₁, the formation
360 of $\text{CaZn}_2(\text{OH})_6 \cdot 2\text{H}_2\text{O}$ is not promoted due to the abrupt change of the pH of EAFD₁ in
361 the natural state (9.60) and its S/S, whose setting process increases the pH to values ap-
362 proximately 12.

363 Although the S/S mortars of EAFD₁ did not meet the mechanical requirement (1 MPa),
364 leaching of the samples CM₁₋₁ and CM₂₋₁ was studied in order to perform a comparison
365 with S/S mortars of EAFD₂ that exceeded this criterion.

366 **3.2.2. Leaching tests of control and S/S mortars.**

367 **3.2.2.1. Leaching assessment of specimens by compliance test.**

368 Table 7 shows the results of the released levels of control mortars (CM₁ and CM₂) ac-
369 cording to the dosage illustrated in Table 3. According to the leaching data, as expected,
370 after crushing the control mortars and testing by the leaching compliance test, they were
371 classified as inert materials.

372 Then, the S/S mortars of EAFD were also crushed (passing through a 10 mm sieve), with
373 the main objective of obtaining comparable data between the pure EAFD₂ and EAFD₁
374 and the S/S mortars.

375 The results obtained according to the compliance test corresponded to Se, Mo, Cd, Pb and
376 chloride anion since this group of elements was the most conflictive in both untreated
377 samples (EAFD₁ and EAFD₂), i.e., these elements exceeded the non-hazardous legal limit
378 according to the Landfill Directive.

379 Table 8 shows the results of the compliance test for S/S crushed mortars of EAFD, ex-
380 pressed in mg/kg dry weight. Additionally, other parameters such as conductivity
381 ($\mu\text{S}/\text{cm}$), temperature ($^{\circ}\text{C}$) and pH are shown.

382 The comparison between the release levels measured in EAFD₁ and EAFD₂ (pure granu-
383 lar samples) and the registered levels in S/S crushed mortars of EAFD was evaluated
384 according to the parameter Reduction Percentage of Granular Leaching (RPGL) calcu-
385 lated according to equation (1).

$$386 \quad RPGL = 100 - \frac{LRV_c \cdot 100}{LRV_w} \quad (1)$$

387 where: LRV_w = Leaching release value of EAFD₁ or EAFD₂waste.

388 LRV_c = Leaching release value of crushed S/S mortars.

389 The results of the S/S mortars of EAFD₁ showed very low leaching values for Se, Cd and
390 chloride compared to untreated EAFD₁, whose maximum reduction percentages of gran-
391 ular leaching (RPGL) were 94.74%, 99.93% and 71.19%, respectively.

392 However, for Mo and Pb, the S/S behaviour was different. When EAFD₁ was incorporated
393 into a cemented matrix (S/S mortar of EAFD₁), Mo and Pb contained in the waste were
394 released into the medium. Mo increased its initial concentration from 1.87 mg/kg to 2.90
395 mg/kg and 2.09 mg/kg in CM₁₋₁ and CM₂₋₁, respectively. In the case of Pb, the initial con-
396 centration of 6.14 mg/kg increased to 36.00 mg/kg and 28.20 mg/kg, respectively, in CM₁₋₁
397 and CM₂₋₁. This fact was attributed to the difference between pH of untreated EAFD₁
398 (9.60) and the pH reached during the S/S phase of EAFD₁ mortars (12.23 and 12.06).

399 As it is known, during the hydration of cement, the measured pH value is in the basic
400 zone (pH value greater than 12), and this situation is favourable for insolubilized metals
401 [51]. However, this pH increase does not favour the immobilization of all metals. This

402 fact was reported by Navarro et al. [52], who found difficulty to immobilize Pb using
403 cement-based materials.

404 Fig. 3a and 3b show that Mo and Pb exhibited greater release when the pH was approxi-
405 mately 12. This fact was corroborated by van der Sloot and Dijkstra [34], whose pH de-
406 pendence results on heavy metals were in accordance with those of the present study.

407 A decrease in the release levels of Se, Mo, Cd, Pb and chloride from S/S mortars of
408 EAFD₂ was observed, with maximum reduction percentages granular leaching (RPGL)
409 of 96.02%, 91.17%, 97.83%, 99.13% and 79.67%, respectively. The high amount of Pb
410 encapsulated is noteworthy. The initial concentration level in EAFD₂ reached 5483.87
411 mg/kg, whereas with the S/S technique, the release of Pb was 108.00 mg/kg and 47.80
412 mg/kg in CM₁₋₂ and CM₂₋₂, respectively.

