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# Synthesis of Geopolymer Using Alkaline Activation of Building-Related Construction and Demolition Wastes

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## Abstract

There are over 2 billion tons of building-related construction and demolition waste (brCDW) produced annually in China, but less than 30% of the waste is presently recycled. The majority of the brCDW is being disposed of in landfills in suburban or rural areas, resulting in a serious waste of resources and environmental pollution issues. This study proposed a high-value utilization approach for the brCDW, which was used as a precursor material to synthesize the high strength and environmentally friendly geopolymer. The brCDW was classified into three main components, namely, brick, ceramic, and concrete. The X-ray fluorescence (XRF), X-ray diffraction (XRD), particle size analyzer, and unconfined compressive strength tests were conducted to evaluate the influences of chemical composition and particle size of precursor materials on the strength of geopolymers derived from single and mixed brCDW components. The results demonstrated that the brick geopolymer exhibited a low strength in the early stage but a significantly high strength in the late stage. The ceramic geopolymer showed a low strength in both the early and late stages. The strength of concrete geopolymer was high in the early stage, but increased slowly later on. The mixed brCDW geopolymer had the highest early strength and the second-highest final strength compared to the single-component derived geopolymers. Furthermore, the XRD, scanning electron microscopy (SEM), and energy dispersive spectroscopy (EDS) tests were conducted to explore their microscopic mechanisms. The precursor materials dissolved in the geopolymerization process, thereby generating gels to coat and bind the unreacted particles. The formed gel type varied due to the different chemical composition ratios of precursor materials. The morphology and structure of brick geopolymer gels differed from other geopolymers, with the main gel products being C-A-S-H and N-A-S-H. The primary gel of ceramic, concrete, and mixed precursor geopolymers was the C-S-H gel, possibly with small amounts of C-A-S-H and N-A-S-H gel. Both the formed gel type and the compactness of the microstructure had significant influences on the geopolymer strength.

40 **Keywords:** Geopolymer, Building-Related Construction and Demolition Wastes,  
41 Alkaline Activation, Microstructure

42

## 43 **1 Introduction**

44 With the rapid urbanization and infrastructure reconstruction, there are over 2  
45 billion tons of building-related construction and demolition wastes (brCDW) produced  
46 annually in China, but less than 30% of the waste is presently recycled (Akhtar and  
47 Sarmah, 2021; Zhang et al. 2019; Zhang et al. 2020). The majority of the brCDW is  
48 being disposed of in landfills in suburban or rural areas, resulting in a serious waste of  
49 resources and environmental pollution issues (Zhang et al. 2019). Recently, the concept  
50 of a “zero-waste city” has been proposed to minimize the amount of produced waste  
51 and strengthen the recycling program. The Chinese Ministry of Ecology and  
52 Environment has also promoted this concept in more than 100 cities, aiming to improve  
53 the recycling rate of brCDW up to 60%. To achieve this goal, one potential solution is  
54 to develop a high-value utilization approach for the brCDW.

55 In road engineering, the recycled brCDW has been widely used as an alternative  
56 for subgrade filler or aggregates in asphalt mixture, cement concrete, granular base, and  
57 subbase (Zhang et al., 2019; Zhang et al., 2020; Hu et al., 2022; Tang et al., 2021; Guo  
58 et al., 2018; Bai et al., 2020; Chen et al., 2021). However, using the brCDW as a  
59 subgrade filler is a low-value approach, especially for the construction projects with  
60 heavy costs of material transportation. While using the brCDW as alternative  
61 aggregates improved its added value, the relevant implementation is limited due to the  
62 poor quality of the recycled aggregates. The existing studies were focused on the  
63 physical-chemical-biological modification of the recycled brCDW, but there was still  
64 lack of reliable technology to modify the brCDW for use in the upper layers of  
65 pavements (Mohammed et al., 2021; Peng et al., 2023; Mistri et al., 2020; Li et al.,  
66 2022; Feng et al., 2022; Tang et al., 2023). Geopolymer is a class of aluminosilicate  
67 materials with a three-dimensional structure ranging from an amorphous to semi-  
68 crystalline state. It is formed by polymerizing different types of aluminosilicate  
69 precursors through an activation process using alkaline activators (Davidovits, 1991).  
70 Compared to Portland cement, the production of geopolymer can approximately reduce  
71 carbon dioxide emissions by 40-60% and energy consumption by 60% (Gu et al., 2019;  
72 Barcelo et al., 2014; McLellan et al., 2011; Neupane, 2022). Additionally, geopolymer  
73 exhibits excellent engineering properties such as rapid setting, high strength, and high  
74 corrosion resistance (Vafaei et al., 2018; Aiken et al., 2018; Aliques-Granero et al., 2019;  
75 Lahoti et al., 2019; Shill et al., 2020; Obeng et al., 2023). The brCDW typically consists  
76 of brick, concrete, and ceramic, which contain large amounts of Si, Al, and Ca elements  
77 (Komnitsas et al., 2015). This indicates that the brCDW can serve as a suitable  
78 precursor for geopolymer, which will be a high-value and possibly environmentally  
79 friendly binder for construction materials.

80 The engineering performance of geopolymer is mainly dependent on the chemical  
81 composition and particle size of precursor materials and the type and dosage of alkaline  
82 activators (Komnitsas et al., 2015; Leong et al., 2016; Özbayrak et al., 2023; Jiang et  
83 al., 2023; Jiang et al., 2023; Zhang et al., 2020). Fly ash, metakaolin, and slag are the  
84 most commonly used precursor materials in geopolymer. The previous studies had  
85 shown that the geopolymers derived from fly ash and metakaolin had an unconfined  
86 compressive strength (UCS) ranging from 30 to 60 MPa after 28 days of curing, which  
87 was comparable to that of cement concrete (Luna-Galiano et al., 2022; Temuujin et al.,  
88 2010; Tchakouté et al., 2017; Chen et al., 2016). By using slag as a precursor material,  
89 the geopolymer had even a higher UCS (i.e., 80 to 100 MPa) (Lemougna et al., 2020;  
90 Komnitsas et al., 2009; Zhang et al., 2020). Due to the high residual alkalinity, many  
91 types of industrial wastes such as red mud, coal gangue, and waste gypsum were also  
92 used as a precursor material in geopolymer production. Although the geopolymers  
93 derived from the industrial wastes had acceptable UCS, the leaching of heavy metals  
94 from the geopolymers impeded their implementation (Nie et al., 2019; Yang et al., 2019;  
95 Zhang et al., 2020).

96 In recent years, some pioneer researchers have used the brCDW as a precursor  
97 material for geopolymer production. They found that the UCS of brCDW-derived  
98 geopolymer had a wide variation, ranging from 10 MPa to 60 MPa (Sun et al., 2013;  
99 Tan et al., 2022; Bassani et al., 2019; Hwang et al., 2019; Tuyan et al., 2018; Moreno-  
100 Maroto et al., 2022; Xiao et al., 2022). This variability is attributed to the current  
101 synthesis approaches, which do not adequately control the chemical composition and  
102 particle size of brCDW. These factors may significantly impact the engineering  
103 properties of geopolymer (Komnitsas et al., 2015; Petrakis et al., 2019; Ahmari et al.,  
104 2012). The strength of the geopolymer is also influenced by the type and dosage of the  
105 alkaline activator (Tchakouté et al., 2017; Kovtun et al., 2015; Helmy et al., 2016). The  
106 most commonly used alkaline activators include sodium hydroxide solution and sodium  
107 silicate solution. Many researchers had demonstrated that using a mixed alkaline  
108 activator, rather than a single component, could significantly enhance the activation  
109 effect, resulting in a higher strength of geopolymer (Ma et al., 2019). Note that  
110 excessively high or low modulus (the mole ratio of  $\text{SiO}_2$  to  $\text{Na}_2\text{O}$ ) and dosage of  
111 alkaline activators adversely affect the geopolymerization process and compromise the  
112 quality of geopolymer. Thus, there is an optimal modulus and dosage for an alkaline  
113 activator that achieves the highest strength of the geopolymer (Sivasakthi et al., 2021;  
114 Ouyang et al., 2019; Luukkonen et al., 2020).

115 To address the aforementioned problems, this study aimed to develop an approach  
116 for synthesizing a high strength geopolymer derived from the brCDW. The recycled  
117 brCDW was classified into three components, namely, brick, ceramic, and concrete, for  
118 analyzing the impact of the chemical composition of each component on the strength  
119 of the geopolymer. Furthermore, this study evaluated the influences of the modulus and

120 dosage of the alkaline activator on the UCS of brCDW geopolymer, which was used to  
121 determine the optimal mix design for the geopolymers with single and mixed brCDW  
122 components. Finally, the microscopic mechanisms of brCDW geopolymer were  
123 revealed via the X-ray diffraction (XRD), scanning electron microscopy (SEM) and  
124 energy dispersive spectroscopy (EDS) tests (Gill et al., 2023).

