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Development of Alkali-Activated Binders froRecycled Mixed Masonry-originated Waste

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29 **Keywords:** Alkali-activated materials (AAMs); Construction and demolition waste (CDW);
30 Masonry, Compressive strength; Microstructure.

31
32 **1. Introduction**

33 Concrete is the most widely used construction material owing to its low cost, ease of
34 applicability, versatility and reliability with yearly consumption levels approaching 30 billion
35 tonnes [1]. As the developing countries invest more and more in their infrastructural
36 development and developed countries upgrade/replace their ageing infrastructure, the demand
37 for concrete increases continuously [1]. However, concrete production is not at a low cost given
38 the fact that majority of the individual constituents forming this material is not eco-friendly and
39 sustainable to manufacture. The most distinctive example of such constituents is Portland
40 cement (PC) which is the main binder for traditional concrete and requires highly energy-
41 intensive stages of production (e.g. fuel combustion and calcination of raw materials) resulting
42 in release of nearly 1 ton of CO₂ per 1 ton of PC manufactured [2]. Currently, PC production is
43 hold responsible for up to 9% of global anthropogenic CO₂ emissions and it is estimated that
44 by 2050, annual PC production will increase by 50% [1]. In this sense, nowadays, the focus of
45 research community is shifting towards the utilization of more eco-friendly binders in concrete.

46 Traditional concrete is reported to be not long-lived due to its brittle nature which triggers
47 high cracking tendency and exacerbation of problems related to durability [3-6]. The
48 insufficiency in the durability performance of conventional concrete which is coupled with the
49 lack of control over cracking generally ends up with costly repair and/or maintenance
50 applications [7] or even with the complete demolition and re-construction of structures, creating
51 large amounts of waste collectively termed as “*Construction and Demolition Waste – CDW*”.
52 CDW industry is regarded to be one of the sectors contributing to the global solid waste
53 production the most [8]. Annual CDW production of major countries is ten billion tons. Three
54 billion of this is contributed by China [9] while, EU-28 and U.S. generate more than 800 and

55 700 million tons, respectively [8]. As exemplified, CDW is a global issue requiring relatively
56 high demand for proper handling. This is not only important to lower the CDW amount going
57 to clean landfills, but also to reduce the amount of concrete (hence, PC and aggregates)
58 production that is, otherwise, going to be used for the construction and repair/maintenance of
59 new and/or existing structures.

60 Due to concerns about the tremendous amounts of concrete and related PC production
61 worldwide, latest research efforts into the development of more eco-friendly binders have been
62 intensified.

63 In this regard, “alkali-activated materials (AAMs)”, including those referred to as
64 “geopolymers” can be regarded as a major advancement towards the realization of greener
65 binders [10]. AAMs can be synthesised by the alkali-activation of a variety of aluminosilicate
66 precursors (source materials) and the majority of the studies up to now were performed using
67 precursors such as fly ash, ground granulated blast furnace slag, silica fume and metakaolin.
68 These precursors are already well-known and have controlled production processes and
69 chemical compositions [11]. However, it is notable that these materials are highly demanded
70 and very successfully utilized in blended PC production and as separate mineral admixtures in
71 concrete mixtures. This therefore pushes researchers all around the world to look for possible
72 aluminosilicate precursors that can be used in the production of AAMs instead of
73 abovementioned main-stream mineral admixtures [12]. In this sense, CDW-based components
74 such as waste concrete, glass, masonry (e.g. bricks, tiles, ceramic) can be regarded as valuable
75 candidates and used as precursors in alkali-activation process which may favour more effective
76 waste management/control and pave the way for novel routes of waste recycling.

77 Studies related to the production of alkali-activated binders with CDW-based precursors
78 have already started although they are significantly limited in number and mostly utilized these
79 precursors singly or in combination with the previously mentioned main-stream precursors to

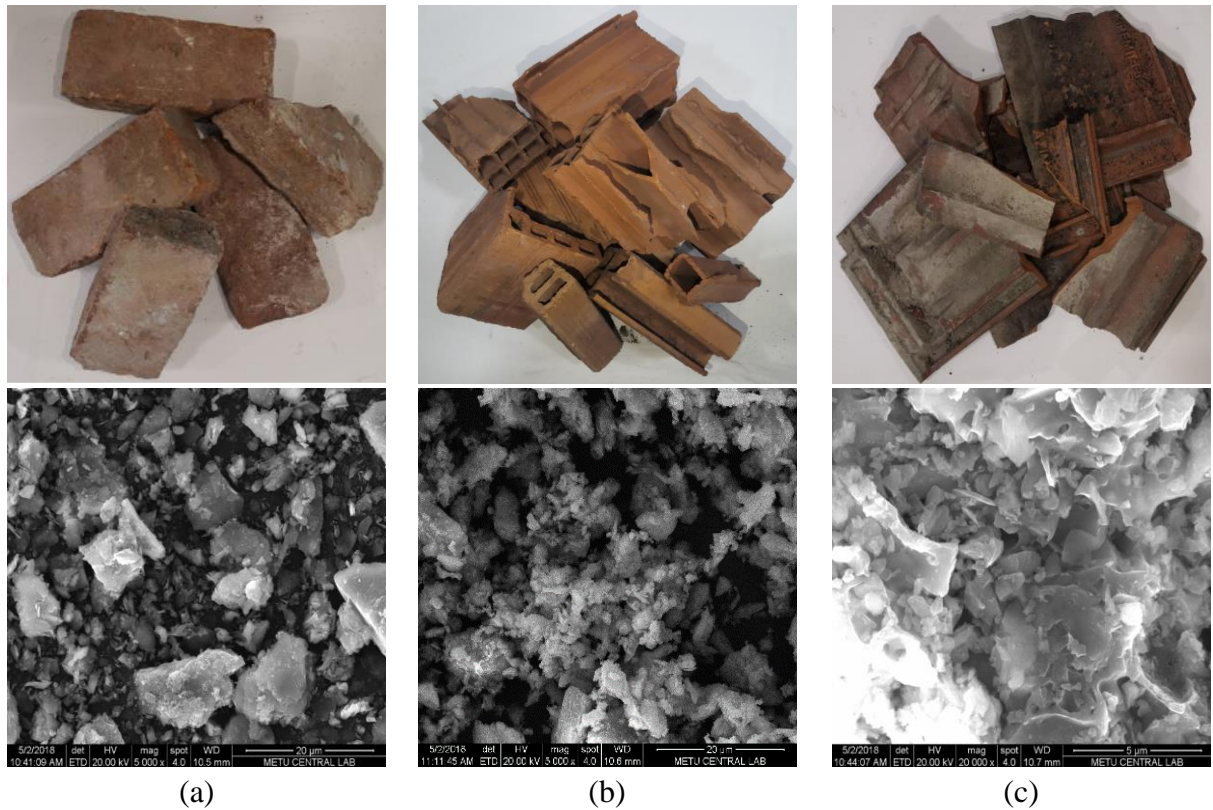
80 control the overall performance of the ultimate material [13-19]. In the incidents of construction
81 and demolition, CDW-based materials are obtained altogether. In order to obtain CDW-based
82 components separately, selective construction and demolition practises need to be employed
83 which may be energy-inefficient and time-consuming. It is therefore much more desirable to
84 use CDW-based precursors simultaneously in the production of alkali-activated binders without
85 elaborative separation to better simulate the first-hand obtained materials. To make an effort in
86 this direction, here, it was aimed to produce and evaluate alkali-activated binders based on
87 mixed CDW-based masonry units different from studies available in the literature. Red clay
88 brick (RCB), hollow brick (HB) and roof tile (RT) were used in this study as different masonry
89 units and in binary combinations by 75-25%, 50-50% and 25-75% of the total weight of the
90 binder. While producing the alkali-activated binders, special attention was paid to curing of
91 specimens produced with different alkaline activator concentrations and cured under different
92 temperature/periods. Performance characterization of alkali-activated binders was made via
93 series of compressive strength measurements which were backed by more in-depth
94 microstructural analyses of X-ray diffraction (XRD) and scanning electron microscopy with
95 energy-dispersive X-ray spectroscopy (SEM/EDX).

96 **2. Experimental Program**

98 **2.1 Materials**

99 In this study, CDW-based masonry units which include red clay brick (RCB), hollow brick
100 (HB) and roof tile (RT) were used as obtained from an urban transformation area located in
101 Ankara, Turkey. These materials were subjected to a non-complex, two-step, crushing-grinding
102 procedure. The first step of the procedure consisted of loading the CDW-based materials into a
103 laboratory-type jaw crusher which provided initial size reduction, followed by the second step
104 which included the loading of the crushed materials into a ball mill and further grinding for an
105 hour. In Fig. 1, views of CDW-based masonry units taken with the help of video camera and

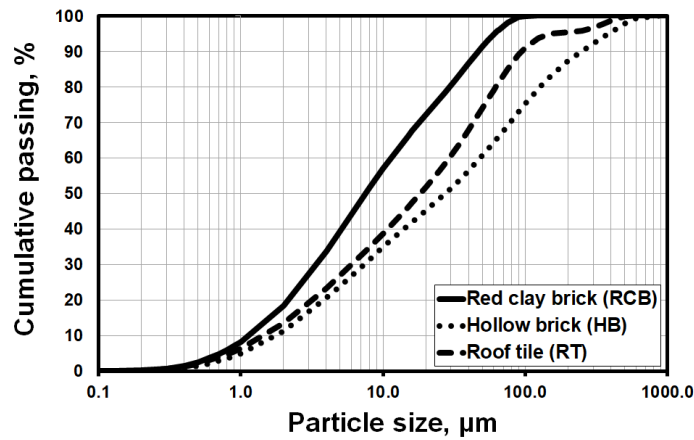
106 scanning electron microscope (SEM) were shown. In Fig. 2, gradation curves which give
107 information about the particle size distributions of the precursors were displayed. Characteristic
108 particle diameters of precursors were shown in Table 1.