413 The conductivity results during the compliance test are reported in Table 8. These results
414 showed that S/S mortars of EAFD₂ obtained higher conductivities than EAFD₁, register-
415 ing a maximum conductivity for CM₁₋₂ (6200 μ S/cm). The eluate temperature showed no
416 elevated changes in the samples.

417 The alkaline pH values recorded in this section (ranging between 12.06 and 13.02) were
418 consistent with previous studies. Laforest and Duchesne [53] controlled the leachability
419 of heavy metals such as Cr, Pb, Ni and Zn, treating them with ground granulated blast
420 furnace slag (GGBFS) and PC clinker. In this research, three types of samples contami-
421 nated with 25% EAFD and with different proportions of PC clinker and GGBFS were
422 manufactured, resulting in a pH range of 11.3 to 13.

423 Thus, regarding the classification of material according to the pollutant behaviour, despite
424 the significant reduction in the release levels of the most hazardous elements, none of the

425 S/S mortars of EAFD₂ tested by the compliance test can be classified as non-hazardous
426 waste due to the level of Pb metal (>10 mg/kg) (Table 4).

427 To clarify the results, it must be noted that a high percentage of substitution (33.33% in
428 weight) was used in the present study (Table 3). Consequently, similar results were ob-
429 served as those obtained by Salihoglu and Pinarli [40] who stabilized Zn and Pb with 30%
430 of EAFD by weight using lime and PC clinker in 1:1 proportion, registering a pH range
431 between 8.2 and 9.4. When the lime Zn samples were encapsulated by a high content of
432 EAFD (70%), this element was below the legal criteria. However, Pb immobilization was
433 insufficient with lime and PC. This proves that a high percentage of encapsulating agent
434 is necessary for immobilization this type of waste. These authors defined an optimum
435 formulation for the stabilization of waste as 30% EAFD + 35% PC clinker + 35% lime.

436 **3.2.2.2. Leaching assessment of specimens by tank leaching test.**

437 Table 9 shows the leaching data for control mortars (CM₁ and CM₂) obtained according
438 to the tank leaching test. There leachate levels obtained for both control mortars were
439 very low and in all cases, the diffusion release levels were lower than the inert legal limit.

440 For developing the evaluation of leaching from the S/S monolithic mortars of EAFD, the
441 most conflictive elements registered from the compliance tests of EAFD₁ and EAFD₂,
442 i.e., Se, Mo, Cd, Pb and chloride, were used.

443 Table 10 shows the tank leaching test results for S/S monolithic mortars of EAFD, ex-
444 pressed in mg/kg dry weight of Se, Mo, Cd, Pb and chloride. Additionally, parameters
445 such as conductivity (μS/cm), temperature (°C) and pH are shown.

446 As expected, the release levels registered by diffusion (Table 10) were lower than the
447 leaching levels registered by the compliance test (Table 8). This was logical since the

448 existing surface contact between the mortar and the leaching fluid was higher in the gran-
449 ular state than in the monolithic state.

450 Mo and Pb behaviours from the tank leaching test were different than the compliance test
451 behaviours. Mo (1.53 mg/kg vs. 1.29 mg/kg) and Pb (1.11 mg/kg vs. 2.82 mg/kg) in S/S
452 mortars of EAFD₁ reduced their leaching compared to untreated EAFD₁ (1.87 mg/kg for
453 Mo and 6.14 mg/kg for Pb). Using the tank leaching test maintained the pH of the S/S
454 mortars of EAFD₁ lower than in the compliance test, which favoured the encapsulation of
455 Mo and Pb.

456 All elements studied in the S/S mortars of EAFD₁ exhibited a release lower than the limit
457 established in the Landfill Directive (Table 4) for classifying waste as hazardous. There-
458 fore, the S/S mortars of EAFD₁ could be classified as non-hazardous waste.

459 Although the reduction of Pb in S/S mortars of EAFD₂ was high, Pb release into the me-
460 dium (14.05 mg/kg and 16.70 mg/kg) was shown to be higher than the legal limit. Again,
461 a clear pH dependence was observed for this element. The pH for S/S mortars of EAFD₁
462 obtained values of 11.44 and 11.82. However, for S/S mortars of EAFD₂, the pH rose to
463 12.57 and 12.38. Due to the high release level of Pb in EAFD₂ in its granular state and the
464 increase of pH in the S/S mortars of EAFD₂, the Pb release can be justified. Authors such
465 as Ledesma et al. [26] and Belebchouche et al. [54] observed a similar Pb release, attrib-
466 uting this phenomenon to the pH dependence.

467 Due to the level of Pb in the leachates exceeding the legal limit of the Landfill Directive
468 (Table 4), the S/S mortars of EAFD₂ were classified as hazardous materials and disposal
469 in conventional landfills without treatment is not possible.

