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Additional Information

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4

5 **Abstract**  
6

7 Reusing ceramic waste as a pozzolanic admixture may offer environmental benefits as it  
8 allows the reduction in the consumption of natural resources and energy, and the reduction  
9 of CO<sub>2</sub> emissions associated with Portland cement (PC) production, while valorising waste  
10 materials with a long biodegradation period. This paper assessed the pozzolanic activity of  
11 three different ceramic waste types: red clay bricks (RCB), ceramic tiles (TCW) and ceramic  
12 sanitaryware (CSW). After adapting their particle size by crushing and milling, each was used  
13 to replace 0 to 50 wt.% PC (CEM I 42.5 R type). The milled powders were characterised by  
14 laser diffraction, field emission scanning electron microscopy (FESEM), X-ray fluorescence  
15 (XRF) and X-ray diffraction (XRD), and consistency and setting time tests were used to  
16 investigate the fresh behaviour of the ceramic waste/PC blended pastes. A basic  
17 sustainability analysis was performed, and the pozzolanic activity of RCB, TCW and CSW  
18 was assessed by compressive strength tests (performed in mortars cured at room  
19 temperature from 3 to 365 days) and microstructural analyses (thermogravimetry, XRD and  
20 FESEM performed in the pastes cured at 20°C for 28 and 90 days). The pozzolanic reaction  
21 of these waste materials improved with curing time, and all the mortars prepared with up to  
22 25 wt.% RCB, TCW or CSW met the mechanical requirements set out for coal fly ashes,  
23 whatever the ceramic waste type used. Among them, these results open up the possibility of  
24 partially replacing PC with the closest available ceramic waste, which would reduce the CO<sub>2</sub>  
25 emissions and economic cost deriving from transporting waste.  
26  
27

28 *Keywords:* Portland cement, ceramic waste, waste management, pozzolanic activity,  
29 compressive strength.  
30

31 **1. Introduction**

32  
33 Sustainable development, based on environmental conservation and responsible use of  
34 resources, is one of the biggest challenges faced today. Thus the construction industry in  
35 general, and cement production in particular, are essential contributors for a transition  
36 towards low carbon economy. Portland cement (PC) is a key construction material that is  
37 widely used worldwide. As reported by the European Cement Association based in Brussels  
38 [1], it is estimated that 4.1 billion tonnes of cement were globally produced in 2017, and  
39 China was the leader producer (56.5% of global production), followed by India (6.8%) and  
40 CEMBUREAU members (6.3%) [1]. As explained by Kajaste and Hurme [2], PC production is  
41 one of the world's highest CO<sub>2</sub>-emitting processes and is responsible for around 5-8% of the  
42 carbon generated worldwide. Approximately 0.75 kg of carbon dioxide per kg of produced  
43 cement is emitted to the atmosphere, which come mainly from limestone (CaCO<sub>3</sub>)  
44 decomposition and the combustion of the fuel required to reach clinkering temperatures  
45 (≈1,450°C) [3-5]. As Imbabi et al. [6] stated, with no changes in the current production rate  
46 and manufacturing processes, it is estimated that CO<sub>2</sub> emissions will have approximately  
47 multiplied those registered in 1990 by 5 by 2050. These predictions were conservative  
48 because, although less than 2 billion tonnes of CO<sub>2</sub> were expected to be emitted to the  
49 atmosphere in 2010, 3.24 billion tonnes were registered in 2012. Thus the increasing use of  
50 PC demands a real urgent change in order to develop sustainable, durable and cost-effective  
51 binders. Both the cement industry and scientific community are actively seeking alternatives  
52 to reduce emissions which, in 2018, had dropped by almost 14% since 1990. The 5C  
53 programme developed by CEMBUREAU compiles the widest possible set of alternatives to  
54 lower the carbon emissions released by the cement industry. The different proposals are  
55 classified in five categories: Clinker, Cement, Concrete, Construction and Carbonation, of  
56 which the last refers to CO<sub>2</sub> capture technologies. Imbabi et al. [6] also summarised four  
57 different approaches that have been developed mainly to minimise CO<sub>2</sub> intensity and to  
58 improve energy efficiency associated with cement production: a) improve manufacturing  
59 processes; b) co-incineration of waste materials during cement production; c) use of  
60 Supplementary Cementitious Materials (SCMs) in cement; d) develop novel resource-  
61 efficient cements. Of these, SCMs have been widely used as fillers or pozzolans, and the  
62 latter implies their reaction with the Ca(OH)<sub>2</sub> released during PC hydration to provide  
63 cementitious products [7,8]. One of the materials satisfactorily used to partially replace PC is  
64 ceramic waste [9-14], which is durable, hard and very resistant to physical, biological and  
65 chemical degradation. As previously explained in [15], large amounts of ceramic waste are  
66 generated yearly from demolition practices or production defects, and dumping it in landfills  
67 leads to environmental problems due to dust pollution and the occupation of vast expanses  
68 of land. Thus reusing ceramic waste materials in PC contributes to both diminish the

69 potential environmental impacts caused by landfill deposits and reduce the use of energy,  
70 natural resources and greenhouse gas emissions associated with PC production.

71  
72 As Ay and Unal [12] or Turanli [16] explained, although clay minerals in their original state do  
73 not present pozzolanic reactivity, they may acquire it when thermally treated given the  
74 destruction of the crystal structure of clay and the formation of amorphous or disordered  
75 aluminosilicate phases. The changes that occur in ceramic materials during the firing process  
76 have been described by Mohammed [3]: the removal of free water from clay (dehydration)  
77 occurs from 20°C to 200°C, and is followed by the dehydroxylation process that consists in  
78 loss of the chemically combined water from the OH groups contained in clay minerals; during  
79 dehydroxylation, the crystalline structure of clay constituents collapses, which results in  
80 unstable silica and alumina with an amorphous or disordered lattice structure. Decarbonation  
81 (release of CO<sub>2</sub>) of compounds, such as calcite or dolomite, occurs within the 700-900°C  
82 range; finally, exothermic recrystallisation takes place at temperatures above 900°C, which  
83 may result in the formation of new thermodynamically stable phases that can reduce the  
84 pozzolanic activity of ceramic materials [3,17]. The study by Baronio and Binda [17], in which  
85 clays were calcined up to 1,000°C, observed that their pozzolanic activity improved when  
86 calcined at temperatures between 600°C and 900°C (depending on the type and amount of  
87 clay used), but diminished when calcined at more than 900°C. However, the study by Turanli  
88 et al. [16] reported slightly better relative strength values (compared to the reference mortar)  
89 when replacing 20 wt.% PC with slab bricks (SB) sintered at 1,000-1,100°C than when using  
90 the same amount of wall bricks (WB) fired at 800-900°C, (86% and 78% strength activity  
91 indices were recorded in the SB- and WB-containing mortars cured for 28 days,  
92 respectively). The review by Mohammed et al. [3], who analysed the pozzolanic behaviour of  
93 clays, clay waste and ceramic products like bricks or tiles, concluded that the reactivity of  
94 calcined clays depended mainly on the degree of dehydroxylation, fineness of particles, and  
95 their SiO<sub>2</sub> + Al<sub>2</sub>O<sub>3</sub> + Fe<sub>2</sub>O<sub>3</sub> content. Binici 2012 [8] and Sánchez de Rojas et al. [13] stated  
96 that the optimum dehydroxylation temperature and time to break the crystalline structure of  
97 clay depended on clay mineral. Accordingly, as the production of ceramic materials is guided  
98 to obtain optimal properties for a specific use (hollow or load-bearing bricks, wall or floor tiles,  
99 sanitaryware units, etc.), sintering temperatures will either presumably differ from the  
100 optimum ones considered for complete clay minerals dehydroxylation or probably lead to the  
101 formation of new crystalline stable phases [3,12,17], which is expected to reduce the  
102 pozzolanic activity of ceramic products. Nonetheless, several studies [9-14] have  
103 successfully proved the pozzolanicity of ceramic waste materials when blended with PC,  
104 along with the environmental benefits that this implies. Puertas et al. [10] evaluated the  
105 pozzolanic activity of six different ceramic tile types. They concluded that they all exhibited  
106 pozzolanic behaviour after 15 curing days, which was strongly influenced by their chemical

107 composition (high silica and alumina contents) and amorphous content (close to 35 wt.% in  
108 three tile types). The studies by Mas et al. [11] and Ay and Unal [12], who used ceramic tiles  
109 as pozzolanic admixtures in PC systems, found that mortars containing up to 35 wt.%  
110 ceramic waste met both the requirements set out in fly ash regulations [11] and those stated  
111 for the cement standard for calcined natural pozzolans (ASTM C 618) [12], respectively.  
112 Sánchez de Rojas et al. [13] also noticed some contribution to compressive strength in PC  
113 mortars cured for 28 days at room temperature when replacing up to 15 wt.% PC with clay  
114 roof tiles. Pereira-de-Oliveira 2012 compared the pozzolanicity of glass, bricks and tiles (10-  
115 40 wt.% PC replacement), and corroborated that it was strongly influenced by calcination  
116 temperature because no pozzolanic behaviour was observed when using brick waste.  
117 However, Toledo-Filho et al. [9] observed a minor influence on the mechanical properties of  
118 mortars when replacing 20 wt.% PC with clay bricks calcined at 850°C.