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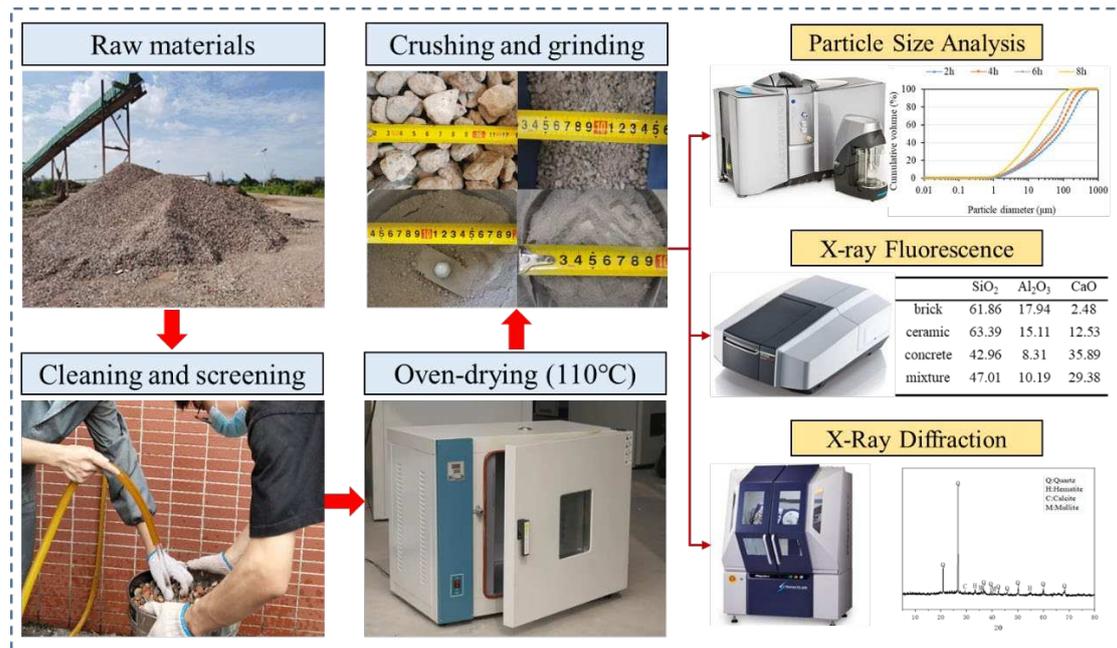
## 126 **2 Materials and test methods**

### 127 ***2.1 Raw materials***

128 The brCDW was collected from a local aggregate recycling plant, where the  
129 brCDW was pre-processed via crushing, separation, screening and stockpiling. In the  
130 laboratory, the brCDW with a particle size above 26.5 mm was cleaned by removing  
131 the debris such as plastic, metal and glass, and then dried overnight in an oven at 110°C.  
132 The processed brCDW consisted of 79% concrete, 16% brick and 5% ceramic in terms  
133 of weight proportion. To investigate the impact of the chemical composition of brCDW  
134 on the mechanical properties of the synthesized geopolymer, the brCDW was separated  
135 into concrete, brick and ceramic stockpiles. Sodium silicate and sodium hydroxide  
136 solutions were used as the alkaline activator. The sodium silicate solution was a  
137 transparent viscous liquid, composed of 27.3% SiO<sub>2</sub>, 8.5% Na<sub>2</sub>O, and 64.2% H<sub>2</sub>O. The  
138 mole ratio of SiO<sub>2</sub> to Na<sub>2</sub>O for sodium silicate was 3.3. The sodium hydroxide was a  
139 white flake solid with a purity level larger than 98%. The deionized water was used for  
140 diluting the alkaline activator solution. In this study, the diluted alkaline activator  
141 solution stood in the laboratory environment for 24 hours before use.

### 142 ***2.2 Preparation of precursor materials***

143 To prepare the precursor materials, the recycled concrete, brick and ceramic  
144 were crushed, pulverized and ground, respectively. As shown in Figure 1, the recycled  
145 materials were crushed by a jaw crusher to a size range of 3-20 mm, and then pulverized  
146 by an ore mill to a size range of 100-600 μm. Finally, the pulverized materials were  
147 ground by a ball mill for a certain period of time. Since the particle size and uniformity  
148 of the precursor significantly affected the mechanical properties of geopolymer, this  
149 study aimed to assess the appropriate grinding time for the preparation of brCDW  
150 precursors. Thereby, four ball milling times (i.e., 2, 4, 6, and 8 hours) were selected to  
151 explore their impact on the particle size distribution of each precursor. The Mastersizer  
152 3000 laser particle size analyzer was employed to determine the particle size  
153 distributions of the processed precursors.



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156

**Figure 1. Pretreatment and research of Precursors**

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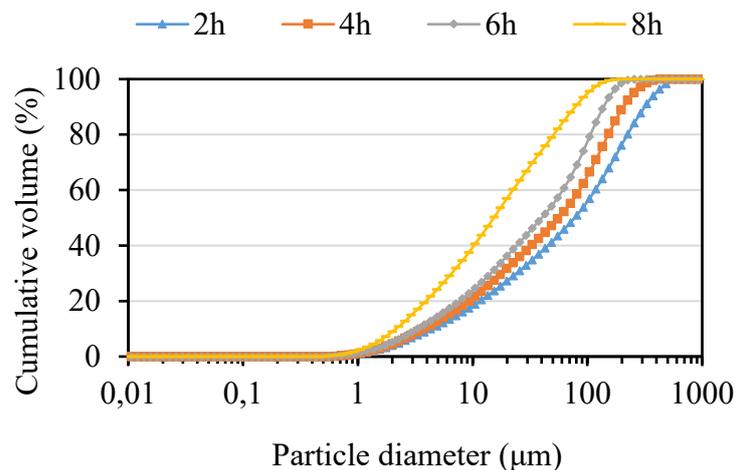
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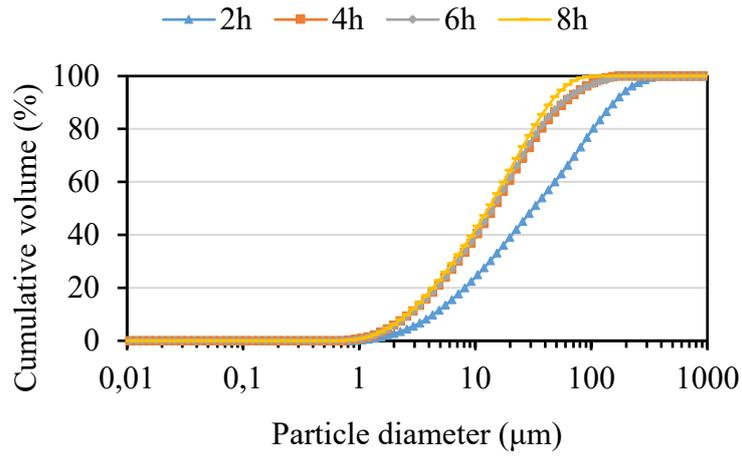
Figure 2 showed the particle size distributions of the processed concrete, brick and ceramic with different milling times. The median particle diameter (D50) was used to quantify the change in particle size of the precursor. As shown in Figure 3, the brick particle was much coarser than the ceramic and concrete particles after 2 hours of milling, and all the ground particles had a D50 smaller than 15 µm after 8 hours of milling.



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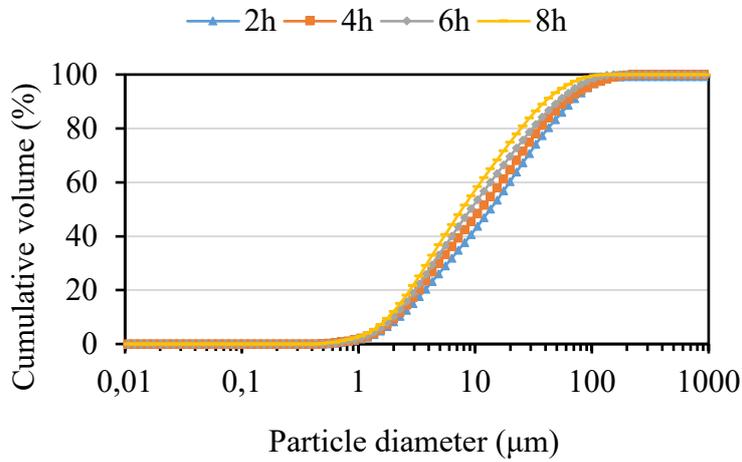
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**a. Brick precursor**



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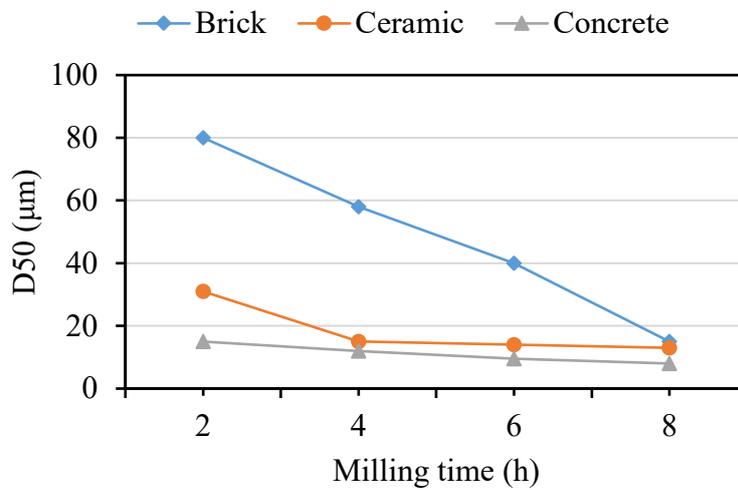
**b. Ceramic precursor**



167  
168

**c. Concrete precursor**

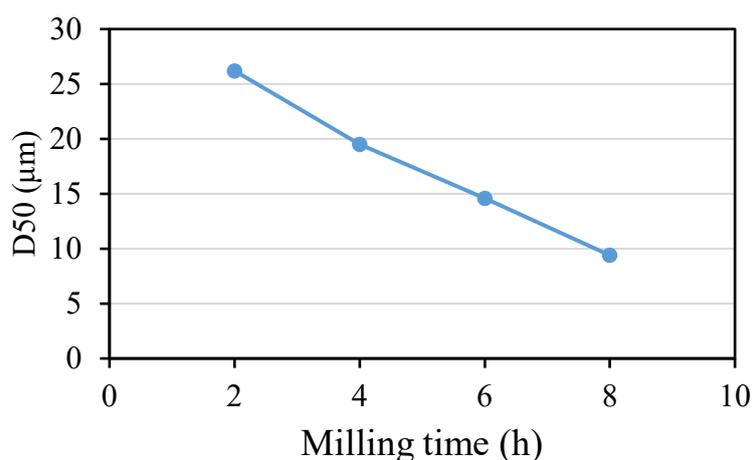
**Figure 2. Particle size distribution of brCDW**



