

1 **Pozzolanic activity of tiles, bricks and ceramic sanitary-ware in eco-friendly Portland**
2 **blended cements**

3
4 A.M. Pitarch^a, L. Reig^a, A.E. Tomás^a, G. Forcada^a, L. Soriano^b, M.V. Borrachero^b, J. Payá^b, J.
5 M. Monzó^b

6
7 ^a EMC - Department of Mechanical Engineering and Construction, Universitat Jaume I,
8 Castellón de la Plana, Spain. pitarcha@uji.es; lreig@uji.es; atomas@uji.es;
9 gforcada@outlook.es

10
11 ^b ICITECH - Instituto de Ciencia y Tecnología del Hormigón, Grupo de Investigación en
12 Química de los Materiales (GIQUIMA), Universitat Politècnica de València, Valencia, Spain.
13 lousomar@upvnet.upv.es; vborrhachero@cst.upv.es; jjpaya@cst.upv.es;
14 jmmonzo@cst.upv.es

15
16 Corresponding author: L. Reig; lreig@uji.es / Tel.: +34 964 729161 / Fax: +34 964 728106

17
18
19
20

21 **Abstract**

22

23 Reusing ceramic waste as a pozzolanic admixture may offer environmental benefits as it
24 allows the reduction in the consumption of natural resources and energy, and the reduction
25 of CO₂ emissions associated with Portland cement (PC) production, while valorising waste
26 materials with a long biodegradation period. This paper assessed the pozzolanic activity of
27 three different ceramic waste types: red clay bricks (RCB), ceramic tiles (TCW) and ceramic
28 sanitaryware (CSW). After adapting their particle size by crushing and milling, each was used
29 to replace 0 to 50 wt.% PC (CEM I 42.5 R type). The milled powders were characterised by
30 laser diffraction, field emission scanning electron microscopy (FESEM), X-ray fluorescence
31 (XRF) and X-ray diffraction (XRD), and consistency and setting time tests were used to
32 investigate the fresh behaviour of the ceramic waste/PC blended pastes. A basic
33 sustainability analysis was performed, and the pozzolanic activity of RCB, TCW and CSW
34 was assessed by compressive strength tests (performed in mortars cured at room
35 temperature from 3 to 365 days) and microstructural analyses (thermogravimetry, XRD and
36 FESEM performed in the pastes cured at 20°C for 28 and 90 days). The pozzolanic reaction
37 of these waste materials improved with curing time, and all the mortars prepared with up to
38 25 wt.% RCB, TCW or CSW met the mechanical requirements set out for coal fly ashes,
39 whatever the ceramic waste type used. Among them, these results open up the possibility of
40 partially replacing PC with the closest available ceramic waste, which would reduce the CO₂
41 emissions and economic cost deriving from transporting waste.

42

43

44 *Keywords:* Portland cement, ceramic waste, waste management, pozzolanic activity,
45 compressive strength.

46

47 1. Introduction

48

49 Sustainable development, based on environmental conservation and responsible use of
50 resources, is one of the biggest challenges faced today. Thus the construction industry in
51 general, and cement production in particular, are essential contributors for a transition
52 towards low carbon economy. Portland cement (PC) is a key construction material that is
53 widely used worldwide. As reported by the European Cement Association based in Brussels
54 [1], it is estimated that 4.1 billion tonnes of cement were globally produced in 2017, and
55 China was the leader producer (56.5% of global production), followed by India (6.8%) and
56 CEMBUREAU members (6.3%) [1]. As explained by Kajaste and Hurme [2], PC production is
57 one of the world's highest CO₂-emitting processes and is responsible for around 5-8% of the
58 carbon generated worldwide. Approximately 0.75 kg of carbon dioxide per kg of produced
59 cement is emitted to the atmosphere, which come mainly from limestone (CaCO₃)
60 decomposition and the combustion of the fuel required to reach clinkering temperatures
61 ($\approx 1,450^{\circ}\text{C}$) [3-5]. As Imbabi et al. [6] stated, with no changes in the current production rate
62 and manufacturing processes, it is estimated that CO₂ emissions will have approximately
63 multiplied those registered in 1990 by 5 by 2050. These predictions were conservative
64 because, although less than 2 billion tonnes of CO₂ were expected to be emitted to the
65 atmosphere in 2010, 3.24 billion tonnes were registered in 2012. Thus the increasing use of
66 PC demands a real urgent change in order to develop sustainable, durable and cost-effective
67 binders. Both the cement industry and scientific community are actively seeking alternatives
68 to reduce emissions which, in 2018, had dropped by almost 14% since 1990. The 5C
69 programme developed by CEMBUREAU compiles the widest possible set of alternatives to
70 lower the carbon emissions released by the cement industry. The different proposals are
71 classified in five categories: Clinker, Cement, Concrete, Construction and Carbonation, of
72 which the last refers to CO₂ capture technologies. Imbabi et al. [6] also summarised four
73 different approaches that have been developed mainly to minimise CO₂ intensity and to
74 improve energy efficiency associated with cement production: a) improve manufacturing
75 processes; b) co-incineration of waste materials during cement production; c) use of
76 Supplementary Cementitious Materials (SCMs) in cement; d) develop novel resource-
77 efficient cements. Of these, SCMs have been widely used as fillers or pozzolans, and the
78 latter implies their reaction with the Ca(OH)₂ released during PC hydration to provide
79 cementitious products [7,8]. One of the materials satisfactorily used to partially replace PC is
80 ceramic waste [9-14], which is durable, hard and very resistant to physical, biological and
81 chemical degradation. As previously explained in [15], large amounts of ceramic waste are
82 generated yearly from demolition practices or production defects, and dumping it in landfills
83 leads to environmental problems due to dust pollution and the occupation of vast expanses
84 of land. Thus reusing ceramic waste materials in PC contributes to both diminish the

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

87

88 As Ay and Unal [12] or Turanli [16] explained, although clay minerals in their original state do
89 not present pozzolanic reactivity, they may acquire it when thermally treated given the
90 destruction of the crystal structure of clay and the formation of amorphous or disordered
91 aluminosilicate phases. The changes that occur in ceramic materials during the firing process
92 have been described by Mohammed [3]: the removal of free water from clay (dehydration)
93 occurs from 20°C to 200°C, and is followed by the dehydroxylation process that consists in
94 loss of the chemically combined water from the OH groups contained in clay minerals; during
95 dehydroxylation, the crystalline structure of clay constituents collapses, which results in
96 unstable silica and alumina with an amorphous or disordered lattice structure. Decarbonation
97 (release of CO₂) of compounds, such as calcite or dolomite, occurs within the 700-900°C
98 range; finally, exothermic recrystallisation takes place at temperatures above 900°C, which
99 may result in the formation of new thermodynamically stable phases that can reduce the
100 pozzolanic activity of ceramic materials [3,17]. The study by Baronio and Binda [17], in which
101 clays were calcined up to 1,000°C, observed that their pozzolanic activity improved when
102 calcined at temperatures between 600°C and 900°C (depending on the type and amount of
103 clay used), but diminished when calcined at more than 900°C. However, the study by Turanli
104 et al. [16] reported slightly better relative strength values (compared to the reference mortar)
105 when replacing 20 wt.% PC with slab bricks (SB) sintered at 1,000-1,100°C than when using
106 the same amount of wall bricks (WB) fired at 800-900°C, (86% and 78% strength activity
107 indices were recorded in the SB- and WB-containing mortars cured for 28 days,
108 respectively). The review by Mohammed et al. [3], who analysed the pozzolanic behaviour of
109 clays, clay waste and ceramic products like bricks or tiles, concluded that the reactivity of
110 calcined clays depended mainly on the degree of dehydroxylation, fineness of particles, and
111 their SiO₂ + Al₂O₃ + Fe₂O₃ content. Binici 2012 [8] and Sánchez de Rojas et al. [13] stated
112 that the optimum dehydroxylation temperature and time to break the crystalline structure of
113 clay depended on clay mineral. Accordingly, as the production of ceramic materials is guided
114 to obtain optimal properties for a specific use (hollow or load-bearing bricks, wall or floor tiles,
115 sanitaryware units, etc.), sintering temperatures will either presumably differ from the
116 optimum ones considered for complete clay minerals dehydroxylation or probably lead to the
117 formation of new crystalline stable phases [3,12,17], which is expected to reduce the
118 pozzolanic activity of ceramic products. Nonetheless, several studies [9-14] have
119 successfully proved the pozzolanicity of ceramic waste materials when blended with PC,
120 along with the environmental benefits that this implies. Puertas et al. [10] evaluated the
121 pozzolanic activity of six different ceramic tile types. They concluded that they all exhibited
122 pozzolanic behaviour after 15 curing days, which was strongly influenced by their chemical