119  
120 All these studies have successfully proved the viability of employing ceramic materials as  
121 pozzolanic admixtures in PC systems, and have consequently encouraged the utilisation of  
122 blended cements with lower clinker contents, moving towards the use of more sustainable  
123 construction materials. However, only some studies have compared the pozzolanic activity of  
124 different ceramic waste types under the same study conditions. Additionally, as the reactivity  
125 of ceramic materials depends mainly on their sintering process, unexpected disadvantages  
126 may appear when using this waste to replace PC, such as less early strength, longer setting  
127 times or delayed cement hydration. Therefore, this research aims to compare the pozzolanic  
128 activity of red clay bricks (RCB), tiles ceramic waste (TCW) and ceramic sanitary-ware  
129 (CSW), and to provide further information on ceramic waste/PC blended binders.

130

131

## 132 **2. Materials and Methods**

133

### 134 *2.1. Materials*

135

136 Figure 1 shows the three herein used ceramic waste materials. TCW was composed of  
137 different ceramic tile types (porcelain stoneware, floor and wall tiles) produced by ceramic  
138 companies located in the province of Castellón (Spain). This waste was supplied as granular  
139 material and its particle size fell within the 4-16 mm range. The RCB and CSW pieces were  
140 collected from dumps filled with construction waste. Portland cement (PC) type CEM I 42.5R,  
141 which complied with the Spanish Cement Reception Instruction [18], was used to assess the  
142 pozzolanic activity of these ceramic waste materials. Mortars were prepared using siliceous  
143 sand with a maximum particle size of 2 mm and a fineness modulus of 2.74.

144



Figure 1. The original RCB, TCW and CSW ceramic waste materials

## 2.2. Ceramic waste preparation and characterisation

RCB and CSW were broken with a hammer. The different ceramic materials were crushed in a jaw crusher (BB200 Retsch) until a particle size under 2 mm was achieved. Crushed particles were milled in alumina media (Orto-Alresa ball mill) to obtain a powder with a fineness close to that of PC. To this end, two jars containing 1,100 g of ceramic waste and 165 balls of alumina, with diameters ranging from 15 to 40 mm (total weight of alumina balls was 4,860 g), were used. The milling time was optimised for each ceramic material to obtain similar granulometric distributions in the shortest grinding time (6 h for the RCB and TCW; and 8 h for the CSW).

The granulometric distribution of the milled powders was determined in a Mastersizer 2000 (Malvern instruments) and their morphology was observed under a field emission scanning electron microscope (FESEM) ZEISS Supra 55, with a working distance of 6-8 mm for the X-ray microanalysis (EDS). The chemical composition of RCB, TCW and CSW was determined by X-ray fluorescence (XRF, Philips Magix Pro spectrometer), and their amorphous content was evaluated following UNE EN 196-2 specifications. The crystalline phases were identified by X-ray diffraction (XRD) in a Brucker AXS D8 Advance using Cu K $\alpha$  radiation at 20 mA and 40 kV from 10° to 70° 2 $\theta$  degrees.

## 2.3. Mortar sample preparation

The compressive strength evolution with increasing waste contents was assessed in mortars. The three ceramic waste materials (RCB, TCW and CSW) were used to replace 0 to 50 wt.% PC (CEM I 42.5R), and a binder:sand:water weight ratio of 1:3:0.5 was used. These samples were produced according to Standard UNE EN 196-1:2005. They were cured in a temperature- and humidity-controlled chamber (20°C and 95%) for up to 365 days. After curing for 24 h, samples were demoulded and then immersed in water with calcium hydroxide until the testing age. The process variables herein used, together with the designations of samples, are summarised in Table 1.

178

179 Table 1. Process variables of the developed RCB, TCW and CSW PC blended samples

Ceramic waste	Designation	binder:sand:water weight ratio	PC replacement, wt.%	Curing temperature, °C	Curing age, days
-	REF		0		
Red clay brick	RCB15		15		
	RCB25		25		
	RCB35		35		
	RCB50		50		
Tile ceramic waste	TCW15	1:3:0.5	15	20	3, 7, 28, 90, 180 and 365
	TCW25		25		
	TCW35		35		
	TCW50		50		
Ceramic sanitaryware	CSW15		15		
	CSW25		25		
	CSW35		35		
	CSW50		50		

180

181 *2.4. Fresh behaviour and mechanical properties of the ceramic waste/PC blended binders*

182

183 Variation in the consistency and setting time with the different amounts of RCB, TCW and  
184 CSW were investigated in pastes according to Standard UNE EN 196-3:2005. All these  
185 pastes were prepared with a constant amount of water (145 g) and, while the reference paste  
186 contained 500 g of PC, they were partially replaced with the different percentages (15 to 50  
187 wt.%) of each ceramic waste type (RCB, TCW and CSW). The compressive strength  
188 evolution with the distinct ceramic waste materials and contents was assessed in mortars  
189 following Standard UNE EN 196-1:2005 and using a MEH-3000 PT/W by Ibertest. Additional  
190 data on the pozzolanic reactivity of RCB, TCW and CSW was provided by the determination  
191 of the strength activity index (SAI) and the strength gain (SG) values. SAI is the relative  
192 strength between the pozzolanic (15 to 50 wt.% PC replacement) and the reference (0 wt.%  
193 ceramic waste) mortars, and the SG attributed to the pozzolanic contribution was calculated  
194 according to Eq. (1).

195

$$196 \quad SG(\%) = \frac{S_{POZ} - (S_{REF} \cdot PC\%)}{S_{REF} \cdot PC\%} \cdot 100 \quad (1)$$

197

198 where:

199  $S_{POZ}$  = compressive strength of the mortar with the ceramic waste (RCB, TCW or CSW);200  $S_{REF}$  = compressive strength of the reference mortar;

201 PC% = percentage of PC in the mortar containing the ceramic waste (per unit).

202 *2.5. Microstructural characterisation of the ceramic waste/PC blended binders*

203  
204 The reference paste (100 wt.% PC) and those containing 25 wt.% RCB, TCW or CSW, cured  
205 at room temperature for 28 and 90 days, were used to assess the microstructural evolution of  
206 the ceramic waste/PC blended cements. Crystalline phases were identified by X-ray  
207 diffractometry (XRD), which were run in a Brucker AXS D4 Endeavor powder diffractometer  
208 equipped with a Lynxeye detector, from 5 to 70 2 $\theta$  degrees, at 40 kV and 40 mA, with Cu K $\alpha$   
209 radiation. The thermogravimetric analyses (TG) performed aimed to assess the compounds  
210 formed after the hydration of the pastes and to determine the percentage of fixed lime. This  
211 value provided information on PC hydration and pozzolanic reactions, and it was determined  
212 as previously explained in [11]. TG tests were run in a Mettler Toledo TGA/DSC3, using  
213 closed aluminium crucibles (100  $\mu$ l) with a perforated lid. These tests were conducted from  
214 35°C to 600°C at a heating rate of 10°C/min in nitrogen atmosphere. In order to confirm the  
215 presence of the compounds previously identified by XRD and TG, and examine the density  
216 of the developed pastes, their microstructures were observed by FESEM, using the  
217 equipment previously described in Section 2.2.

