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Development of Ambient-cured Geopolymer Mortars with Construction and Demolition Waste-based Materials

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Abstract

Degrading infrastructure and applications of structural demolition create tremendous amounts of construction and demolition waste (CDW) all around the world. To address this issue in an effective way, recycling CDW in a most appropriate way has become a global concern in recent years. To this end, this study focused on the valorization of CDW-based materials such as tile, bricks, glass, and concrete in the development of geopolymer mortars. CDWs were first collected from demolition zone and then subjected to crushing-milling operations. To investigate the influence of slag (S) addition to the mixtures, 20% S substituted mixture designs were also made. Fine recycled concrete aggregates (FRCA) obtained from crushing and sieving of the waste concrete were used as the aggregate. A series of mixtures were designed using different proportions of three distinct alkali activators such as sodium hydroxide (NaOH), sodium silicate (Na_2SiO_3), and calcium hydroxide (CH; $Ca(OH)_2$). To improve their applicability, the mixtures were left to cure at room temperature rather than the heat curing which is frequently applied in the literature. After 28 days ambient curing, the 100% CDW-based geopolymer mortar activated with three different activators reached a compressive strength of 31.6 MPa, whereas the 20% S substituted geopolymer mortar showed a 51.9 MPa compressive strength. While the geopolymer mortars activated with only NaOH exhibited poor performance, it was found that the use of Na_2SiO_3 and CH improved the mechanical performance. Main geopolymerization products were related to NASH (Sodium alumino-silicate hydrate), CASH (Calcium alumino-silicate hydrate), and C(N)ASH gel formations. Results demonstrated that mixed CDWs can be employed in the manufacturing geopolymers, making them potential alternatives to Portland cement (PC)-based systems by being eco-friendly, energy-efficient, and comparable in compressive strength.

Keywords

Construction and demolition waste, Ambient curing, Geopolymer, Compressive strength, Microstructure

Introduction

The construction sector is one of the largest energy-intensive and non-eco-friendly sectors globally. It is responsible for 35% of all GHG emissions and 42% of overall energy consumption in Europe [1]. According to Rahman et al. [2], clinker, the primary ingredient of PC, consumes 1.7 t of clean raw materials during production suggesting that the construction industry needs fundamental changes, particularly in material selection.

Geopolymers/alkali-activated materials are an eco-friendly alternative to traditional cement-based materials and produced by the activation of aluminosilicates [3]. The aluminosilicate-based materials used in geopolymer production are mostly industrial wastes and/or by-products (e.g., S and fly ash) generally known as supplementary cementitious materials (SCMs). On the other hand, SCMs are also used in PC and concrete production and were noted as very beneficial in improving mechanical and durability characteristics. These materials are therefore in high demand and sold at high prices nowadays. Correspondingly, the current research efforts in geopolymer technology have shifted towards finding locally available and unpopular wastes to be used in geopolymer production. One such option is CDW, which arises from new construction applications, infrastructural operations, natural disasters, and demolition processes. In Europe, 800 Mt of CDW are generated annually [4], and this amount accounts for 25 - 30% of the total waste [5]. The high alumina and silica content along with the wide raw material potential, makes CDW a perfect candidate for geopolymer synthesis.

