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Characterisation and standardisation of different-origin end-of-life building materials for assessment of circularity

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Abstract

Construction and demolition waste (CDW) management and recycling practices are crucial for transitioning to a circular economy. This study focuses on the detailed characterization of CDWs, including hollow brick (HB), red clay brick (RCB), roof tile (RT), concrete (C), and glass (G), collected from seven different sites. The CDWs were characterized based on particle size distribution, chemical composition, and crystalline nature. Pozzolanic activity was evaluated through compressive strength measurements of cement mortars with 20% cement replacement by CDWs at 7, 28, and 90 days. The results showed that clayey CDWs exhibited similar physical/chemical properties and crystalline structures. Compositions of Cs varied significantly based on their original materials. CDWs satisfied the minimum strength activity index for supplementary cementitious materials, with pozzolanic activity influenced by fineness and SiO₂+Al₂O₃ contents. The average strength activity indexes for HB, RCB, RT, C, and G were 84.5%, 86.3%, 83.4%, 80.7%, and 75.8%, respectively. Clayey CDWs contributed to mechanical strength development, while Cs' contribution was related to hydration of unreacted cementitious particles. G exhibited the weakest pozzolanic activity due to its coarser particle size. Overall, CDWs demonstrated suitable properties for use as supplementary cementitious materials in PC-based systems.

Keywords: Construction and demolition waste (CDW); Characterization; Physical and chemical properties; Circular economy; Pozzolanic activity; UN SDG 11 and 13.

1. Introduction

In today's world, in addition to the designs that make human life easier, the development of eco-friendly materials is of great importance. Every step that needs to be taken regarding the environmental, socio-economic and sustainability aspects is therefore critical to avert the issues like rapid consumption of natural resources, greenhouse gas emissions, and global warming.

31 Within this context, one of the leading materials to consider is concrete, which is the second
32 most used material in the world after water (Scrivener and Kirkpatrick, 2008). Within the
33 composition of traditional concrete, Portland cement (PC), which acts as the main binder for
34 the material, is critically important, since its production requires high energy and raw materials
35 consumption creating huge amounts of carbon emissions. For instance, a ton of manufactured
36 PC clinker requires 850 kcal of energy and 1.7 tons of natural resources and is responsible for
37 0.94 tons of CO₂ release (Rashad and Zeedan, 2011; Praneeth et al., 2020). PC production is
38 accountable for 5-8% of the total anthropogenic CO₂ in the atmosphere (Turner and Collins,
39 2013; Chan et al., 2015; Bakhtyar et al., 2017). Despite its drawbacks, the place of traditional
40 concrete, and relatedly PC production, did not change over the years and the material is still
41 the most commonly used construction material in the world (Stafford et al., 2016). Considering
42 the increased number of activities related to the construction and demolition sector along with
43 the requirements for the repetitive repair/retrofit/rehabilitation activities creating additional
44 waste and necessitating more and more PC-based concrete-like materials, the development of
45 more eco-friendly and sustainable alternative materials is necessary (Monteiro et al., 2017).

46 As abovenoted, the construction and demolition sector and activities related to it are
47 booming in recent years. This can be mainly related to the continuously growing urban
48 population and developing industries/economies of countries around the globe. Despite its
49 commonness, the construction and demolition sector is one of the biggest contributors to solid
50 waste production globally (Eurostat, 2015). The sector leads to different waste streams, which
51 can be collectively termed as, “Construction and Demolition Waste (CDW)”. It has been
52 reported that over 10 billion tons of CDW are generated worldwide each year (Wu et al., 2019),
53 of which about 700 million tons are produced by the United States (Jain et al., 2015), 800
54 million tons by Europe (Ajayi et al., 2016) and 2.3 billion tons by China (Zheng et al., 2017).
55 These wastes, which are likely to incorporate toxic substances, are usually dumped in clean
56 landfills, thus jeopardizing the health of individuals and contaminating the surrounding
57 environment (Roussat et al., 2008). Although the recycling of CDW has attracted the attention
58 of the whole world in recent years, it is nevertheless estimated that more than 35% of CDWs
59 are routinely diverted directly to clean landfills without any further process (Menegaki and
60 Damigos, 2018). Therefore, it is crucial to handle CDW in an appropriate manner for the sake
61 of environmental, social, and economic benefits. This is important both for reducing the
62 quantities of CDW going to clean landfills and the amount of concrete material that is,
63 otherwise, going to be used to build/repair/maintain new and/or existing infrastructures.

64 In order to tackle the CDW problem, transforming CDW-based materials into sustainable
65 construction materials has attracted the growing attention of researchers. One of the recently
66 trending ways to utilize CDWs is producing alkali-activated materials/geopolymers (Yildirim
67 et al., 2021,2022; Ilcan et al., 2023; Mir et al., 2023; Ozcelikci et al., 2023a). A geopolymer is
68 a binder obtained by the alkaline activation of solid alumina- and silica-containing precursors
69 (Zhang et al., 2014). Considering that the most of CDW-based components are rich in alumina
70 and silica, components derived from CDW can preferably be used in geopolymerization. As
71 another trending method to recycle CDWs, partial replacement of CDWs with the clinker
72 and/or PC is considered. A study replaced clinker with waste brick at various rates (0-20%)
73 and found that higher waste brick content reduced setting times and grinding requirements
74 (Naceri and Hamina, 2009). While the mechanical properties decreased after 7 and 28 days,
75 they improved after 90 days, particularly with a 10% brick replacement ratio. In a study by Li
76 et al. (2019), the effects of adding clay brick waste to mortars were evaluated. The compressive
77 strength increased when cement paste was replaced by the waste, while the opposite trend was
78 observed with cement powder replacement. The authors concluded that replacing paste with
79 clay brick waste maintained a constant water/cement ratio, benefiting from the filler effect and
80 promoting cement hydration. However, replacing cement powder led to a higher effective
81 water/cement ratio, resulting in a negative effect.

82 Research conducted worldwide on the use of CDWs in cement-based and geopolymer
83 systems has revealed variations in the performance of the final products, even when the mixture
84 designs and curing processes are kept consistent. Notably, studies involving red clay brick
85 activation using 10% Na_2O and thermal curing at around 70°C for 24 hours resulted in
86 compressive strengths of approximately 10 MPa (Robayo-Salazar et al., 2017) and 6 MPa
87 (Ulugöl et al., 2021) for geopolymers, respectively. These discrepancies can be attributed to
88 the differences in the chemical composition of the red clay bricks obtained from different
89 regions, despite similar volume mean diameters (both under $25\ \mu\text{m}$). Similarly, studies on brick
90 waste-based geopolymer pastes using sodium hydroxide and sodium silicate as alkaline
91 activators demonstrated compressive strengths of around 25 MPa (Komnitsas et al., 2015) and
92 38 MPa (Mahmoodi et al., 2020) under similar curing conditions. Differences were also
93 observed in studies where CDWs were used as partial replacements in cement-based systems.
94 For instance, the compressive strength of a mixture with a 15% replacement ratio outperformed
95 a control sample (Oliveira et al., 2020), while another study reported that mixtures with
96 replacement ratios of 20% and 30% exceeded the control mixture, reaching a maximum

97 compressive strength of 53 MPa after 28 days (Mucsi et al., 2021). The optimum replacement
98 rate of CDWs is highly dependent on their origin and properties, underscoring the need for
99 standardized characterization and technological guidelines (Cheng, 2016).