123 composition (high silica and alumina contents) and amorphous content (close to 35 wt.% in
124 three tile types). The studies by Mas et al. [11] and Ay and Unal [12], who used ceramic tiles
125 as pozzolanic admixtures in PC systems, found that mortars containing up to 35 wt.%
126 ceramic waste met both the requirements set out in fly ash regulations [11] and those stated
127 for the cement standard for calcined natural pozzolans (ASTM C 618) [12], respectively.
128 Sánchez de Rojas et al. [13] also noticed some contribution to compressive strength in PC
129 mortars cured for 28 days at room temperature when replacing up to 15 wt.% PC with clay
130 roof tiles. Pereira-de-Oliveira 2012 compared the pozzolanicity of glass, bricks and tiles (10-
131 40 wt.% PC replacement), and corroborated that it was strongly influenced by calcination
132 temperature because no pozzolanic behaviour was observed when using brick waste.
133 However, Toledo-Filho et al. [9] observed a minor influence on the mechanical properties of
134 mortars when replacing 20 wt.% PC with clay bricks calcined at 850°C.

135

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

146

147

148 **2. Materials and Methods**

149

150 *2.1. Materials*

151

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

160

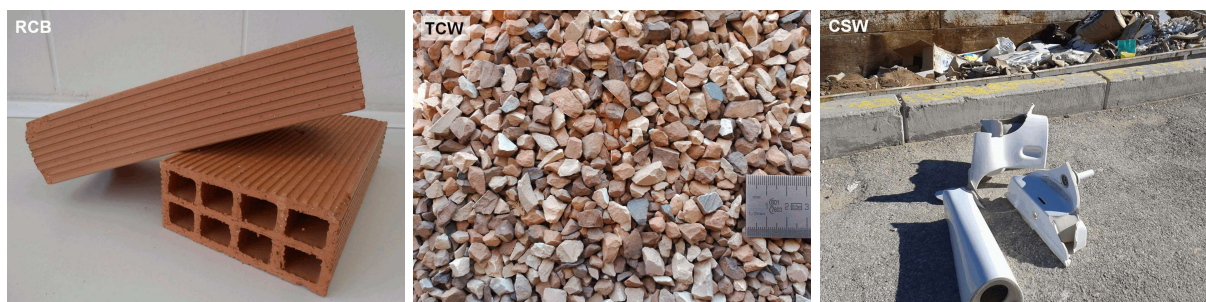


Figure 1. The original RCB, TCW and CSW ceramic waste materials

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 Bruker AXS D8 Advance using Cu K α radiation at 20 mA and 40 kV from 10° to 70° 2 θ 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.

194

195

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		

196

197 *2.4. Fresh behaviour and mechanical properties of the ceramic waste/PC blended binders*

198

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

211

$$212 \quad SG(\%) = \frac{S_{POZ} - (S_{REF} \cdot PC\%)}{S_{REF} \cdot PC\%} \cdot 100 \quad (1)$$

213

214 where:

215 S_{POZ} = compressive strength of the mortar with the ceramic waste (RCB, TCW or CSW);216 S_{REF} = compressive strength of the reference mortar;

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

218 *2.5. Microstructural characterisation of the ceramic waste/PC blended binders*

219

220 The reference paste (100 wt.% PC) and those containing 25 wt.% RCB, TCW or CSW, cured
221 at room temperature for 28 and 90 days, were used to assess the microstructural evolution of
222 the ceramic waste/PC blended cements. Crystalline phases were identified by X-ray
223 diffractometry (XRD), which were run in a Brucker AXS D4 Endeavor powder diffractometer
224 equipped with a Lynxeye detector, from 5 to 70 2θ degrees, at 40 kV and 40 mA, with Cu K α
225 radiation. The thermogravimetric analyses (TG) performed aimed to assess the compounds
226 formed after the hydration of the pastes and to determine the percentage of fixed lime. This
227 value provided information on PC hydration and pozzolanic reactions, and it was determined
228 as previously explained in [11]. TG tests were run in a Mettler Toledo TGA/DSC3, using
229 closed aluminium crucibles (100 μ l) with a perforated lid. These tests were conducted from
230 35°C to 600°C at a heating rate of 10°C/min in nitrogen atmosphere. In order to confirm the
231 presence of the compounds previously identified by XRD and TG, and examine the density
232 of the developed pastes, their microstructures were observed by FESEM, using the
233 equipment previously described in Section 2.2.

234

235 *2.6. Sustainability analysis*

236

237 To estimate the environmental impact of reusing RCB, TCW or CSW as a partial
238 replacement of PC, a basic sustainability analysis was conducted. A gate-to-gate life cycle
239 analysis approach was followed, which contemplated only the CO₂ emissions associated with
240 binder production. This simplified calculation contemplated neither the transport of the
241 materials from their origin to the laboratory, nor the mixing or curing of pastes and mortars, or
242 other mortar life cycle stages, such as maintenance or demolition. To calculate the embodied
243 specific energy (e-energy) and embodied specific CO₂ (e-CO₂) emissions of the ceramic
244 waste/PC blended cements, the e-energy and e-CO₂ values previously reported for CEM I
245 42.5 (5,500 MJ/ton [19] and 750 kg CO₂/ton [5], respectively) were used. Moreover the
246 percentage of PC replacement, and the energy and CO₂ emissions generated when crushing
247 and milling ceramic waste were considered. The jaw crusher BB200 Retsch, employed to
248 crush the ceramic waste materials, is driven by a 1,4914 kW power unit, and it took 2
249 minutes and 30 seconds to crush 1 kg of ceramic waste. The Orto-Alresa ball mill used to
250 reduce the size of waste was equipped with a 150 W power unit, and two jars containing 1.1
251 kg of crushed ceramic particles were employed in each batch. An emission factor of 0.241 kg
252 CO₂ eq/kWh was utilised to convert the used energy into CO₂ emissions (data obtained from
253 the Spanish Ministry for the Ecological Transition and Demographic Challenge for 2019 [20]).

254

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

258

$$259 \quad E_s = \frac{e\text{-energy}}{S_{POZ_{28d}}} \quad (2)$$

260

$$261 \quad C_s = \frac{e\text{-CO}_2}{S_{POZ_{28d}}} \quad (3)$$

262 where:

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

265 C_s = Embodied CO₂ in relation to the 28-day compressive strength of the pozzolanic mortar,
 266 kg CO₂/ton · MPa

267 $S_{POZ_{28d}}$ = compressive strength of the pozzolanic mortar cured for 28 days, MPa.

268

269

270 3. Results and Discussion

271

272 3.1. Ceramic waste material characterisation

273

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

279

280 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

281

282 Table 3 shows the chemical composition and amorphous content of the three ceramic
 283 wastes herein employed. All the ceramic materials were composed mainly of SiO₂ and Al₂O₃,
 284 with bigger quantities in CSW (the sum of 89.6%) compared to those recorded for TCW and
 285 RCB (the sum of 79.8% and 66.5%, respectively). RCB and TCW presented relatively high
 286 Fe₂O₃ contents, and the sum of SiO₂, Al₂O₃ and Fe₂O₃ in them all was above 70%, which
 287 Mohammed et al. [3] pointed out to be an important parameter that promotes pozzolanic

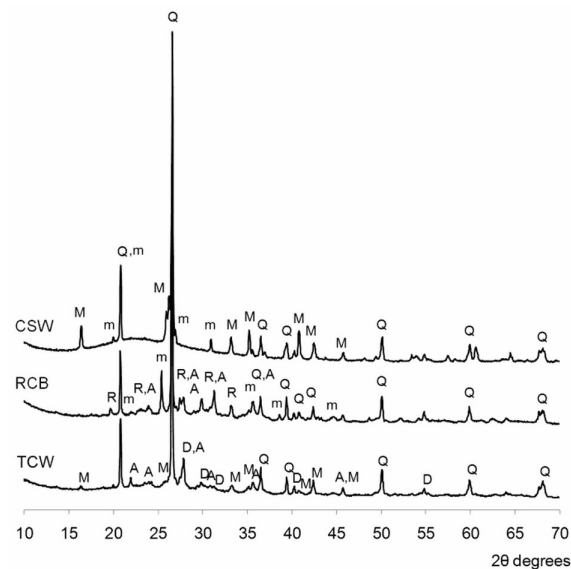
288 activity. Amorphous content varied from 35 to 60 wt.% (RCB and TCW, respectively), and the
 289 amount of vitreous phases recorded for TCW and CSW fell within the range previously
 290 reported by Zanelli et al. [22] for porcelain stoneware tiles (40-75 wt.%, a range obtained
 291 after analysing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