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171

**Figure 3. Relationship between D50 of brCDW and milling time**

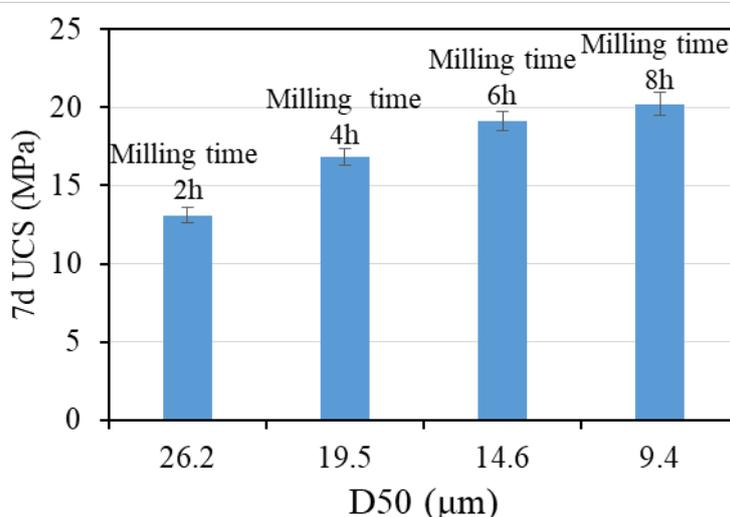
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174

**Figure 4. Relationship between D50 of Mixed precursor and milling time**



175

176

**Figure 5. Relationship between UCS of geopolimer and D50**

177 In general, a smaller particle size yields a larger specific surface area, which  
 178 will increase the reaction rate between the brCDW and the alkaline activator (Petrakis  
 179 et al., 2019; Ahmari et al., 2012). In order to explore the influence of particle size of  
 180 precursor materials on geopolimer strength, the mixed precursor materials with  
 181 different particle sizes were obtained after various milling times. According to the  
 182 original proportion of each component in brCDW (brick: ceramic: concrete =16:5:79),  
 183 the D50 of mixed precursor materials under different milling time was calculated using  
 184 the weighted average method, as shown in Figure 4. The geopolimer was prepared with  
 185 mixed precursors for 7-day UCS test. Herein, the modulus of the alkaline activator was  
 186 1.0, the dosage of the alkaline activator was 35%, and the water-precursor ratio was  
 187 0.35. Figure 5 presented the UCS test results. It was shown that the strength of the  
 188 geopolimer was well correlated with the particle size of the precursor material. When  
 189 the D50 of the precursor material was smaller than 15 μm, the change in the strength of

190 geopolymer became negligible. Therefore, by controlling the milling time, each  
191 precursor material was ground to a certain size with D50 equal to 15  $\mu\text{m}$ .

## 192 **2.3 Test methods**

### 193 **2.3.1 XRF test**

194 The X-ray Fluorescence (XRF) test was performed via Shimadzu UV2700  
195 fluorescence spectrometer under vacuum to determine the chemical composition of  
196 concrete, brick, ceramic and the mixed brCDW. The principle of XRF is to irradiate  
197 atoms with X-rays, causing the electrons of the irradiated atoms to undergo transitions  
198 and emit characteristic secondary X-rays. By testing and analyzing the characteristic  
199 radiation and its intensity for various elements, the types and concentrations of the  
200 elements can be determined.

### 201 **2.3.2 XRD test**

202 The mineralogical composition of concrete, brick, and ceramic brCDW was  
203 determined by XRD analysis. The diffraction patterns were obtained using a Rigaku  
204 Smartlab diffractometer equipped with Cu K $\alpha$  115 radiation, operating at 40 kV and 30  
205 mA. The samples were scanned between 5° and 80° 2 $\theta$ , with a step size of 0.02° 2 $\theta$  and  
206 a scan rate of 4 °/min (Moreno-Maroto et al., 2022). The obtained diffraction data were  
207 analyzed using Jade software to establish the relationship between the angle and the  
208 strength of the test sample, thereby determining the phase composition of the materials.

### 209 **2.3.3 UCS test**

210 The UCS test was conducted by the YAW-300D microcomputer controlled  
211 pressure testing machine, with a loading rate of 2.4 kN/s. Prior to the test, the specimens  
212 were prepared with dimensions of 40 mm×40 mm×40 mm, and conditioned in a  
213 chamber with 20±2°C and 95% relative humidity for 7, 14 and 28 days. For the same  
214 condition, six replicates were prepared for unconfined compressive strength (UCS)  
215 testing.

### 216 **2.3.4 SEM-EDS**

217 The EVO-10 scanning electron microscope produced by Carl Zeiss was used  
218 for the experiments. Before the test, the selected samples were dried and cleaned using  
219 high-pressure blower to remove dust and other loose particles from the surface. The  
220 sample surface was plated with palladium before being fixed onto the sample table for  
221 imaging.

222 To identify the gel types in the different geopolymers, the EDS test was  
223 performed on the selected points of the geopolymer samples. This allowed for  
224 determination of the composition of the main elements in the gel. Note that the EDS  
225 test was conducted simultaneously with the SEM test using the same instrument.

226

227 **3 Results and discussion**228 **3.1 Chemical composition of brCDW**

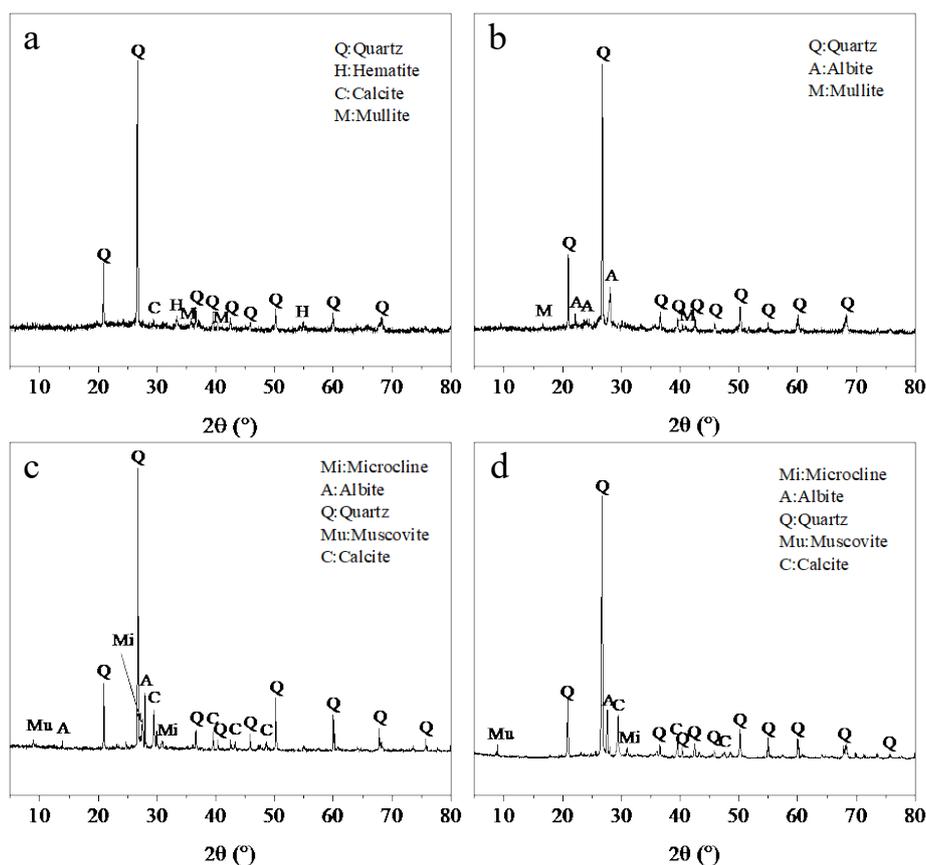
229 The XRF results were listed in Table 1. The chemical component of mixture  
 230 was calculated based on the proportion of each component. The main oxides of each  
 231 brCDW component were SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and CaO, which were the primary raw materials  
 232 for geopolymer. Thus, the brCDW was highly suitable for geopolymer production. It  
 233 was shown that SiO<sub>2</sub> always had the highest content, but its composition varied among  
 234 the brCDW components. Brick had an extremely low CaO content but the highest Fe<sub>2</sub>O<sub>3</sub>  
 235 content, whereas ceramic had a relatively high CaO content but the lowest Fe<sub>2</sub>O<sub>3</sub>  
 236 content. Concrete had the highest amount of CaO, but a relatively low Al<sub>2</sub>O<sub>3</sub> content.  
 237 This was because the cement paste attached to the recycled concrete contained rich  
 238 calcium and the limestone in the recycled concrete also provided calcium. Since the  
 239 concrete was a dominant component in the brCDW, the chemical compositions of the  
 240 mixed precursor were similar to those of concrete, which contained substantial amounts  
 241 of SiO<sub>2</sub> and CaO.