100 Considering the fact that construction and demolition is one of the five priority sectors
101 designated by the European Action Plan, the necessary steps to shift CDWs from the "make-
102 use-dispose" linear economy model to the circular economy model must be taken properly. In
103 order to the integration of CDW into the circular economy, it must go through stages such as
104 being properly managed, identified and ultimately included in the value chain by demonstrating
105 the mechanical, economical, and environmental advantages of recycling/upcycling activities
106 (Zhang et al., 2022; Ozcelikci et al., 2023b). Therefore, as the major target of this study, the
107 characterization of different-origin CDWs for their further identification can be clearly
108 considered one of the most important steps. The significance of this research lies in examining
109 and uncovering the underlying mechanisms behind the variations in mechanical performance
110 observed by researchers worldwide, despite similar formulations. By comprehensively
111 investigating the effects of CDWs with different origins on the characteristics of the final
112 product, this study aims to shed light on the factors influencing these variations. This, in turn,
113 paves the way for optimizing CDW recycling practices by gaining a better understanding of
114 the material and achieving maximum performance efficiency. In this study, hollow brick, red
115 clay brick, roof tile, concrete and glass wastes from seven different demolition sites were
116 considered under the same preparation and testing conditions. Particle size distribution,
117 chemical composition and crystalline nature of each sample of CDW materials were thoroughly
118 analyzed and compared to its strength activity indexes at 7, 28 and 90 days to determine their
119 effectiveness as cementitious constituents. Microstructural analyses such as Scanning electron
120 microscopy and Energy-dispersive X-ray spectroscopy (SEM/EDX) were conducted on the
121 selected mixtures to validate and correlate obtained results. Additionally, the
122 Thermogravimetry (TG/DTG) analysis was carried out to examine the reactivity and thermal
123 behavior of CDW substitution in the PC system. The results obtained from this analysis provide
124 valuable insights into the changes in reactivity, hydration characteristics, and thermal
125 decomposition patterns, enabling a better understanding of CDW's performance as a PC
126 substitute. The results are expected to provide an important database for industrials and
127 researchers interested in the mass recycling of CDW materials, especially with the confusion
128 and disagreement caused by the inconsistent results in the literature.

129 **2. Experimental Program**

130 **2.1. Materials**

131 CDW-based materials, including hollow brick (HB), red clay brick (RCB), roof tile (RT),
132 concrete (C) and glass (G), were collected from seven urban transformation sites in Turkey
133 after selective demolition. Fig. 1 shows a typical site with different CDW-based materials such
134 as HB, RCB, RT, C and G. The same crushing/grinding procedures were applied for all raw
135 CDWs. No special measures were taken to bring the waste materials to the same particle size
136 distribution. As can be expected, in most demolition sites, the waste materials are present
137 together and it is much less labor- and energy-intensive to recycle them without a specific
138 preparation method. They were first subjected to initial crushing in a jaw crusher and reduced
139 in size. Each CDW-based material was then loaded into a ball mill and grinded for an hour.
140 Since the study deals with the characterization of different CDWs, the physical, chemical and
141 mechanical properties of the wastes were presented in the following sections.



142

143 **Fig. 1** A typical demolition site in Turkey and representative images of CDW-based materials
144 before crushing, after crushing and after grinding

145 **2.2. Methods**

146 Analyses and tests for the determination of chemical composition, particle size distribution,
147 crystalline structure and pozzolanic activity of powdery CDWs were performed. X-ray
148 fluorescence (XRF) analysis was utilized to determine the chemical composition of the
149 materials. Particle size distributions were determined by using dry laser granulometry method
150 (Malvern Mastersizer Scirocco 2000 assembled with a hopper instrument). X-ray diffraction
151 (XRD) analysis was performed to observe the crystalline nature of the materials, in which
152 parameters such as scan range of $5^{\circ} \leq 2\theta \leq 55^{\circ}$, 2θ step length of 0.033° , a scanning step time of
153 30.48 s, and a wavelength $K\alpha_1$ of copper ($\lambda=1.5406\text{\AA}$) were used. Mortar specimens were
154 produced in accordance with the ASTM C618-19 standard to measure the pozzolanic activity
155 of materials (ASTM C618, 2019). The test was carried out on a control mixture proportioned
156 with 500 g of Portland cement, 1375 g of sand and 242 g of water. Then, at the same amount
157 of water usage as the control mixture, CDW-based compositions were designed by substituting
158 20% of PC with HB, RCB, RT, C or G. Compressive strength measurements were performed
159 after 7, 28 and, 90 days by using three separate 50-mm cubic specimens for each age, in
160 accordance with the ASTM C39-2021 standard (ASTM C39/39M, 2021). A loading device
161 with 100-ton capacity and a loading rate of 0.9 kN/s was used for these tests. The strength
162 activity index (SAI – %) was calculated as the ratio between the compressive strength of the
163 CDW-substituted mortars and the control mixture. For in-depth microstructural
164 characterization of selected mixtures, Scanning electron microscopy and Energy-dispersive X-
165 ray spectroscopy (SEM/EDX) and Thermogravimetry (TG/DTG) were conducted. Specimens
166 with dimensions less than 1 cm were used for the SEM/EDX analysis. To ascertain the
167 temperatures at which the materials degraded, TG/DTG analyzes were carried out on the 15
168 mg powder samples at incremental rates of $10^{\circ}\text{C}/\text{min}$ from 25 to 1000°C .

169 **3. Results and Discussion**

170 ***3.1. Chemical composition of CDW-based materials***

171 Table 1 presents the chemical compositions of CDW-based materials obtained from seven
172 different demolition sites. The main elements detected in the composition of HBs were oxygen,
173 silicon, aluminum, iron and calcium. SiO_2 content of HBs varied between 42.1-57.7%, while
174 Al_2O_3 was in the range of 13.7-16.2%. RCBs had SiO_2 and Al_2O_3 contents in the range of 45.8-
175 59.9% and 13.5%-18.3%, respectively. The chemical composition of RTs was also found to be
176 close to other clayey materials with SiO_2 and Al_2O_3 ranges between 46.2-55.3% and 12.6-

177 16.3%, respectively. The properties of bricks and tiles largely depend on the manufacturing
178 method and properties of raw materials used in their production (Brick Industry Association,
179 2006). Raw materials of bricks and tiles are soils containing high amounts of clay together with
180 some silt. Bricks and tiles generally contain 50-60% silica, 20-30% alumina, 2-5% lime, <7%
181 iron oxide and <1% magnesia (Pumnia et al., 2003). In the selection of raw materials, the
182 quality of clay-based soils is one of the most important parameters controlling the performance
183 of tiles and bricks. Illite, kaolinite and a reduced amount of montmorillonite are usually used
184 as clay minerals (Sverguzova et al., 2016). Based on the aforementioned statements, the small
185 differences registered in the chemical compositions of the clayey CDWs can be due to the type
186 of raw materials used in materials' production together with impurities that may have been
187 contained throughout such products' service life.

188 Unlike clayey CDWs, clear differences can be seen in the chemical compositions of concrete
189 wastes. This was expected since the original concretes are mostly with diverse strength classes
190 and components and subjected to different service exposure conditions. For example, the
191 aggregates, which constitute a significant portion of the collected concrete waste, are very
192 likely to be obtained from diverse sources and origins (*e.g.*, siliceous, calcareous). Considering
193 that limestone (calcareous) aggregates are with low contents of SiO₂ and high contents of CaO
194 and loss on ignition (LoI) (Aquino et al., 2010), concrete wastes collected from K and L sites
195 seem to be produced with calcerous aggregates. As for materials from X, Y, and Z sites, which
196 contain SiO₂ in the range of 54.7-56.9%, the original aggregates are likely to be siliceous, such
197 as quartz and albite (Medina et al., 2015; Bui et al., 2018). Regarding the concrete wastes from
198 M and N sites with close SiO₂ and CaO contents, they are likely to be prepared with silico-
199 calcareous aggregates. It needs to be stated that the compositions of concrete wastes are not
200 only dependent on the possible types of aggregates, but also on the aggregate-to-cement ratio,
201 type/chemical composition of PC, and physical/chemical changes that can take place during
202 the service life of these materials (Sánchez-Cotte et al., 2020).

203 Considering the limited availability of glass in demolition sites compared to other CDWs,
204 only a single type of glass obtained from the K site was characterized in the study. Another
205 reason for the selection of glass from a single site was due to that comparable glass types were
206 utilized in residential constructions in general, with no significant differences in the materials'
207 chemical compositions according to the literature (Jani and Hogland, 2014; Olofinnade et al.,
208 2017). The chemical composition of glass waste comprised SiO₂, CaO and Na₂O
209 concentrations of 65.4%, 9.0%, and 12.0% respectively, together with other minor oxides. In

210 accordance with the literature, this composition was found to be rather similar to that of other
 211 glass wastes obtained from different urban transformation and demolition sites (Varshneya and
 212 Mauro, 2019). As inferred from Table 1, the amount of silica in glass waste is slightly higher
 213 than other CDWs, although the amount of alumina is remarkably lower than all CDWs.