292
 293
 294 Table 3. Chemical composition of RCB, TCW and CSW ceramic wastes, wt.%

Ceramic waste	SiO ₂	Al ₂ O ₃	CaO	MgO	K ₂ O	Fe ₂ O ₃	SO ₃	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

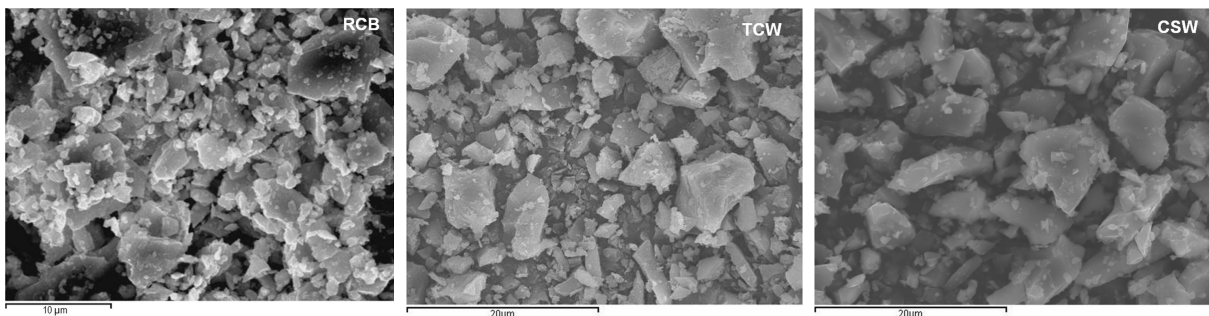
295 *At 1,000°C

296
 297 The mineralogical composition of RCB, TCW and CSW is presented in Figure 2. All the
 298 ceramic materials showed signals due to quartz (Q, SiO₂; PDFcard 331161), while mullite (M,
 299 Al₆Si₂O₁₃; PDFcard 150776) was identified only within the spectra of TCW and CSW (bigger
 300 amounts in CSW, denoted by higher-intensity signals). This was attributed to the lower
 301 sintering temperatures applied to produce RCB (usually fired up to approx. 900°C) [23]
 302 compared to those used to make ceramic tiles, whose peak temperatures usually vary within
 303 the 1,120–1,220°C range [11], or CSW, whose plateau in the heating curve generally falls
 304 within the 1,200-1,280°C range [24]. Small amounts of sodium feldspar albite (A, NaAlSi₃O₈;
 305 PDFcard 190926) were distinguished in the TCW and RCB diffractograms, and signals due
 306 to microcline (m, KAlSi₃O₈, PDFcard090466) appeared in the RCB and CSW patterns.
 307 Traces of diopside (D, CaMg(SiO₃)₂; PDFcard 190239) and rankinite (R, 2SiO₂·3CaO,
 308 PDFcard220539) were also identified in TCW and RCB, respectively. These crystalline
 309 phases typically form in ceramic materials, and have also been distinguished in previous
 310 studies, where TCW was used to partially replace PC [11], and RCB or CSW were utilised as
 311 precursors in alkali-activated binders [25,26]. The deviation from the baseline observed
 312 within the 15-30° 2θ degree range corroborates the presence of amorphous phases in all the
 313 ceramic waste materials, as previously quantified in Table 3. In the RCB waste these
 314 disordered phases are explained by the dehydroxylation of the clay [3,23], while in the TCW
 315 and CSW their presence is attributed to the formation of new vitreous phases from the
 316 melting of feldspars which, according to Zanelli et al. [22], occurs from approximately
 317 1050°C.



318
 319 Figure 2. X-ray diffractogram of the ceramic waste materials. Q: Quartz (SiO_2); M: Mullite ($\text{Al}_6\text{Si}_2\text{O}_{13}$);
 320 A: Albite ($\text{NaAlSi}_3\text{O}_8$); m: Microcline (KAlSi_3O_8); D: Diopside ($\text{CaMg}(\text{SiO}_3)_2$); R: Rankinite
 321 ($2\text{SiO}_2 \cdot 3\text{CaO}$)
 322

323 The micrographs of the milled ceramic waste materials (see Figure 3) display irregular
 324 particles with a smooth surface. No significant differences were observed among them.
 325

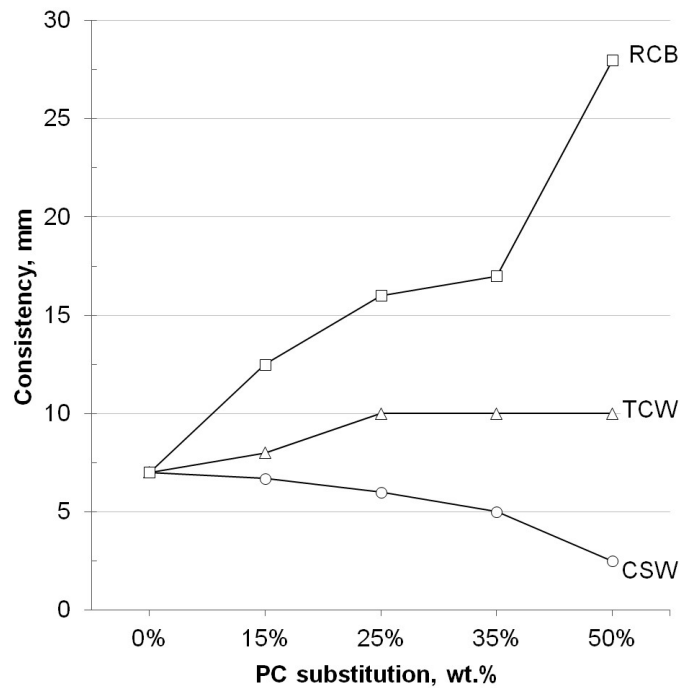


326
 327 Figure 3. Field emission scanning electron microscope images of the milled RCB, TCW and CSW
 328 particles
 329

330 3.2. Consistency

331
 332 Variation in consistency, reported as the distance between the lower side of the 1 cm-
 333 diameter plunger and the bottom of Vicat's apparatus, depending on the different ceramic
 334 waste types and contents, is plotted in Figure 4. Minor variations in consistency were
 335 observed when replacing PC with TCW, and the results slightly lowered (better workability)
 336 with increasing CSW additions, and significantly increased with RCB content, which denotes
 337 loss of workability in the RCB/PC blended systems. As all the herein used milled ceramic
 338 particles presented a similar morphology (Fig. 3), the obtained results were attributed mainly
 339 to water absorption differences. The results agree with those previously reported by Pitarch

340 et al. [27], who employed the same RCB, TCW and CSW waste materials to replace natural
 341 aggregates in structural concrete. In their study [27], water absorption values of 15.76 wt.%,
 342 6.28 wt.% and 0.69 wt.% were recorded for the RCB, TCW and CSW particles (up to 4 mm
 343 in size, used as recycled sand), respectively. Our results also agree with those previously
 344 reported by Mas et al. [11] in PC/TCW blended systems, whose consistency values
 345 (determined by the flow table test) varied within a narrow range (150-160 mm) in mortars
 346 containing up to 50 wt.% TCW. The authors [11] attributed these minor variations to the low
 347 porosity and smooth surface of the tile ceramic particles.
 348