242 **Table 1. Main chemical components and contents of brCDW (%)**

	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	CaO	Fe <sub>2</sub> O <sub>3</sub>	MgO	K <sub>2</sub> O	TiO <sub>2</sub>
brick	61.86	17.94	2.48	11.09	0.81	3.63	1.42
ceramic	63.39	15.11	12.53	2.72	1.40	3.03	0.62
concrete	42.96	8.31	35.89	5.71	1.24	2.61	0.73
mixture	47.01	10.19	29.38	6.42	1.18	2.79	0.84

243

244 Figure 6 showed the XRD patterns of each brCDW component. As presented,  
 245 the brick mainly consisted of quartz, hematite, calcite, and mullite. The ceramic was  
 246 primarily comprised of quartz, albite, and mullite. The main phases of concrete included  
 247 quartz, calcite, albite, microcline, and muscovite. Since the mixed brCDW contained  
 248 no new material, its primary phases included quartz, calcite, albite, microcline, and  
 249 muscovite. Due to the low content of hematite in bricks and the fact that bricks only  
 250 accounted for 16% of the mixed precursor, hematite was not detected in the XRD  
 251 pattern of the mixed precursor. Thus, XRD may not be able to detect the phases with  
 252 low content. The highest content in all materials was quartz, whose main component  
 253 was SiO<sub>2</sub>, as confirmed by the XRF test results. Furthermore, calcite in concrete was  
 254 the main source of calcium, which was also proved by the XRF results. The brick and  
 255 ceramic contained more aluminum elements that were primarily from mullite and albite.  
 256 In addition, the high content of Fe<sub>2</sub>O<sub>3</sub> in the brick may be mainly derived from hematite.



257  
 258 **Figure 6. XRD pattern of brCDW: (a): brick, (b): ceramic, (c): concrete, (d):**  
 259 **mixed precursor**

### 260 **3.2 Synthesis of geopolymer**

#### 261 **3.2.1 Precursor material**

262 To eliminate the influence of precursor particle size on geopolymer strength, it  
 263 is important to control the particle size of the precursor materials since it greatly affects  
 264 the reactivity rate. To achieve this, the D50 of each precursor material was controlled  
 265 at 15 $\mu$ m by adjusting the milling time. The ball milling time for brick, ceramic and  
 266 concrete was set at around 8 hours, 4 hours and 2 hours, respectively. Apart from  
 267 preparing single-component geopolymer using each precursor material, the mixed-  
 268 components geopolymer was also developed. Note that the mixed precursor followed  
 269 the original proportion of each component of brCDW, with a specific ratio of brick:  
 270 ceramic: concrete = 16: 5: 79.

#### 271 **3.2.2 Design of mix proportion**

272 The alkaline activator's modulus can significantly impact the strength of  
 273 geopolymer. To investigate this effect, it is necessary to adjust the modulus of the  
 274 alkaline activator by adding sodium hydroxide. The initial modulus of the sodium  
 275 silicate solution was 3.3. The relationship between the amount of sodium hydroxide ( $x$ )

276 to be added per 100 grams of sodium silicate solution and the target modulus of the  
277 alkaline activator ( $M_x$ ) was described by Equation 1.

$$M_x = \frac{m_s/M_s}{m_n/M_n + x/(2M_h)} \quad (1)$$

278 where  $M_x$  is the modulus of target activator;  $m_s$  is the mass proportion of  $\text{SiO}_2$  in water  
279 glass;  $M_s$  is the relative molecular weight of  $\text{SiO}_2$ ;  $m_n$  is the mass proportion of  $\text{Na}_2\text{O}$   
280 in water glass;  $M_n$  is the relative molecular weight of  $\text{Na}_2\text{O}$ ;  $M_h$  is the relative molecular  
281 mass of  $\text{NaOH}$ .

282 The amount of alkaline activator was another crucial factor affecting the  
283 strength of geopolymer. In this study, we referred to previous research and designed  
284 three different dosages of alkaline activator (30%, 35% and 40%) for each modulus of  
285 alkaline activator (1.0, 1.2 and 1.4) (Huo et al., 2021; Zhang et al., 2020; Luna-Galiano  
286 et al., 2022), which was presented in Table 2. To ensure the proper fluidity of  
287 geopolymer, we considered the water-precursor weight ratio as 0.35, based on the  
288 findings from Huo et al. (2021).

289 **Table 2. Mix proportion of geopolymer**

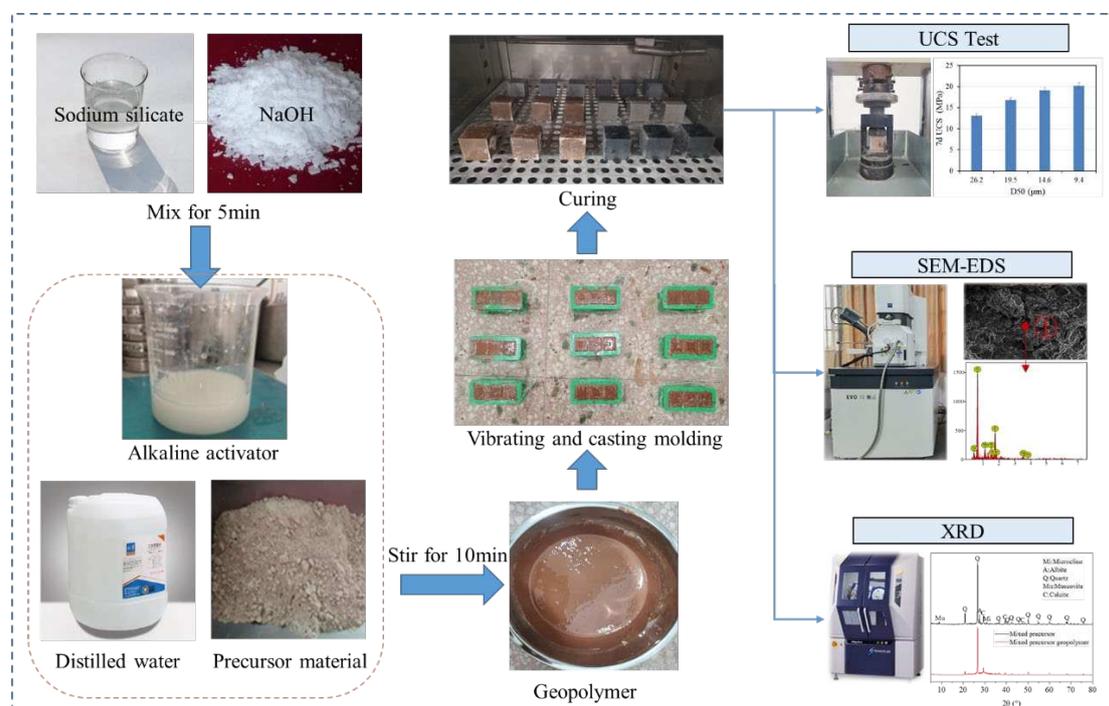
Water-precursor weight ratio	Modulus of alkaline activator	Dosage of alkaline activator
0.35	1.0	30%
		35%
		40%
	1.2	30%
		35%
		40%
	1.4	30%
		35%
		40%

290

### 291 **3.2.3 Production and curing**

292 The production of geopolymer was shown in Figure 7. Firstly, the required mass  
293 of sodium hydroxide was calculated and added to the sodium silicate solution. The  
294 resulting alkaline activator solution stood for 24 hours. Next, the precursor material was  
295 mixed with the alkaline activator solution and stirred for 5-10 minutes. The geopolymer  
296 slurry was poured into a mold in three layers and each layer was vibrated for 2 minutes  
297 to remove air. The specimens were then covered with film to prevent water evaporation  
298 and cured for 24 hours at room temperature. After that, the specimens were demolded  
299 and placed in a constant temperature and humidity curing box for further curing  
300 (Moreno-Maroto et al., 2022; Patil et al., 2014). The curing condition was  $20 \pm 2^\circ\text{C}$

301 with 95% relative humidity. The specimens were tested at the designated curing time to  
 302 assess their engineering properties. To ensure reliable results, each formulation yielded  
 303 18 cubic specimens (40mm×40mm×40mm), with six parallel specimens prepared for  
 304 UCS test at each curing time.



305  
 306 **Figure 7. Production and testing of geopolymer**

### 307 3.3 UCS test results

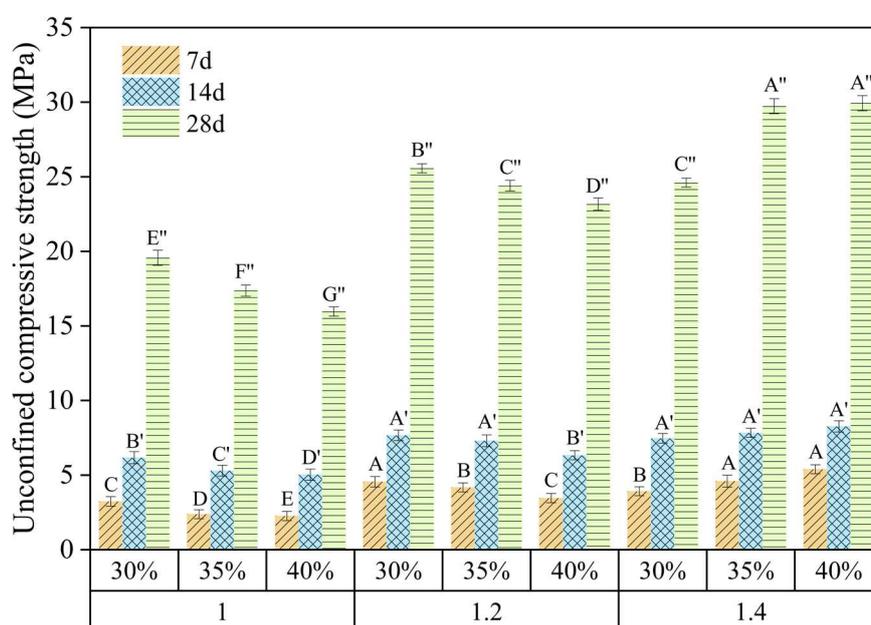
#### 308 3.3.1 Geopolymer of brick precursor

309 Figure 8 presented the UCS of brick geopolymer. In this figure, 1.0, 1.2, and 1.4  
 310 represented the modulus of the alkaline activator and 30%, 35%, and 40% represented  
 311 the dosage of the alkaline activator. A statistical analysis was conducted using the  
 312 analysis of variance (ANOVA) with Tukey honestly significant difference test, with a  
 313 confidence level set at 95% ( $\alpha=0.05$ ). Labels A, A', A'' were used to represent the group  
 314 of samples with the statistically highest strength at 7, 14, and 28 days, respectively.  
 315 Samples with different letters indicated a significant difference in strength, with lower  
 316 alphabetical letters indicating lower strength grades.