470 **4. Conclusions.**

471 The study focused on two different Electric Arc Furnace Dust (EAFDs). Both EAFDs
472 exhibited similar physico-chemical properties, and the pH value was the main difference
473 observed. Due to the release levels of Se, Cd, Mo, Pb and chloride in accordance with the
474 compliance test, the legal limits values were exceeded, and both EAFDs were classified
475 as hazardous waste, which cannot be landfilled without adequate treatment.

476 The pH dependence test of EAFD detected different leaching behaviours for each of the
477 elements. Pb and Mo showed similar pH variation curves, resulting in elevated releases
478 with higher pH values. The Cd leaching behaviour was the opposite. This fact justified
479 the different concentrations of these elements in the two EAFDs.

480 Two types of control mortars were used (CM₁ and CM₂). CM₁ and CM₂ showed good
481 compressive strengths, with CM₂ being larger due to the mineral skeleton of the mortar.
482 Regarding their leaching behaviour, the control mortars were classified as inert materials
483 according to the Landfill Directive for granular and monolithic states.

484 The imposed criterion of compressive strength (1 MPa) was only satisfied by the S/S
485 mortars of EAFD₂. This achievement of compressive strength was attributed to the for-
486 mation of CaZn₂(OH)₆·2H₂O whose presence was not detected in the S/S mortars of
487 EAFD₁. CaZn₂(OH)₆·2H₂O formation in the S/S mortars of EAFD₂ was favourable due
488 to natural pH of EAFD₂ (13.28) and the basic pH achieved during the mortar setting pro-
489 cess.

490 Regarding the compliance test of S/S mortars of EAFD performed after crushing them,
491 Se, Mo, Cd and chloride showed lower release levels than the values obtained for the pure
492 samples of EAFD. Despite this fact, none of the S/S crushed mortars of EAFD could be
493 classified as non-hazardous waste due to the high Pb level released in the leachates (>10
494 mg/kg). Thus, although the greatest Reduction Percentage of Granular Leaching (RPGL)

495 in Pb was 99.13%, the basic pH values achieved during the hydration of cement and com-
496 pliance test were not favourable for the immobilization of this metal.

497 The pH difference between untreated EAFD₁ (9.60) and during the compliance test (12.23
498 and 12.06) of S/S mortars of EAFD₁ caused a higher release of Mo and Pb in the leachate
499 than untreated EAFD₁. This fact was not observed in the S/S mortars of EAFD₂ because
500 the difference between the pH values of the original waste (13.28) and that reached during
501 the compliance test (13.02 and 12.8) was small.

502 Although the S/S mortars of EAFD₁ did not exhibit good compressive strengths, they
503 were classified as non-hazardous materials in the monolithic state because the tank leach-
504 ing test results showed releases lower than the legal limits established by the Landfill
505 Directive.

506 Despite the high reduction value of Pb, the release values in the S/S mortars of EAFD₂
507 (14.05 mg/kg and 16.70 mg/kg) exceeded the legal limits of the Landfill Directive to be
508 classified as non-hazardous materials and could not be landfilled in conventional landfills.
509 New techniques to improve Pb encapsulation with alternative materials should be the aim
510 of future research.

511 Leaching factors, especially pH dependence, are important for the elemental release of
512 EAFD encapsulated in cement base materials. Therefore, it is not possible to establish
513 general rules for encapsulation technique of EAFD because the complex and varied ty-
514 pologies of EAFD.

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Table 1. Control mortars dosage.

Composition (% weight)	C	LF	NS
CM ₁	60	40	-
CM ₂	30	30	40

C: cement; LF: limestone filler; NS: natural sand

Table 2. Mortars nomenclature.

Control mortars	EAFD	S/S mortars
CM ₁	EAFD ₁	CM ₁₋₁
	EAFD ₂	CM ₁₋₂
CM ₂	EAFD ₁	CM ₂₋₁
	EAFD ₂	CM ₂₋₂

Table 3. Mortars dosage by mixture (g), water/cement ratio and consistency (mm).

S/S mortars	Dosage by mixture (g)						w/c*	Consistency (mm)
	C	NS	LF	EAFD ₁	EAFD ₂	Water		
CM ₁	2400.00	-	1600.00	-	-	1380.00	0.58	230
CM ₁₋₁	1600.20	-	1666.80	1333.00	-	1640.00	1.02	235
CM ₁₋₂	1600.20	-	1666.80	-	1333.00	1674.40	1.05	226
CM ₂	1200.00	1600.00	1200.00	-	-	1060.00	0.88	236
CM ₂₋₁	800.10	1666.80	800.10	1333.00	-	1300.00	1.62	238
CM ₂₋₂	800.10	1666.80	800.10	-	1333.00	1381.60	1.73	223

*water/cement ratio

Table 4. Legal limits of European Council Decision 2003/33/EC (L/S = 10 l/Kg).