349 Figure 4. Consistency of the blended RCB, TCW and CSW PC pastes

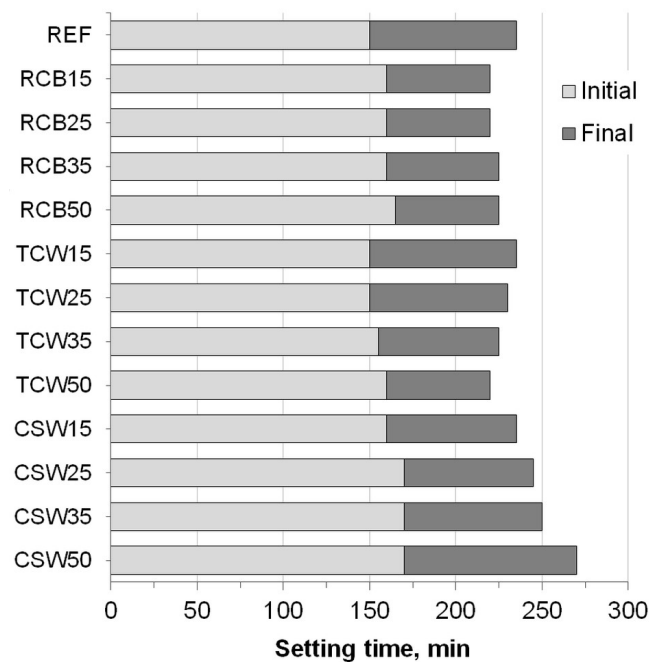
350
 351

352 3.3. Setting time

353

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

365 content (up to 270 min with 50 wt.%). This implied that the time in which the sample
 366 remained under plastic condition, required for handling the paste, mortar or concrete
 367 samples, hardly varied in the RCB/PC samples, became progressively shorter in the TCW-
 368 blended pastes (from 85 min with 15 wt.% TCW to 60 min with 50 wt.% TCW) and prolonged
 369 in those prepared with CSW (from 75 min with 15 wt.% to 100 min with 50 wt.%). The results
 370 of the PC/CSW blended systems coincided with those previously reported by Medina et al.
 371 [28], who observed that partially replacing PC with CSW (10 wt.% or 20 wt.%) modified the
 372 rheology of pastes and retarded their hydration. The RCB/PC system results came close to
 373 those previously reported by Naceri et al. [29], who observed shorter IST and FST in the
 374 cements developed by partially replacing clinker with brick ceramic waste (up to 20 wt.%).
 375 However, differences in the results herein obtained (the IST was slightly longer in the
 376 RCB/PC systems) were attributed mainly to the increasing gypsum-to-clinker ratios used by
 377 Naceri et al. [29] with higher ceramic contents, because those authors mixed a set amount of
 378 gypsum no matter what the clinker replacement was.



379
 380 Figure 5. Setting time of the blended RCB, TCW and CSW PC pastes

381
 382 **3.4. Compressive strength of pozzolanic mortars**

383
 384 The evolution of compressive strength (MPa), strength activity index (SAI, %) and strength
 385 gain (SG, %) with the different study parameters (curing age, types and amount of ceramic
 386 waste used) are plotted in Figures 6 to 8. To correlate the strength parameters with curing
 387 age, simple linear equations were established according to Equation (4) for each ceramic
 388 waste and PC replacement percentage.

390 $S_{PARAMETER} = a \cdot \ln(t) + b$ (4)

391

392 Where:

393 $S_{PARAMETER}$ = Strength parameter: compressive strength (MPa), SG (%) or SAI (%);

394 t = curing age, days

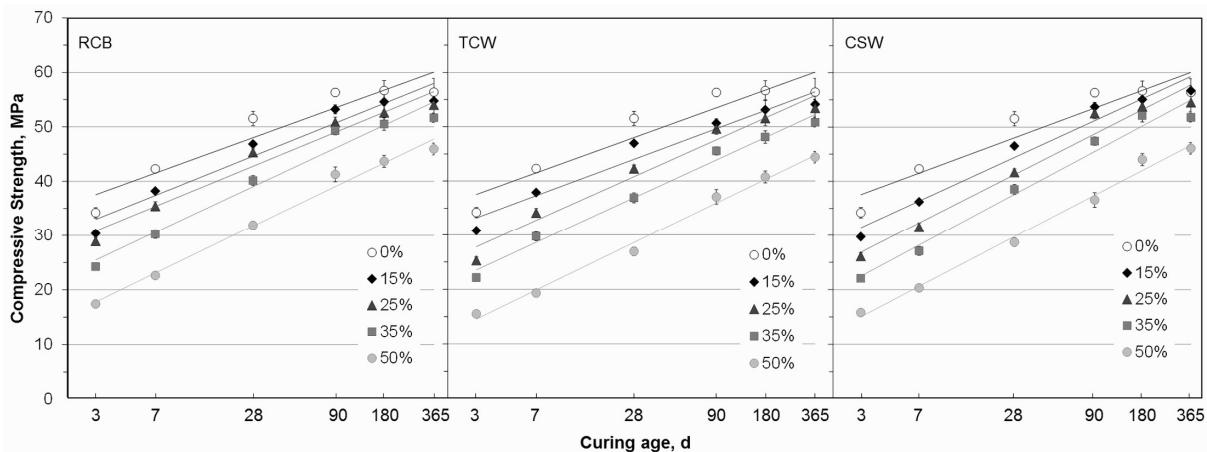
395 \ln = natural (based-e) logarithm

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

397

398 As Figure 6 shows, similar compressive strength results were obtained for a given
 399 replacement percentage, no matter what ceramic waste was used. Given that the three
 400 ceramic waste materials herein used had a close particle size distribution and similar $\text{SiO}_2 +$
 401 $\text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$ contents (Section 3.1), these results are mainly attributed to the pozzolanic
 402 reactivity of the amorphous phases formed during their production processes which, as
 403 explained in Section 3.1, derive from the dehydroxylation of the clay (RCB) and the melting
 404 of feldspars (TCW, CSW). Although the strength values recorded at short curing ages (3 or 7
 405 days) significantly lowered compared to the reference mortar, they came closer with the
 406 curing time, especially in the samples containing up to 25 wt.% ceramic waste (close to 50
 407 MPa after 90 curing days).

408



409

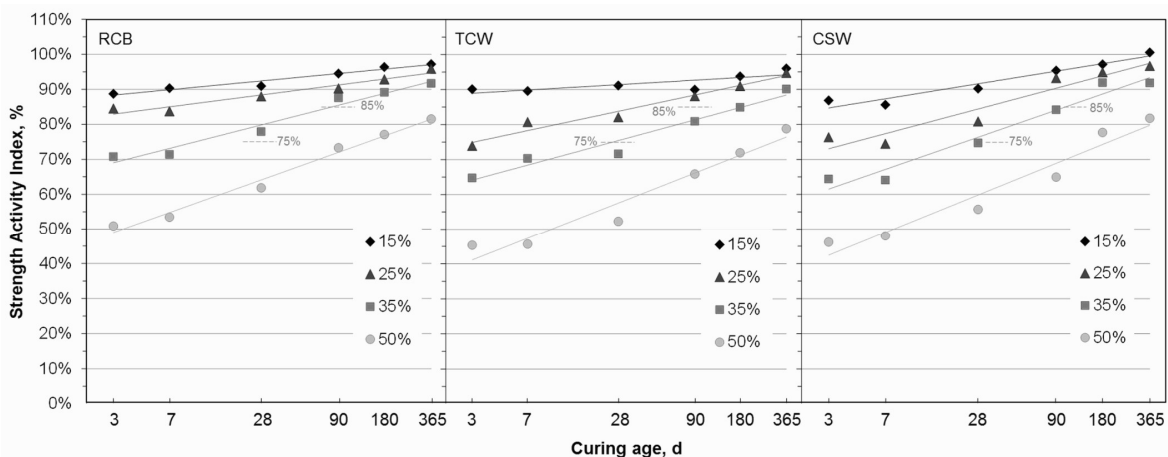
410 Figure 6. Compressive strength of the mortars prepared with 0 to 50 wt.% RCB, TCW or CSW, cured
 411 at 20°C from 3 to 365 days