218

219 *2.6. Sustainability analysis*

220  
221 To estimate the environmental impact of reusing RCB, TCW or CSW as a partial  
222 replacement of PC, a basic sustainability analysis was conducted. A gate-to-gate life cycle  
223 analysis approach was followed, which contemplated only the CO<sub>2</sub> emissions associated with  
224 binder production. This simplified calculation contemplated neither the transport of the  
225 materials from their origin to the laboratory, nor the mixing or curing of pastes and mortars, or  
226 other mortar life cycle stages, such as maintenance or demolition. To calculate the embodied  
227 specific energy (e-energy) and embodied specific CO<sub>2</sub> (e-CO<sub>2</sub>) emissions of the ceramic  
228 waste/PC blended cements, the e-energy and e-CO<sub>2</sub> values previously reported for CEM I  
229 42.5 (5,500 MJ/ton [19] and 750 kg CO<sub>2</sub>/ton [5], respectively) were used. Moreover the  
230 percentage of PC replacement, and the energy and CO<sub>2</sub> emissions generated when crushing  
231 and milling ceramic waste were considered. The jaw crusher BB200 Retsch, employed to  
232 crush the ceramic waste materials, is driven by a 1,4914 kW power unit, and it took 2  
233 minutes and 30 seconds to crush 1 kg of ceramic waste. The Orto-Alresa ball mill used to  
234 reduce the size of waste was equipped with a 150 W power unit, and two jars containing 1.1  
235 kg of crushed ceramic particles were employed in each batch. An emission factor of 0.241 kg  
236 CO<sub>2</sub> eq/kWh was utilised to convert the used energy into CO<sub>2</sub> emissions (data obtained from  
237 the Spanish Ministry for the Ecological Transition and Demographic Challenge for 2019 [20]).  
238



239 As previously proposed by Xiao et al. [21], in order to take into account the 28-day  
 240 compressive strength of the developed binders in the sustainability analysis, two additional  
 241 factors were calculated,  $E_s$  and  $C_s$ , determined according to Eq. (2) and (3), respectively.

$$243 E_s = \frac{e\text{-energy}}{S_{\text{POZ,28d}}} \quad (2)$$

$$245 C_s = \frac{e\text{-CO}_2}{S_{\text{POZ,28d}}} \quad (3)$$

246 where:

247  $E_s$  = Embodied energy in relation to the 28-day compressive strength of the pozzolanic  
 248 mortar, MJ/ton · MPa;

249  $C_s$  = Embodied CO<sub>2</sub> in relation to the 28-day compressive strength of the pozzolanic mortar,  
 250 kg CO<sub>2</sub>/ton · MPa

251  $S_{\text{POZ,28d}}$  = compressive strength of the pozzolanic mortar cured for 28 days, MPa.

### 254 3. Results and Discussion

#### 256 3.1. Ceramic waste material characterisation

257  
 258 The milling time selected for each ceramic waste, together with the granulometric parameters  
 259 obtained after the milling process, are summarised in Table 2. All the powders presented a  
 260 close granulometric distribution and, although CSW particles were slightly larger than RCB or  
 261 TCW, they all had a mean diameter that came close to 20 µm, 90 vol.% of particles below 56  
 262 µm and 10 wt.% vol. under 1.62 µm.

264 Table 2. Milling time and granulometric parameters of the ceramic milled powders

Ceramic waste	Milling time, h	Mean diameter, µm	$d_{10}$ µm	$d_{50}$ µm	$d_{90}$ µm
RCB	6	19.87	1.31	11.24	52.31
TCW	6	19.67	1.60	14.01	46.81
CSW	8	22.26	1.62	14.09	55.88

265  
 266 Table 3 shows the chemical composition and amorphous content of the three ceramic  
 267 wastes herein employed. All the ceramic materials were composed mainly of SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>,  
 268 with bigger quantities in CSW (the sum of 89.6%) compared to those recorded for TCW and  
 269 RCB (the sum of 79.8% and 66.5%, respectively). RCB and TCW presented relatively high  
 270 Fe<sub>2</sub>O<sub>3</sub> contents, and the sum of SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub> in them all was above 70%, which  
 271 Mohammed et al. [3] pointed out to be an important parameter that promotes pozzolanic

272 activity. Amorphous content varied from 35 to 60 wt.% (RCB and TCW, respectively), and the  
 273 amount of vitreous phases recorded for TCW and CSW fell within the range previously  
 274 reported by Zanelli et al. [22] for porcelain stoneware tiles (40-75 wt.%, a range obtained  
 275 after analysing 93 different samples).

276  
 277  
 278

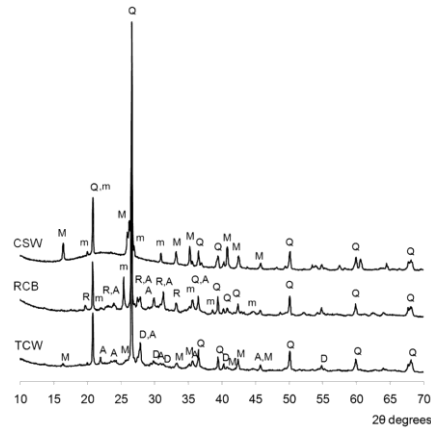
Table 3. Chemical composition of RCB, TCW and CSW ceramic wastes, wt.%

Ceramic waste	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	CaO	MgO	K <sub>2</sub> O	Fe <sub>2</sub> O <sub>3</sub>	SO <sub>3</sub>	LOI*	Other	Amorph.
RCB	49.9	16.6	9.7	5.5	4.4	6.5	3.3	2.4	1.7	35
TCW	61.2	18.6	5.8	1.8	3.3	5.0	0.09	0.7	3.5	60
CSW	66.0	23.6	1.2	0.7	2.9	1.3	0.07	0.3	4.0	46

279 \*At 1,000°C

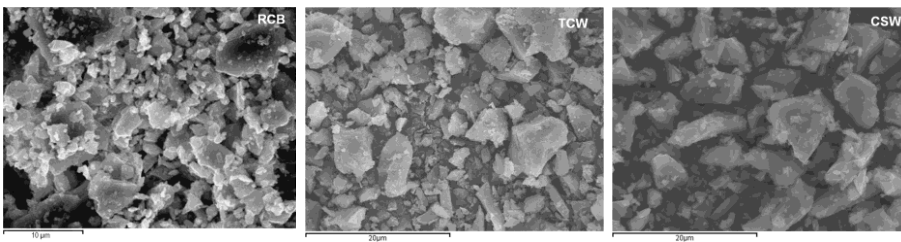
280

281 The mineralogical composition of RCB, TCW and CSW is presented in Figure 2. All the  
 282 ceramic materials showed signals due to quartz (Q, SiO<sub>2</sub>; PDFcard 331161), while mullite (M,  
 283 Al<sub>6</sub>Si<sub>2</sub>O<sub>13</sub>; PDFcard 150776) was identified only within the spectra of TCW and CSW (bigger  
 284 amounts in CSW, denoted by higher-intensity signals). This was attributed to the lower  
 285 sintering temperatures applied to produce RCB (usually fired up to approx. 900°C) [23]  
 286 compared to those used to make ceramic tiles, whose peak temperatures usually vary within  
 287 the 1,120–1,220°C range [11], or CSW, whose plateau in the heating curve generally falls  
 288 within the 1,200-1,280°C range [24]. Small amounts of sodium feldspar albite (A, NaAlSi<sub>3</sub>O<sub>8</sub>;  
 289 PDFcard 190926) were distinguished in the TCW and RCB diffractograms, and signals due  
 290 to microcline (m, KAlSi<sub>3</sub>O<sub>8</sub>, PDFcard090466) appeared in the RCB and CSW patterns.  
 291 Traces of diopside (D, CaMg(SiO<sub>3</sub>)<sub>2</sub>; PDFcard 190239) and rankinite (R, 2SiO<sub>2</sub>·3CaO,  
 292 PDFcard220539) were also identified in TCW and RCB, respectively. These crystalline  
 293 phases typically form in ceramic materials, and have also been distinguished in previous  
 294 studies, where TCW was used to partially replace PC [11], and RCB or CSW were utilised as  
 295 precursors in alkali-activated binders [25,26]. The deviation from the baseline observed  
 296 within the 15-30° 2θ degree range corroborates the presence of amorphous phases in all the  
 297 ceramic waste materials, as previously quantified in Table 3. In the RCB waste these  
 298 disordered phases are explained by the dehydroxylation of the clay [3,23], while in the TCW  
 299 and CSW their presence is attributed to the formation of new vitreous phases from the  
 300 melting of feldspars which, according to Zanelli et al. [22], occurs from approximately  
 301 1050°C.