	Components concentration (mg/kg dry matter)														
	Cr	Ni	Cu	Zn	As	Se	Mo	Cd	Sb	Ba	Hg	Pb	Fluoride	Chloride	Sulphate
Inert	0.5	0.4	2	4	0.5	0.1	0.5	0.04	0.06	20	0.01	0.5	10	800	1000
Non-Hazardous	10	10	50	50	2	0.5	10	1	0.7	100	0.2	10	150	15000	20000
Hazardous	70	40	100	200	25	7	30	5	5	300	2	50	500	25000	50000

Table 5. EAFD chemical composition and comparison with values from previous literature.

Compound (% weight)	EAFD ₁	EAFD ₂	Oustadakis et al. [4]	Sapiña et al. [9]	López and López- Delgado [10]
ZnO	36.28	32.77	25.29	33.49	34.11
Fe ₂ O ₃	28.32	30.48	24.78	32.37	19.05
Cl	4.82	11.37	3.62	1.38	4.53
CaO	4.56	2.93	18.60	5.97	3.49
SiO ₂	3.17	2.65	1.15	4.35	3.47
Na ₂ O	3.07	2.59	2.44	3.05	2.73
MnO	2.92	2.51	2.45	2.64	1.48
PbO	2.77	2.23	6.02	0.12	7.03
SO ₃	2.46	2.21	3.21	1.19	2.23
K ₂ O	2.13	1.96	1.80	0.90	1.75
MgO	1.50	1.67	3.95	2.16	1.93
Al ₂ O ₃	0.99	0.94	0.52	1.31	1.06
Cr ₂ O ₃	0.53	0.50	0.19	0.32	0.19
F	0.43	0.43	-	-	-
P ₂ O ₅	0.29	0.20	-	0.24	-
SnO ₂	0.14	0.08	-	-	-
Br	0.14	0.07	-	-	0.07
TiO ₂	0.09	0.07	-	0.12	0.11
CdO	0.08	0.04	-	-	0.09
BaO	0.06	0.03	-	-	-
NiO	0.03	0.02	-	0.03	0.03
MoO ₃	0.01	0.01	-	-	-
ZrO ₂	ND*	ND*	-	-	-

*Not detected

Table 6. Release levels of EAFD₁ and EAFD₂ according to UNE-EN 12457-4:2003.

Concentration (mg/kg dry matter)	EAFD ₁	EAFD ₂
Cr	0.02	1.97
Ni	0.04	0.05
Cu	0.08	2.16
Zn	1.29	24.05
As	< 0.05	<0.05
Se	4.56 _H	2.76 _H
Mo	1.87	20.49 _H
Cd	2.71 _H	0.14
Sb	ND*	ND*
Ba	1.86	6.94
Hg	0.13	0.18
Pb	6.14	5483.87 _H
Fluoride	71.40	65.80
Chloride	41300.00 _H	24100.00 _H
Sulphate	10000.00	16300.00
Test sample conditions		
Conductivity (µS/cm)	7780.00	8560.00
Temperature (°C)	19.40	19.80
pH	9.60	13.28

H = Hazardous material

*Not detected

Table 7. Release levels of control mortars according to UNE-EN 12457-4:2003.

Concentration (mg/kg dry matter)	CM ₁	CM ₂
Cr	0.11	0.10
Ni	0.03	0.03
Cu	0.01	ND*
Zn	0.07	0.05
As	0.05	0.05
Se	ND*	ND*
Mo	0.03	0.02
Cd	ND*	ND*
Sb	ND*	ND*
Ba	18.60	10.00
Hg	ND*	ND*
Pb	0.01	0.01
Fluoride	10.00	10.00
Chloride	41.60	50.00
Sulphate	51.60	55.80
Test sample conditions		
Conductivity (µS/cm)	5060.00	4270.00
Temperature (°C)	18.10	18.50
pH	13.00	12.90

*Not detected

Table 8. Release levels of S/S mortars according to UNE-EN 12457-4:2003 and Reduction Percentage of Granular Leaching (RPGL).