412

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

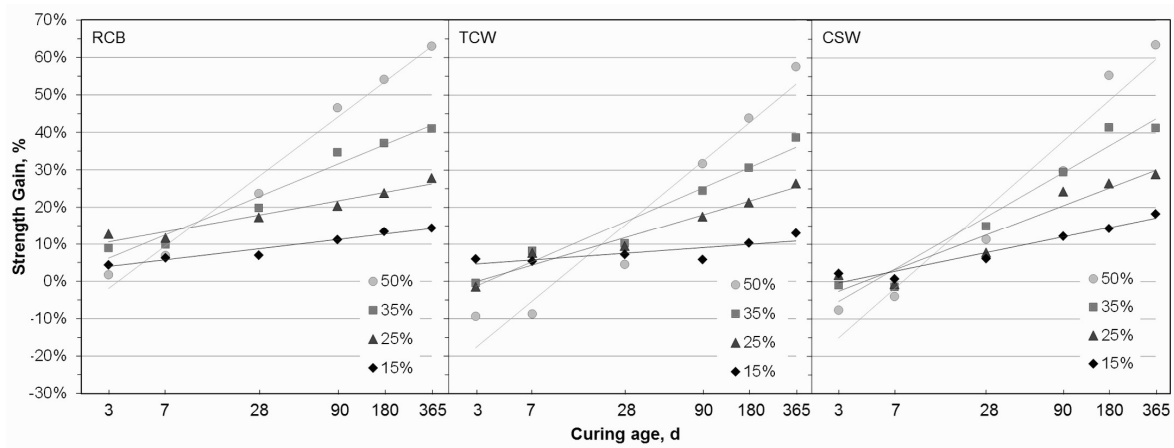
419 absorption when compared with that registered for TCW and CSW (15.76%, 6.28% and
 420 0.69%, respectively [27]). The relatively high RCB water absorption progressively reduced
 421 the workability of the blended cements with increasing RCB contents and also diminished the
 422 effective water (that available to react with cement, determined as the total amount of water
 423 minus that absorbed by the ceramic waste particles). Thus, the reduction in the water to
 424 cement ratio, as widely known, improves strength and durability. However, the contribution
 425 on strength of these ceramic waste-based materials improved with curing time and all the
 426 ceramic waste/PC blended mortars presented similar long-term strength: all the mortars
 427 blended with 35 wt.% ceramic waste had SAI indices over 90% after being cured for 1 year.
 428 In other words, the strength of the mortars prepared with 65 wt.% PC reduced by only 10%
 429 after 365 curing days respect to the reference mortar (100% PC as binder). This means that
 430 these blended mortars are an interesting alternative that contributes to sustainable
 431 development in applications that do not require high strength at short curing times.

432
 433



434
 435 Figure 7. Strength activity index of the mortars prepared with 0 to 50 wt.% RCB, TCW or CSW, cured
 436 at 20°C from 3 to 365 days
 437

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



444
 445 Figure 8. Strength gain of the mortars prepared with 0 to 50 wt.% RCB, TCW or CSW, cured at 20°C
 446 from 3 to 365 days

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

458
 459 Table 4. Linear regression data for the compressive strength, SG and SAI of RCB, TCW and CSW PC
 460 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
σ_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

461
462 The obtained results agree with those previously reported by Wild et al. [31], who
463 investigated the influence of firing temperature (600 to 1,100°C) on the pozzolanic activity of
464 clays. These authors [31] observed that the mortars containing up to 20% of clay that had
465 been calcined at 600-800°C gave good strength results after 28 curing days, while those
466 prepared with clays calcined at higher temperatures (from 800°C to 1,100 °C) exhibited
467 better strengths after 90 curing days. Mas et al. [11] observed low pozzolanic activity for
468 TCW at short curing ages, which became significant after 28 curing days (SG values from
469 10% to 32% with up to 50 wt.% PC replacement). Puertas et al. [10], who investigated the
470 pozzolanic behaviour of six different ceramic tile types, also concluded that they all
471 presented pozzolanic activity after 15 curing days. The strength results came close to those
472 previously reported by Lavat et al. [32], who used three different roof tile types to replace up
473 to 40 wt.% PC. These authors concluded that the partial replacement of PC with up to 20-30
474 wt.% did not significantly affect the compressive strength of the developed mortars. Similarly,
475 no significant variations in the strength values were observed by Toledo-Filho et al. [9] when
476 replacing up to 20 wt.% PC with ceramic brick waste (93% SAI after 28 days). The herein
477 obtained SAI results also came close to those previously reported by Pereira de Oliveira et
478 al. [14], who recorded relative strength values of 85% and 80% in mortars containing 25 wt.%
479 bricks and 25 wt.% tiles, respectively, after 28 curing days.

480

481 *3.5. Microstructural evolution of the ceramic waste/PC blended binders*

482

483 The derivative thermogravimetric curves of the reference paste (REF) and those containing
484 25 wt.% RCB, TCW or CSW, cured at room temperature for 28 and 90 days, are plotted in
485 Figure 9. The total weight loss values (TWL) have been indicated in percentage. No
486 significant differences were observed among the DTG curves recorded for the cements
487 blended with the three different ceramic waste materials. The signals arising from 100°C to
488 180°C are attributed to the dehydration of ettringite and calcium silicate hydrates (CSH),
489 while those appearing at slightly higher temperatures (180-240°C) are assigned to the
490 dehydration of calcium aluminate and calcium aluminosilicate hydrates (CAH and CASH)
491 [11,33]. As expected, the intensity of these bands grew with curing time. The signal that
492 arose due to the dehydroxylation of $\text{Ca}(\text{OH})_2$ (520 to 600°C) was weaker on the DTG curve
493 of the blended pastes (from -2.17 to -2.36 wt.% and from -2.54 to -2.77 wt.% in the 25 wt.%
494 pastes cured for 28 and 90 days, respectively) than in the reference sample (-3.12 wt.% and
495 -3.69 wt.% after 28 and 90 curing days, respectively). This was explained by the lower PC
496 content (dilution effect), and by the pozzolanic contribution and particle effect of RCB, TCW
497 and CSW which modifies the total amount of portlandite released during PC hydration.

498

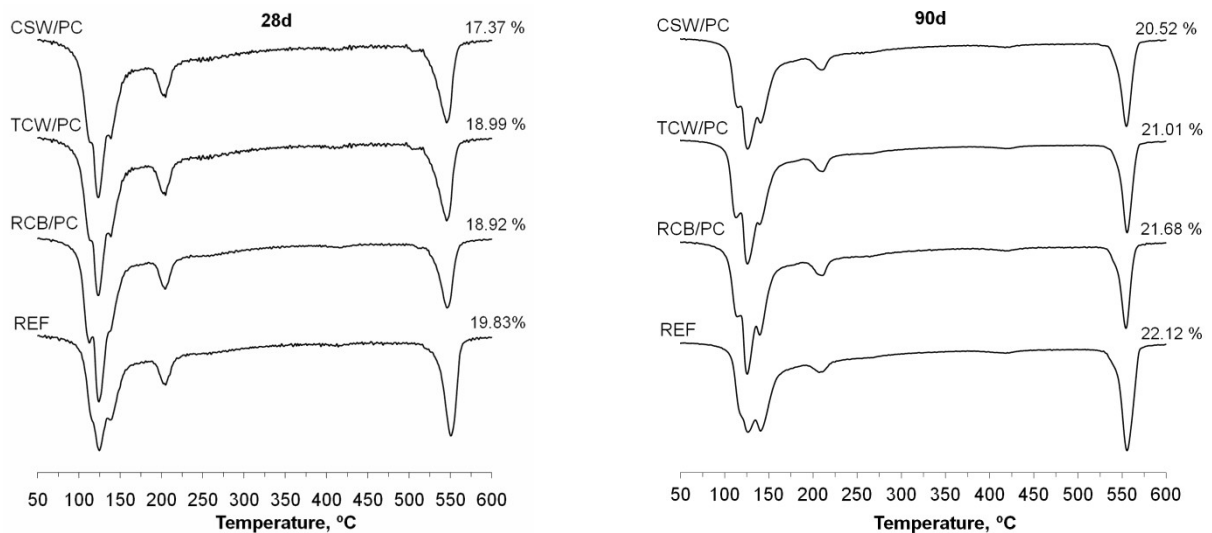


Figure 9. DTG curves of the pastes prepared with 0 and 25 wt.% RCB, TCW or CSW, cured at 20°C for 28 and 90 days.