302  
 303 Figure 2. X-ray diffractogram of the ceramic waste materials. Q: Quartz ( $\text{SiO}_2$ ); M: Mullite ( $\text{Al}_6\text{Si}_2\text{O}_{13}$ );  
 304 A: Albite ( $\text{NaAlSi}_3\text{O}_8$ ); m: Microcline ( $\text{KAlSi}_3\text{O}_8$ ); D: Diopside ( $\text{CaMg}(\text{SiO}_3)_2$ ); R: Rankinite  
 305 ( $2\text{SiO}_2 \cdot 3\text{CaO}$ )  
 306

307 The micrographs of the milled ceramic waste materials (see Figure 3) display irregular  
 308 particles with a smooth surface. No significant differences were observed among them.  
 309

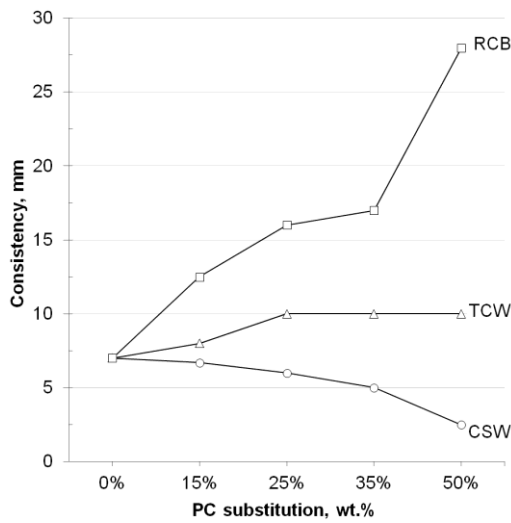


310  
 311 Figure 3. Field emission scanning electron microscope images of the milled RCB, TCW and CSW  
 312 particles  
 313

### 314 3.2. Consistency

315  
 316 Variation in consistency, reported as the distance between the lower side of the 1 cm-  
 317 diameter plunger and the bottom of Vicat's apparatus, depending on the different ceramic  
 318 waste types and contents, is plotted in Figure 4. Minor variations in consistency were  
 319 observed when replacing PC with TCW, and the results slightly lowered (better workability)  
 320 with increasing CSW additions, and significantly increased with RCB content, which denotes  
 321 loss of workability in the RCB/PC blended systems. As all the herein used milled ceramic  
 322 particles presented a similar morphology (Fig. 3), the obtained results were attributed mainly  
 323 to water absorption differences. The results agree with those previously reported by Pitarch

324 et al. [27], who employed the same RCB, TCW and CSW waste materials to replace natural  
 325 aggregates in structural concrete. In their study [27], water absorption values of 15.76 wt.%,  
 326 6.28 wt.% and 0.69 wt.% were recorded for the RCB, TCW and CSW particles (up to 4 mm  
 327 in size, used as recycled sand), respectively. Our results also agree with those previously  
 328 reported by Mas et al. [11] in PC/TCW blended systems, whose consistency values  
 329 (determined by the flow table test) varied within a narrow range (150-160 mm) in mortars  
 330 containing up to 50 wt.% TCW. The authors [11] attributed these minor variations to the low  
 331 porosity and smooth surface of the tile ceramic particles.  
 332

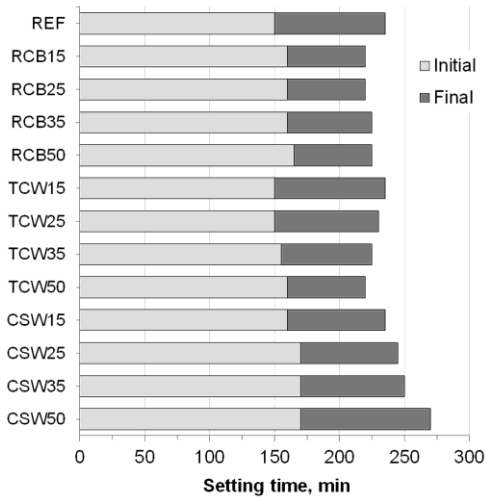


333  
 334 Figure 4. Consistency of the blended RCB, TCW and CSW PC pastes  
 335

### 336 3.3. Setting time

337  
 338 The evolution of the initial and final setting times (IST and FST, respectively) with the  
 339 different ceramic waste types and contents is shown in Figure 5. Minor variations in both IST  
 340 and FST were generally observed, and the largest differences were recorded in the PC/CSW  
 341 system. Replacement of PC with RCB, TCW or CSW slightly prolonged the time when  
 342 pastes started losing their plasticity (IST), and the maximum variation was recorded in the  
 343 pastes containing 25 wt.% CSW, or more (170 min, 20 min later than the REF paste). The  
 344 slightly prolonged IST facilitates the transportation, placing and compaction operations of the  
 345 concrete prepared with these blended cements. Although the FST lowered by 10-15 minutes  
 346 in the RCB samples (220-225 min vs. the 235 min recorded for the REF paste), it hardly  
 347 varied with the amount of RCB. The FST became progressively shorter with increasing  
 348 amounts of TCW (up to 220 min with 50 wt.%) and, contrarily, gradually prolonged with CSW

349 content (up to 270 min with 50 wt.%). This implied that the time in which the sample  
 350 remained under plastic condition, required for handling the paste, mortar or concrete  
 351 samples, hardly varied in the RCB/PC samples, became progressively shorter in the TCW-  
 352 blended pastes (from 85 min with 15 wt.% TCW to 60 min with 50 wt.% TCW) and prolonged  
 353 in those prepared with CSW (from 75 min with 15 wt.% to 100 min with 50 wt.%). The results  
 354 of the PC/CSW blended systems coincided with those previously reported by Medina et al.  
 355 [28], who observed that partially replacing PC with CSW (10 wt.% or 20 wt.%) modified the  
 356 rheology of pastes and retarded their hydration. The RCB/PC system results came close to  
 357 those previously reported by Naceri et al. [29], who observed shorter IST and FST in the  
 358 cements developed by partially replacing clinker with brick ceramic waste (up to 20 wt.%).  
 359 However, differences in the results herein obtained (the IST was slightly longer in the  
 360 RCB/PC systems) were attributed mainly to the increasing gypsum-to-clinker ratios used by  
 361 Naceri et al. [29] with higher ceramic contents, because those authors mixed a set amount of  
 362 gypsum no matter what the clinker replacement was.



363  
 364 Figure 5. Setting time of the blended RCB, TCW and CSW PC pastes

365

366 **3.4. Compressive strength of pozzolanic mortars**

367

368 The evolution of compressive strength (MPa), strength activity index (SAI, %) and strength  
 369 gain (SG, %) with the different study parameters (curing age, types and amount of ceramic  
 370 waste used) are plotted in Figures 6 to 8. To correlate the strength parameters with curing  
 371 age, simple linear equations were established according to Equation (4) for each ceramic  
 372 waste and PC replacement percentage.