The use of precursors derived from CDWs in the development of geopolymers has found its way in literature and related studies have been published, although the subject is quite new compared to studies utilizing mainstream SCMs. Moreover, the majority of studies focus on developing geopolymers based on single CDW precursors or CDW precursors together with some part of PC and/or SCMs [6-8]. For example, in the work of Vafaei and Allahverdi [9], geopolymers were developed by the combined use of glass powder and calcium aluminate cement (CAC). Higher compressive strengths were noted when the CAC content increased in the compositions of geopolymer mortars. Reig et al. [10] focused on developing geopolymer mortars with waste ceramic tiles both as precursors and aggregates. They applied curing at 65 °C for 3 and 7 days by using PC, CAC, and CH for the activation of waste ceramic tiles. The compressive strength range recorded at the end of 7-day curing at 65 °C was 25 - 40 MPa and the results increased with the increments in the contents of PC, CAC, and CH. The compressive strength range was 5 - 12 MPa after 28 days when curing temperature was 20 °C. In another work of Reig et al. [11], after 7-day curing of clay brick wastes at 65 °C, compressive strength approaching to 30 MPa was obtained from mortars and with further optimization of mixture proportions, results were capable of increasing up to 50 MPa. Concrete waste was used in single or hybrid (with metakaolin or PC) form to obtain geopolymers activated with Na_2SiO_3/NaOH and cured for 28 days in Vásquez et al. [12]. It was recorded that for the single use of concrete waste, the maximum compressive strength reached was 25 MPa while for hybrid systems, compressive strength results reached up to 33 and 46.4 MPa for PC- and metakaolin-replaced mixtures, respectively. Robayo-Salazar et al. [13] focused on the development of building materials after alkaline activation of brick, concrete, and glass derived from CDW singly and/or in combination with PC. They reported better properties for mixtures having blends of CDW and PC, although adequate properties were acquirable from mixtures with CDW only. In another work, hybrid cements with the mixture of red clay brick and PC were manufactured and compressive strength levels reaching 102 MPa after 28 days were achievable [14]. Wastes of concrete, tile, and brick were used to manufacture geopolymers in the study of Komnitsas et al. [7] and it was found that tiles were the best-performing materials by achieving compressive strength of 57.8 MPa.

Literature studies clearly show that precursors derived from CDW are suitable to produce geopolymeric materials, although they also highlight that majority of the published works focus on either single utilization of CDWs and/or combined use of CDWs with SCMs/PC. CDWs are usually generated collectively and therefore, require upcycling collectively from the perspectives of practicality and time, workmanship, energy, and cost requirements [15]. Moreover, despite the availability of studies on CDW-based geopolymers, there are a very small number of research to date that concentrates on scaling up such materials to the extent of their application as mortar, concrete, building material, and/or practice [16]. Additionally, based on the previous research findings of the authors, it has been established that in order to achieve high performance in CDW-based geopolymers when ambient curing is applied, a combination of Na_2SiO_3 and NaOH is necessary due to the low reactivity of CDW materials [15, 17]. It has been observed that NaOH alone is not sufficient for ambient curing and can only be used solely in CDW-based geopolymers that undergo high-temperature curing. Therefore, different combinations of alkali activators were applied in this study, as ambient curing was targeted. The current study aims to investigate the compressive strength and microstructural properties of entirely CDW-based geopolymer mortars with at least 30 MPa compressive strength, which can then be improved in the form of completely CDW-based concretes in the future. Unlike the majority of literature reports, CDW was both used in the binder and aggregate phases. In the binder phase, CDW containing a mixture of HB (Hollow brick), RCB (Red clay brick), RT (Roof tile), G (Glass), and C (Concrete) was used, while in half of the mixtures, some part of CDW (20%, by weight) was replaced by S. In the aggregate phase, untreated FRCA (0 - 2 mm) was used for all mixtures. Compressive strength measurements and SEM/EDX (Scanning electron microscope/ Energy dispersive X-ray) analyzes were conducted for a general evaluation of the mixtures.

Experimentation

Materials

In this study, CDW-based materials including HB, RCB, RT, G, and C were taken from the demolition zone and used as precursors in mixed form. These wastes were subjected to identical two-stage size reduction process, which included initial crushing with a jaw crusher and followed by ball-mill grinding for an hour. Figure 1 illustrates the views of the precursors before and after the size reduction process. In half of the mixtures, mixed CDW-based materials were replaced with S (20%, by weight) to see the changes in compressive strength results. The particle size distributions and chemical compositions of the precursors are given in figure 2a and table 1, respectively. The CDW were nanoscale materials.