317 The strength of brick geopolymer initially developed slowly, with a low UCS  
 318 of only 2-8 MPa. However, the strength improved significantly in the later stage, with  
 319 the highest strength of 29.9 MPa. There was no significant difference in the strength of  
 320 the brick geopolymer prepared using the two mix proportions: a modulus of alkaline  
 321 activator of 1.4 with a dosage of alkaline activator of 35% and 40%. However, the  
 322 optimal mix proportion should have a lower alkali content, resulting in a more  
 323 environmentally-friendly and cost-effective material for preparing brick geopolymer.  
 324 Thus, a modulus of alkaline activator of 1.4 with a dosage of alkaline activator of 35%

325 was recommended as the optimal mix proportion. The low early-stage strength of brick  
 326 geopolymer might be attributed to the low calcium content, which resulted in a low  
 327 alkali consumption and generated only a small amount of C-S-H and C-A-H gels in the  
 328 early stage. While the curing continued, most of the alkali reacted with the active silicon  
 329 and aluminum components to form N-A-S-H and C-A-S-H gels, which provided the  
 330 brick geopolymer high strength at the final stage.

331 The UCS of the brick geopolymer increased with the modulus of the alkaline  
 332 activator, and the strength gain was greater with a lower modulus. For instance, the 28-  
 333 day strength increased by an average of 6.8 MPa when the modulus increased from 1.0 to  
 334 1.2, but the average strength gain was only 3.7 MPa when the modulus increased  
 335 from 1.2 to 1.4. This was likely because increasing the modulus of the alkaline activator  
 336 improved the dissolution efficiency of active silicon, aluminum, and calcium  
 337 components in precursor materials, particularly when the modulus was low. As the  
 338 modulus of the alkaline activator increased, the dissolution efficiency of these  
 339 components gradually decreased. When the modulus was 1.0 or 1.2, the strength of the  
 340 brick geopolymer decreased with an increase in alkaline activator dosage, likely  
 341 because the alkali content was high under these conditions. An excessive amount of  
 342 alkali negatively affected geopolymerization, leading to a reduction in reaction products  
 343 and structural damage.



344  
 345 **Figure 8. UCS of brick geopolymer**

### 346 3.3.2 Geopolymer of ceramic precursor

347 Figure 9 showed the UCS of ceramic geopolymer. The 7-day UCS of ceramic  
 348 geopolymer ranged from 6-9 MPa. Compared to the brick geopolymer, the early  
 349 strength of ceramic geopolymer was generally higher. However, from 7 days to 14 days,

350 the strength only increased by 2-3 MPa. After 28 days of curing, the UCS only increased  
351 by 1 MPa compared to the strength at 14 days. Ceramic geopolymer had lower strength  
352 compared to other geopolymers and was not suitable for the production of high-strength  
353 geopolymers. The reason for this phenomenon might be the calcium of the ceramic was  
354 rapidly consumed by the alkaline activator at the early stage, and the geopolymer of  
355 ceramic formed certain strength gels such as C-S-H and C-A-H. However, the calcium  
356 of the ceramic consumed the alkaline activator and affected the progress of the  
357 geopolymerization reaction, resulting in the formation of limited amounts of high-  
358 strength geopolymer gels (N-A-S-H, C-A-S-H). Another reason could be the low  
359 content of active silicon and aluminum components in ceramic, as most of these  
360 components did not participate in the geopolymerization reaction. The highest strength  
361 of ceramic geopolymer was observed when the modulus of the alkaline activator was  
362 1.0 and the dosage of the alkaline activator was 35%, which was recommended as the  
363 optimal mix proportion.

364 The UCS of ceramic geopolymer initially increased and then decreased with an  
365 increase in the modulus of the alkaline activator. The strength of ceramic-based  
366 geopolymer showed little increase when the modulus of the alkaline activator increased  
367 from 1.0 to 1.2, but it decreased significantly when the modulus of the alkaline activator  
368 increased from 1.2 to 1.4. For example, at 28 days, the UCS increased by an average of  
369 0.9 MPa when the modulus of the alkaline activator increased from 1.0 to 1.2, while it  
370 decreased by an average of 2.9 MPa when the modulus of the alkaline activator  
371 increased from 1.2 to 1.4. This phenomenon may be attributed to the fact that most of  
372 active silicon and aluminum components in ceramic dissolved efficiently at a modulus  
373 of alkaline activator of 1.2. A further reduction in the modulus of the alkaline activator  
374 did not significantly improve the dissolution efficiency and resulted in excess alkali.  
375 The excess alkali affected the geopolymerization process and reduced the reaction  
376 products. The remaining alkali also destroyed the formed gel structure, leading to a  
377 reduction in geopolymer strength. When the modulus of the alkaline activator increased  
378 from 1.2 to 1.4, the strength of the geopolymer decreased significantly because the  
379 active silicon and aluminum components in the precursor materials could not dissolve  
380 fully due to the low alkali concentration.

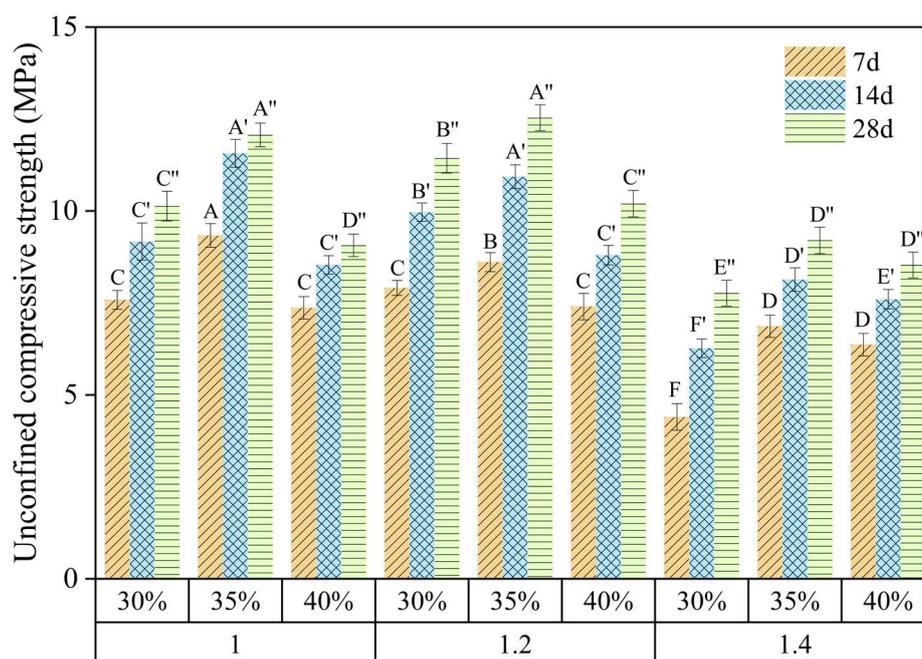


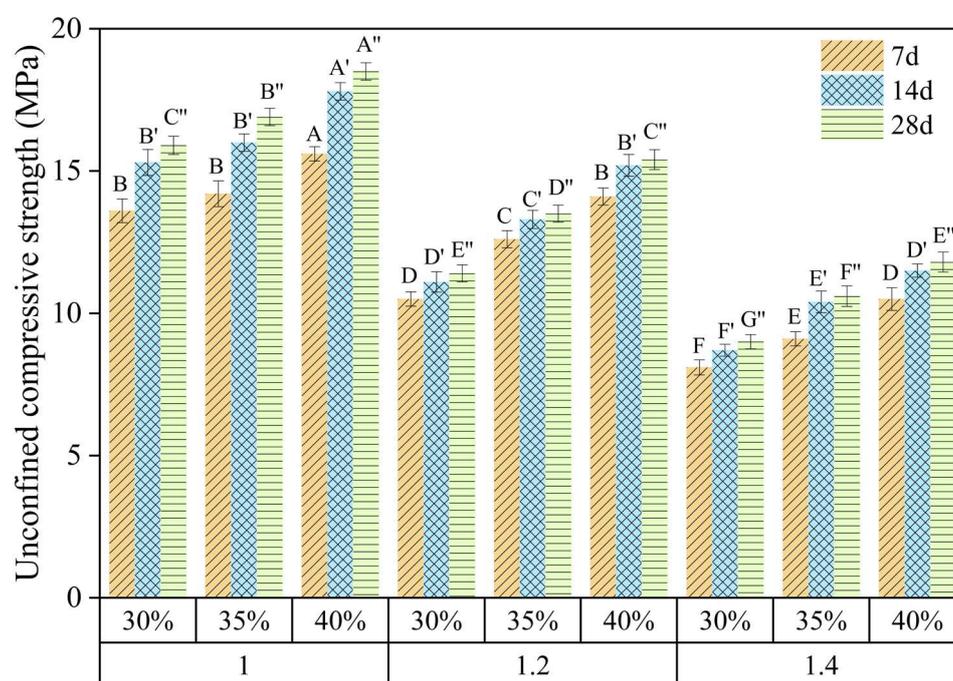
Figure 9. UCS of ceramic geopolymer

### 3.3.3 Geopolymer of concrete precursor

As shown in Figure 10, the early-stage strength of concrete geopolymer exhibited rapid development and surpassed that of brick and ceramic geopolymer. However, the rate of strength development gradually slowed down in the later stages. The highest strength of concrete geopolymer was achieved when the modulus of the alkaline activator was 1.0 and the dosage of the alkaline activator was 40%, which was recommended as the optimal mix proportion. Note that this mix proportion had the highest alkali content. The abundance of calcium components in concrete necessitated a substantial amount of alkali for reaction. During the early stage, a larger quantity of gels such as C-S-H and C-A-H, were generated. In addition, a small amount of unhydrated cement particles could also undergo the hydration reaction. Consequently, concrete geopolymer exhibited significant early-stage strength. However, due to the high calcium content, a considerable amount of alkali was consumed, resulting in insufficient alkali for reaction with the active silicon and aluminum components in concrete, or a low content of active silicon and aluminum components. As a result, the formation of N-A-S-H and C-A-S-H gels was limited, leading to a slow strength growth in the later stage.