109 **Fig. 1.** Video camera and SEM views of CDW-based precursors (a) red clay brick (RCB),
110 (b) hollow brick (HB), (c) roof tile (RT).
111

112 As can be seen from both Fig. 2 and Table 1, RCB is the finest CDW-based precursor
113 followed by RT and HB. According to Komnitsas et al. [13], significant improvements can be
114 achieved in the compressive strength of alkali-activated binders when fractions with $D_{50} < 15$
115 μm are used. However, only RCB was found to have a D_{50} value less than $15 \mu\text{m}$ (see Table 1).
116 Further grinding to reach finer particle fractions for precursors was not intended in this study,
117 since further milling is more labourious and energy-intensive. Another reason for the avoidance
118 of further grinding of the precursors is related to more realistic simulation of the situation in
119 practice. Upon construction/demolition of a certain structure, different waste types are obtained

120 collectively and it is therefore more logical and representative of the actual situation to keep the
 121 grinding period constant, if no special separation of wastes is intended.



122
 123 **Fig. 2.** Gradation curves of CDW-based precursors.
 124

125
 126 **Table 1** Characteristic particle diameters of different CDW-based materials (units are in µm).

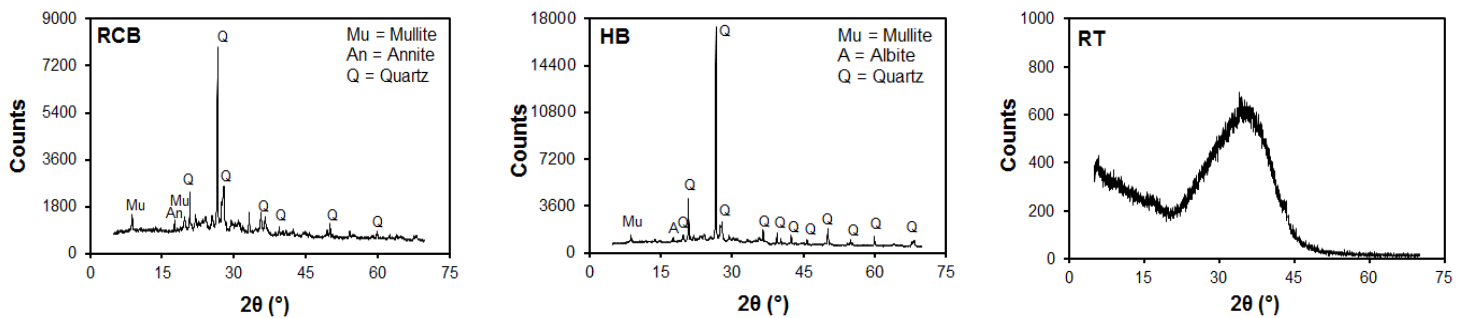
CDW-based material	Surface-weighted	Volume-weighted	D_{10}	D_{50}	D_{90}
	mean diameter ($D_{3,2}$)	mean diameter ($D_{4,3}$)			
Red clay brick (RCB)	2.9	16.4	1.2	7.6	46.9
Hollow brick (HB)	4.7	82.0	1.8	27.5	246.7
Roof tile (RT)	3.9	44.4	1.5	18.3	94.6

127
 128 **Table 2** Chemical compositions of CDW-based precursors as determined by XRF.

Chemical composition, %	CDW-based precursor		
	Red clay brick (RCB)	Hollow brick (HB)	Roof tile (RT)
Loss on ignition	2.18	1.99	2.11
SiO ₂	53.4	61.6	54.0
Al ₂ O ₃	20.5	17.3	15.9
Fe ₂ O ₃	7.77	6.70	8.93
CaO	4.75	3.31	7.42
MgO	3.70	2.66	4.84
SO ₃	1.16	0.38	0.68
Na ₂ O	1.53	1.61	1.41
K ₂ O	3.42	2.80	2.30

129
 130 X-ray fluorescence (XRF) analysis was performed to determine the chemical composition
 131 of the CDW-based precursors and the results are shown in Table 2. The analysis showed that
 132 all precursors were rich in siliceous and aluminous oxides which are fundamentally important
 133 oxides for the alkali-activation. The total of siliceous and aluminous oxide compositions of

134 RCB (73.9%) and RT (69.9%) were found to be similar to each other while higher for HB
135 (78.9%).



136 **Fig. 3.** XRD diffractograms of CDW-based precursors (Powder diffraction file [PDF]
137 numbers: Mullite ($\text{Al}_{2.2}\text{Si}_{0.7}\text{O}_{4.8}$) PDF No: 96-900-1568, Albite ($\text{NaAlSi}_3\text{O}_8$) PDF No: 96-900-
138 2201, Annite ($\text{K}_{0.94}\text{Fe}_{2.43}\text{Al}_{2.15}\text{Si}_{2.43}\text{O}_{12}$) PDF No: 96-900-2310, Quartz (SiO_2) PDF No: 96-
139 101-1160.

140 X-ray diffraction (XRD) technique was also used to analyse the crystalline nature of the
141 CDW-based precursors. As shown in Fig. 3, main crystalline peaks of RCB and HB were related
142 to quartz (SiO_2 , PDF No: 96-101-1160), mullite ($\text{Al}_{2.2}\text{Si}_{0.7}\text{O}_{4.8}$, PDF No: 96-900-1568), albite
143 ($\text{NaAlSi}_3\text{O}_8$, PDF No: 96-900-2201) and annite ($\text{K}_{0.94}\text{Fe}_{2.43}\text{Al}_{2.15}\text{Si}_{2.43}\text{O}_{12}$, PDF No: 96-900-
144 2310) while RT was amorphous with a broad hump centred between 2θ values of approximately
145 32° and 36° , although its production process involves very similar steps to that of RCB and HB.
146 As it is well-known, clayey materials do not have pozzolanic activity and are crystalline/semi-
147 crystalline in nature. However, when calcined at temperature ranges of $600\text{--}900^\circ\text{C}$, they lose
148 their combined water and their crystallographic structure collapses, leading to the formation of
149 silica and alumina in an amorphous state or in a state characterized by disorder in the lattice
150 structure. Under such circumstances, clay can exhibit pozzolanic activity since silica and
151 alumina can react with calcium hydroxide [20]. However, when the calcination temperature
152 exceeds 900°C , reorganization of alumina and silica can take place, which leads to the
153 formation of thermodynamically stable compounds such as mullite, tridimite etc. These
154 compounds are also crystalline in nature and non-reactive with calcium hydroxide [20]. Today's
155 so-called clayey building materials are rarely pozzolanic not only because they are calcined at
156 higher temperatures but also they are composed of ingredients that contain no or low amounts

157 of clayey ingredients [20]. Based on these statements, it can be therefore stated that differences
158 in the XRD diffractograms of CDW-based precursors can be largely related to the differences
159 in the production processes, calcination temperature and raw materials used in these building
160 materials [17,20]. Since the initial producer, production stages and the sources of CDW-based
161 raw materials are not known, no further details related to the differences in the mineralogy of
162 these materials can be provided.

163 CDW-based precursors were activated using a sodium hydroxide (NaOH) solution. The
164 NaOH used for the preparation of the solution was in flake form and included a minimum 98%
165 of sodium hydroxide, maximum 0.4% of sodium carbonate, 0.1% of sodium chloride and a
166 maximum of 15 ppm iron.

167

168 **2.2 Proportioning and mixing of alkali-activated binders**

169 Preparation of alkali-activated binders essentially consisted of two steps: (i) preparation of
170 the alkaline activator and (ii) the mixing of alkaline activator with the precursors. While
171 preparing the alkaline activator, NaOH flakes were first dissolved in tap water at varying Na^+
172 concentrations of 7.36, 11.04 and 13.98% which corresponded to NaOH molarities of 10, 15
173 and 19M, respectively. After dissolving, the solutions were allowed to cool down in a laboratory
174 environment set at room temperature. As mentioned previously, alkali-activated binders were
175 produced by the binary combinations of different CDW-based precursors. All mixtures were
176 produced with a constant water/binder ratio of 0.32 and incorporated a constant total amount of
177 binder (1000 g). Depending on the mixture composition, each binary combination of CDW-
178 based precursors (i.e. RCB-RT, RCB-HB and RT-HB) changed by 75-25%, 50-50% and 25-
179 75% of the total weight of the binder. The mixture proportions of the alkali-activated binders
180 are provided in Table 3 in more detail. No additional chemical admixtures were used in the
181 mixtures to avoid any interactions that may occur with the alkaline solution.

182 **Table 3** Mixture proportions of CDW-based alkali-activated binders.