C was also used as FRCA in the experimental program. C

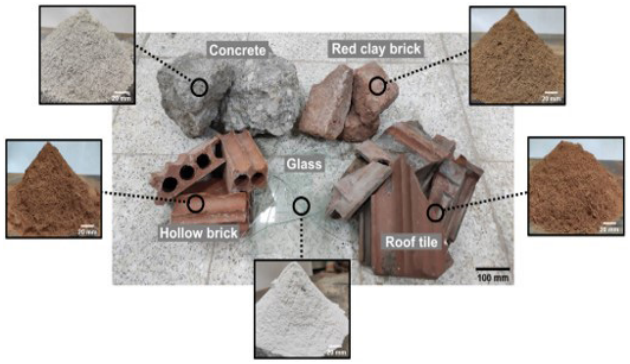

Figure 1: The views of the precursors before and after grinding (the powders, from left to right, are RT, RCB, HB, C, and G).

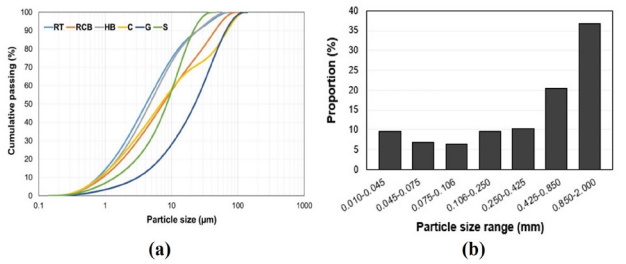

Figure 2: The particle size distibutions of the materials **(a)** precursors and **(b)** recycled concrete aggregates.

was crushed and sieved, and the passed materials (0 - 2 mm) were used as FRCA. Figure 2b presents the view and particle size distribution of FRCA used in this study. The FRCA had the following physical properties: a water absorption of 11.1%, porosity of 21.7%, compacted unit weight of 1352.5 kg/m^3 and dry specific gravity of 1.95. As alkali activators, commercially available Na_2SiO_3, NaOH, and CH were preferred.

Mixtures and method

The mixture proportions of mixed CDW-based geopolymer mortars are listed in table 2. As previously noted, in half of the mixtures, S was used instead of mixed CDW-based precursors, by 20% of the weight of the CDW-based precursors. The remaining mixtures contained 100% CDW in their binder phase. Based on preliminary studies of the authors, mixed CDW was composed of 30% RT, 23% HB, 20% C, 17% RCB, and 10% G. In all mixtures, the water/precursor and FRCA/precursor ratios were adjusted to 0.35.

To dissolve CDW-based materials, NaOH was used in all mixtures [18]. Additionally, Na_2SiO_3 was added to M1, M2, M3, and M4 mixtures to accelerate the geopolymerization [19]. Apart from those, CH was used as an additional activator in M3, M4, M5 and M6 mixtures to constitute Ca-based structures, which help with the strength development. Three different alkali activator combinations were designed in the mixtures. The first two mixtures (M1 and M2) were activated by NaOH with a Na concentration of 6% and Na_2SiO_3 has twice the quantity of NaOH. In the next series (M3 and M4), CH by 2% of overall precursor weight was used in addition to the alkali activators used in the first group. In the third group (M5 and M6), NaOH (Na concentration of 10%) was employed along with the CH, by 6% of the overall precursor weight.

Table 1: Chemical compositions of the precursors.

Chemical Composition (%)	RT	RCB	HB	C	G	S
SiO_2	49.3	52.4	53.5	37.4	72.5	32.1
Al_2O_3	20.0	19.9	19.3	10.7	0.93	11.2
Fe_2O_3	8.16	7.92	7.45	3.82	0.25	0.62
CaO	5.16	4.18	4.21	21.2	10.5	36.1
MgO	3.29	2.84	2.61	1.29	0.43	5.64
SO_3	0.79	0.95	1.46	0.54	0.24	1.21
Na_2O	1.23	1.58	1.50	1.96	12.6	0.31
K_2O	3.67	3.72	3.58	2.22	0.20	0.83
Loss on ignition	6.64	4.68	4.91	19.7	2.15	9.09