373

374  $S_{PARAMETER} = a \cdot \ln(t) + b$  (4)

375

376 Where:

377  $S_{PARAMETER}$  = Strength parameter: compressive strength (MPa), SG (%) or SAI (%);

378  $t$  = curing age, days

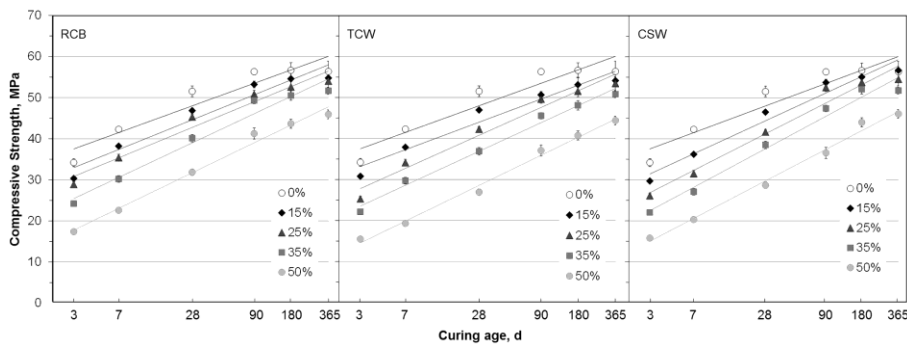
379  $\ln$  = natural (based-e) logarithm

380  $a$  and  $b$  = constants for each ceramic waste type and PC replacement percentage

381

382 As Figure 6 shows, similar compressive strength results were obtained for a given  
 383 replacement percentage, no matter what ceramic waste was used. Given that the three  
 384 ceramic waste materials herein used had a close particle size distribution and similar  $\text{SiO}_2 +$   
 385  $\text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$  contents (Section 3.1), these results are mainly attributed to the pozzolanic  
 386 reactivity of the amorphous phases formed during their production processes which, as  
 387 explained in Section 3.1, derive from the dehydroxylation of the clay (RCB) and the melting  
 388 of feldspars (TCW, CSW). Although the strength values recorded at short curing ages (3 or 7  
 389 days) significantly lowered compared to the reference mortar, they came closer with the  
 390 curing time, especially in the samples containing up to 25 wt.% ceramic waste (close to 50  
 391 MPa after 90 curing days).

392



393

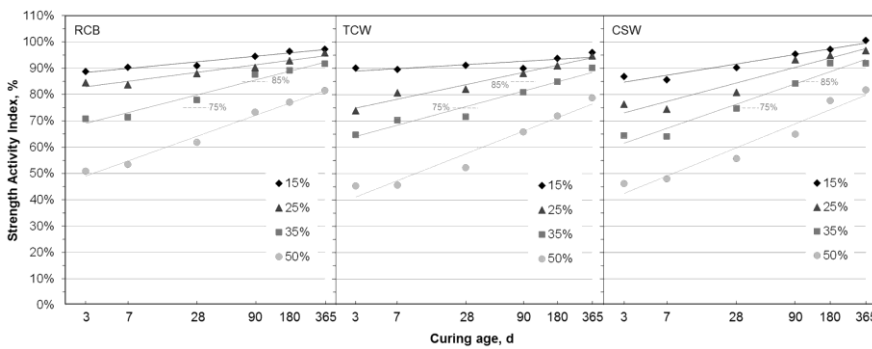
394 Figure 6. Compressive strength of the mortars prepared with 0 to 50 wt.% RCB, TCW or CSW, cured  
 395 at 20°C from 3 to 365 days

396

397 As plotted in Figure 7, all the mortars blended with up to 25 wt.% ceramic waste met the  
 398 requirements set out in UNE EN 450-1 for fly ash [30], and their SAI values were over 75%  
 399 and 85% after 28 and 90 curing days, respectively. The 35 wt.% RCB mortars met the  
 400 standard requirements; the 35 wt.% CSW mortars were at the limit and up to 25 wt.% TCW  
 401 could be used according to these specifications. In addition to the pozzolanic effect, the  
 402 slightly better results obtained when using RCB are also attributed to its higher water

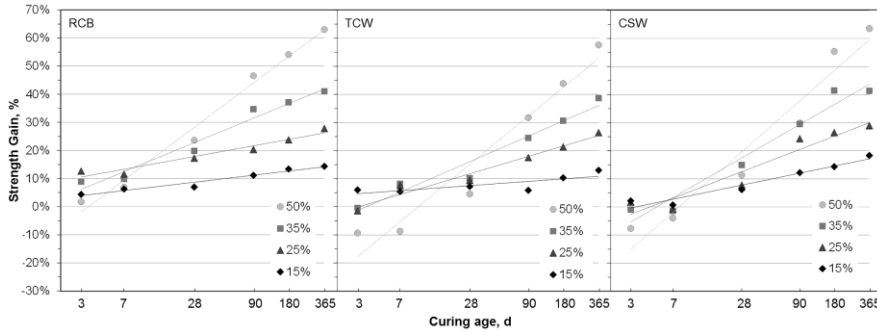
403 absorption when compared with that registered for TCW and CSW (15.76%, 6.28% and  
 404 0.69%, respectively [27]). The relatively high RCB water absorption progressively reduced  
 405 the workability of the blended cements with increasing RCB contents and also diminished the  
 406 effective water (that available to react with cement, determined as the total amount of water  
 407 minus that absorbed by the ceramic waste particles). Thus, the reduction in the water to  
 408 cement ratio, as widely known, improves strength and durability. However, the contribution  
 409 on strength of these ceramic waste-based materials improved with curing time and all the  
 410 ceramic waste/PC blended mortars presented similar long-term strength: all the mortars  
 411 blended with 35 wt.% ceramic waste had SAI indices over 90% after being cured for 1 year.  
 412 In other words, the strength of the mortars prepared with 65 wt.% PC reduced by only 10%  
 413 after 365 curing days respect to the reference mortar (100% PC as binder). This means that  
 414 these blended mortars are an interesting alternative that contributes to sustainable  
 415 development in applications that do not require high strength at short curing times.

416  
 417



418  
 419 Figure 7. Strength activity index of the mortars prepared with 0 to 50 wt.% RCB, TCW or CSW, cured  
 420 at 20°C from 3 to 365 days  
 421

422 The strength gain (SG) results reported in Figure 8 show that RCB contributed some strength  
 423 to the system with short curing ages (3 and 7 days). Although TCW and CSW presented  
 424 some negative SG values after 3 and 7 curing days, they became positive with curing time,  
 425 especially with increasing waste contents. In agreement with the compressive strength and  
 426 SAI results, similar SG values were recorded after 365 curing days for a given replacement  
 427 percentage whatever the ceramic waste material used.



428  
429 Figure 8. Strength gain of the mortars prepared with 0 to 50 wt.% RCB, TCW or CSW, cured at 20°C  
430 from 3 to 365 days

431 The regression data for the different ceramic waste materials and replacement percentages  
432 are presented in Table 4. Constant 'a' illustrates the slope of the linear regression, while 'b'  
433 indicates the intercept of the strength parameter (compressive strength, SAI or SG) with the  
434 Y axis. The generally obtained high coefficient of determination ( $R^2$  typically higher than 0.94)  
435 denotes good simple linear regressions between strength parameters and the based-e  
436 logarithm of curing age. The slope of the linear regressions generally increased with the  
437 replacement percentage, which indicates a stronger influence with curing time with larger  
438 amounts of waste. Contrarily, the b-intercept values lowered with increasing ceramic  
439 contents, which agrees with the lower strength results and activity indices observed at short  
440 curing times. The positive b-intercept SG values obtained with up to 25 wt.% RCB confirmed  
441 some contribution of this waste at short curing ages.

442  
443 Table 4. Linear regression data for the compressive strength, SG and SAI of RCB, TCW and CSW PC  
444 blended mortars

Strength	Regression	RCB					TCW					CSW				
		PC replacement, wt.%					PC replacement, wt.%					PC replacement, wt.%				
		0	15	25	35	50	0	15	25	35	50	0	15	25	35	50
$\sigma_c$ , MPa	a	4.69	5.21	5.34	6.07	6.25	4.69	4.84	5.82	5.98	6.29	4.69	5.81	6.43	6.77	6.59
	b	32.38	27.25	24.97	18.79	10.91	32.38	27.87	21.42	16.98	7.63	32.38	25.05	19.79	15.01	7.79
	$R^2$	0.89	0.94	0.96	0.97	0.99	0.89	0.95	0.97	0.99	0.99	0.89	0.97	0.97	0.98	0.99
SAI, %	a	-	0.018	0.025	0.048	0.068	-	0.011	0.040	0.051	0.074	-	0.031	0.051	0.066	0.078
	b	-	0.86	0.80	0.64	0.42	-	0.88	0.71	0.59	0.33	-	0.81	0.67	0.54	0.34
	$R^2$	-	0.95	0.94	0.97	0.99	-	0.63	0.96	0.95	0.95	-	0.94	0.92	0.96	0.94
SG, %	a	-	0.021	0.033	0.074	0.135	-	0.013	0.053	0.078	0.147	-	0.036	0.068	0.102	0.155
	b	-	0.02	0.07	-0.02	-0.17	-	0.03	-0.06	-0.10	-0.34	-	-0.04	-0.10	-0.17	-0.32
	$R^2$	-	0.95	0.94	0.97	0.99	-	0.63	0.96	0.95	0.95	-	0.94	0.92	0.96	0.94