Mixture ID.	Precursor, g			Alkaline solution				Alkaline activator/binder ratio
	RCB	HB	RT	Na, %	NaOH molarity, M	NaOH, g	Water, g	
75RCB-25RT	750	-	250	7.36	10	128.0	320	0.448
50RCB-50RT	500	-	500	11.04	15	192.0	320	0.512
25RCB-75RT	250	-	750	13.98	19	243.2	320	0.563
75RCB-25HB	750	250	-	7.36	10	128.0	320	0.448
50RCB-50HB	500	500	-	11.04	15	192.0	320	0.512
25RCB-75HB	250	750	-	13.98	19	243.2	320	0.563
75RT-25HB	-	250	750	7.36	10	128.0	320	0.448
50RT-50HB	-	500	500	11.04	15	192.0	320	0.512
25RT-75HB	-	750	250	13.98	19	243.2	320	0.563

183

184 At the mixing stage, the selected binary combinations of precursors were first loaded into a
185 mortar mixer and mixed for 60 s. Then, the solution of the alkaline activator was slowly added
186 to the mixer during the course of 30 s. After that, mixing was continued for 210 s at low speed.
187 Finally, after a 15 s of waiting period during which the blade and cone of the mixer were
188 cleaned, mixing was continued for 60 s at high speed to complete the mixing.

189

190 **2.3 Specimen preparation, curing and testing**

191 Current work included compressive strength testing and detailed microstructural assessment
192 of alkali-activated binders for the performance characterization. For compressive strength tests,
193 fresh mixtures were cast into cubic moulds with 50 mm dimensions and immediately after the
194 completion of molding, specimens with their molds were placed into an oven and subjected to
195 heat curing at the temperatures of 95, 105, 115 and 125°C for a period of 1, 2 and 3 days. Upon
196 completion of curing, the cubic specimens were directly tested under uniaxial compressive
197 loading applied at a rate of 0.9 kN/s. For a certain alkaline activator molarity, curing
198 temperature/period and combination of CDW-based precursors, six specimens were tested and
199 the obtained results were averaged for the determination of the compressive strength.

200 Microstructural characterization of the alkali-activated binders included the performance of
201 X-ray diffraction (XRD) analyses and scanning electron microscopy observations coupled with
202 energy-dispersive X-ray spectroscopy (SEM/EDX). XRD is a non-destructive method that is

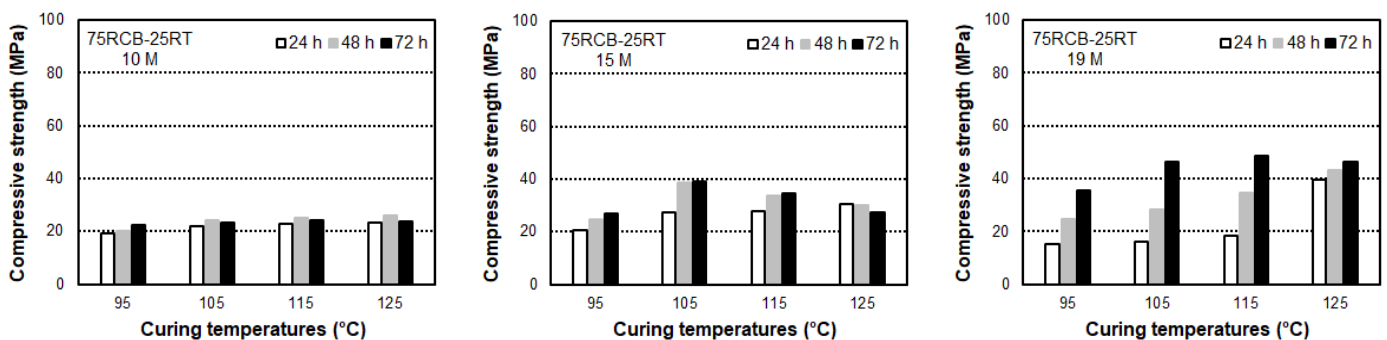
203 able to analyse the crystalline structure of materials with the diffraction of X-rays. The
 204 diffraction profile identifies the crystalline phases and thereby the chemical composition of the
 205 material. In the experiments, XRD analyses were performed at a scan range of $5^\circ \leq 2\theta \leq 80^\circ$,
 206 with a 2θ step length of 0.033° , scanning step time of 30.48 s and for the wavelength $K\alpha_1$ of
 207 copper ($\lambda = 1.5406 \text{ \AA}$). Powder samples weighing approximately 20 mg were obtained from
 208 selected cubic alkali-activated binder specimens left from the compressive strength testing and
 209 used for the XRD analyses. Comparisons were then made between the XRD results of raw
 210 precursors and the alkali-activated binders. Similarly, samples with dimensions less than 1 cm
 211 were obtained from the selected specimens of compressive testing and used for SEM/EDX
 212 analyses. SEM micrographs were recorded and chemical formulations of the selected areas
 213 were further analysed by EDX.

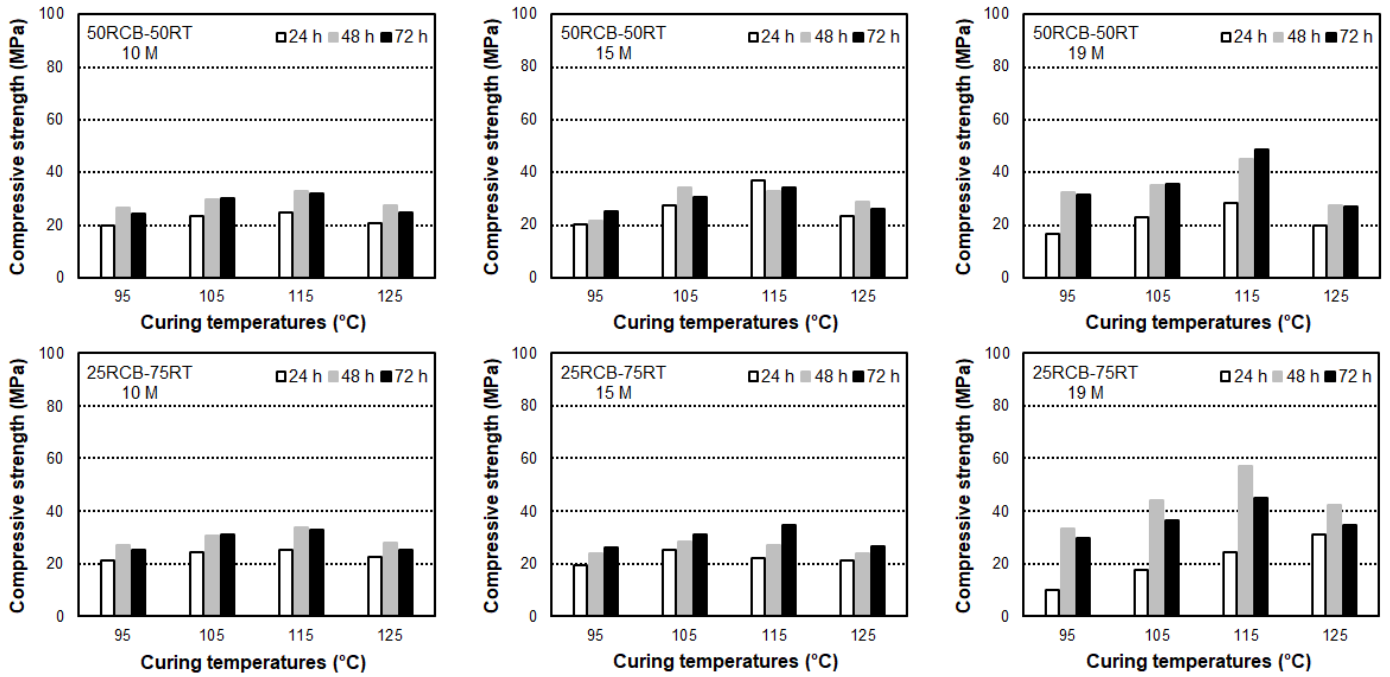
214
 215 **3. Results and Discussion**

216 **3.1 Compressive strength**

217 The average compressive strength results of the alkali-activated binders with different
 218 substitution rates of RCB-RT, RCB-HB and RT-HB are shown in Figs. 4-6, respectively with
 219 respect to the differences in curing temperature/periods and NaOH solution molar ratios.

220 According to the data presented in Figs. 4-6, the lowest compressive strength results were
 221 recorded at the curing temperature of 95 °C for all proposed systems. However, despite the
 222 lower values, results well higher than 30 MPa were easily obtainable depending on the
 223 parameters of the alkali-activated binders.