The UCS of concrete geopolymer decreased with an increase in the modulus of the alkaline activator at each stage. This phenomenon may be due to the fact that the active silicon and aluminum components in the concrete were fully dissolved when the modulus of the alkaline activator was 1.0, allowing the alkaline activator to react adequately with the precursor material. However, an increase in the modulus of the alkaline activator led to a decrease in alkali concentration, resulting in decreased

406 dissolution efficiency of the active silicon and aluminum components. Moreover,  
 407 increasing the modulus of the alkaline activator led to insufficient geopolymerization  
 408 and a reduction of reaction products, which ultimately lowered the geopolymer strength.



409

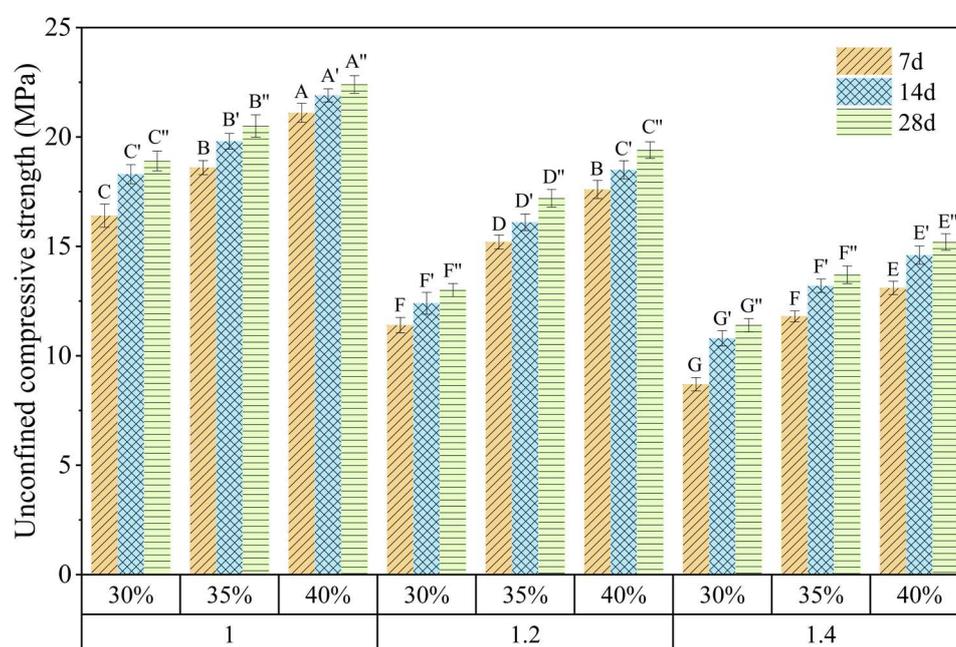
410

**Figure 10. UCS of concrete geopolymer**

#### 411 3.3.4 Geopolymer of mixture precursor

412 Figure 11 showed the UCS of the mixed brCDW geopolymer. The strength  
 413 growth of mixed brCDW geopolymer followed a similar trend to that of concrete  
 414 geopolymer, and its early-stage strength of geopolymer was greater than that of brick,  
 415 ceramic and concrete geopolymer. From 7 to 14 days, the UCS only increased by  
 416 approximately 1-2 MPa, and from 14 to 28 days, the UCS only increased by around 1  
 417 MPa. The reason for this phenomenon may be that concrete accounted for 79% of the  
 418 mixed precursor, with calcium comprising nearly 36% of it. The high calcium content  
 419 results in the generation of a large amount of C-S-H and C-A-H in the early stage,  
 420 leading to a rapid strength development. Furthermore, there may be unhydrated cement  
 421 particles in the mixed precursor, which could undergo hydration reaction to provide  
 422 additional strength. The mixed precursor also contained a small amount of brick and  
 423 ceramic, which supplemented the silicon and aluminum components in the precursor  
 424 materials, further enhancing the early strength of the mixed precursor geopolymer.  
 425 However, the low content of brick and ceramic implied that the active silicon and  
 426 aluminum components were not enough for an incomplete geopolymerization.  
 427 Consequently, there was little formation of high-strength geopolymer gels (N-A-S-H,  
 428 C-A-S-H) in the later stage, resulting in a slow strength growth.

429 The UCS of the mixed brCDW geopolymer decreased as the modulus of the  
 430 alkaline activator increased. This phenomenon may be due to the fact that when the  
 431 modulus of the alkaline activator was 1.0, the active silicon and aluminum components  
 432 in the mixed precursor were fully dissolved, resulting in the highest strength. Increasing  
 433 the modulus of the alkaline activator reduced the alkali concentration and the  
 434 dissolution efficiency of the active components, leading to an insufficient  
 435 geopolymerization and a lower geopolymer strength. For all moduli of alkaline  
 436 activator, the strength of the mixed precursor geopolymer increased with the increase  
 437 of alkaline activator dosage, and the strength of the mixed brCDW geopolymer was  
 438 highest when the modulus of alkaline activator was 1.0 and the dosage of alkaline  
 439 activator was 40%, which was recommended as the optimal mix proportion.



440

441

Figure 11. UCS of mixed precursor geopolymer

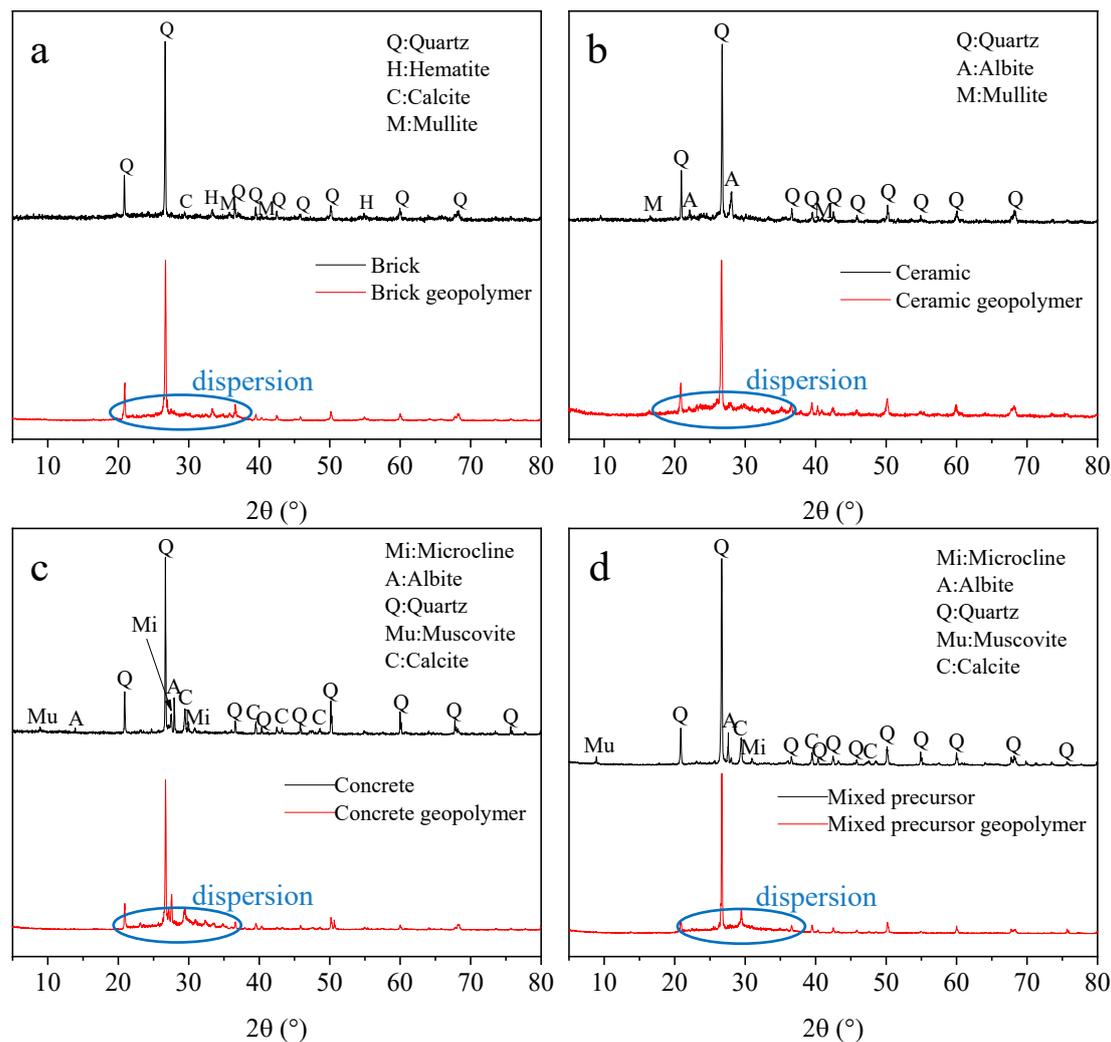
### 442 3.4 Microscopic mechanism of geopolymers

#### 443 3.4.1 XRD test results

444 In this section, we conducted the XRD test to analyze the phase composition of  
 445 brCDW precursor and geopolymer, which was then used to explore the changes in the  
 446 material phase before and after geopolymerization. Figure 12 presented the XRD  
 447 patterns of brCDW components and the derived geopolymers. The detected phases in  
 448 the geopolymers included quartz, muscovite, calcite, albite, and microcline, which were  
 449 originally present in the precursor materials, and no new phases were generated after  
 450 geopolymerization. Because geopolymer gels existed in an amorphous to semi-  
 451 crystalline state, there were no obvious crystallization peaks of reaction products in the  
 452 XRD pattern of geopolymer. Nevertheless, the XRD pattern of geopolymer showed a  
 453 dispersion peak at around  $20^{\circ}$ - $40^{\circ}$   $2\theta$ , which was the characteristic peak of geopolymer

454 gels. Compared to the XRD pattern of the precursor material, an obvious change was  
455 that the strength of partially crystallized peaks decreased, and some peaks disappeared  
456 in the geopolymer pattern. This indicated that some phases dissolved and participated  
457 in the geopolymerization, and other phases completely dissolved due to their low  
458 content and participated in the reaction, causing their characteristic peak to disappear  
459 in the XRD pattern of geopolymer.