214 **Table 1** Chemical composition of CDW-based clayey materials.

Demolition site notation	Chemical composition (%)								
	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	K ₂ O	Na ₂ O	SO ₃	LoI*
K-HB	54.0	16.2	7.62	6.82	2.58	2.99	0.34	0.69	3.43
L-HB	52.5	14.2	12.1	4.53	5.10	1.76	0.80	1.38	0.59
M-HB	56.8	15.8	8.24	3.13	2.41	2.88	0.32	1.48	0.77
N-HB	42.1	13.8	11.8	6.42	6.45	1.55	1.45	1.46	1.43
X-HB	48.9	13.7	11.0	7.44	4.82	2.07	0.37	1.47	3.02
Y-HB	52.5	14.9	8.59	4.06	2.65	2.74	0.44	0.52	1.99
Z-HB	57.7	14.7	7.90	3.97	2.24	2.43	-	0.42	3.15
K-RCB	55.1	16.7	8.20	3.59	2.63	2.94	0.53	3.70	1.51
L-RCB	55.0	15.5	11.7	2.79	2.86	2.42	0.26	0.21	0.26
M-RCB	59.2	18.3	7.72	2.78	2.35	2.77	0.42	1.41	0.73
N-RCB	45.8	13.5	11.9	7.81	5.55	1.88	0.98	1.88	3.90
X-RCB	52.6	14.8	8.31	4.64	2.78	2.45	0.69	0.95	2.92
Y-RCB	57.6	14.4	8.28	4.70	2.53	2.14	0.77	1.93	1.45
Z-RCB	59.9	13.6	7.40	5.20	2.27	2.02	0.93	2.31	1.30
K-RT	47.7	13.7	14.0	7.62	6.54	1.18	0.69	0.72	0.69
L-RT	51.6	13.2	8.95	9.83	4.30	1.98	0.59	0.86	2.73
M-RT	53.4	16.3	9.49	5.27	3.64	2.78	0.39	1.61	1.67
N-RT	46.2	12.6	12.1	9.88	5.41	1.16	1.13	0.40	2.59
X-RT	50.8	14.4	10.5	7.09	4.55	2.28	0.46	0.83	2.13
Y-RT	55.3	14.8	8.28	6.69	4.72	2.11	1.04	1.16	1.45
Z-RT	54.7	13.2	9.28	6.90	4.01	1.92	0.68	0.54	2.20
K-C	4.34	1.24	0.76	49.7	0.81	0.17	0.02	-	37.7
L-C	4.16	1.34	0.87	50.2	0.82	0.16	0.02	-	38.7
M-C	37.4	10.7	3.82	21.2	1.29	2.22	1.96	0.54	19.7
N-C	34.8	4.36	3.45	27.2	4.46	0.79	0.19	1.32	22.0
X-C	56.9	8.76	4.01	11.8	1.50	1.53	1.16	0.85	9.76
Y-C	56.0	8.88	4.23	11.8	1.51	1.51	1.03	0.72	8.84
Z-C	54.7	9.76	4.22	11.5	1.52	1.53	2.02	0.80	8.94
K-G	65.4	0.98	0.54	8.97	3.73	0.14	12.0	0.65	1.60

215 *: Loss on ignition

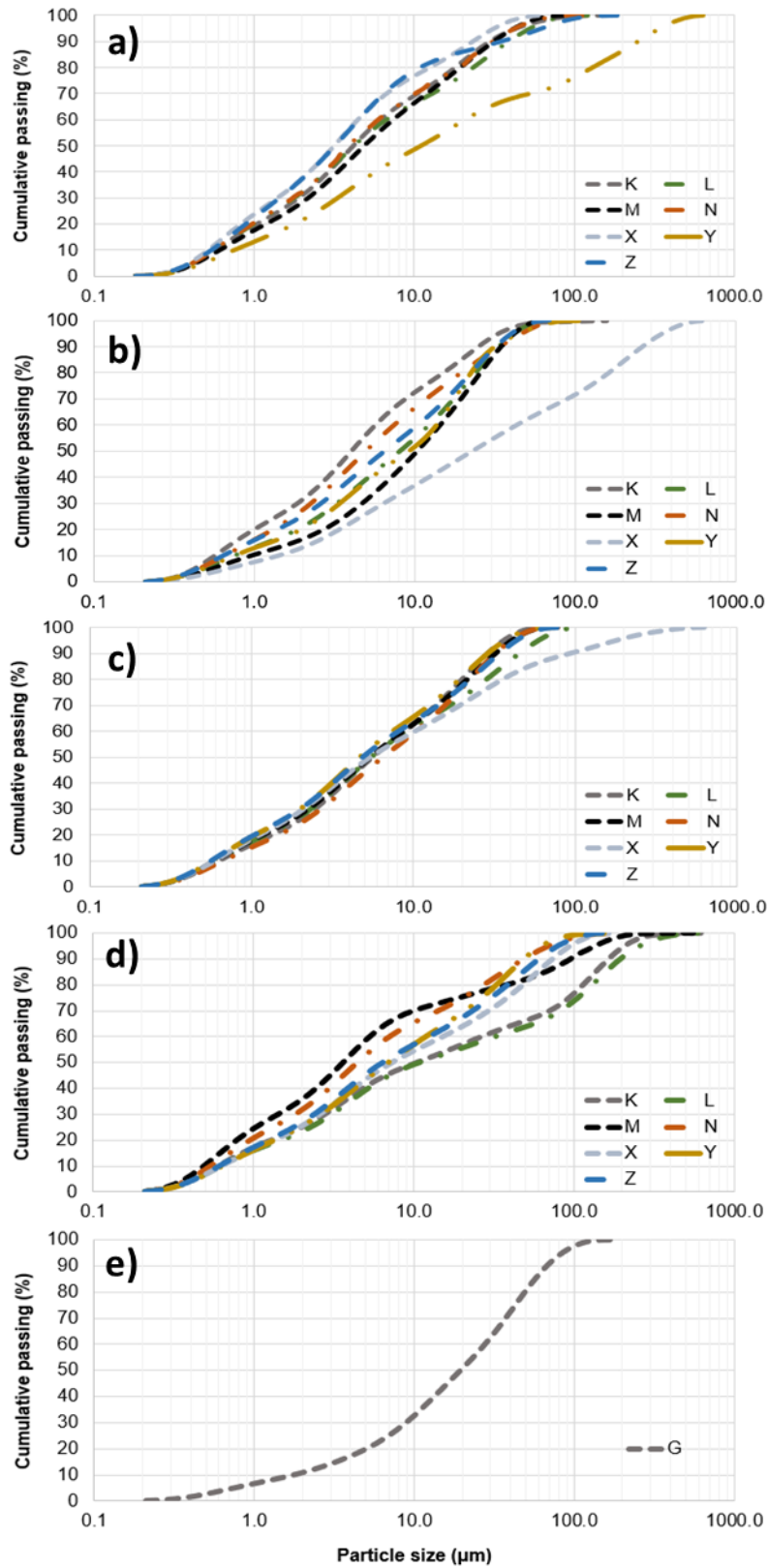
216 3.2. Particle size distribution of CDW-based materials

217 Particle size distributions of CDW-based materials subjected to the same crushing and
 218 grinding processes are presented in Fig. 2. In addition, their related d(0.1), d(0.5), and d(0.9)
 219 particle sizes are presented in Table 2. The particle size distributions of most clayey materials
 220 were found to be similar, except for HB, RCB, and RT obtained from sites Y, X, and X,
 221 respectively, which presented some differences. This can be also seen from the particle
 222 diameter results shown in Table 2. For example, the d(0.5) values of HB from Y site and RCB

223 from X site were more than three times higher than those of other HBs and RCBs; whereas the
224 $d(0.9)$ value of RT from X site was almost three times higher than other RTs. These differences
225 registered under the same crushing and grinding conditions can be due to various brittleness
226 levels and impurities of each collected sample, since no clear relation was noticed with their
227 respective chemical compositions. It can be stated that the grindability and its associated
228 particle size distribution can be greatly affected by material characteristics such as strength and
229 nature, brittleness, hardness, smoothness, stickiness and also moisture content (Meghwal and
230 Goswami, 2013). Also, the grindability is highly influenced by the presence of different phases
231 such as quartz in varied amounts and intensities (Asensio et al., 2016).