499

500

501

502

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

514

515

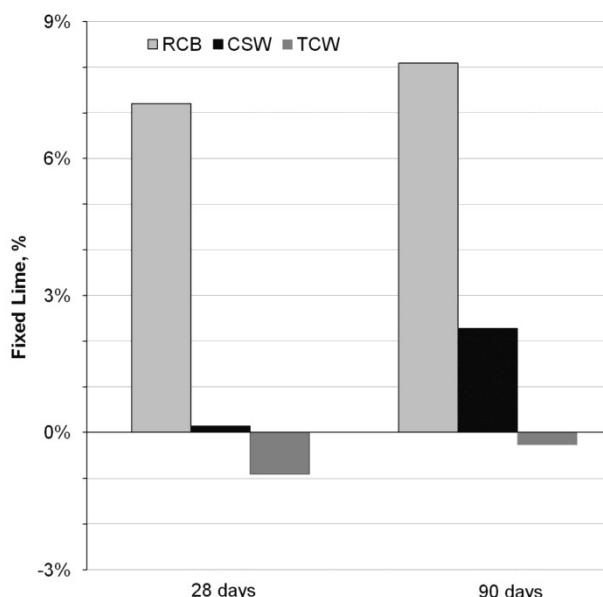


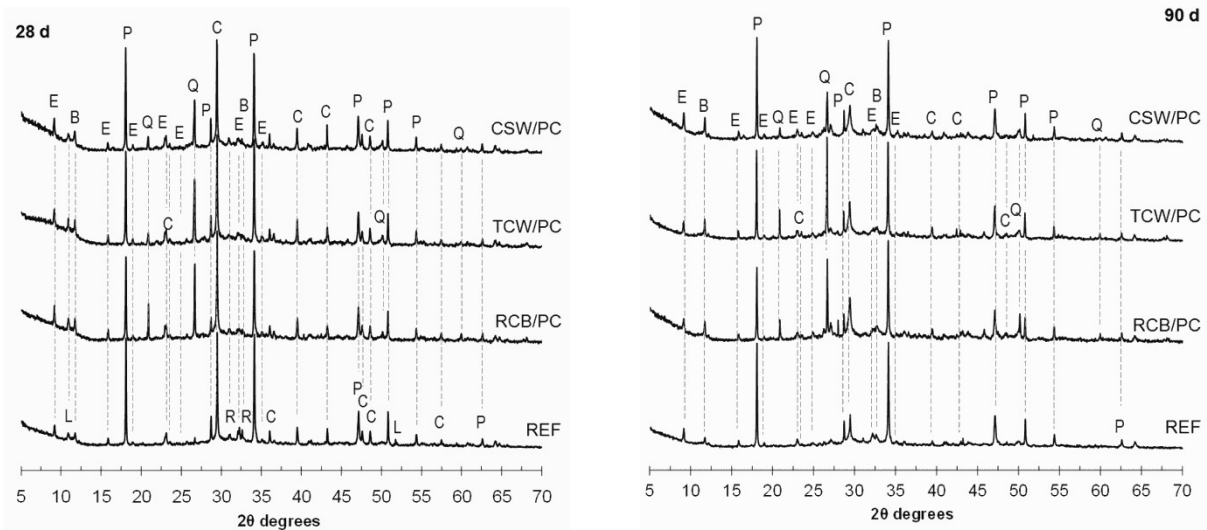
Figure 10. Percentage of fixed Ca(OH)_2 determined for the ceramic waste/PC blended pastes prepared with 25 wt.% RCB, TCW or CSW, cured at 20°C for 28 and 90 days.

516
517
518
519
520
521
522
523
524
525
526
527
528
529
530
531
532
533
534
535
536
537
538
539
540
541

The XRD spectra for the PC reference paste and those blended with 25 wt.% RCB, TCW or CSW, cured at room temperature for 28 and 90 days, are plotted in Figure 11. In line with the XRD spectra of the raw ceramic waste materials, signals due to quartz (Q, SiO_2 , PDF #331161) appeared in all the blended pastes. Although some crystalline phases as mullite (M, $\text{Al}_6\text{Si}_2\text{O}_{13}$, PDF #150776), albite (A, $\text{NaAlSi}_3\text{O}_8$), microcline (m, KAlSi_3O_8), diopside (D, $\text{CaMg}(\text{SiO}_3)_2$) and rankinite (R, $2\text{SiO}_2 \cdot 3\text{CaO}$) had also previously been identified in the original RCB, TCW or CSW (Section 3.1, Figure 2), they were not clearly distinguished in the spectra of the blended pastes. This was attributed to the small amount of ceramic waste in the paste (25 wt.%), the essentially amorphous nature of the formed CSH gel, and the high crystallinity of quartz, whose peaks partially hid those brought about by other minor phases.

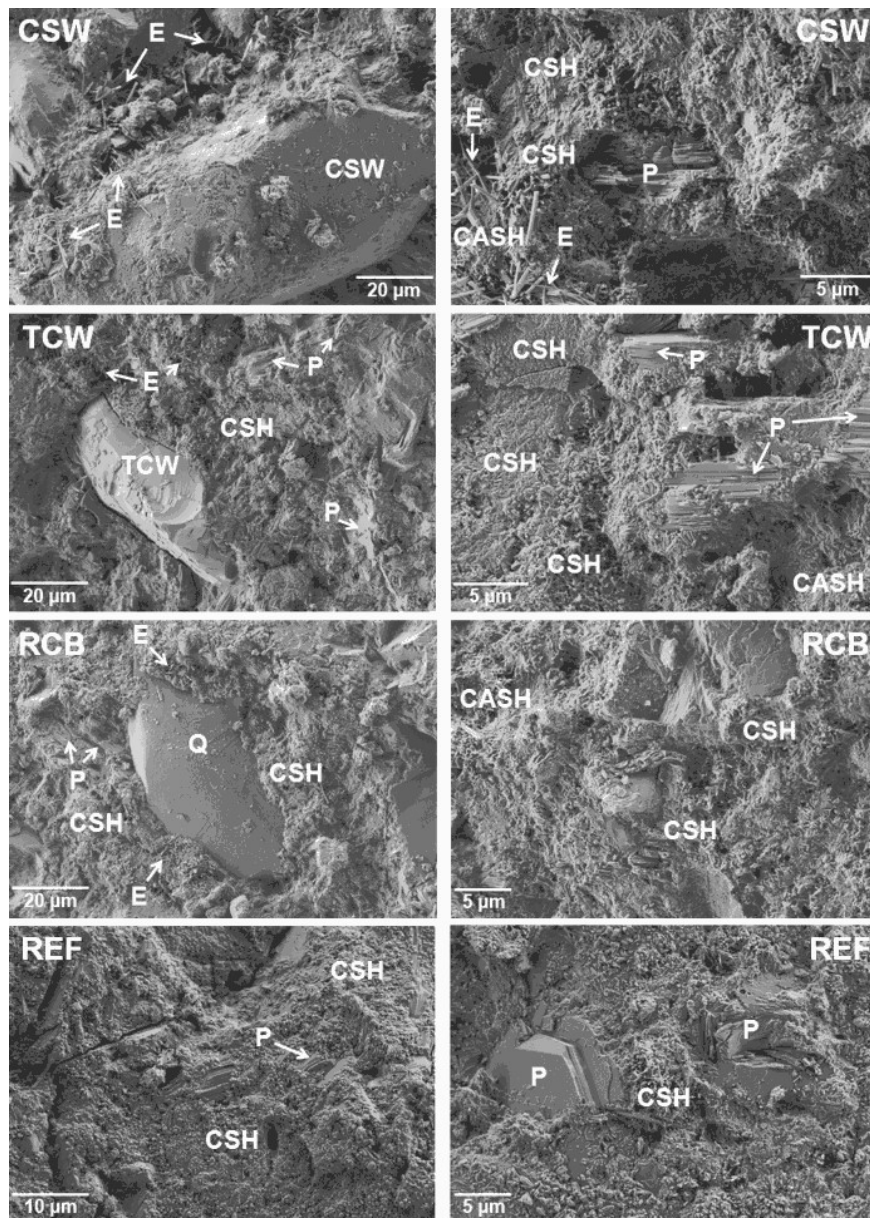
The 28-days XRD diffractograms contained signals attributed to unreacted larnite (R, $\beta\text{-Ca}_2\text{SiO}_4$, PDF #330302). The peaks originated by the diffraction of Portlandite (P, Ca(OH)_2 , PDF #040733) showed a similar intensity in all the pastes except that containing RCB. Given the lower amount of PC in the blended pastes, this corroborated that the particle effect originated by the ceramic waste accelerated cement hydration. In agreement with the fixed lime values (Figure 10), XRD results confirmed that RCB partially consumed Ca(OH)_2 via pozzolanic reactions.