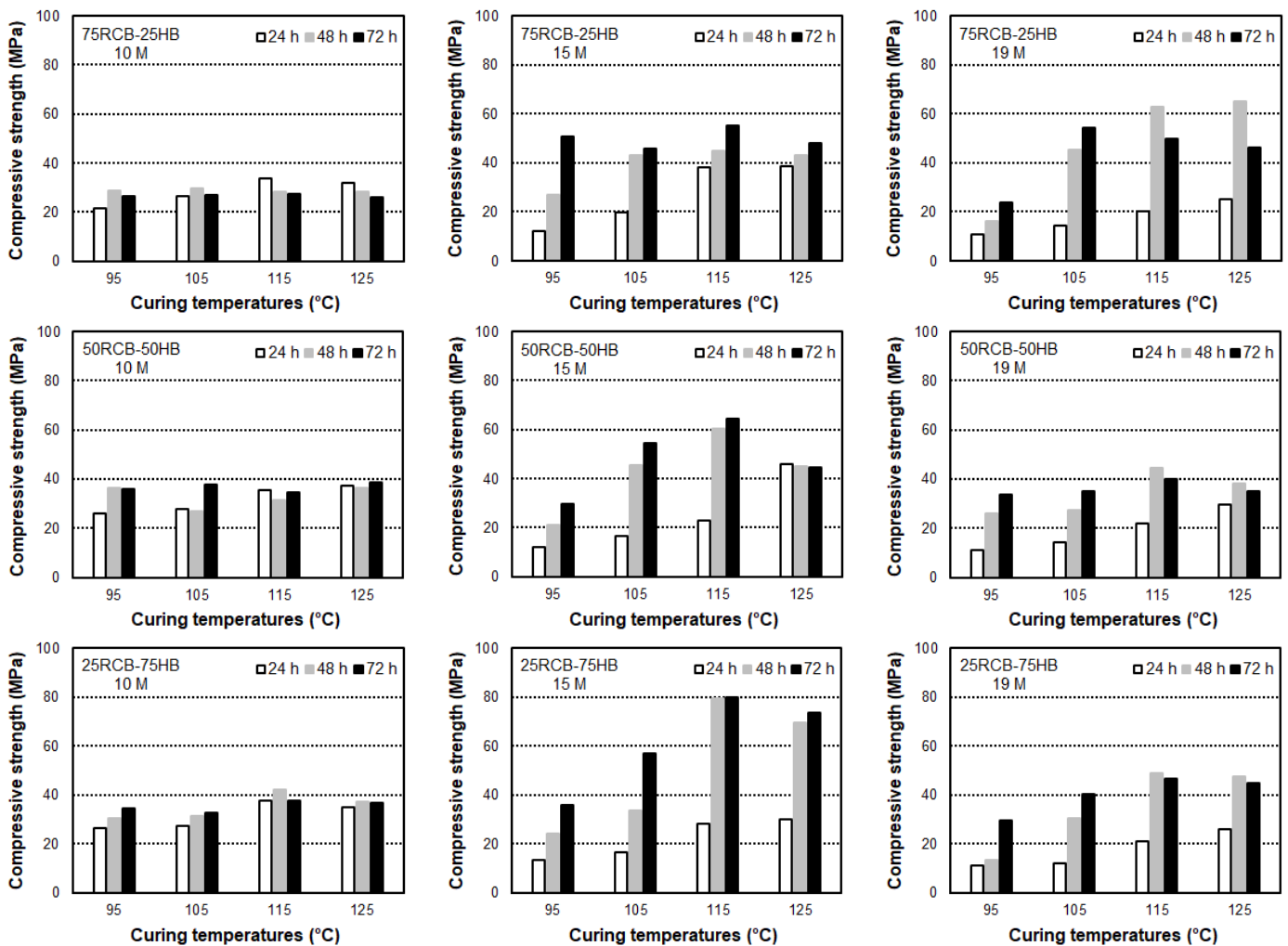




224 **Fig. 4.** Average compressive strength results of alkali-activated binders with different
 225 substitution rates of RCB and RT.

226 When the curing temperature was increased, the results mostly exhibited an incremental
 227 trend irrespective of the differences in precursors (Figs. 4-6). The effect of increased
 228 temperatures on the compressive strength of alkali-activated binders can be more
 229 understandable by looking at the processes involved in alkali activation process. On a general
 230 perspective, three main steps can be mentioned related to the processes of the formation of an
 231 alkali-activated (geopolymer) system. In the first step, continuous dissolution takes place from
 232 the surface of the aluminosilicate precursors to form tetrahedral units of SiO_4 and AlO_4 as a
 233 result of the breakage of $\text{Si} - \text{O} - \text{Si}$ and $\text{Si} - \text{O} - \text{Al}$ bonds in the presence of hydroxide ions.
 234 Secondly, the precursors and alkali polysilicates partially restructure and tetrahedral units of
 235 SiO_4 and AlO_4 are linked alternately to yield amorphous alkali-activated binders. Finally, the
 236 entire system re-precipitates leading to the settlement of an inorganic cross-linked 3-
 237 dimensional network [21]. At higher temperatures, initial dissolution of the aluminosilicate
 238 precursors gets faster due to increased activity of hydroxide ions of alkaline solution [22,23],
 239 which also accelerates the polycondensation reactions, hard structure formation and relatedly
 240 compressive strength results [23,24]. As the supersaturation point gets nearer, the process of

241 polycondensation takes the place of dissolution predominantly, which expels the water
 242 available in the solidified gels. At this stage, the dissolved phases very rapidly polymerize and
 243 reprecipitate, the rearrangement/polymerization of precursors take place and some of the alkali
 244 cations (Na^+) are bonded to the aluminosilicate gel network. It appears that at higher
 245 temperatures of curing, the processes mentioned above are completed earlier [23], which results
 246 in higher grades of compressive strength.



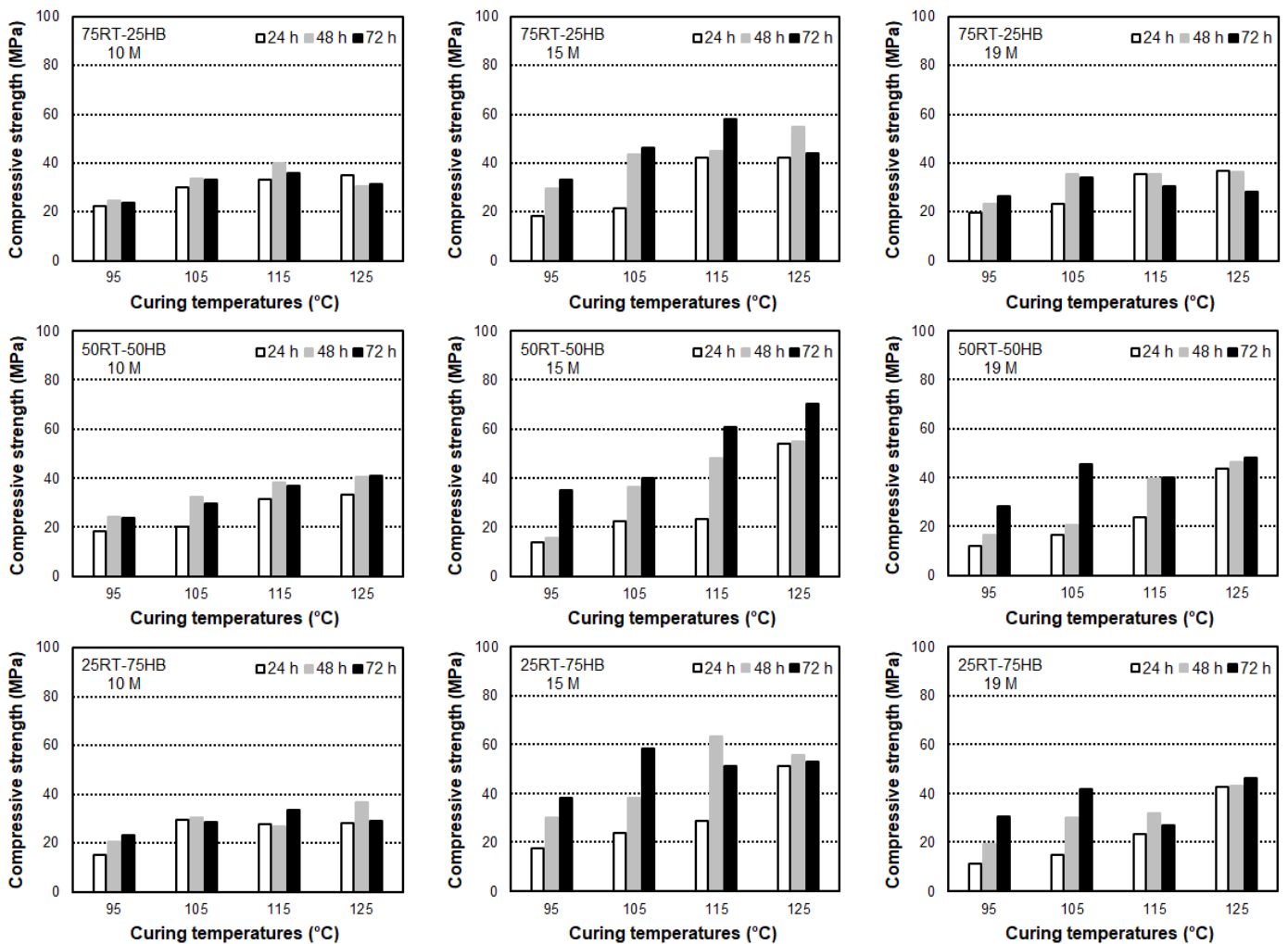
247 **Fig. 5.** Average compressive strength results of alkali-activated binders with different
 248 substitution rates of RCB and HB.

249
 250 For most of the alkali-activated binder mixtures, continuous increments in the compressive
 251 strength results did not continue with the continuous increments in curing temperatures. At a
 252 certain curing temperature, the compressive strength results peaked and after exceeding this
 253 level of curing temperature, results either did not significantly change or decreased (Figs. 4-6).

254 This finding implies that temperature is a parameter which needs to be optimized to reach the
255 highest level of improvement in the microstructures of alkali-activated binders. For alkali-
256 activated binders with different substitution rates of RCB-RT, RCB-HB and RT-HB the level
257 of curing temperature for the achievement of the highest compressive strength results was
258 recorded to be mostly 115 °C. Some of the alkali-activated binders, exhibited maximum
259 compressive strength results at the curing temperature of 125 °C as well (Figs. 4-6), however,
260 in most of the cases, results either did not increase too much or decreased at this temperature
261 level.