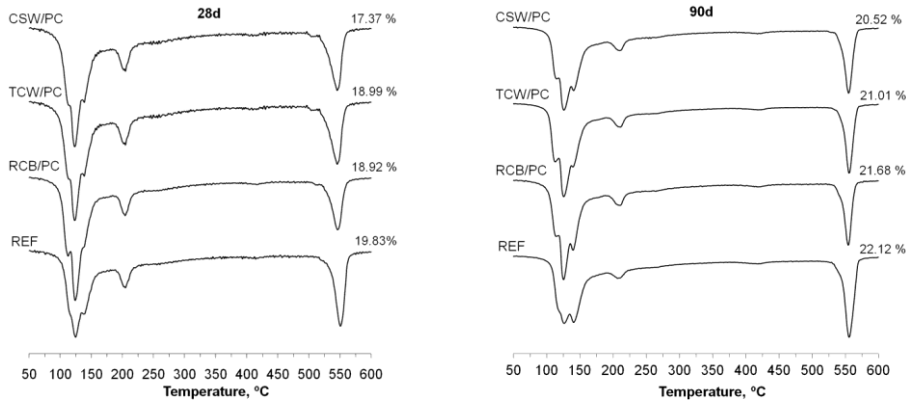


445  
446 The obtained results agree with those previously reported by Wild et al. [31], who  
447 investigated the influence of firing temperature (600 to 1,100°C) on the pozzolanic activity of  
448 clays. These authors [31] observed that the mortars containing up to 20% of clay that had  
449 been calcined at 600-800°C gave good strength results after 28 curing days, while those  
450 prepared with clays calcined at higher temperatures (from 800°C to 1,100 °C) exhibited  
451 better strengths after 90 curing days. Mas et al. [11] observed low pozzolanic activity for  
452 TCW at short curing ages, which became significant after 28 curing days (SG values from  
453 10% to 32% with up to 50 wt.% PC replacement). Puertas et al. [10], who investigated the  
454 pozzolanic behaviour of six different ceramic tile types, also concluded that they all  
455 presented pozzolanic activity after 15 curing days. The strength results came close to those  
456 previously reported by Lavat et al. [32], who used three different roof tile types to replace up  
457 to 40 wt.% PC. These authors concluded that the partial replacement of PC with up to 20-30  
458 wt.% did not significantly affect the compressive strength of the developed mortars. Similarly,  
459 no significant variations in the strength values were observed by Toledo-Filho et al. [9] when  
460 replacing up to 20 wt.% PC with ceramic brick waste (93% SAI after 28 days). The herein  
461 obtained SAI results also came close to those previously reported by Pereira de Oliveira et  
462 al. [14], who recorded relative strength values of 85% and 80% in mortars containing 25 wt.%  
463 bricks and 25 wt.% tiles, respectively, after 28 curing days.

### 464 465 *3.5. Microstructural evolution of the ceramic waste/PC blended binders*

466  
467 The derivative thermogravimetric curves of the reference paste (REF) and those containing  
468 25 wt.% RCB, TCW or CSW, cured at room temperature for 28 and 90 days, are plotted in  
469 Figure 9. The total weight loss values (TWL) have been indicated in percentage. No  
470 significant differences were observed among the DTG curves recorded for the cements  
471 blended with the three different ceramic waste materials. The signals arising from 100°C to  
472 180°C are attributed to the dehydration of ettringite and calcium silicate hydrates (CSH),  
473 while those appearing at slightly higher temperatures (180-240°C) are assigned to the  
474 dehydration of calcium aluminate and calcium aluminosilicate hydrates (CAH and CASH)  
475 [11,33]. As expected, the intensity of these bands grew with curing time. The signal that  
476 arose due to the dehydroxylation of  $\text{Ca}(\text{OH})_2$  (520 to 600°C) was weaker on the DTG curve  
477 of the blended pastes (from -2.17 to -2.36 wt.% and from -2.54 to -2.77 wt.% in the 25 wt.%  
478 pastes cured for 28 and 90 days, respectively) than in the reference sample (-3.12 wt.% and  
479 -3.69 wt.% after 28 and 90 curing days, respectively). This was explained by the lower PC  
480 content (dilution effect), and by the pozzolanic contribution and particle effect of RCB, TCW  
481 and CSW which modifies the total amount of portlandite released during PC hydration.

482



483  
 484 Figure 9. DTG curves of the pastes prepared with 0 and 25 wt.% RCB, TCW or CSW, cured at 20°C  
 485 for 28 and 90 days.

486  
 487 The percentages of fixed lime calculated for the blended pastes prepared with 25 wt.% RCB,  
 488 TCW or CSW, cured at 20°C for 28 and 90 days, are shown in Figure 10. The negative or  
 489 nearly-zero values recorded in the 25 wt.% CSW and TCW pastes cured for 28 days denote  
 490 higher  $\text{Ca}(\text{OH})_2$  contents than those theoretically expected in a paste prepared with 75 wt.%  
 491 PC and without ceramic waste. In agreement with the compressive strength results, these  
 492 negative fixed lime values indicate that, until 28 curing days, the particle effect generally  
 493 prevailed over the pozzolanic reaction, and provided new nucleation sites that facilitated  
 494 cement hydration [33]. The bigger amounts of lime fixed by the RCB waste fall in line with the  
 495 scarcely higher SAI values recorded for these mortars. These results confirmed the slightly  
 496 better pozzolanic activity exhibited by this waste up to 90 curing days, compared to that  
 497 shown by the TCW and CSW powders.

498  
 499

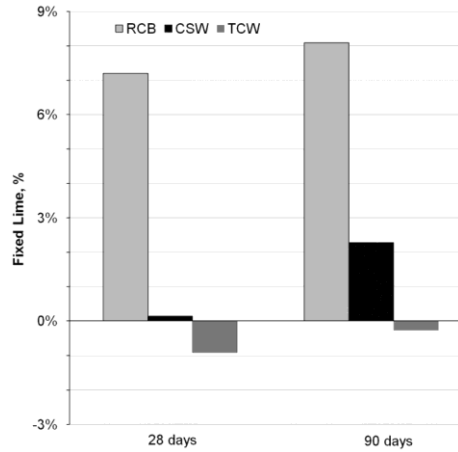
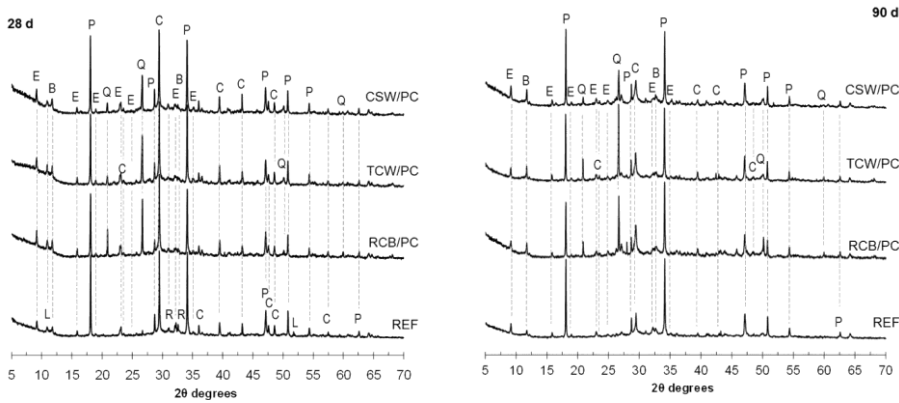


Figure 10. Percentage of fixed  $\text{Ca(OH)}_2$  determined for the ceramic waste/PC blended pastes prepared with 25 wt.% RCB, TCW or CSW, cured at 20°C for 28 and 90 days.