232 According to the particle size distributions of concrete wastes presented in Fig. 2, samples
233 from K and L sites showed small dissimilarities than other concrete wastes. Although the $d(0.1)$
234 values were similar for all concrete wastes, the samples from K and L sites presented higher
235 $d(0.5)$ and $d(0.9)$ values, implying also coarser particle sizes (Table 2). By comparing these
236 results with the chemical composition of concrete wastes, samples from K and L sites were
237 shown to be produced with calcareous aggregates. Thus, the coarser particle size distribution
238 of concrete wastes from K and L sites can be related to the possible agglomeration of limestone
239 particles during pulverization, which reduced their grinding efficiency (Ghiasvand et al., 2015).

240 According to the particle size distribution results (Fig. 2), glass waste has coarser particle
241 size distribution than most of other CDW-based materials. This can be confirmed from the
242 $d(0.5)$ value, which is more than two times higher than those of all CDWs, except for RCB
243 obtained from X site. The amorphous nature of glass waste is likely to be one of the important
244 reasons for its coarser particle size obtained under the same crushing/grinding procedures
245 applied to other CDWs. In accordance with (Stekla, 2016), brittle and amorphous nature of
246 glass causes it to have a sharp-edged grain morphology causing it to have a higher tendency of
247 agglomeration during grinding. In conclusion, its grindability is poorer compared to other
248 CDW-based materials, which are softer than glass (Ulugöl et al., 2021).



249

250 **Fig. 2** Particle size distribution of CDW-based clayey materials, a) HB; b) RCB; c) RT; d) C;

251

e) G.

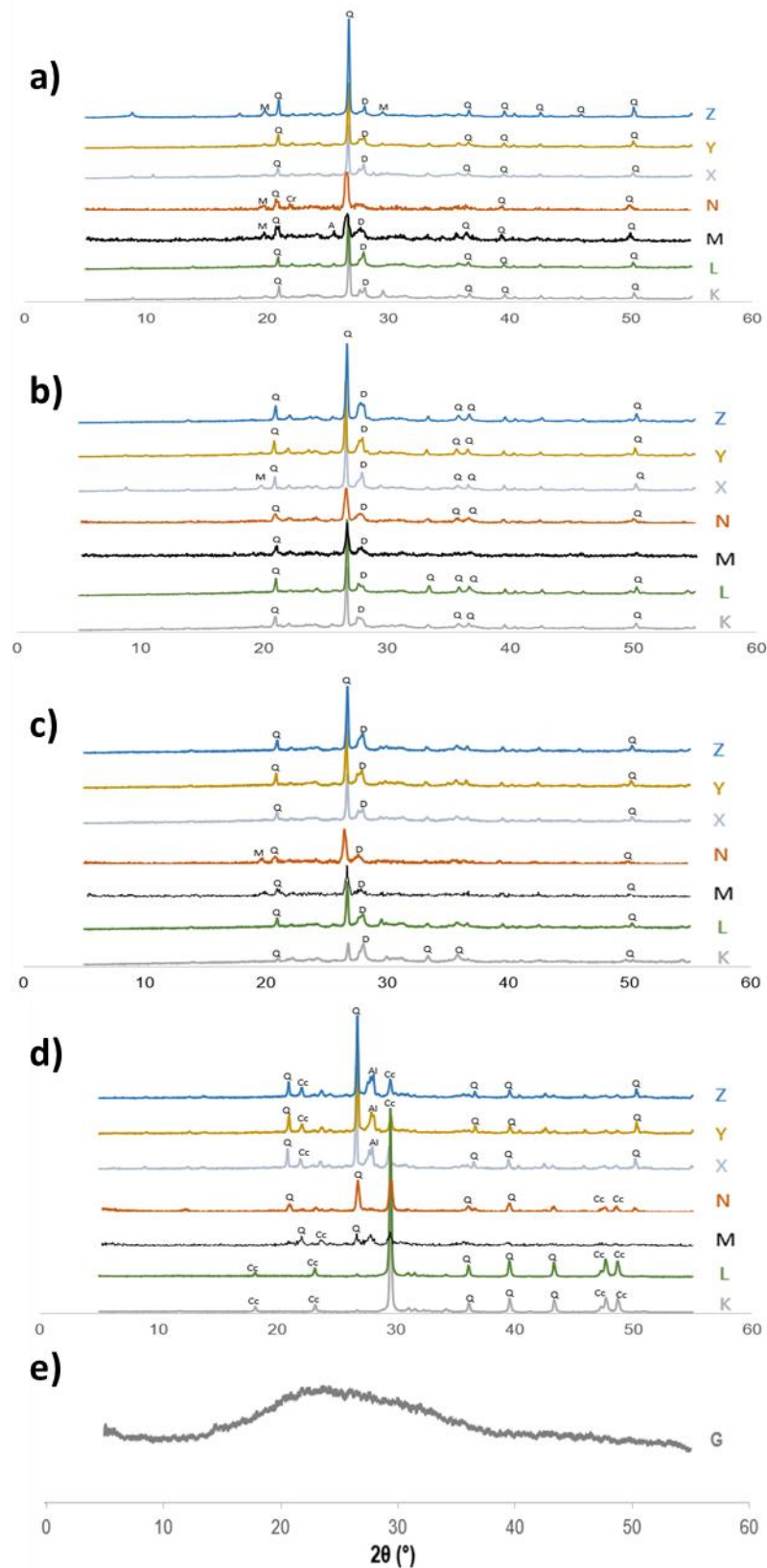
252

Table 2 Characteristic particle diameters of CDW-based materials (units are in μm).

Demolition site notation	d(0.1)	d(0.5)	d(0.9)	Demolition site notation	d(0.1)	d(0.5)	d(0.9)
K-HB	0.596	4.18	29.6	K-RT	0.621	5.55	28.6
L-HB	0.567	4.20	38.6	L-RT	0.603	5.50	45.4
M-HB	0.612	4.88	31.4	M-RT	0.592	5.41	32.1
N-HB	0.557	4.19	31.2	N-RT	0.663	6.49	32.3
X-HB	0.519	3.18	23.2	X-RT	0.587	5.40	93.4
Y-HB	0.754	10.1	247.4	Y-RT	0.566	4.64	28.5
Z-HB	0.543	3.21	34.6	Z-RT	0.559	4.85	34.0
K-RCB	0.566	4.03	24.9	K-C	0.597	10.4	163.5
L-RCB	0.742	8.02	33.2	L-C	0.620	10.8	193.4
M-RCB	0.951	10.3	34.6	M-C	0.486	3.60	95.0
N-RCB	0.651	4.97	34.2	N-C	0.536	4.43	48.6
X-RCB	1.403	22.7	260.2	X-C	0.626	7.29	73.3
Y-RCB	0.740	9.12	31.3	Y-C	0.635	6.91	48.4
Z-RCB	0.634	6.48	32.0	Z-C	0.610	6.36	60.1
				K-G	1.75	19.4	67.2

255 3.3. Crystalline nature of CDW-based materials

256 Structures observed as a result of XRD analyzes of CDW-based materials are presented in
257 Fig. 3. Details of the observed peaks including the chemical formula and particle diffraction
258 file (PDF) numbers, are also given in Table 3. According to XRD results, all clayey CDWs
259 studied were with a semi-crystalline structure, with the main peaks being related to quartz and
260 a broad hump centered between 2θ values of approximately 21° and 26° . Besides quartz, minor
261 peaks related to other crystalline formations such as cristobalite, diopside, mullite, and
262 akermanite, which consist of silica, alumina and calcium-based components were also
263 observed. As was also shown in literature, quartz, which represents a substantial portion of the
264 crystalline structure of clayey materials, is mostly unreactive and undergoes almost no change
265 in intensity after interaction with the alkalis (Lee and Deventer, 2003). However, when these
266 materials were calcined by heat treatment up to 800-1000 $^\circ\text{C}$, their crystalline structure was
267 disturbed, which results in the formation of amorphous alumina and silica and positively
268 contributes to their pozzolanic activity (Tantawy, 2015; Cao et al., 2016). Although applying a
269 further calcination treatment on the collected HB, RCB and RTs can ensure their use with
270 greater mechanical performance, additional cost, energy, and time required for the calcination
271 process may highly limit the outcomes of recycling applications.