262 At highly elevated curing temperatures (e.g. 125 °C herein), fast increments in the viscosity
263 of alkali-activated binder structure can take place with the start of polycondensation and the
264 capture and reaction of aluminosilicate species released after the precursors' dissolution. These
265 phenomena occur rapidly and cause the alkali-activated binders to set promptly. The chance
266 for clotting of the alkali-activated binder slurry increases with the rapid setting, which also
267 increases the chance for undissolved aluminosilicate precursors to be covered. By this way,
268 further dissolution of the precursors is prevented and the transformation from diffluent to a hard
269 and more compact structure is limited. Beyond the optimum curing temperatures (mostly 115
270 °C in this study), elevated temperatures can also cause the contraction of alkali-activated binder
271 gels due to excessive shrinkage/dehydration [24] and consequently cracking at microscale, all
272 of which could be responsible for the stabilization or reduction of the compressive strength.
273 Furthermore, the stabilization or reduction of the compressive strength can also be associated
274 with the possible effects of highly elevated temperatures on the quality of alkali-activation
275 products. When samples are cured at optimum curing temperatures that are not excessively high
276 for a certain mixture proportion, an alkali-activated binder structure that is low-porosity and
277 high-toughness forms since the reaction products find enough time to slowly fill the pores. On
278 the other hand, when the curing temperature is higher than the optimum curing temperature,

279 although the reactions take place very rapidly, a less ordered and more porous structure with
 280 lower-quality reaction products forms which may lower the compressive strength [23]. It can
 281 be stated here that although the negative and positive effects of high temperature curing are
 282 expected to be present for all the curing temperatures utilized in the study, for highly elevated
 283 temperatures (125 °C), negative effects seem to outweigh the positive effects and lower the
 284 compressive strength results.



285 **Fig. 6.** Average compressive strength results of alkali-activated binders with different
 286 substitution rates of RT and HB.
 287

288 When the curing period was extended, compressive strength results of the alkali-activated
 289 binders mostly increased. The increments in the results were clearer from 24 h to 48 h. When
 290 the curing period was extended from 48 h to 72 h, there were quite a number of cases where
 291 either slight changes or reductions in the compressive strength results of alkali-activated binders

292 were noted (Figs. 4-6). Moreover, for the majority of specimens tested, 24 h of heat curing was
293 adequate to obtain the large share of the ultimate compressive strength that will be reached after
294 72 h of heat curing. This therefore implied that curing at elevated temperatures for longer
295 periods is not always an effective way to improve the compressive strength of alkali-activated
296 binders [25]. The positive outcomes of curing for longer periods are related to the beneficial
297 effects of curing at higher temperatures on the previously-noted steps of alkali activation
298 process. Possible reasons for the slight changes or reductions noted in the compressive strength
299 results of certain specimens with the prolonged periods of heat curing can be related to the
300 breakdown of the gelular structure of alkali-activated binder, occurrence of gel contraction,
301 dehydration and excessive shrinkage without transforming into a more semi-crystalline form,
302 the changes in the amorphous phase of gel formation as the crystalline part of the alkali-
303 activated binder is not affected by the longer periods of exposure to heat curing and the
304 inadequate availability of certain amount of water to eliminate cracking and maintain structural
305 integrity [26-28].

306 When the molarity of NaOH solution was increased, compressive strength results generally
307 increased, although there were also clear deviations from this trend depending on the curing
308 temperature/period and the selected precursor type/substitution ratios, especially when the
309 molarity of the alkaline solution was increased from 15M to 19M (Fig. 4). In most of the models
310 proposed for the alkali activation, consecutive events of dissolution, orientation and
311 reprecipitation of precursors and alkaline activators are reported [29]. Among these events,
312 dissolution is regarded to be the most critical since it plays certain roles in relation to the
313 liberation of substances to be used in the formation of SiO_4 and AlO_4 tetrahedral units and in
314 activating the surface bonding reaction (polymerization). The latter strongly contributes to the
315 final strength of the alkali-activated binder structure [30,31]. In line with these statements, the
316 higher compressive strengths of alkali-activated binders produced with higher molarities of

317 NaOH solution can be attributed to the better ability of the highly concentrated NaOH solution
318 to dissolve the CDW-based precursors and form a polymerized network having strengthened
319 link with the dissolved particles of the precursors [32]. On the other hand, reductions noted in
320 the compressive strength results with the increased NaOH solution molarities depending on the
321 other mixture parameters were attributed to the coagulation of silica [33,34] and faster setting
322 which does not allow for a homogenous mixing resulting in a poor and incipient polymerization
323 [35].

324 Activators are usually the most expensive components of alkali-activated binders or
325 geopolymers [12] and their optimization is therefore desirable, taking the cost and
326 environmental impact considerations into account. It is important to state that although
327 increased compressive strength results were recorded from alkali-activated binders with NaOH
328 solutions having higher molarities on a general perspective, reasonable compressive strength
329 results which were in the range of 25-35 MPa were easily obtainable from the proposed alkali-
330 activated binders even at NaOH molarity of 10M, depending on the combinations of precursors
331 (Figs. 4-6).

332 Despite the certain differences in the physical properties (particle size and diameter [see Fig.
333 2 and Table 1]) and chemical compositions (see Table 2) of RCB and RT, as detailed in Section
334 2.1, the utilization of these CDW-based precursors at different substitution rates did not create
335 distinctive differences in the compressive strength results. As can be seen from Fig. 4, for each
336 binary combination of RCB and RT, the compressive strength results are close when other
337 mixture/production parameters are kept the same. This suggests that RCB and RT compensated
338 for each other's drawbacks as precursors and more or less acted as a single precursor.

339 At similar substitution ratios, the average compressive strength results of RCB-HB-based
340 alkali-activated binders were found to be comparably higher than those of RCB-RT-based
341 alkali-activated binders, especially when the curing periods were extended, which showed that

342 the utilization of HB instead of RT promoted the achievement of higher strength grades.
343 Depending on the other mixture parameters, it was possible to reach an average compressive
344 strength value of 80 MPa for RCB-HB-based alkali-activated binders (e.g. 25RCB-75HB
345 specimens produced with 15M of NaOH solution and cured at 115 °C for 48h and 72h) (Fig.
346 5). Alkali activation is affected to a great extent by the chemical composition, solubility, particle
347 size distribution/fineness and the degree of amorphousness of precursors. The alkali activation
348 capability of the aluminosilicate precursors is usually perceived to be more pronounced when
349 highly amorphous structure, high amounts of siliceous/aluminous oxides and smaller grain size
350 are present. In addition, it has been reported that significantly higher compressive strength
351 results could be obtained from alkali-activated binders when the particle fractions of precursors
352 are kept under 150 μm and the D_{50} value is less than 15 μm [13]. As can be seen in Table 1,
353 Fig. 2 and Fig. 3, among all CDW-based precursors used in this work, HB had D_{50} value of 27.5
354 μm , which was the coarsest in terms of the grain size and exhibited more distinctive crystalline
355 peaks in the XRD analysis, all of which contradicting with the abovementioned parameters that
356 nourish a better alkali activation capability in general. On the other hand, HB possessed a higher
357 total amount of siliceous/aluminous oxides (78.9%) compared to RCB (73.9%) and RT
358 (69.9%). The relationship between the compressive strength results, physical properties and
359 chemical composition of precursors, therefore showed that despite the importance of other
360 properties of the precursors, their chemical composition (i.e. aluminosiliceous material
361 composition) has a more decisive influence on the compressive strength results. This result was
362 also found to be in concordance with the study of De Silva et al. [22] who stated that even the
363 minor changes in the available Si and Al concentrations during alkali activation can drastically
364 affect the properties of the ultimate alkali-activated binder.

365 As the chemical compositions of different CDW-based precursors are quite similar to each
366 other (Table 2), one might still consider if this much of a small difference can lead to such

367 differences in the compressive strength results of the alkali-activated binders. In this regard, the
368 study of Van Jaarsveld et al. [36] also concluded similar findings for fly ash-based geopolymers.
369 They reported that two types of fly ash which were acquired from the same source and had
370 nearly identical average particle sizes with only slight differences in their CaO contents, showed
371 significantly different setting behaviours. This difference was associated with the greater
372 tendency of one of the fly ashes to aggregate more within the aqueous media due to the
373 differences in the surface charge of particles. It was concluded in this sense that zeta-potential
374 of fly ash can be decisive on the observed differences in the setting and dissolution rate of fly
375 ash [36]. This behaviour mentioned in [36] is also likely to cause the differences noted in the
376 compressive strength results of alkali-activated binders with CDW-based precursors having
377 rather similar chemical compositions.

378 With the increased substitution rates of HB, a general incremental trend in the compressive
379 strength results of RT-HB-based alkali-activated binders was observed, especially for
380 specimens cured for longer periods. In addition, the compressive strength results of RT-HB-
381 based alkali-activated binders (Fig. 6) were higher than RCB-RT-based alkali-activated binders
382 (Fig. 4), which again showed the positive effects of the utilization/increased amounts of HB in
383 the alkali-activated binder production. When the plots shown in Figs. 5 and 6 are compared, it
384 can be seen that the compressive strength results of RCB-HB-based alkali-activated binders are
385 slightly higher than RT-HB-based alkali-activated binders, which suggested that RCB worked
386 better with HB and favoured the strength evolution. The better synergy of RCB with HB
387 compared to RT can be explained by the smaller particle diameter/gradation (see Table 1 and
388 Fig. 2) and the higher total amount of siliceous/aluminous oxides (Table 2) in RCB compared
389 to RT. The comparatively higher amount of CaO (7.42%) available in the composition of RT
390 with respect to other precursors having maximum CaO content of 4.75% may have also
391 contributed to the slightly lower compressive strength results in the RT-HB-based geopolymers.