In the production stage, NaOH solution was prepared 1 day before the casting to cool down and kept at room temperature. The mixing process started with the loading of CDW-based precursors and FRCA into the mixer and mixing for 60 s at 100 rpm. Then, the NaOH solution was gradually poured into the mixer over 60 s and mixed for 120 s at 100 rpm. Meanwhile, if found in the mixture design, other alkali activators were added to the mixer following the same procedure. Finally, the mixing process ended after mixing for 180 s at 150 rpm. Subsequently, the mixture was molded, and their surfaces were covered for 24 h at laboratory (23 ± 2 °C and 50 ± 5% Relative Humidity). After 1 day, the mortar specimens were demolded and moved into the plastic bags for curing (23 ± 2 °C and 95 ± 5% Relative Humidity) until the test date.

In the compressive strength test, the ASTM C109 standard was followed. Nine 50 mm-diameter cubic specimens were produced for each mixture. The 7, 28, and 90 days of compressive strength were measured by crushing three specimens for each age and the obtained results were averaged. SEM/EDX analysis was conducted to see the microstructural characterization of the 1 cm dimension samples.

Table 2: Mixture designs.

Materials		Mixture ID					
		M1	M2	M3	M4	M5	M6
Precursors*	Mixed CDW	80	100	80	100	80	100
	S	20	-	20	-	20	-
Aggregates*	FRCA	35	35	35	35	35	35
Alkali activators*	Na	6	6	6	6	10	10
	NaOH	10.44	10.44	10.44	10.44	17.40	17.40
	Na_2SiO_3	20.88	20.88	20.88	20.88	-	-
	CH	-	-	2	2	6	6

Note: By the total weight of the precursor (%).

Results and Discussion

Compressive strength

The compressive strength results of CDW-based geopolymer are shown in figure 3. The results ranged between 10.3 and 34.4 MPa after 7 days. With further curing, results increased progressively and after 90 days, the mixtures achieved a minimum of 28.2 MPa and a maximum of 56.4 MPa compressive strength. The individual compressive strength results for each mixture were consistent, and the maximum standard deviation was found to be 3.08 MPa. S substitution and alkali activator selection significantly affected the compressive strength. Considering the average compressive strength results obtained after 90 days, S inclusion led to a strength increase of up to 60.6% in M3 and M4 mixtures. These strength increments were recorded as 39.9% and 19.5% for mixtures activated with NaOH + Na_2SiO_3 (M1 and M2) and NaOH + CH (M5 and M6), respectively. The strength enhancement with the S substitution could be related to the increase in reactivity and Ca content [20]. NASH gels are the main reaction products that yield strength in CDW-based geopolymers [15-17]. However, the additional Ca minerals that come with the S substitution also led to Ca-based structures such as calcium silicate hydrate (CSH) and CASH [21]. Therefore, the formation of these products along with the NASH gel might result in denser matrices and higher mechanical performance. On the other hand, the mixtures containing full CDW in their binder phase (M2, M4, and M6) also achieved promising results (22.6 - 31.6 MPa) after 28 days of ambient curing. This strength range meets standard requirements for most construction applications.

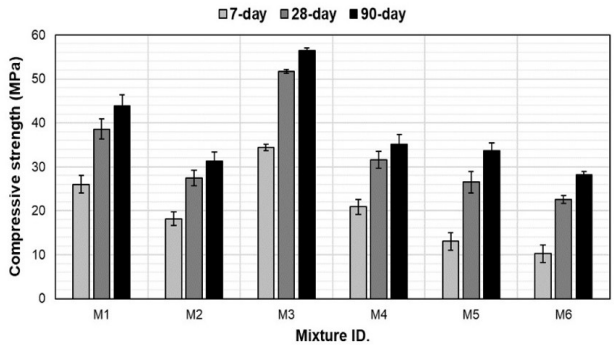

Figure 3: The compressive strength results of the mixtures.