Waste (mg/kg) [×]	Se	Mo		Cd		Pb		Chloride		Test sample conditions			
										Conductivity (μ S/cm)	T ^a (°C)	pH	
EAFD ₁	4.56	1.87		2.71		6.14		41300		7780.00	19.40	9.60	
EAFD ₂	2.76	20.49		0.14		5483.87		24100		8560.00	19.80	13.28	
S/S mortars (mg/kg) ⁺	RPGL		RPGL		RPGL		RPGL		RPGL				
	%		%		%		%		%				
CM ₁₋₁	0.24	94.74	2.90	-	< 0.01	99.89	36.00	-	11900	71.19	3640	18.40	12.23
CM ₂₋₁	0.29	93.64	2.09	-	< 0.01	99.93	28.20	-	12400	69.98	3610	18.50	12.06
CM ₁₋₂	0.11	96.02	1.81	91.17	< 0.01	97.83	108.00	98.03	4900	79.67	6200	17.40	13.02
CM ₂₋₂	0.14	94.93	4.43	78.38	< 0.01	97.10	47.80	99.13	5600	76.76	4160	17.60	12.8

[×]LRV_w

⁺LRV_c

Table 9. Release levels of control mortars according to tank leaching test: XP X31-211:2012.

Concentration (mg/kg dry matter)	CM ₁	CM ₂
Cr	0.02	0.02
Ni	0.01	0.02
Cu	ND*	0.01
Zn	0.08	0.08
As	0.05	0.05
Se	ND*	ND*
Mo	0.02	0.01
Cd	ND*	ND*
Sb	ND*	ND*
Ba	2.01	0.61
Hg	ND*	ND*
Pb	0.02	0.01
Fluoride	10.00	10.00
Chloride	7.46	12.83
Sulphate	19.79	25.25
Test sample conditions		
Conductivity (µS/cm)	711.00	400.50
Temperature (°C)	20.70	20.00
pH	11.70	11.50

*Not detected

Table 10. Release levels of S/S mortars according to XP X31-211:2012.

Waste (mg/kg) [×]	Se	Mo	Cd	Pb	Chloride	Test sample conditions		
						Conductivity (μ S/cm)	T ^a (°C)	pH
EAFD ₁	4.56	1.87	2.71	6.14	41300	7780.00	19.40	9.60
EAFD ₂	2.76	20.49	0.14	5483.87	24100	8560.00	19.80	13.28
S/S mortars (mg/kg) ⁺								
CM ₁₋₁	0.22	1.53	< 0.01	1.11	11425	2865	24.50	11.44
CM ₂₋₁	0.24	1.29	< 0.01	2.82	11325	3065	25.45	11.82
CM ₁₋₂	0.10	0.50	< 0.01	14.05	3250	2850	19.20	12.57
CM ₂₋₂	0.10	0.87	< 0.01	16.70	3800	2280	21.40	12.38

[×]LRV_w

⁺LRV_m

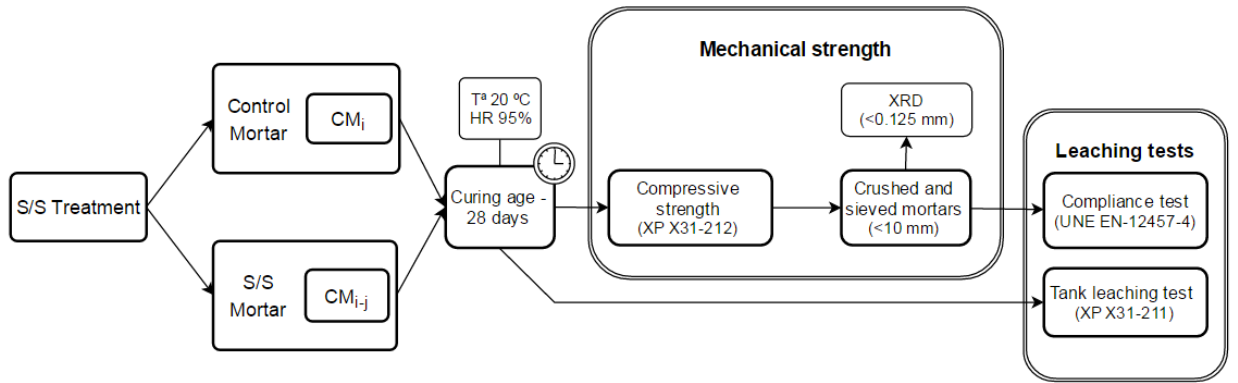


Fig. 1. Diagram of the experimental procedure.