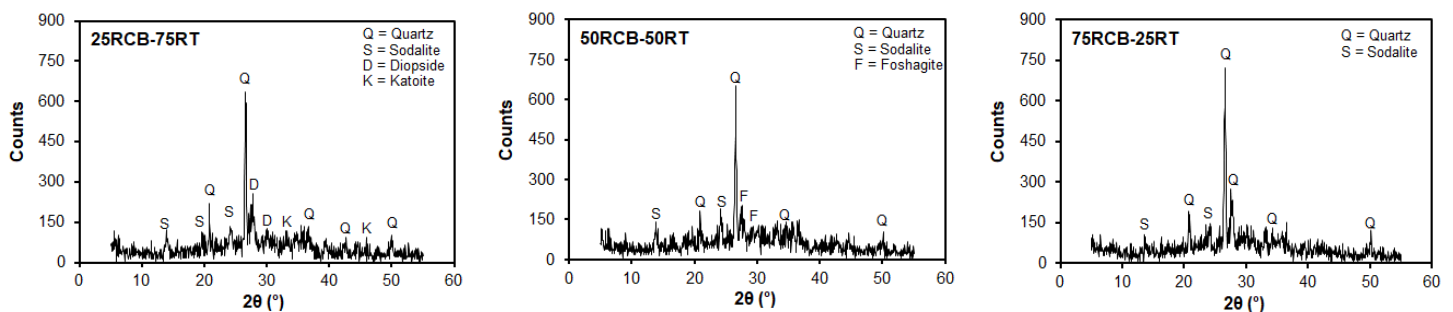
392 As the Ca^{+2} species dissolve from aluminosilicate material in high alkaline medium and react
393 with OH^- ions in the system, a decrease in strength results can be observed as a result of
394 lowering the pH of the system and precipitation as $\text{Ca}(\text{OH})_2$ [13,37]. CaO can also react with
395 SiO_2 phases and result in calcium-silicate-hydrate (CSH) gels similar to the case observed in
396 Portland cement-based systems. However, the amount of CaO, even in RT, is relatively low for
397 the precursors used herein to assure stable CSH gel formation therefore, calcium-based gel
398 formation was not observed as also explained in the following sections.

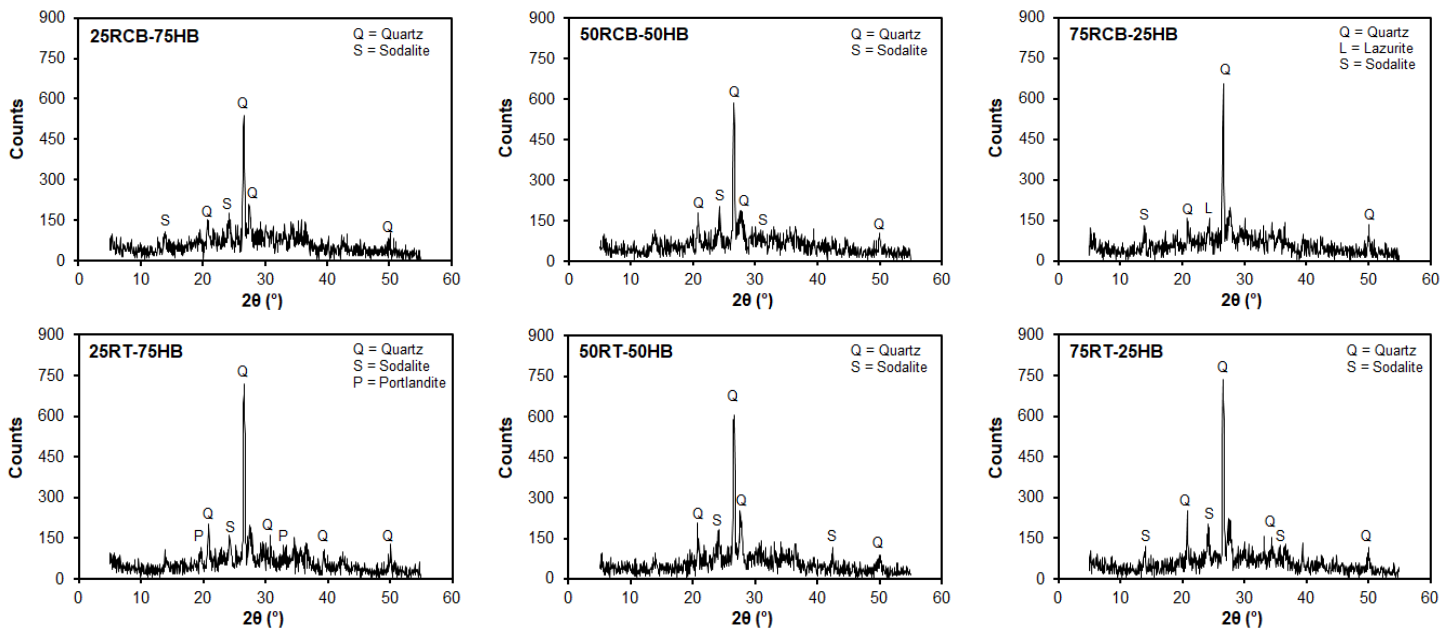
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400 3.2 Microstructural characterization

401 3.2.1 X-ray diffraction (XRD)

402 In Fig. 7, the selected XRD diffractograms of alkali-activated binders produced with
403 different substitution rates of CDW-based precursors are shown. For further discussions, the
404 mixture parameters that generally yielded optimum compressive strength results for the alkali-
405 activated binders are chosen. Therefore, the XRD diffractograms belonging to alkali-activated
406 binders activated with 15M NaOH solution and cured at 115 °C for 48 hours are the ones
407 incorporated in Fig. 7. It needs to be stated here that although the intensity of peaks and the
408 ultimate alkali activation products changed with regard to the rest of the mixture design
409 parameters, these changes were not dramatic, suggesting that the results presented in Fig. 7 are
410 representative of a general behaviour.





411 **Fig. 7.** XRD diffractograms of alkali-activated binders having different substitution rates of
 412 CDW-based precursors, activated with 15M NaOH solution and cured at 115 °C for 48 hours.
 413 [Quartz (SiO₂) PDF No: 96-101-1160, Sodalite (Na₄Al₃Si₃O₁₂Cl) PDF No: 96-900-3326,
 414 Diopside (CaMg_{0.7}Si_{1.7}O₆) PDF No: 96-900-5280, Katoite (Ca₃Al₂Si₃O₁₂H₁₂) PDF No: 96-
 415 900-2709, Foshagite (Ca₄Si₃O₁₁H₂) PDF No: 96-901-1044, Lazurite
 416 (Na_{3.48}Ca_{0.6}Al_{2.91}Si_{3.09}O_{11.52}S) PDF No: 96-901-1357, Portlandite (Ca(OH)₂) PDF No: 96-900-
 417 6836]
 418

419 As can be seen from the XRD diffractograms of the raw precursors (Fig. 3), RCB and HB
 420 are crystalline in nature while RT is amorphous. Alkali-activated binders (geopolymers) are
 421 amorphous to X-rays, although there is also clear evidence in the literature that final alkali
 422 activation products can show structure ranging from amorphous to near-crystalline [26]. In the
 423 case of current research, for all of the studied mixtures, the XRD diffractograms of the alkali-
 424 activated binders exhibited clear crystalline peaks. However, evidently, the intensity of peaks
 425 of the raw precursors (Fig. 3) either significantly decreased or changed into crystals with
 426 different nature (Fig. 7). Decrements and/or changes in the XRD patterns of alkali-activated
 427 binders were anticipated and suggestive of the occurrence of alkali-activation, since these
 428 minerals of precursors are used in the dissolution/reorganization steps of alkali activation.
 429 Studies available in the literature also found clear peaks visible to X-rays after XRD analyses
 430 [19,38]. Those studies attributed the availability of such crystalline phases in alkali-activated
 431 binders to the unreacted feedstock of crystal phases of the precursors and/or formation of

432 zeolitic structures, overlapping the amorphous baseline, particularly when highly concentrated
433 NaOH is used as the activator [19,38].

434 It was reported in [39] that the formation of different zeolitic reaction products is affected
435 by the molarity of the NaOH solution and when the molarities higher than 5M are targeted
436 (similar to the cases here), cancrinite group of minerals (e.g. herschelite, hydroxysodalite or
437 hydroxycancrinite) are usually generated. These minerals, in terms of their crystal structure,
438 show similarities to the zeolitic minerals. Similar findings are also reported in [40,41] where it
439 was concluded that sodium aluminosilicate (N-A-S-H) gel is the main reaction product for fly
440 ash-based alkali-activated materials. N-A-S-H is also the main reaction product of the alkali-
441 activated binders of the current study, as will be detailed in the following section. Due to their
442 medium/long-range disorder, N-A-S-H gels were reported to be X-ray amorphous, however,
443 the gels were also found to show zeolite-like three-dimensional structure at nano-scale [40]. In
444 accordance with [42,43], this confirms that N-A-S-H gel is a zeolite precursor with a
445 thermodynamic tendency and has a high likelihood to crystallize into a zeolite, which may
446 explain the evident crystalline peaks observed in this study after alkali activation process.