542 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
 543 #411451), which came about through the reaction between tricalcium aluminate and calcium
 544 sulphate, both contributed by PC. Signals originated by calcite (C, CaCO_3 , PDF #050586)
 545 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
 546 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
 547 #360129) only appeared in the 28-days samples.
 548



549
 550 Figure 11. X-ray diffractograms of the pastes prepared with 0 and 25 wt.% RCB, TCW or CSW, cured
 551 at 20°C for 28 and 90 days. Q, quartz (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
 552 ($\text{Ca}(\text{OH})_2$); R, larnite ($\beta\text{-Ca}_2\text{SiO}_4$); C, calcite (CaCO_3); B, carboaluminate ($\text{Ca}_4\text{Al}_2\text{O}_6\text{CO}_3\cdot 11\text{H}_2\text{O}$); L,
 553 carboaluminate ($\text{Ca}_8\text{Al}_4\text{O}_{14}\text{CO}_2\cdot 24\text{H}_2\text{O}$)
 554

555 Figure 12 shows the microstructure of the reference paste and those containing 25 wt.%
 556 RCB, TCW or CSW, cured at 20°C for 28 days. As expected from the TGA and XRD tests,
 557 unreacted ceramic particles coexisted with plane hexagonal portlandite, ettringite needles
 558 and amorphous hydration products (CSH and CASH gels). These phases were differentiated
 559 both by their morphology and by their composition, determined by EDS microanalysis. The
 560 ceramic waste particles were completely surrounded by reaction products, and calcium
 561 silicate hydrate was the main reaction product observed.
 562



563
 564 Figure 12. FESEM images of the reference paste and those prepared with 25 wt.% RCB, TCW or
 565 CSW, cured at 20°C for 28 days. E: Ettringite; P: Portlandite; CSH: Calcium silicate hydrate; CASH:
 566 Calcium aluminosilicate hydrate; Q: Quartz; TCW: Tile ceramic waste; CSW: ceramic sanitary-ware
 567 waste

568 **3.6. Sustainability analysis**

569 The results obtained after running the basic sustainability analysis of the developed ceramic
 570 waste/PC blended binders are summarised in Table 5. As observed, the e-energy and e-CO₂
 571 values for the blended cements progressively lowered with increasing RCB, TCW or CSW
 572 contents. The E_s and C_s parameters, which considered the 28-day compressive strength,
 573 were calculated from the linear regression data (Table 4). In general terms, the E_s values for
 574 the blended cements were similar or slightly lower than those calculated for CEM I 42.5R,
 575 and were only higher when replacing 50 wt.% PC with ceramic waste. This was attributed
 576 mainly to two factors: 1) the relatively low compressive strength values exhibited by the 50
 577 wt.% blended mortars, in which the dilution effect involved small amounts of portlandite

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

582

583 The e-CO₂ values also lowered with increasing percentages of ceramic waste, which yielded
 584 an interesting reduction for the C_s parameter, especially when replacing 25 wt.% and 35
 585 wt.% PC. More specifically, the RCB/PC blended cements reduced C_s by 11.6% and 13.5%
 586 (compared to CEM I 42.5R) when using 25 wt.% and 35 wt.% RCB, respectively. These
 587 reductions were 7.3% and 8.6% with TCW and 7.0% and 8.2% with CSW (same PC
 588 replacement percentages). Due to the pozzolanic reaction evolution, these CO₂ emissions
 589 per tonne of cement and MPa further reduced with curing time. Consequently after 365
 590 curing days, the following C_s reductions were determined: 16.2% and 22.7% for 25 wt.% and
 591 35 wt.% RCB; 15.1% and 19.2% for the same amounts of TCW; 16.9% and 21.5% for the
 592 corresponding cements blended with CSW.

593

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

601

Table 5. E-energy, e-CO₂, E_s and C_s of the ceramic waste/PC blended cements

Cement	PC Replacement, wt. %	e-Energy, MJ/ton	e-CO ₂ , kg CO ₂ /ton	E_s *, MJ/ton · MPa	C_s *, kg 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

602

*Values obtained for 28-days curing mortars.

603 4. Conclusions

604

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

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

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

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

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

625 - The embodied energy and CO₂ emissions reduced with increasing replacements of
626 PC with RCB, TCW or CSW. After 365 curing days, the calculated CO₂ emissions per tonne
627 of used cement and MPa diminished from 15.1% to 16.9% (compared to PC CEM I 42.5R)
628 when replacing 25 wt.% PC with any these ceramic waste materials.

629

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

640

641 **Acknowledgements**

642 They also thank the Electron Microscopy Service of the Universitat Politècnica de València
643 and the Central Service for Scientific Instrumentation of the Universitat Jaume I of Castellón
644 for helping with the microstructural characterisation tests.

645 The authors are grateful to the Spanish Ceramic Tile Manufacturers' Association (ASCER)
646 for supporting this study through the collaboration agreement with Universitat Jaume I (*Aula*
647 *Cerámica*)

648

649 **References**

650 [1] CEMBUREAU, Activity report 2018. Built in concrete, made with cement, viewed online on 13
651 March 2020 < <https://cembureau.eu/news-views/publications/>>

652 [2] R. Kajaste, M. Hurme, Cement industry greenhouse gas emissions - Management options and
653 abatement cost, *J. Clean. Prod.* 112 (2016) 4041–52. doi:10.1016/j.jclepro.2015.07.055.

654 [3] S. Mohammed, Processing, effect and reactivity assessment of artificial pozzolans obtained from
655 clays and clay wastes: A review, *Constr. Build. Mater.* 140 (2017) 10–19.
656 doi:10.1016/j.conbuildmat.2017.02.078.

657 [4] L. Rodier, H. Savastano, Use of glass powder residue for the elaboration of eco-efficient
658 cementitious materials, *J. Clean. Prod.* 184 (2018) 333–341. doi:10.1016/j.jclepro.2018.02.269.

659 [5] R. Maddalena, J.J. Roberts, A. Hamilton, Can Portland cement be replaced by low-carbon
660 alternative materials? A study on the thermal properties and carbon emissions of innovative cements,
661 *J. Clean. Prod.* 186 (2018). doi.org/10.1016/j.jclepro.2018.02.138.

662 [6] M. S. Imbabi, C. Carrigan, S. McKenna, Trends and developments in green cement and concrete
663 technology, *Int. J. Sustain. Built Environ.* 1 (2012), 194–216.
664 <https://doi.org/10.1016/j.ijsbe.2013.05.001>.

665 [7] P.C. Jacoby, F. Pelisser, Pozzolan effect of porcelain polishing residue in Portland cement, *J.*
666 *Clean. Prod.* 100 (2015) 84–88. doi:10.1016/j.jclepro.2015.03.096.

667 [8] H. Binici, S. Kapur, J. Arocena, H. Kaplan, Cem. Concr. Compos. The sulphate resistance of
668 cements containing red brick dust and ground basaltic pumice with sub-microscopic evidence of intra-
669 pore gypsum and ettringite as strengtheners, *Cem.Concr.Compos.* 34 (2012) 279–287.
670 doi:10.1016/j.cemconcomp.2011.10.001.

671 [9] R.D.T. Filho, J.P. Gonçalves, B.B. Americano, E.M.R. Fairbairn, Potential for use of crushed waste
672 calcined-clay brick as a supplementary cementitious material in Brazil, *Cem. Concr. Res.* 37(9):1357-
673 1365 (2007). doi:10.1016/j.cemconres.2007.06.005.

674 [10] F. Puertas, A. Barba, M. F. Gazulla, M. P. Gómez, M. Palacios, S. Martínez-Ramírez. Ceramic
675 wastes as raw materials in portland cement clinker fabrication: characterization and alkaline activation
676 *Mater. Constr.*, Vol. 56, 281, 73-84, enero-marzo 2006 ISSN: 0465-2746.
677 <https://doi.org/10.3989/mc.2006.v56.i281.94>

678 [11] M.A. Mas, J. Monzó, J. Payá, L. Reig, M.V. Borrachero, Ceramic tiles waste as replacement
679 material in Portland cement, *Adv. Cem. Res.*, 2016, 28(4), 221–232.
680 <http://dx.doi.org/10.1680/jadcr.15.00021>.

681 [12] N. Ay, M. Ünal, The use of waste ceramic tile in cement production, *Cem.Concr. Res.* 30 (2000)
682 497–499.

683 [13] M. I. Sánchez de Rojas, F. Marín, J. Rivera, M. Frías, Morphology and Properties in Blended
684 Cements with Ceramic Wastes as a Pozzolan Material, *J. Am. Ceram. Soc.*, 89 [12] 3701–3705
685 (2006), DOI: 10.1111/j.1551-2916.2006.01279.x.