272

273 **Fig. 3** XRD diffractograms of CDW-based materials, a) HB; b) RCB; c) RT; d) C; e) G

274 XRD diffractograms of different concrete wastes are given in Fig. 3 together with the

275 chemical formula and PDF numbers of the identified peaks in Table 3. Fig. 3 shows that the

276 concrete wastes have also crystalline nature, with principal peaks of quartz, calcite and albite.
 277 However, significant differences were observed in the crystalline structures of concrete wastes
 278 compared to the clayey wastes with largely similar structures. Quartz peaks with higher
 279 intensity were observed in samples from X, Y, and Z sites, especially at around 2θ of 26° .
 280 While calcite was the dominant crystalline structure for the concrete wastes from K and L sites,
 281 a mixture of quartz and calcite was noticed at higher intensity in concrete wastes from M and
 282 N sites. It is notable that the intensities of different peaks are quite compatible with the chemical
 283 compositions of each sample. Another aspect to consider is that calcite peaks were found not
 284 only in concrete wastes produced with the calcareous aggregates but in all concrete wastes,
 285 which may be formed as a result of the carbonation of cement hydration products (Moreno-
 286 Pérez et al., 2018). According to the XRD diffractograms, the glass waste had a completely
 287 amorphous nature. As is known, the pozzolanic activity and alkaline activation efficiency of
 288 the materials are largely driven by their amorphousness (Ulugöl et al., 2021). However, the
 289 complete amorphousness of a material does not ensure that it will have excellent pozzolanic
 290 activity, because this property is also highly dependent on the chemical composition and
 291 fineness

292 **Table 3** Types, PDF numbers, and chemical formulas of crystalline phases

Crystalline phase	Symbol	PDF number	Chemical formula
Quartz	Q	96-101-1160	SiO ₂
Crystobalite	Cr	96-900-8230	SiO ₂
Diopside	D	96-900-5280	Al _{0.6} CaMg _{0.7} O ₆ Si _{1.7}
Mullite	M	96-900-5502	Al ₂ O ₅ Si
Akermanite	A	96-900-6115	AlCa ₂ Mg _{0.4} O ₇ Si _{1.5}
Calcite	Cc	96-900-0968	CaCO ₃
Albite	Al	96-154-0705	Ca ₃ O ₅ Si

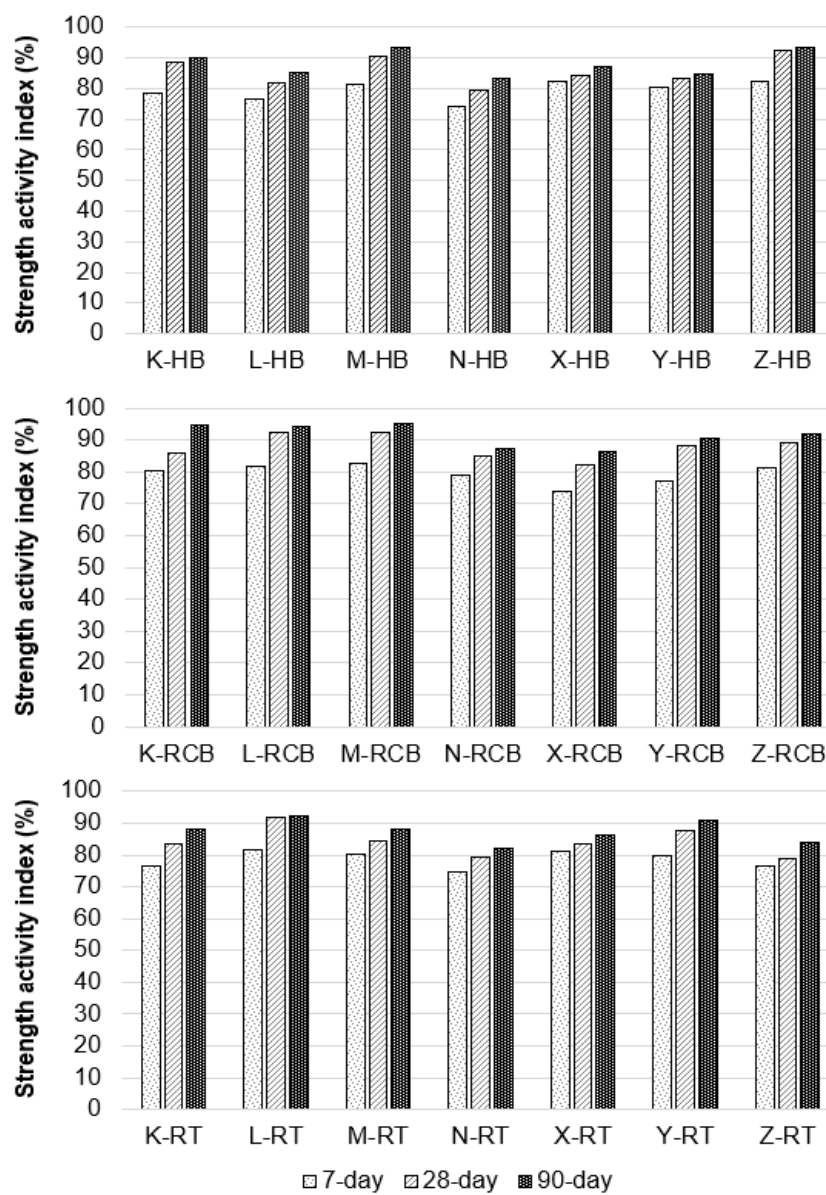
293

294 *3.4. Pozzolanic activity of CDW-based materials*

295 *3.4.1. Clayey materials*

296 7-, 28-, and 90-day compressive strengths of the mortar samples were measured and their
 297 SAIs were calculated relative to the control samples (Fig. 4). All HB, RCB, and RTs had lower
 298 SAIs than the control samples with SAI of 100%, regardless of the curing age and demolition
 299 site from where they were collected. Comparing the results of different clayey materials, HB,
 300 RCB, and RT showed average SAI values of all ages of 84.5%, 86.3%, and 83.4%, respectively.
 301 In general, these differences are believed to be in relation with the previously shown variances

302 noted in the CDW material properties such as fineness, chemical composition and crystalline
 303 nature. Another reason for the differences noted in the pozzolanic activity of clayey CDWs can
 304 be the kiln temperature utilized in producing these materials, since lower firing temperatures
 305 have been reported to yield to lower pozzolanic activity (Pereira-de-Oliveira et al., 2012). As
 306 expected, the pozzolanic activity of clayey CDWs generally increased with the increased curing
 307 ages and contributed more to the compressive strength development at later ages (Zhao et al.,
 308 2020). This is related to the increased efficiency of materials with sufficient amounts of
 309 aluminosilicate oxides and fineness in binding Ca(OH)_2 in the presence of moisture over time
 310 to form C-S-H gels (O'Farrell et al., 2006; Pereira-de-Oliveira et al., 2012).



311

312

Fig. 4 Strength activity indexes of CDW-based clayey materials

313 According to the Fig. 4, among all HBs, although one of the weakest pozzolanic activity of
314 HB from Y site (Y-HB) was in line with the material's coarser particle size distribution, X-HB
315 with the finest particle size distribution did not exhibit the optimum pozzolanic activity. The
316 highest SAI of M-HB at 90 days was both related to its highest $\text{SiO}_2+\text{Al}_2\text{O}_3$ content and
317 fineness. It can generally be stated that for HBs, the applied crushing/grinding procedures were
318 more efficient in reaching SAIs higher than 80% with the achievement of maximum $d(0.5)$ and
319 $d(0.9)$ values of around $4\ \mu\text{m}$ and $40\ \mu\text{m}$, respectively. Together with the high fineness of HB,
320 $\text{SiO}_2+\text{Al}_2\text{O}_3$ content greater than 70% are the likely parameters to achieve an average SAI of
321 at least 90% at 90 days.