447 As can be seen from Fig. 7, after alkali activation, irrespective of the different substitution
448 rates of precursors, peaks of mullite, annite and albite in the precursors disappeared entirely and
449 crystalline peaks related to quartz (SiO_2 , PDF No: 96-101-1160), sodalite ($\text{Na}_4\text{Al}_3\text{Si}_3\text{O}_{12}\text{Cl}$,
450 PDF No: 96-900-3326), diopside ($\text{CaMg}_{0.7}\text{Si}_{1.7}\text{O}_6$, PDF No: 96-900-5280), katoite
451 ($\text{Ca}_3\text{Al}_2\text{Si}_3\text{O}_{12}\text{H}_{12}$, PDF No: 96-900-2709) foshagite ($\text{Ca}_4\text{Si}_3\text{O}_{11}\text{H}_2$, PDF No: 96-901-1044),
452 lazurite ($\text{Na}_{3.48}\text{Ca}_{0.6}\text{Al}_{2.91}\text{Si}_{3.09}\text{O}_{11.52}\text{S}$, PDF No: 96-901-1357 and portlandite ($\text{Ca}(\text{OH})_2$, PDF
453 No: 96-900-6836) were visible.

454 Among alkali activation products which were visible under X-rays, only sodalite constitutes
455 an ion (chloride [Cl^-]) which was not available in the compositions of CDW-based precursors
456 (Table 2, Fig. 3). Available Cl^- ions in sodalite mineral are most likely to be originated from the

457 NaOH solution which incorporates certain amount of sodium chloride, as noted in Section 2.1.
458 As seen from Fig. 7, peaks of quartz and sodalite were both clearer and higher in intensity for
459 all alkali-activated binders in addition to the less intensified peaks of lazurite, diopside, katoite,
460 foshagite and portlandite. Peaks related to quartz and zeolite type minerals such as sodalite and
461 lazurite were noted in alkali-activated binders produced with Al-rich precursors [39,44]. Peaks
462 related to lazurite, diopside, katoite, foshagite and portlandite were associated with the reactions
463 between precursors and NaOH solution involving the participation of CaO and MgO, depending
464 on the resultant mineral type. As discussed in the literature, it is assumed that the calcium
465 species dissolved from aluminosilicate based precursors will precipitate as portlandite at the
466 initial dissolution stage in an alkaline environment, and will participate in the formation of C-
467 S-H gel at later stages [10,45]. However, these peaks were not of high intensity, therefore
468 minerals related to these peaks did not play a decisive role in changing the overall nature of
469 ultimate alkali activation reaction products, as discussed in the following section. Calcination
470 of clayey materials at temperatures higher than 950 °C leads to the formation of mullite crystals,
471 which are stated to be non-reactive in alkaline activation [46]. In this study, however, mullite
472 crystals available in some of the precursors disappeared after alkali activation, which was an
473 outcome concordant with several literature studies [42,47] that stated it was possible to change
474 the mullite crystals under strongly alkaline environments.

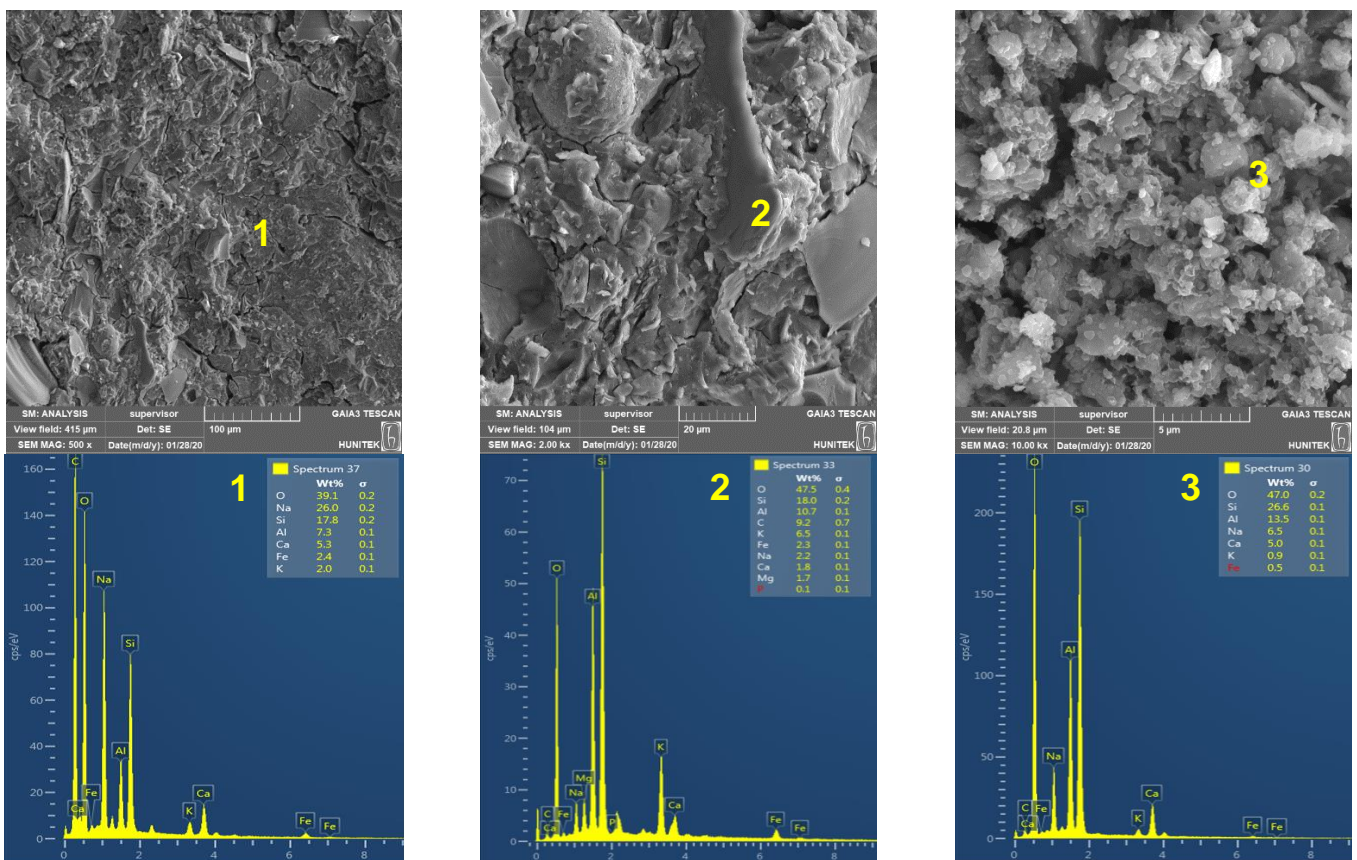
475 When compared to each other, it was not easy to clearly comment on the relationship
476 between the values of compressive strength and the XRD diffractograms of the alkali-activated
477 binders produced with different binary substitutions of CDW-based precursors. The intensities
478 and types of crystalline peaks were similar to each other. However, it can be stated based on
479 Fig. 7 that when the amount of HB was increased in the alkali-activated binder mixtures
480 (especially for RCB-HB binary combination), the intensity of the main quartz peaks started to

481 be less pronounced, suggesting better alkali activation process. This fact can be related to the
482 higher compressive strength results of corresponding specimens as discussed formerly.

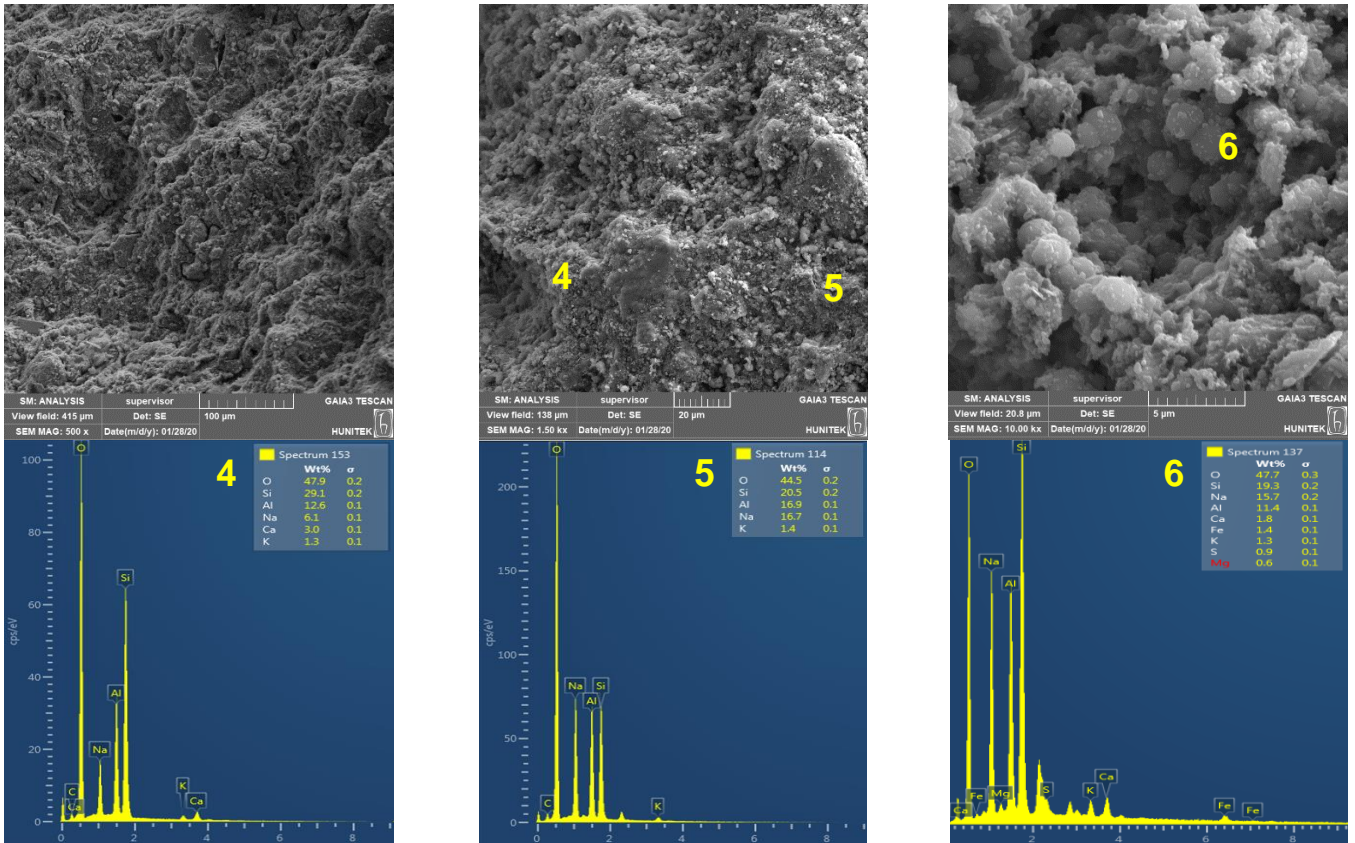
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484 **3.2.2 Scanning electron microscopy (SEM/EDX)**