500  
 501  
 502  
 503  
 504 The XRD spectra for the PC reference paste and those blended with 25 wt.% RCB, TCW or  
 505 CSW, cured at room temperature for 28 and 90 days, are plotted in Figure 11. In line with the  
 506 XRD spectra of the raw ceramic waste materials, signals due to quartz (Q,  $\text{SiO}_2$ , PDF  
 507 #331161) appeared in all the blended pastes. Although some crystalline phases as mullite  
 508 (M,  $\text{Al}_6\text{Si}_2\text{O}_{13}$ , PDF #150776), albite (A,  $\text{NaAlSi}_3\text{O}_8$ ), microcline (m,  $\text{KAlSi}_3\text{O}_8$ ), diopside (D,  
 509  $\text{CaMg}(\text{SiO}_3)_2$ ) and rankinite (R,  $2\text{SiO}_2 \cdot 3\text{CaO}$ ) had also previously been identified in the  
 510 original RCB, TCW or CSW (Section 3.1, Figure 2), they were not clearly distinguished in the  
 511 spectra of the blended pastes. This was attributed to the small amount of ceramic waste in  
 512 the paste (25 wt.%), the essentially amorphous nature of the formed CSH gel, and the high  
 513 crystallinity of quartz, whose peaks partially hid those brought about by other minor phases.  
 514  
 515 The 28-days XRD diffractograms contained signals attributed to unreacted larnite (R,  $\beta$ -  
 516  $\text{Ca}_2\text{SiO}_4$ , PDF #330302). The peaks originated by the diffraction of Portlandite (P,  $\text{Ca(OH)}_2$ ,  
 517 PDF #040733) showed a similar intensity in all the pastes except that containing RCB. Given  
 518 the lower amount of PC in the blended pastes, this corroborated that the particle effect  
 519 originated by the ceramic waste accelerated cement hydration. In agreement with the fixed  
 520 lime values (Figure 10), XRD results confirmed that RCB partially consumed  $\text{Ca(OH)}_2$  via  
 521 pozzolanic reactions.

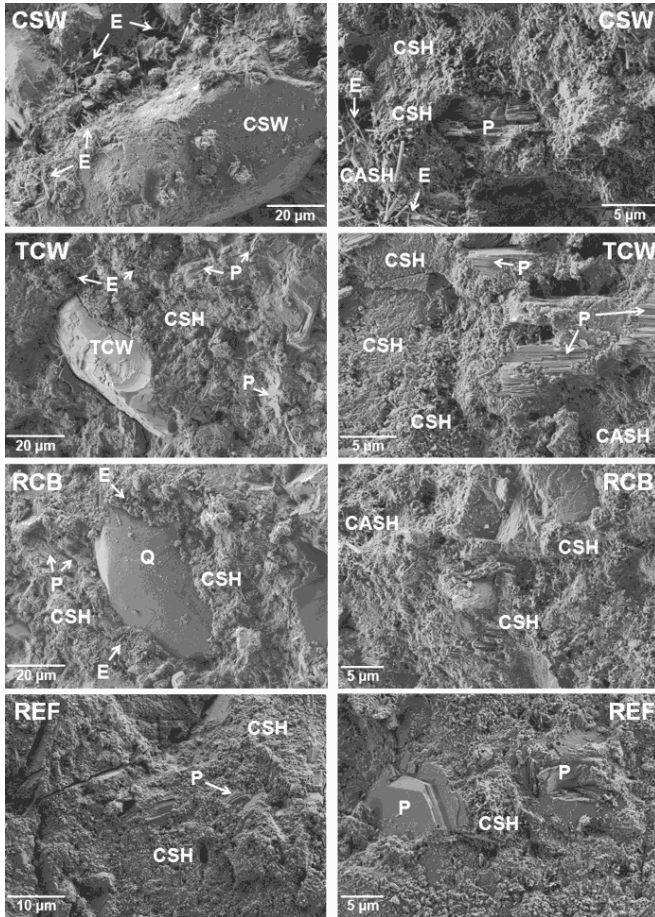
522  
 523  
 524  
 525

526 All the pastes showed peaks that arose due to ettringite (E,  $\text{Ca}_6\text{Al}_2(\text{SO}_4)_3(\text{OH})_{12}\cdot 26\text{H}_2\text{O}$ , PDF  
 527 #411451), which came about through the reaction between tricalcium aluminate and calcium  
 528 sulphate, both contributed by PC. Signals originated by calcite (C,  $\text{CaCO}_3$ , PDF #050586)  
 529 and  $\text{Ca}_4\text{Al}_2\text{O}_6\text{CO}_3\cdot 11\text{H}_2\text{O}$  carboaluminate (B, PDF #410219) were also distinguished in all  
 530 the pastes, while those arising due to  $\text{Ca}_8\text{Al}_4\text{O}_{14}\text{CO}_2\cdot 24\text{H}_2\text{O}$  carboaluminate (L, PDF  
 531 #360129) only appeared in the 28-days samples.  
 532



533  
 534 Figure 11. X-ray diffractograms of the pastes prepared with 0 and 25 wt.% RCB, TCW or CSW, cured  
 535 at 20°C for 28 and 90 days. Q, quartz ( $\text{SiO}_2$ ); E, ettringite ( $\text{Ca}_6\text{Al}_2(\text{SO}_4)_3(\text{OH})_{12}\cdot 26\text{H}_2\text{O}$ ); P, portlandite  
 536 ( $\text{Ca}(\text{OH})_2$ ); R, larnite ( $\beta\text{-Ca}_2\text{SiO}_4$ ); C, calcite ( $\text{CaCO}_3$ ); B, carboaluminate ( $\text{Ca}_4\text{Al}_2\text{O}_6\text{CO}_3\cdot 11\text{H}_2\text{O}$ ); L,  
 537 carboaluminate ( $\text{Ca}_8\text{Al}_4\text{O}_{14}\text{CO}_2\cdot 24\text{H}_2\text{O}$ )  
 538

539 Figure 12 shows the microstructure of the reference paste and those containing 25 wt.%  
 540 RCB, TCW or CSW, cured at 20°C for 28 days. As expected from the TGA and XRD tests,  
 541 unreacted ceramic particles coexisted with plane hexagonal portlandite, ettringite needles  
 542 and amorphous hydration products (CSH and CASH gels). These phases were differentiated  
 543 both by their morphology and by their composition, determined by EDS microanalysis. The  
 544 ceramic waste particles were completely surrounded by reaction products, and calcium  
 545 silicate hydrate was the main reaction product observed.  
 546



547  
 548 Figure 12. FESEM images of the reference paste and those prepared with 25 wt.% RCB, TCW or  
 549 CSW, cured at 20°C for 28 days. E: Ettringite; P: Portlandite; CSH: Calcium silicate hydrate; CASH:  
 550 Calcium aluminosilicate hydrate; Q: Quartz; TCW: Tile ceramic waste; CSW: ceramic sanitary-ware  
 551 waste

552 **3.6. Sustainability analysis**

553 The results obtained after running the basic sustainability analysis of the developed ceramic  
 554 waste/PC blended binders are summarised in Table 5. As observed, the e-energy and e-CO<sub>2</sub>  
 555 values for the blended cements progressively lowered with increasing RCB, TCW or CSW  
 556 contents. The E<sub>s</sub> and C<sub>s</sub> parameters, which considered the 28-day compressive strength,  
 557 were calculated from the linear regression data (Table 4). In general terms, the E<sub>s</sub> values for  
 558 the blended cements were similar or slightly lower than those calculated for CEM I 42.5R,  
 559 and were only higher when replacing 50 wt.% PC with ceramic waste. This was attributed  
 560 mainly to two factors: 1) the relatively low compressive strength values exhibited by the 50  
 561 wt.% blended mortars, in which the dilution effect involved small amounts of portlandite

562 available for the pozzolanic reaction; 2) the crushed ceramic particles were milled under  
 563 laboratory conditions. Thus, the employed energy would probably be lower if milling would be  
 564 carried out in industrial installations, with less required energy and, consequently, lower  $E_s$   
 565 values would be achieved.