322 Considering the pozzolanic activity of RCBs, it was observed that the M-RCB presented the
323 greatest average SAIs with 82.7%, 92.6%, and 95.3% in 7, 28, and 90 days, respectively. This
324 can be related to material's highest $\text{SiO}_2+\text{Al}_2\text{O}_3$ content (77.4%), finer particle size distribution
325 and crystallinity level compared to most of other RCBs. Another point to stress is that, although
326 the $\text{SiO}_2+\text{Al}_2\text{O}_3$ content of N-RCB was moderate, and L-RCB was with a relatively coarse
327 particle size, SAIs of these materials were still higher than 87% after 90 days. This can be either
328 due to suppression of the effect of relatively coarse particle size distribution (although below
329 $45\ \mu\text{m}$) by adequate $\text{SiO}_2+\text{Al}_2\text{O}_3$ content thanks to better chemical interactions (Xiao et al.,
330 2018), or suppression of relatively lower $\text{SiO}_2+\text{Al}_2\text{O}_3$ content by relatively finer particle size
331 distribution thanks to the micro-filler effect besides pozzolanic activity (Cordeiro et al., 2008).

332 Considering the pozzolanic activity of RTs, it can be surprisingly seen that L-RT has the
333 highest SAI values, especially at the early ages, although it is neither with the finest particle
334 size distribution nor with the highest $\text{SiO}_2+\text{Al}_2\text{O}_3$ content. The supplementary cementitious
335 materials can contribute to compressive strength not only thanks to pozzolanic activity but also
336 with micro-aggregate effect, especially at the early ages (Zhao et al., 2020). Y-RT with the
337 highest $\text{SiO}_2+\text{Al}_2\text{O}_3$ content (70.1%) showed the second-best pozzolanic activity after L-RT,
338 and N-RT with the lowest $\text{SiO}_2+\text{Al}_2\text{O}_3$ content (58.8%) exhibited the worst pozzolanic activity
339 at later ages. Unlike other RTs, mullite crystals observed in N-RT might also have caused poor
340 reactivity.

341 **3.4.2. Concrete waste**

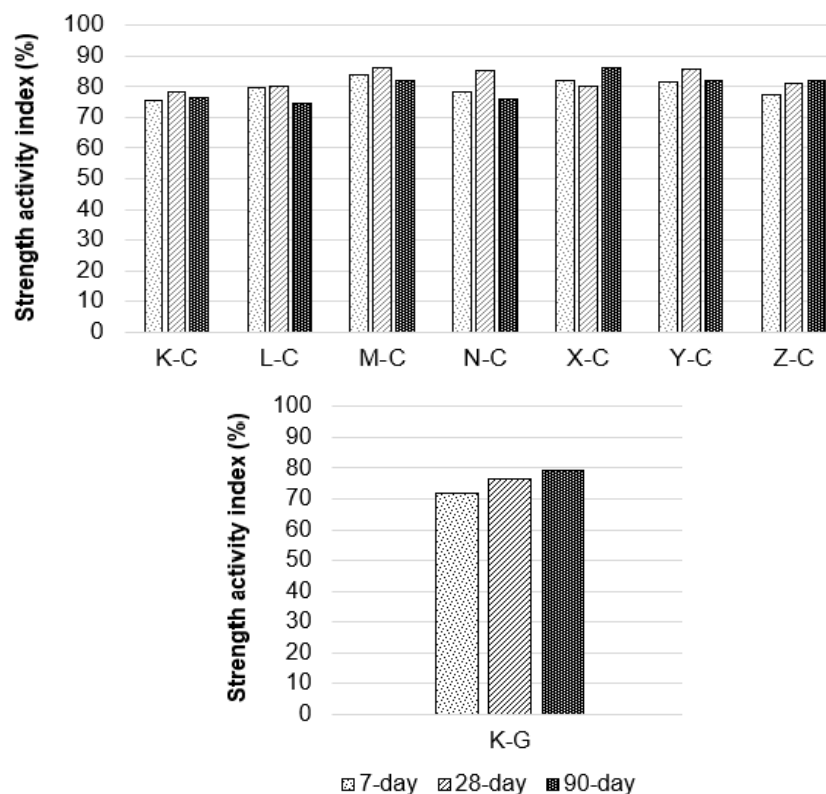
342 SAIs of concrete wastes were presented in Fig. 5. Similar to clayey CDWs, concrete wastes
343 also showed lower SAIs than the control mixture. The average results of all ages for the diverse
344 concrete wastes were 80.7%. The 90-day SAIs of concrete wastes were generally lower than

345 the 7- and 28-day values. This can be related to the fact that waste concrete contributes to the
346 achievement of compressive strength at early ages thanks to unhydrated cementitious materials
347 available in their surface leading to the formation of extra C-S-H gels (Horsakulthai, 2021). In
348 a recent study, similar results were reached and it was concluded that waste concrete powder
349 improved the hydration reaction by accelerating the formation of C-S-H gels due to its nucleus
350 effect at early ages (*i.e.*, 7- and 28-day) (Huashan and Yujun, 2021). Based on these findings,
351 it therefore seems that the amount of attached cement/mortar particles on the surface of
352 concrete wastes highly control the contribution of these materials to the strength development
353 of mortars incorporating them. Contrary to clayey CDWs, concrete wastes with high calcium
354 and low silica contents were expected to be with poor pozzolanic activity (Kim and Choi,
355 2012). For example, according to (Antony and Nair, 2016), concrete waste powder was shown
356 to exhibit substantially lower reactivity than roof tile powder in terms of lime reactivity test. In
357 this study, concrete wastes were with lower average SAIs compared to clayey CDWs,
358 especially at 90 days. However, some concrete wastes, such as M-C and Y-Cs, presented close
359 or even better strength activity indexes than clayey CDWs at 7 and 28 days. Thus, the
360 pozzolanic activity of this waste is closely related to its composition incorporating large shares
361 of unhydrated cement grains, which likely contributes to the compressive strength development
362 of the new mortars through further cement hydration rather than pozzolanic activity (Mas et
363 al., 2012; Zhu et al., 2019; Juan-Valdes et al., 2021). This statement was approved by the
364 mostly lower SAIs of concrete wastes at later curing age of 90 days. However, K-C and L-C
365 with lower $\text{SiO}_2 + \text{Al}_2\text{O}_3$ contents exhibited lower SAIs than other concrete wastes, implying for
366 a possible contribution of this parameter to the strength development of C-based concretes. The
367 important difference between the activity indexes of K-C and L-C, despite their similar
368 chemical composition, particle size distribution and crystalline structures, also hints for the
369 possible effects of impurities available on the surface of waste concrete particles.

370 **3.4.3. Glass waste**

371 According to the Fig. 5, G had lower SAIs compared to all other CDW-based materials. The
372 values for 7, 28, and 90 days of curing were 71.8%, 76.5%, and 79.1%, respectively. Greater
373 pozzolanic activity and reactivity were expected from the G due to its high silica content and
374 amorphous nature. Furthermore, several investigations in the literature reported that G could
375 exhibit very high pozzolanic activity at different ages (even at 90 days) (Shi et al., 2005;
376 Afshinnia and Rangaraju, 2015). However, the G herein was with comparatively coarser
377 particle size distribution compared to those in literature and other CDW-based materials, which

378 may be the main reason for the poor pozzolanic activity observed. This has been supported by
 379 a study in literature which stated that the level of fineness of G is the most fundamental
 380 parameter in determining its pozzolanic activity (Pereira-de-Oliveira et al., 2012). Here, it
 381 needs to be emphasized that G was not subjected to further grinding procedure for an easier
 382 comparison with other CDW-based materials, although it is highly likely to obtain better results
 383 at lower particle size of glass. The prepared powder presented a lower pozzolanic activity
 384 although it was with completely amorphous structure and this was mostly due to low Al₂O₃
 385 content and relatively coarse particle size of the G.



386
 387 **Fig. 5** Strength activity indexes of CDW-based concrete and glass.