Footnote: i: mortar type (1,2); j: waste type (1,2)

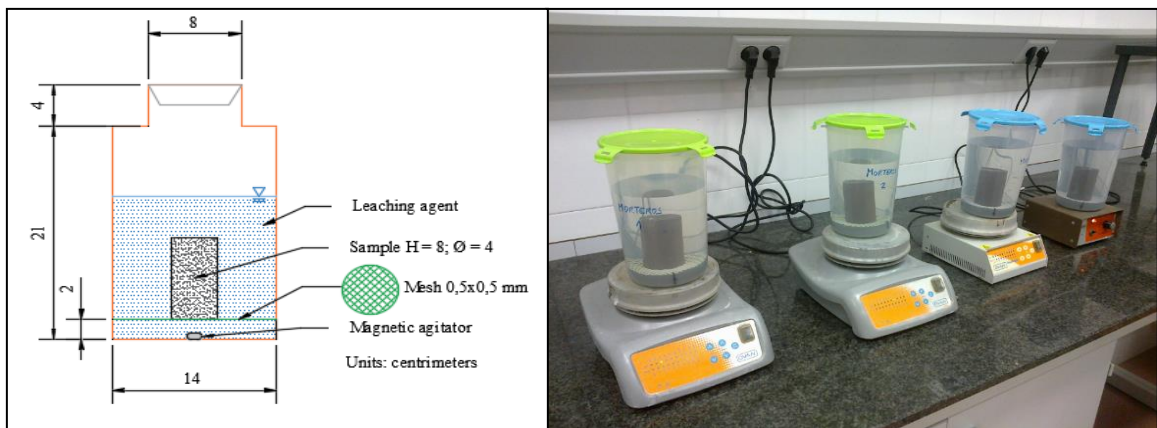


Fig. 2. Left: Tank leaching test scheme. Right: Tank leaching test laboratory.

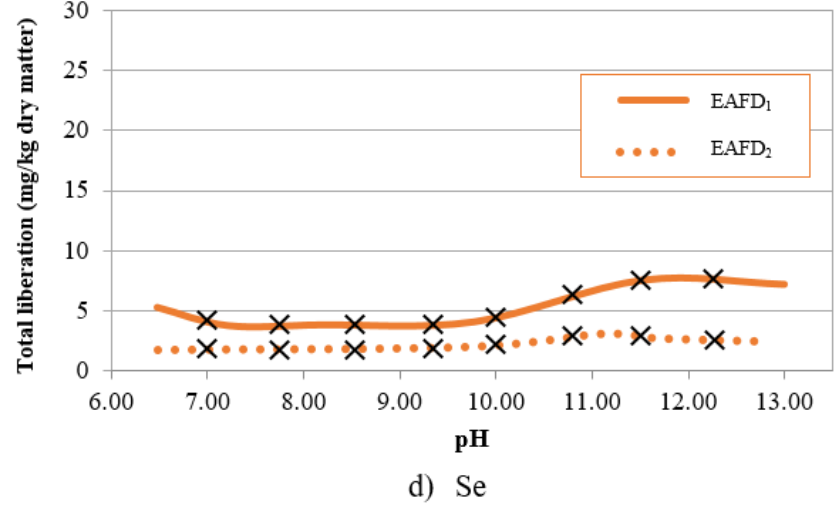
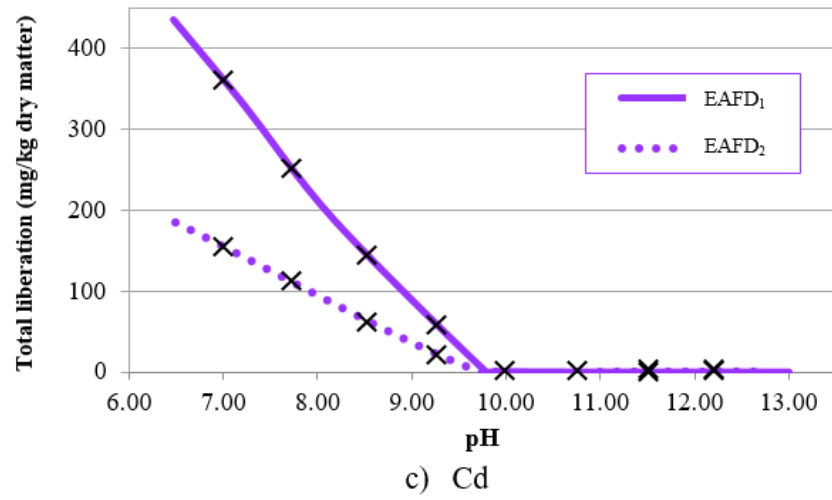
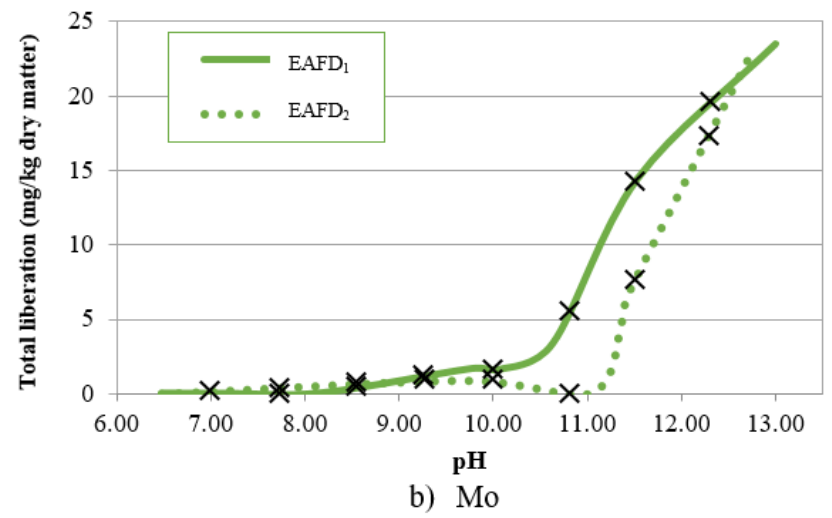
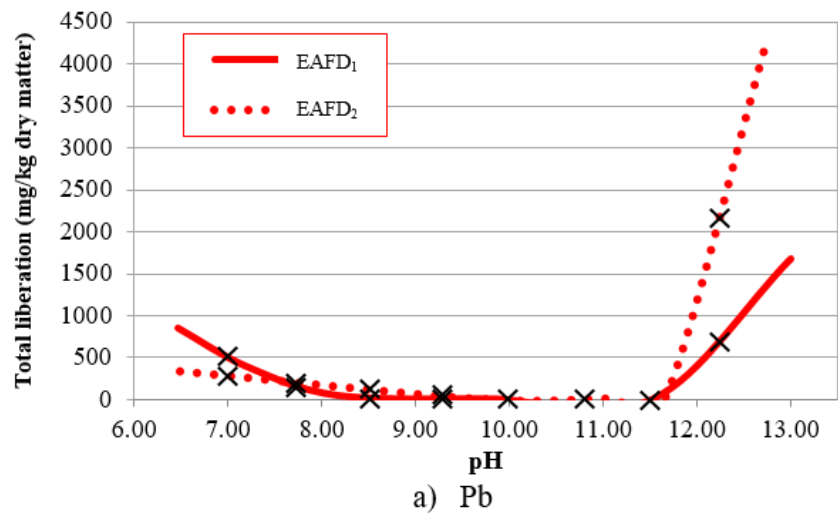