566  
 567 The  $e\text{-CO}_2$  values also lowered with increasing percentages of ceramic waste, which yielded  
 568 an interesting reduction for the  $C_s$  parameter, especially when replacing 25 wt.% and 35  
 569 wt.% PC. More specifically, the RCB/PC blended cements reduced  $C_s$  by 11.6% and 13.5%  
 570 (compared to CEM I 42.5R) when using 25 wt.% and 35 wt.% RCB, respectively. These  
 571 reductions were 7.3% and 8.6% with TCW and 7.0% and 8.2% with CSW (same PC  
 572 replacement percentages). Due to the pozzolanic reaction evolution, these  $\text{CO}_2$  emissions  
 573 per tonne of cement and MPa further reduced with curing time. Consequently after 365  
 574 curing days, the following  $C_s$  reductions were determined: 16.2% and 22.7% for 25 wt.% and  
 575 35 wt.% RCB; 15.1% and 19.2% for the same amounts of TCW; 16.9% and 21.5% for the  
 576 corresponding cements blended with CSW.

577  
 578 In short, the partial replacement of PC with RCB, TCW or CSW allowed new binders to be  
 579 designed with green chemistry principles. The developed ceramic waste/PC binders are  
 580 environmentally friendly cements that allow waste materials to be reused, while lowering  
 581 greenhouse gas emissions and the use of natural resources and energy associated with PC  
 582 production. Additionally, a reduction in the cost of the newly-designed blended cements is  
 583 expected because the emissions and costs linked with extracting and preparing natural raw  
 584 materials also diminish, along with green taxes related to climate change policies.

585 Table 5. E-energy,  $e\text{-CO}_2$ ,  $E_s$  and  $C_s$  of the ceramic waste/PC blended cements

Cement	PC Replacement, wt. %	e-Energy, MJ/ton	e- $\text{CO}_2$ , kg $\text{CO}_2$ /ton	$E_s$ *, MJ/ton · MPa	$C_s$ *, kg $\text{CO}_2$ /ton · MPa
PC CEM I 42.5R	0	5500	750	114.6	15.62
RCB/PC	15	4929	655	110.5	14.67
	25	4549	591	106.4	13.82
	35	4169	527	106.8	13.51
	50	3598	432	113.4	13.61
TCW/PC	15	4929	655	112.0	14.88
	25	4549	591	111.5	14.48
	35	4169	527	113.0	14.29
	50	3598	432	125.9	15.10
CSW/PC	15	5003	659	112.7	14.85
	25	4672	599	113.3	14.54
	35	4341	539	115.5	14.34
	50	3844	448	129.2	15.07

586 \*Values obtained for 28-days curing mortars.

587 **4. Conclusions**

588

589 This research assessed the influence of RCB, TCW and CSW ceramic waste materials as  
590 pozzolanic admixtures in PC-blended systems. According to the obtained results, the  
591 following conclusions were drawn:

592 - Consistency of pastes significantly increased with RCB content, hardly varied with  
593 TCW replacements and slightly improved with larger amounts of CSW. These findings were  
594 attributed mainly to the water absorption of the ceramic waste materials.

595 - Replacing PC with these ceramic waste materials slightly prolonged the IST of the  
596 blended systems (up to 20 additional minutes). The FST was cut when RCB or TCW were  
597 used as pozzolanic admixtures (up to 15 minutes shorter than the reference paste), and  
598 progressively prolonged with CSW contents (up to 35 min longer).

599 - No significant differences in the mechanical properties of the developed mortars were  
600 observed when partially replacing PC with RCB, TCW or CSW. The TCW- and CSW-blended  
601 mortars containing up to 25 wt.% ceramic waste met the mechanical requirements set out for  
602 fly ashes, and presented SAI values over 75% and 85% after 28 and 90 curing days,  
603 respectively. RCB somewhat contributed to short-term compressive strength, and provided  
604 these SAI values with up to 35 wt.% contents.

605 - The mechanical contribution of all the ceramic materials improved with curing time.  
606 For a given percentage of replacement, similar compressive strength results were obtained  
607 after 365 curing days (over 50 MPa with up to 35 wt.% RCB, TCW or CSW, close to the 56  
608 MPa recorded for the reference sample).

609 - The embodied energy and CO<sub>2</sub> emissions reduced with increasing replacements of  
610 PC with RCB, TCW or CSW. After 365 curing days, the calculated CO<sub>2</sub> emissions per tonne  
611 of used cement and MPa diminished from 15.1% to 16.9% (compared to PC CEM I 42.5R)  
612 when replacing 25 wt.% PC with any these ceramic waste materials.

613

614 The obtained results show that the three different herein used ceramic waste types are  
615 potential candidates to partially replace PC. Although further research must be conducted to  
616 effectively transfer these results to industry, *a priori* up to 25 wt.% PC can be replaced with  
617 any of these ceramic waste materials. This opens up the possibility of reusing regionally  
618 available ceramic waste as supplementary cementing material, helping to further reduce the  
619 carbon footprint associated with excessive PC use, as it minimizes emissions from  
620 transporting materials. Reusing and valorising RCB, TCW or CSW in PC-blended systems  
621 contributes to reduce not only the environmental impact caused by PC production, but also  
622 the accumulation of waste materials. This will promote sustainable development based on  
623 the design of green building materials.

624

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726 Figure Captions:  
727  
728 Figure 1. The original RCB, TCW and CSW ceramic waste materials  
729  
730 Figure 2. X-ray diffractogram of the ceramic waste materials. Q: Quartz ( $\text{SiO}_2$ ); M: Mullite  
731 ( $\text{Al}_6\text{Si}_2\text{O}_{13}$ ); A: Albite ( $\text{NaAlSi}_3\text{O}_8$ ); m: Microcline ( $\text{KAlSi}_3\text{O}_8$ ); D: Diopside ( $\text{CaMg}(\text{SiO}_3)_2$ ); R:  
732 Rankinite ( $2\text{SiO}_2 \cdot 3\text{CaO}$ )  
733  
734 Figure 3. Scanning electron microscope images of the milled RCB, TCW and CSW particles  
735  
736 Figure 4. Consistency of the blended RCB, TCW and CSW PC pastes  
737  
738 Figure 5. Setting time of the blended RCB, TCW and CSW PC pastes  
739  
740 Figure 6. Compressive strength of the mortars prepared with 0 to 50 wt.% RCB, TCW or  
741 CSW, cured at  $20^\circ\text{C}$  from 3 to 365 days  
742  
743 Figure 7. Strength activity index of the mortars prepared with 0 to 50 wt.% RCB, TCW or  
744 CSW, cured at  $20^\circ\text{C}$  from 3 to 365 days  
745  
746 Figure 8. Strength gain of the mortars prepared with 0 to 50 wt.% RCB, TCW or CSW, cured  
747 at  $20^\circ\text{C}$  from 3 to 365 days  
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749 Figure 9. DTG curves of the pastes prepared with 0 and 25 wt.% RCB, TCW or CSW, cured  
750 at  $20^\circ\text{C}$  for 28 and 90 days.  
751  
752 Figure 10. Percentage of fixed  $\text{Ca}(\text{OH})_2$  determined for the ceramic waste/PC blended  
753 pastes prepared with 25 wt.% RCB, TCW or CSW, cured at  $20^\circ\text{C}$  for 28 and 90 days  
754  
755 Figure 11. X-ray diffractograms of the pastes prepared with 0 and 25 wt.% RCB, TCW or  
756 CSW, cured at  $20^\circ\text{C}$  for 28 and 90 days. Q, quartz ( $\text{SiO}_2$ ); E, ettringite  
757 ( $\text{Ca}_6\text{Al}_2(\text{SO}_4)_3(\text{OH})_{12} \cdot 26\text{H}_2\text{O}$ ); P, portlandite ( $\text{Ca}(\text{OH})_2$ ); R, larnite ( $\beta\text{-Ca}_2\text{SiO}_4$ ); C, calcite  
758 ( $\text{CaCO}_3$ ); B, carboaluminate ( $\text{Ca}_4\text{Al}_2\text{O}_6\text{CO}_3 \cdot 11\text{H}_2\text{O}$ ); L, carboaluminate  
759 ( $\text{Ca}_8\text{Al}_4\text{O}_{14}\text{CO}_2 \cdot 24\text{H}_2\text{O}$ )  
760  
761 Figure 12. FESEM images of the reference paste and those prepared with 25 wt.% RCB,  
762 TCW or CSW, cured at  $20^\circ\text{C}$  for 28 days. E: Ettringite; P: Portlandite; CSH: Calcium silicate

763 hydrate; CASH: Calcium aluminosilicate hydrate; Q: Quartz; TCW: Tile ceramic waste; CSW:  
764 ceramic sanitary-ware waste