388 3.5. Scanning electron microscopy and Energy-dispersive X-ray spectroscopy analysis

389 SEM micrographs and point EDX analysis results of mixtures produced with M-coded
 390 CDW-based materials and K-G were presented in Fig. 6 and Table 4, respectively. SEM
 391 micrographs of the specimens including M-coded CDW-based materials were found to be
 392 compact and homogeneous. While the reaction products of the specimens produced with clayey
 393 CDWs (*i.e.*, HB, RCB and RT) were generally well dispersed and showed a smooth surface,
 394 local crack formation in the specimen including C and partial agglomeration of the products in
 395 the specimen including G was observed. To interpret these circumstances and make it clear,
 396

397 EDX analysis was performed at various points to reveal the presence of possible unreacted
398 CDWs and their agglomeration and observe the reaction products whether formed as a result
399 of cement hydration or pozzolanic reaction of CDW grains. The EDX analysis results showed
400 that while the chemical compositions varied in several regions, the matrices generally had
401 comparable atomic distributions. For instance, in the HB-included specimens, while similar
402 chemical composition was observed at points 1,3 and 5, oxide values were lower and Ca values
403 were higher at points 2 and 4. This indicates the presence of calcium-rich structures such as
404 $\text{Ca}(\text{OH})_2$ at points 2 and 4. Also, higher Al detection in the point 3 of the RCB-included mixture
405 and in the point 1 of the RT-included mixture might be attributed to formation of C-A-S-H gel
406 type. Also, significantly higher Si value at point 5 in the C-included mixture compared to other
407 points might be attributed to the presence of unreacted C particles. In the EDX analysis of G-
408 included mixture, all selected points from different locations exhibited identical chemical
409 compositions.

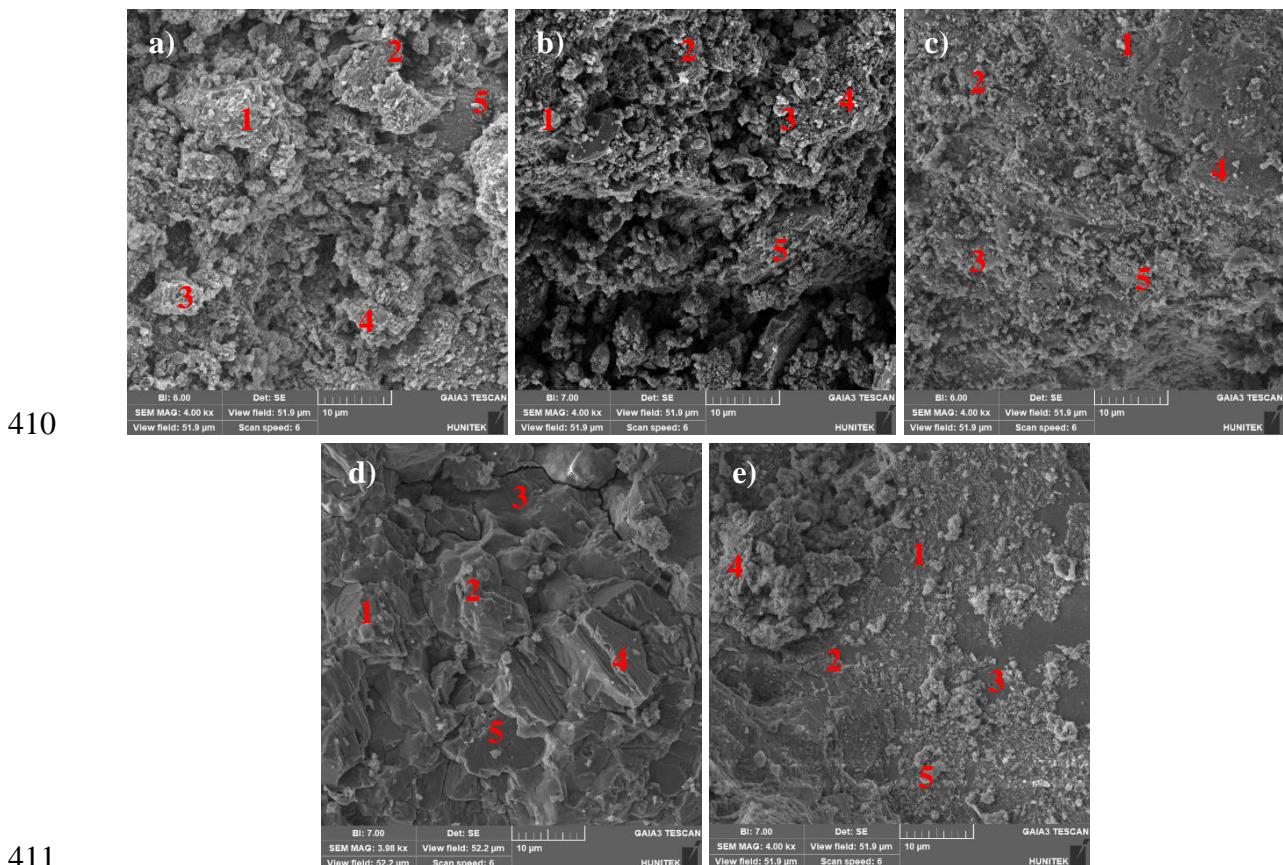


Fig. 6 SEM micrographs of the mixtures, a) M-HB; b) M-RCB; c) M-RT; d) M-C; e) K-G.

Table 4 EDX analysis results of the mixtures including M-coded CDW-based materials

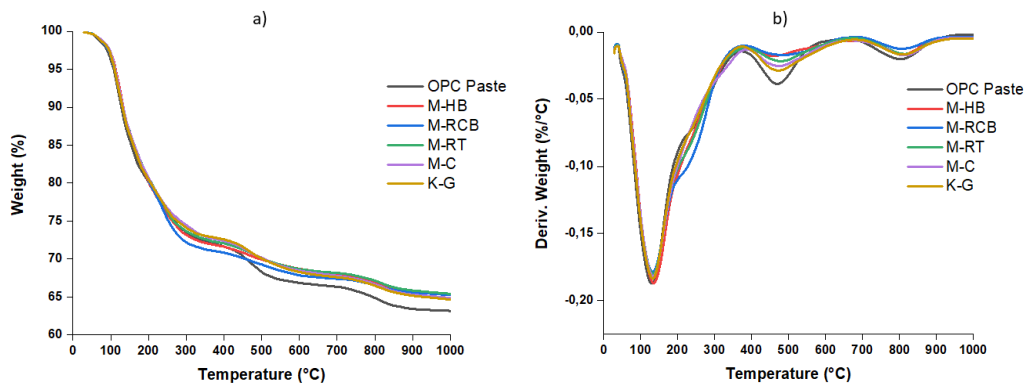
CDW Component	Point No.	O	Ca	C	Si	Al	Fe
		(wt%)					
HB	1	53.8	29.8	16.3	0.10	0.00	0.10
	2	33.6	54.2	5.50	2.10	0.90	3.80
	3	48.2	29.9	11.3	6.80	2.20	1.70
	4	51.4	28.5	14.4	4.50	0.50	0.60
	5	25.9	63.4	4.20	4.20	0.70	1.60
RCB	1	44.6	25.6	15.3	9.30	3.10	2.00
	2	52.8	25.7	14.5	4.20	1.60	1.20
	3	59.6	8.90	13.2	10.0	7.00	1.30
	4	56.6	19.1	16.3	4.60	1.60	1.80
	5	55.2	22.9	13.4	7.00	0.80	0.70
RT	1	31.5	7.50	15.6	27.1	14.5	3.80
	2	54.5	29.6	14.6	0.90	0.40	0.10
	3	46.3	40.7	10.2	1.80	0.60	0.30
	4	50.8	31.4	16.0	1.60	0.20	0.00
	5	53.3	29.3	12.4	3.90	1.00	0.10
C	1	56.6	26.9	15.8	0.50	0.20	0.00
	2	45.1	43.3	11.5	0.00	0.00	0.00
	3	56.2	25.8	17.9	0.01	0.00	0.20
	4	57.6	20.5	17.9	3.00	0.50	0.40
	5	58.1	6.6	16.4	10.9	7.60	0.50
G	1	56.9	5.30	8.70	28.2	0.60	0.30
	2	52.6	7.80	11.4	27.2	0.70	0.30
	3	53.2	8.20	10.7	27.3	0.70	0.00
	4	47.3	11.7	11.5	28.4	0.80	0.60
	5	57.1	6.00	10.2	25.8	0.70	0.20