485 In this section, the products that were formed after the alkali activation is detailed
486 considering the SEM micrographs, supported by EDX analyses, performed on the selected
487 regions. Similar to the XRD results, the analyses are focused on alkali-activated binders having
488 different substitution rates of CDW-based precursors, activated with 15M NaOH solution and
489 cured at 115 °C for 48 hours. To be more concise, Figs 8-10 show the SEM/EDX analyses on
490 alkali-activated binders with the precursor combinations of 25RCB-75RT, 25RCB-75HB,
491 25RT-75HB mixtures that showed different levels of compressive strength for the selected
492 molarity and curing conditions, respectively.

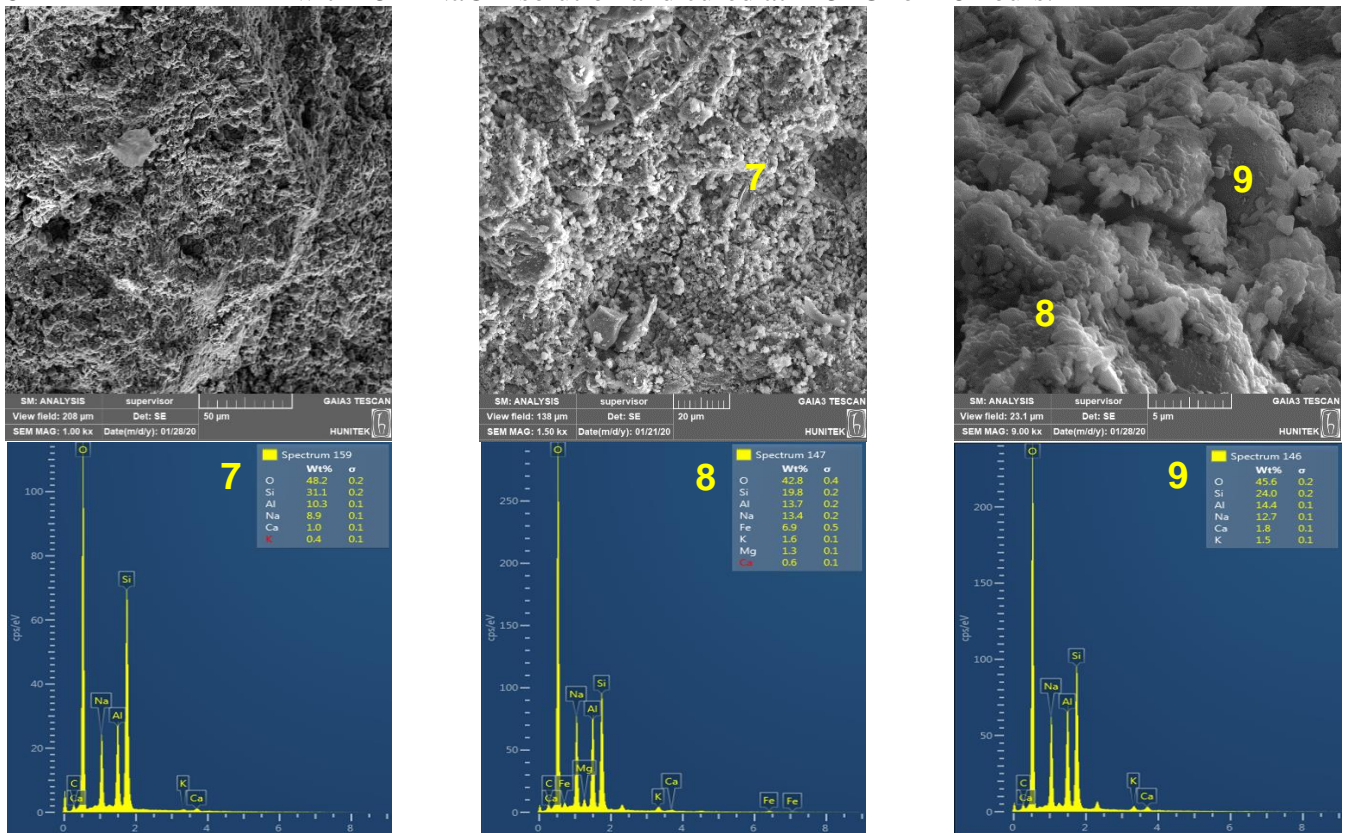
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494 **Fig. 8.** SEM micrographs with EDX spectra of 25RCB-75RT alkali-activated binder activated
495 with 15M NaOH solution and cured at 115 °C for 48 hours.
496



497 **Fig. 9** SEM micrographs with EDX spectra of 25RCB-75HB alkali-activated binder activated
 498 with 15M NaOH solution and cured at 115 °C for 48 hours.



499 **Fig. 10.** SEM micrographs with EDX spectra of 25RT-75HB alkali-activated binder activated
 500 with 15M NaOH solution and cured at 115 °C for 48 hours.
 501

502 When compared to the SEM photos of precursors shown in Fig. 1, clearly, significant
503 improvements and densifications took place in the microstructures of the mixtures after alkali
504 activation, irrespective of the way the precursors were combined. Furthermore, there were
505 differences noted in the microstructures of different alkali-activated binders, although not that
506 substantial. As can be seen in Fig. 8, there are certain microcracks, irregularities in the
507 distribution of the ultimate products and also undissolved raw materials available in the
508 microstructure of the 25RCB-75RT-based alkali-activated binders. However, the
509 microstructures of the 25RCB-75HB and 25RT-75HB-based alkali-activated binders did not
510 show microcracking presence. Those alkali-activated binders were more compact, with more
511 uniformly distributed products and with less undissolved raw materials (especially those with
512 25RCB-75HB [Fig. 9]). These findings for different alkali-activated binders are in line with the
513 compressive strength results discussed previously.

514 For all the samples, clear peaks of Na, Al and Si with different intensities were observed in
515 the EDX spectra, in addition to crystal-like structures observed in the SEM micrographs. These
516 findings therefore suggest that the main reaction products of the alkali-activated binders
517 produced here are N-A-S-H gels, which are combined with zeolitic polytypes (e.g. sodalite,
518 lazurite) having a structure ranging from amorphous to polycrystalline. It must be also noted
519 that there are slightly visible Ca peaks in the 25RCB-75RT-based alkali-activated binders,
520 which were also evidenced in the XRD results in the form of CaO bearing minerals such
521 diopside and katoite and was found attributable to the higher CaO content of RT.

522

523 **4. Conclusions**

524 This study showed that precursors obtained from CDW-based masonry units can be used
525 together in the production of alkali-activated binders. It is notable that this can be achieved by
526 a simple combination of these units at different proportions rather than using them singly, which
527 requires selective demolishing practises/individual separation and can be time-

528 consuming/energy-inefficient. Additionally, the following conclusions were drawn from the
529 current research:

- 530 • The compressive strength of alkali-activated binders from precursors of CDW-based
531 masonry tends to increase with the increments in curing temperature/period, molarity of
532 NaOH used as the alkaline activator and the increased amounts of HB in precursor
533 combinations. The highest compressive strength results were generally reached at curing
534 temperature/period and NaOH solution molarity of 115°C/48 hours and 15M. Using these
535 mixture design parameters, compressive strength values as high as 80 MPa could be
536 obtained. Depending on different design parameters, compressive strength results ranging
537 from 25 to 30 MPa are easily obtained even after 24 h of curing at 95 °C (which were the
538 lowest curing temperature and shortest curing period used herein).
- 539 • The compressive strength results were largely concordant with the microstructural analyses.
540 For all the alkali-activated binders, the main alkali activation products were N-A-S-H gels
541 containing different zeolitic polytypes (e.g. sodalite, lazurite), which had a structure ranging
542 from amorphous to polycrystalline nature.

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