Fig. 3. Influence of pH on the release level of Pb, Mo, Cd and Se (UNE-EN 14429:2015)

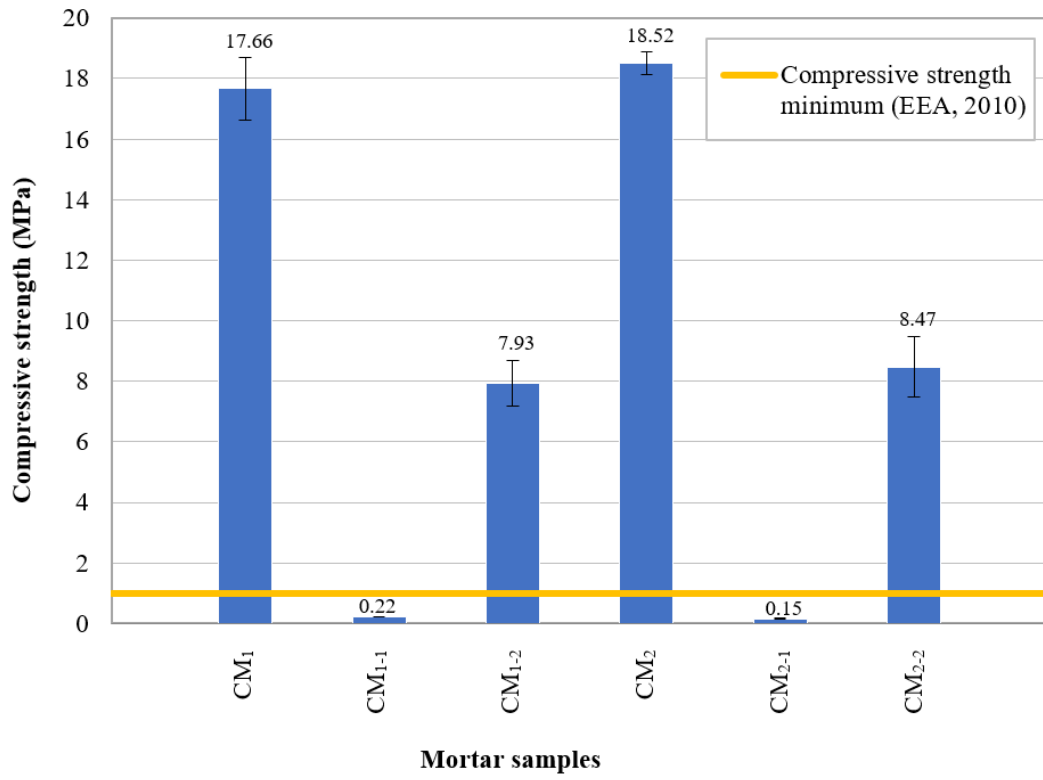


Fig. 4. Compressive strength of control and S/S mortars.