418

419 3.6. Thermogravimetry (TG/DTG) analysis

420 Thermogravimetry (TG/DTG) analysis of the mixtures included M-coded HB, RCB, RT, C
421 and K-coded G and OPC paste were presented in Fig. 7. According to the TG/DTG analysis
422 results, four different temperature regions were detected where remarkable differences
423 appeared in weight change. In the first region where the temperature range is between
424 approximately 30-110 °C, similar weight losses were encountered for all mixtures. The weight
425 losses observed in this range can be attributed to the loss of evaporable water in the system
426 (Alarcon-Ruiz et al., 2005). In the second region where the temperature range is between
427 approximately 180-300 °C, it was observed that the M-RCB coded mixture experienced the
428 highest weight loss, followed by M-RT, M-HB, K-G, M-C and OPC paste, respectively. As it
429 is known from the literature, considering that the weight loss in the 180-300 °C range is
430 generally caused by the loss of bound water from the decomposition of the C-S-H and loss of
431 carboaluminate hydrates (Alarcon-Ruiz et al., 2005; Thongsanitgarn et al., 2014), it can be

432 concluded that, in the M-RCB coded mixture, the gel structure formation that will cause water
 433 loss is slightly higher than the other mixtures at the end of the 90-day curing. The significant
 434 pozzolanic activity of CDW-based materials contributed to the formation of extra C-S-H in the
 435 matrix in the long term. Next, the weight loss of OPC paste increased sharply in the temperature
 436 range around 450–550 °C, while it was found that other mixtures incorporating CDW-based
 437 material exhibited significantly lower weight losses. The weight loss observed in this
 438 temperature range, where indicates the dehydroxylation of the calcium hydroxyde in general
 439 (Lorca et al., 2014), showed that CDW-based materials pozzolanically combined with calcium
 440 hydroxyde which is the secondary hydration product of Portland cement and caused the
 441 decreasing amount of it. Finally, significant weight losses were also observed in the
 442 temperature range of 700-900 °C. The most severe weight loss of OPC paste in this temperature
 443 range, which is associated with the decarbonation of calcium carbonates formed as a result of
 444 carbonation of calcium hydroxide (Huang et al., 2019), indicates that it contains a higher
 445 proportion of calcium hydroxide than other mixtures containing CDW-based materials. This
 446 can be explained by the consumption of calcium hydroxide in mixtures containing CDW-based
 447 materials as a result of pozzolanic reactions.



448
 449 **Fig. 7** TG/DTG analyses of the mixtures, a) TGA b) DTG.

450 **4. Pearson correlation analysis**

451 In order to statistically examine the influences of chemical composition (*i.e.*, SiO₂, Al₂O₃
 452 and CaO) and fineness degrees (*i.e.*, d[0.1], d[0.5] and d[0.9]) of CDWs on the 7-, 28- and 90-
 453 day SAI values, Pearson's Correlation Coefficient analysis was performed. The results shown
 454 in Table 5 demonstrated that there is a strong positive relationship between the SiO₂ and Al₂O₃
 455 concentration and the SAI values. In particular, when the relationship of SiO₂ and Al₂O₃
 456 content with the 90-day SAI values is examined, it was seen that the Pearson correlation
 457 coefficient has a very high positive correlation with values of 0.65 and 0.835, respectively. On

458 the other hand, CaO had a negative relationship with the SAI values of CDW-substituted
 459 mixtures. Also, the fineness degrees of the CDW-based materials (*i.e.*, d[0.1], d[0.5], d[0.9])
 460 showed a negative correlation with SAI values at all ages, as discussed above. Another
 461 interesting finding is that while the influence of d(0.1) and d(0.5) reduced with age, the
 462 influence of d(0.9) increased with age. When comparing the results in Table 5, it can be seen
 463 that fineness had a significant impact on SAI values at early ages (*i.e.*, 7- and 28-day), but the
 464 influence of chemical composition became more prominent in 90-day SAI values.

465 **Table 5** Pearson correlation analysis of the selected parameters

		SiO ₂	Al ₂ O ₃	CaO	d(0.1)	d(0.5)	d(0.9)
7-day SAI	Pearson corr.	0.117	0.332	-0.1	-0.499**	-0.484**	-0.16
	Sig. (2-tailed)	0.547	0.078	0.607	0.008	0.008	0.406
	N	29	29	29	29	29	29
28-day SAI	Pearson corr.	0.35	0.567**	-0.375*	-0.23	-0.309	-0.342
	Sig. (2-tailed)	0.063	0.001	0.045	0.229	0.103	0.07
	N	29	29	29	29	29	29
90-day SAI	Pearson corr.	0.65**	0.835**	-0.731**	-0.094	-0.246	-0.439*
	Sig. (2-tailed)	0	0	0	0.627	0.198	0.017
	N	29	29	29	29	29	29

466 * : Correlation is significant at the 0.01 level (2-tailed)

467 ** : Correlation is significant at the 0.05 level (2-tailed)

468 5. Conclusions

469 A detailed characterization of CDW-based hollow brick, red clay brick, roof tile, concrete,
 470 and glass obtained from various demolition sites was performed in this study.
 471 Physical/chemical properties and crystalline structures of these materials were determined and
 472 possible interrelationships of these properties and the pozzolanic activity of each material were
 473 evaluated. The following conclusions can be made:

- 474 • Chemical compositions of HB, RCB, and RTs were found almost in the same range,
 475 except for some differences related to their original raw materials/production method.
 476 Larger differences were reported in the chemical compositions of Cs, which highly
 477 depend on the type/content of cement and aggregates used in the original concretes. G
 478 sample was with slightly higher silica content and lower alumina than all CDWs.
- 479 • Most of HB, RCB, RT, and Cs have reached to similar particle size distributions under
 480 the same crushing/grinding procedures, although some materials presented coarser
 481 particles because of the presence of the impurities and inherent hardness. Compared to
 482 other CDWs, G showed coarser particle size distribution after same crushing/grinding.

- 483
- XRD analyses revealed a semi-crystalline structure for all HB, RCB, and RTs while a
484 crystalline structure for all Cs, however, with clear differences in the quartz and calcite
485 peaks between clayey materials and Cs. G presented a clear amorphous structure unlike
486 other CDWs.
 - Average SAIs of all ages were 84.5%, 86.3%, 83.4%, 80.7%, and 75.8% for HB, RCB,
487 RT, C, and G, respectively. Clayey CDWs contributed to mechanical strength through
488 their sufficient fineness and aluminosilicate oxide contents. Also, clayey CDWs can be
489 utilized together without any selection/separation process since their SAIs are quite
490 comparable to each other with only slight differences. However, the contribution of Cs
491 was more related to the possible effects of hydration of the unreacted cementitious
492 particles. On the other hand, G was found to have the weakest pozzolanic activity due
493 to its coarser grain size distribution compared to other CDWs.
 - The SAIs of all CDWs, irrespective of the origin and type of CDWs, were found
494 acceptable for their use as SCMs in the PC-based systems, since their minimum average
495 SAI was higher than 75% specified in ASTM C618.
 - It was micromechanically confirmed by SEM/EDX and TGA/DTG analysis that CDW-
496 based materials are pozzolanically combined with calcium hydroxide and contribute to
497 the formation of extra C-S-H gel by consuming calcium hydroxide.
- 498
- 499
- 500

501 Although one of the important criteria controlling the pozzolanic activity is the level of
502 fineness reached for each CDW, materials' $\text{SiO}_2+\text{Al}_2\text{O}_3$ contents seem to highly influence
503 the results. Particularly, when CDWs with similar finenesses were compared, materials
504 with higher $\text{SiO}_2+\text{Al}_2\text{O}_3$ contents displayed better pozzolanic activity. It was suggested that
505 the SAI of G sample can be improved by achieving higher fineness for this material. In this
506 study, no further grinding was applied to materials since it would be more economical and
507 ecological to prepare all CDW-based materials together in an optimum way without sorting
508 and separation procedures. However, this seems inappropriate, especially when the nature
509 of CDWs is different. Thus, it is recommended to separately test each collected material
510 from various sites. It is also important to reach the adequate fineness for each material for
511 an optimized performance of the newly developed materials.

512

513

514

515 **Declaration of competing interest**

516 The author declares that they have no known competing financial interests or personal
517 relationships that could have appeared to influence the work reported in this paper.

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