686 [14] L.A. Pereira-De-Oliveira, J.P. Castro-Gomes, P.M.S. Santos, The potential pozzolanic activity of
687 glass and red-clay ceramic waste as cement mortars components, *Constr. Build. Mater.* 31 (2012)
688 197–203. doi:10.1016/j.conbuildmat.2011.12.110.

689 [15] L. Reig, M. M. Tashima, L. Soriano, M. V. Borrachero, J. Monzó, J. Payá, Alkaline Activation of
690 Ceramic Waste Materials, *Waste Biomass Valor.* (2013) 4:729–736. DOI 10.1007/s12649-013-9197-z.

691 [16] L. Turanli, F. Bektas, P.J.M. Monteiro, Use of ground clay brick as a pozzolanic material to reduce
692 the alkali – silica reaction, *Cem. Concr. Res.* 33 (2003) 1539–1542. doi:10.1016/S0008-
693 8846(03)00101-7.

694 [17] G. Baronio, L. Binda, Study of the pozzolanicity of some bricks and clays, *Constr. Build. Mater.* 11
695 (1) (1997) 41-46.

696 [18] Ministry of Public Works. RC-16. Instrucción para la Recepción de Cementos (Cement Reception
697 Instruction). Madrid, Spain; 2016.

698 [19] G. Hammond, C. Jones. Embodied carbon. The inventory of carbon and energy (ICE), 2011.

699 [20] <https://energia.gob.es/es-es/Paginas/index.aspx> (accessed on 13 June 2020)

700 [21] R. Xiao, Y. Ma, X. Jiang, M. Zhang, Y. Zhang, Y. Wang, B. Huang, Q. He, Strength,
701 microstructure, efflorescence behavior and environmental impacts of waste glass geopolymers cured
702 at ambient temperature, *J. Clean. Prod.* 252 (2020). doi.org/10.1016/j.jclepro.2019.119610.

703 [22] C. Zanelli, M. Raimondo, G. Guarini, M. Dondi, The vitreous phase of porcelain stoneware:
704 Composition, evolution during sintering and physical properties, *J. Non. Cryst.Solids.* 357 (2011)
705 3251–3260. doi:10.1016/j.jnoncrysol.2011.05.020.

706 [23] The Brick industry association, Technical notes on brick construction. Manufacturing of brick.
707 December 2006. Available online: <http://www.gobrick.com/portals/25/docs/technical%20notes/tn9.pdf>.
708 (accessed on 07 June 2018).

709 [24] A. Bernasconi, V. Diella, A. Pagani, A. Pavese, F. Francescon, K. Young, J. Stuart, L. Tunnicliffe, The
710 role of firing temperature, firing time and quartz grain size on phase-formation, thermal dilatation and
711 water absorption in sanitary-ware vitreous bodies, *J. Eur. Ceram. Soc.*, Volume 31, Issue 8, July 2011,
712 Pages 1353-1360, <https://doi.org/10.1016/j.jeurceramsoc.2011.02.006>

713 [25] L. Reig, M.M. Tashima, M.V. Borrachero, J. Monzó, C.R. Cheeseman, J. Payá, Properties and
714 microstructure of alkali-activated red clay brick waste, *Constr. Build. Mater.* 43 (2013) 98-106.
715 <https://doi.org/10.1016/j.conbuildmat.2013.01.031>.

716 [26] L. Reig, L. Soriano, M. M. Tashima, M. V. Borrachero, J. Monzó, J. Payá, Influence of calcium
717 additions on the compressive strength and microstructure of alkali-activated ceramic sanitary-ware, *J.*
718 *Am. Ceram. Soc.* 101 (7) (2018) 3094-3104. <https://doi.org/10.1111/jace.15436>.

719 [27] A. M. Pitarch, L. Reig, A. E. Tomás, F. J. López, Effect of tiles, bricks and ceramic sanitary-ware
720 recycled aggregates on structural concrete properties, *Waste Biomass Valor.*,
721 <https://doi.org/10.1007/s12649-017-0154-0>.

722 [28] C. Medina, P.F.G. Banfill, M.I.S. De Rojas, M. Frías, Rheological and calorimetric behaviour of
723 cements blended with containing ceramic sanitary ware and construction/demolition waste, *Constr.*
724 *Build. Mater.* 40 (2013) 822–831. doi:10.1016/j.conbuildmat.2012.11.112.

- 725 [29] A. Naceri, M.C. Hamina, Use of waste brick as a partial replacement of cement in mortar, *Waste*
726 *Manag.* 29 (2009) 2378–2384. doi:10.1016/j.wasman.2009.03.026.
- 727 [30] UNE (2013) EN 450-1:2013: Fly ash for concrete. Part 1: Definition, specifications and conformity
728 criteria.
- 729 [31] S. Wild, J.M. Khatib, M. O'Farrell. Sulphate resistance of mortar, containing ground brick clay
730 calcined at different temperatures. *Cem. Concr. Res.*, Vol. 27, No. 5, pp. 697.709,1997.
731 [https://doi.org/10.1016/S0008-8846\(97\)00059-8](https://doi.org/10.1016/S0008-8846(97)00059-8).
- 732 [32] A.E. Lavat, M. a. Trezza, M. Poggi, Characterization of ceramic roof tile wastes as pozzolanic
733 admixture, *Waste Manag.* 29 (2009) 1666–1674. doi:10.1016/j.wasman.2008.10.019.
- 734 [33] J. Payá, J. Monzó, M. V. Borrachero, S. Velázquez, Evaluation of the pozzolanic activity of fluid
735 catalytic cracking catalyst residue (FC3R). Thermogravimetric analysis studies on FC3R-Portland
736 cement pastes, *Cem. Concr. Res.* 33 (4) (2003) 603–609.
- 737
- 738
- 739

740 Figure Captions:
741
742 Figure 1. The original RCB, TCW and CSW ceramic waste materials
743
744 Figure 2. X-ray diffractogram of the ceramic waste materials. Q: Quartz (SiO_2); M: Mullite
745 ($\text{Al}_6\text{Si}_2\text{O}_{13}$); A: Albite ($\text{NaAlSi}_3\text{O}_8$); m: Microcline (KAlSi_3O_8); D: Diopside ($\text{CaMg}(\text{SiO}_3)_2$); R:
746 Rankinite ($2\text{SiO}_2 \cdot 3\text{CaO}$)
747
748 Figure 3. Scanning electron microscope images of the milled RCB, TCW and CSW particles
749
750 Figure 4. Consistency of the blended RCB, TCW and CSW PC pastes
751
752 Figure 5. Setting time of the blended RCB, TCW and CSW PC pastes
753
754 Figure 6. Compressive strength of the mortars prepared with 0 to 50 wt.% RCB, TCW or
755 CSW, cured at 20°C from 3 to 365 days
756
757 Figure 7. Strength activity index of the mortars prepared with 0 to 50 wt.% RCB, TCW or
758 CSW, cured at 20°C from 3 to 365 days
759
760 Figure 8. Strength gain of the mortars prepared with 0 to 50 wt.% RCB, TCW or CSW, cured
761 at 20°C from 3 to 365 days
762
763 Figure 9. DTG curves of the pastes prepared with 0 and 25 wt.% RCB, TCW or CSW, cured
764 at 20°C for 28 and 90 days.
765
766 Figure 10. Percentage of fixed $\text{Ca}(\text{OH})_2$ determined for the ceramic waste/PC blended
767 pastes prepared with 25 wt.% RCB, TCW or CSW, cured at 20°C for 28 and 90 days
768
769 Figure 11. X-ray diffractograms of the pastes prepared with 0 and 25 wt.% RCB, TCW or
770 CSW, cured at 20°C for 28 and 90 days. Q, quartz (SiO_2); E, ettringite
771 ($\text{Ca}_6\text{Al}_2(\text{SO}_4)_3(\text{OH})_{12} \cdot 26\text{H}_2\text{O}$); P, portlandite ($\text{Ca}(\text{OH})_2$); R, larnite ($\beta\text{-Ca}_2\text{SiO}_4$); C, calcite
772 (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
773 ($\text{Ca}_8\text{Al}_4\text{O}_{14}\text{CO}_2 \cdot 24\text{H}_2\text{O}$)
774
775 Figure 12. FESEM images of the reference paste and those prepared with 25 wt.% RCB,
776 TCW or CSW, cured at 20C for 28 days. E: Ettringite; P: Portlandite; CSH: Calcium silicate

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