460 The XRD patterns also demonstrated that the magnitude of the crystallization  
461 peaks of the concrete and mixed precursors decreased most sharply. This suggested that  
462 the amounts of dissolved and reactive substances in these materials were greater than  
463 the other precursor materials. Additionally, the concrete geopolymer showed the most  
464 distinct dispersion peak at  $20^{\circ}$ - $40^{\circ}2\theta$ , indicating an abundance of gels. The significant  
465 decrease in peak magnitude of calcite suggested that a large amount of calcium was  
466 involved in the geopolymerization, and the resulting gels were primarily C-S-H and C-  
467 A-H, which was confirmed in the EDS analysis later. In contrast, the XRD patterns of  
468 ceramic and brick and their respective geopolymers showed a small decrease in peak  
469 magnitude, indicating that fewer dissolved substances participated in the  
470 geopolymerization. However, the brick geopolymer, despite having a smaller amount  
471 of substances involved, had a more stable structure and higher strength than other  
472 precursor materials.

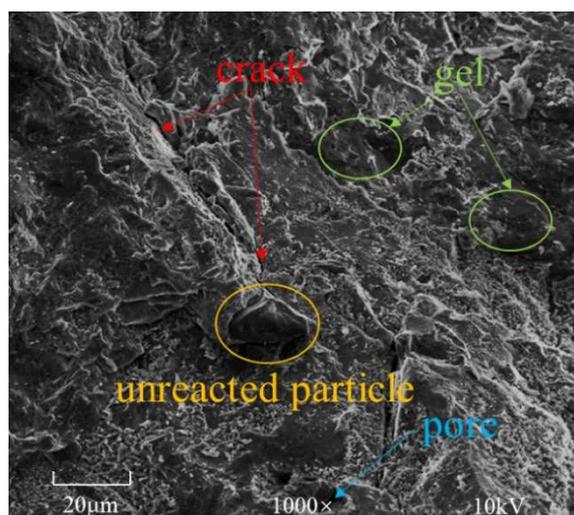


473

474 **Figure 12. XRD pattern of brCDW geopolymer: (a): brick geopolymer, (b):**  
 475 **ceramic geopolymer, (c): concrete geopolymer, (d): mixed precursor geopolymer**

### 476 3.4.2 SEM test results

477 We also performed the SEM test on the optimal mix proportion of brCDW  
 478 geopolymers at 7 and 28 curing days for analyzing their microstructures. Figure 13  
 479 illustrated the microstructure of geopolymers, which is comprised of gels, unreacted  
 480 particles, cracks, and pores. Since the brCDW had low activity, some particles did not  
 481 dissolve and participate in the geopolymerization. Other precursor materials dissolved  
 482 and reacted to form gels, which covered the unreacted particles and bound them  
 483 together. The strength of the geopolymer was formed after the gel hardened. The pores  
 484 and cracks had an adverse impact on the strength of the geopolymer. The formation of  
 485 pores may be due to the presence of water and air in the geopolymer. As water and air  
 486 dissipated, the space they occupied initially evolved into these pores. The cracks may  
 487 have formed due to the loss of water in the geopolymer, resulting in a shrinkage issue.



**Figure 13. Microstructure of geopolymer**

488

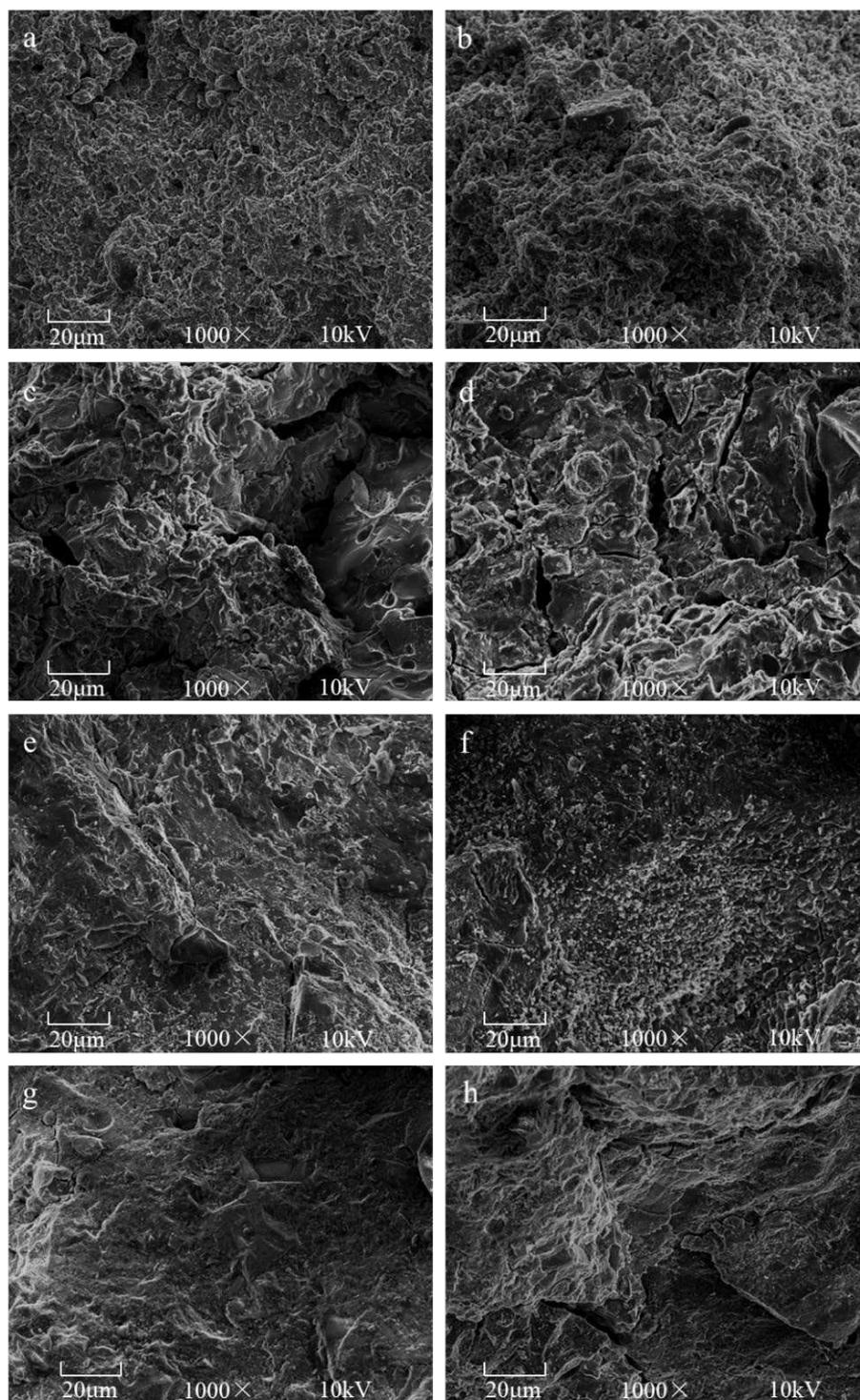
489

490 As shown in Figures 14a and 14b, the brick geopolymer had a cluster structure  
 491 and a rough surface, indicating that the type of gel in brick geopolymer differed from  
 492 that in other geopolymers. At 7 days of curing, brick geopolymer generated more gels  
 493 but had more pores with a loose structure, resulting in low early strength. However, at  
 494 28 days of curing, the number of pores in the brick geopolymer decreased notably,  
 495 leading to a high final strength. As illustrated in Figures 14c and 14d, the structure of  
 496 ceramic geopolymer was highly uneven with numerous pores and cracks, and the gels  
 497 were not completely bound together, resulting in low early strength. Although the  
 498 number of large pores in ceramic geopolymer slightly improved from 7 to 28 days of  
 499 curing, there were still many pores and cracks existing, which explained why the final  
 500 strength remained low. The concrete geopolymer (Figures 14e and 14f) contained a  
 501 large amount of gels and had a dense structure with no obvious pores or cracks, resulting  
 502 in high strength. From 7 to 28 days of curing, the coating effect of the concrete  
 503 geopolymer gel slightly improved, and no unreacted particles were detected.

504 The mixed precursor geopolymer (Figures 14g and 14h) also had a dense  
 505 structure, leading to a relatively high strength. After 7 days of curing, there were more  
 506 unreacted particles than the concrete. The consumption of alkaline activator by the  
 507 concrete may adversely impact the dissolution of some brick and ceramic particles.  
 508 From 7 to 28 days of curing, the number of unreacted particles decreased significantly  
 509 as gels continued to form and coat these particles. Consequently, the strength of the  
 510 mixed precursor geopolymer slightly improved in the later stage.