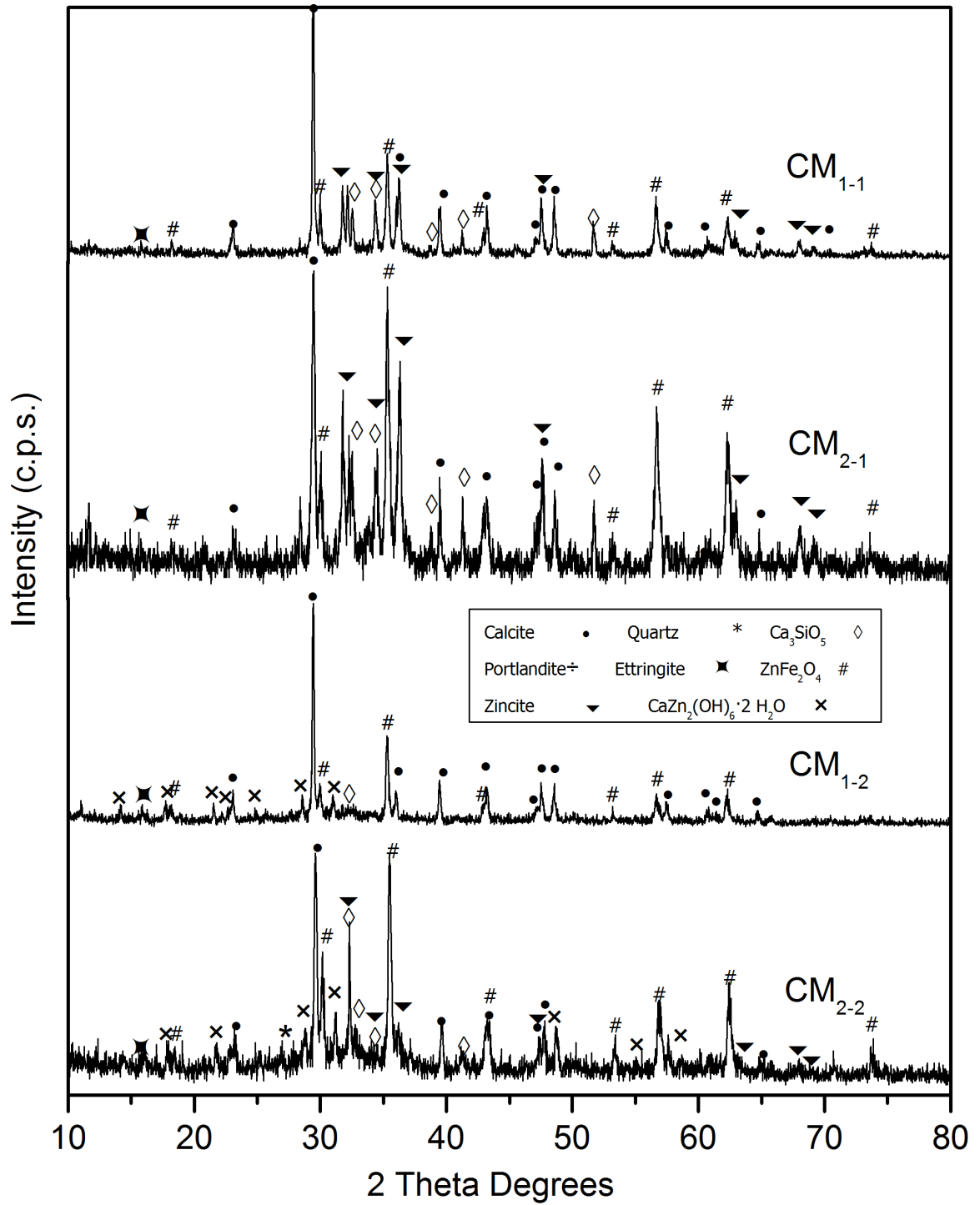


Fig. 5. XRD patterns of S/S mortars of EAFD₁ and EAFD₂.