511 In sum, the microstructure of the mixed precursor geopolymer was the densest  
 512 compared to the single component-derived geopolymers. The mixed precursor  
 513 geopolymer had the highest strength, indicating that the strength of the geopolymer was  
 514 closely related to the density of the structure. Although the microstructure of brick  
 515 geopolymer was not dense, it formed clustered gels, which also remarkably influenced

516 the geopolymer strength. Therefore, it was deduced that both the type of gels and the  
517 compactness of the structure significantly affected the strength of the geopolymer.



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**Figure 14. SEM images of different brCDW based geopolymers at two curing times: (a): 7d brick geopolymer, (b): 28d brick geopolymer, (c): 7d ceramic geopolymer, (d): 28d ceramic geopolymer, (e): 7d concrete geopolymer, (f): 28d concrete geopolymer, (g): 7d mixed precursor geopolymer, (h): 28d mixed precursor geopolymer**

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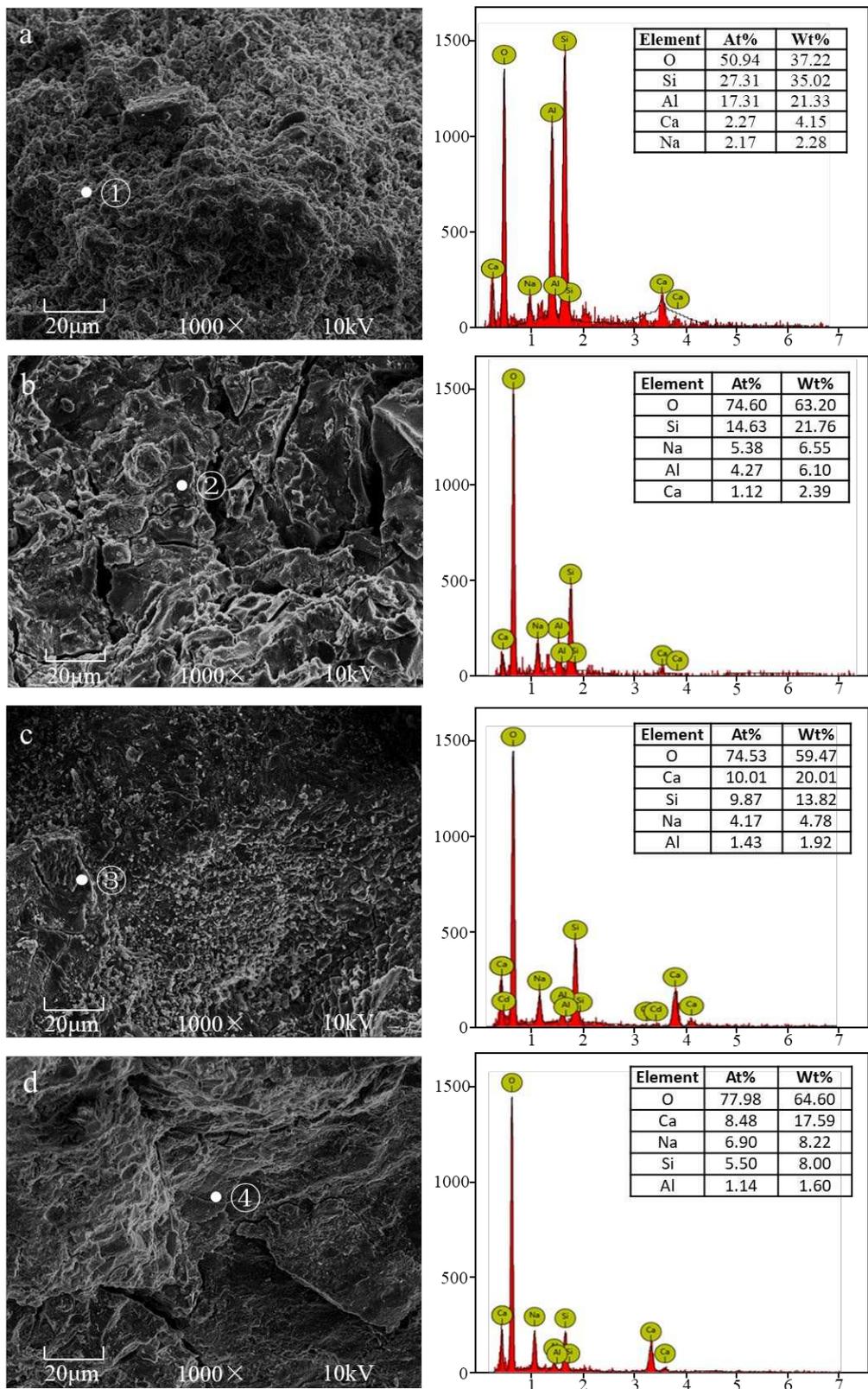
525 **3.4.3 EDS test results**

526 In addition to the SEM analysis, the EDS test was performed to determine the  
527 elemental composition of the gels. As shown in Figure 15a, the proportion sequences  
528 of the main elements in the gel of brick geopolymer were O, Si, Al, Ca, and Na. The  
529 elements of Si and Al had relatively high percentages, while the proportion of Ca was  
530 low, indicating a low probability of forming C-S-H and C-A-H gels. The likely gel types  
531 in the brick geopolymer were C-A-S-H and N-A-S-H gels, which coexisted to strength  
532 the material.

533 As presented in Figure 15b, the proportion sequences of the main elements in  
534 the gel of ceramic geopolymer were O, Si, Na, Al, and Ca. The Si element accounted  
535 for a relatively high proportion, while the Al element had a low proportion. Given the  
536 morphology of ceramic geopolymer, it was inferred that the amount of C-A-S-H and  
537 N-A-S-H gels formed in ceramic geopolymer was relatively small, and the major gel  
538 type was the C-S-H gel.

539 Figure 15c demonstrated that the proportion sequences of the main elements in  
540 the gel of concrete geopolymer were O, Ca, Si, Na, and Al. The elements of Ca and Si  
541 accounted for a relatively high proportion, while Al had a low proportion. This  
542 suggested that the gel type in concrete geopolymer was most likely to be the C-S-H gel.  
543 Additionally, the presence of Na and Al indicated that the gel might also contain an  
544 extremely low amount of C-A-S-H and N-A-S-H gels.

545 Figure 15d showed that the proportion sequences of the main elements in the  
546 gel of the mixed brCDW geopolymer were O, Ca, Na, Si, and Al. The elements of Ca,  
547 Na, and Si accounted for a relatively high proportion, while the Al element had a low  
548 proportion. Therefore, it was inferred that the dominant gel type in the mixed brCDW  
549 geopolymer was likely to be the C-S-H gel. Furthermore, the high proportion of Na and  
550 Ca suggested that the gel also contained the C-A-S-H and N-A-S-H gels.



551

552 **Figure 15. SEM-EDS test results of different geopolymers after 28 days of**

553 **curing: (a): brick geopolymer; (b): ceramic geopolymer; (c): concrete**

554 **geopolymer; (d): mixed precursor geopolymer**

555

556

557 **4 Conclusions**

558 In this study, the building-related construction and demolition waste (brCDW) was  
559 divided into three components, namely, brick, ceramic, and concrete. The influences of  
560 chemical composition and particle size of brCDW as precursors on the strength of the  
561 subsequent geopolymer were evaluated. The X-ray diffraction (XRD), scanning  
562 electron microscopy (SEM) and energy dispersive spectroscopy (EDS) tests were  
563 performed to investigate the strength evolution and mechanism of the single-component  
564 and mixed-component derived geopolymers. The major conclusions were summarized  
565 as follows.

- 566 • The brCDW components contained large amounts of Si, Al, and Ca elements,  
567 suggesting that the brCDW was a suitable precursor for synthesizing the  
568 geopolymer. Nonetheless, the chemical composition of each brCDW  
569 component differed, leading to differences in the strength of the resulting  
570 geopolymer. This indicated that the chemical composition of precursor material  
571 significantly affected the geopolymer strength. In addition, the strength of the  
572 geopolymer is significantly influenced by the modulus and dosage of the  
573 alkaline activator.
- 574 • The brick geopolymer exhibited a low strength in the early stage but a  
575 significantly high strength in the late stage. The ceramic geopolymer showed a  
576 low strength in both the early and late stages, indicating that it is not suitable  
577 for high-strength geopolymer production. The strength of concrete geopolymer  
578 was high in the early stage, but increased slowly later on. The strength  
579 evolution of the mixed brCDW geopolymer followed a similar pattern to that  
580 of the concrete geopolymer, as the mixed brCDW consisted of 79 % of concrete.  
581 The mixed brCDW geopolymer had the highest early strength and the second-  
582 highest final strength compared to the single component-derived geopolymers.
- 583 • The results of XRD, SEM and EDS analyses indicated that some precursor  
584 materials dissolved in the geopolymerization process, which then formed gels  
585 to coat and bind the unreacted particles and thereby resulting in a hardening  
586 strength. The formed gel type varied due to the different chemical composition  
587 ratios of each precursor material. The morphology and structure of brick  
588 geopolymer gels differed from other geopolymers, with the main gel products  
589 being C-A-S-H and N-A-S-H. The primary gel of ceramic, concrete, and mixed  
590 precursor geopolymers was the C-S-H gel, possibly with small amounts of C-  
591 A-S-H and N-A-S-H gel. Both the formed gel type and the compactness of the  
592 microstructure had significant influences on the geopolymer strength.

593

594

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601

## 602 Credit authorship contribution statement

603 **Fan Gu:** Conceptualization, Methodology, Project Administration, Funding  
604 Acquisition, Writing-Drafting, Reviewing and Editing. **Jianwei Xie:** Methodology,  
605 Investigation, Writing-Original Draft. **Cedric Vuye:** Methodology, Funding acquisition,  
606 Writing-Reviewing and Editing. **Ya Wu:** Project administration, Funding acquisition,  
607 Writing-Reviewing and Editing. **Junhui Zhang:** Methodology, Project administration,  
608 Writing-Reviewing and Editing.

609

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