Alkali activator selection also played a key role in affecting compressive strength. In S-substituted mixtures, the compressive strength changed between 28.8 - 67.4% when different alkali activators were used. M3 mixture containing NaOH + Na_2SiO_3 + CH achieved the maximum compressive strength. The addition of CH facilitated the formation of a gel predominantly composed of Ca within the matrix, thereby enhancing the viscosity of the mixtures [22]. This, in turn, contributed significantly to the solidification process and exerted a beneficial impact on the compressive strength. The presence of Na_2SiO_3 increased the soluble silicate content in the system, which accelerated geopolymerization [19]. In contrast, compared to other alkali activator combinations, the NaOH + CH combination contributed less to compressive strength. This was possibly due to the high NaOH content, which slowed ion transfer and polymerization [23]. Furthermore, although NaOH can be effectively used to activate CDWs with low reactivity in heat-curing applications, it was demonstrated that NaOH alone is inadequate for ambient curing. This highlights the necessity of using combinations of alkali activators in the ambient curing applications of CDW-based geopolymers.

SEM/EDX analysis

SEM/EDX tests were made on the M3 mixture cured for 90 days, which showed the highest compressive strength. Considering the findings obtained from the compressive strength results, the additional Ca content was found to be beneficial for strength improvement. For a detailed investigation, it was aimed to focus on regions containing low, medium, and high amounts of Ca in the specimens. Figure 4 presents the SEM images and EDX analyses acquired from various regions depending on the differences in Ca concentration. In figure 4a, NASH gels were seen with rectangular-faced and prismatic structures [24]. As it is known, NASH gel type, which is responsible for the strength, is formed as a result of geopolymerisation reactions, especially in CDW-based materials with high Si content [17]. The prominent alumina, silica, and sodium elements seen in the EDX analysis also confirmed the NASH formation. However, in addition to CDW-based materials

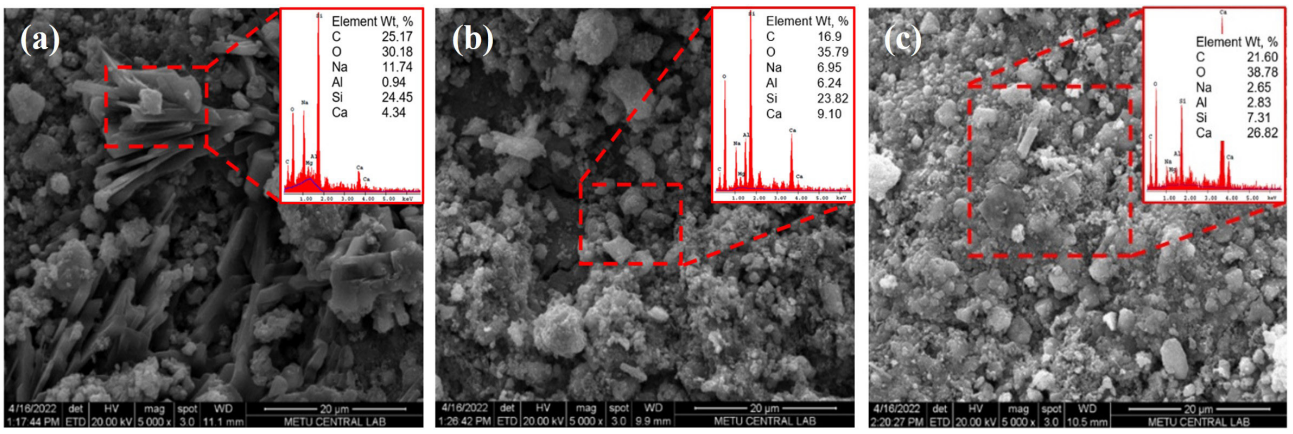

Figure 4: SEM images and EDX analyses of M3 mixture obtained from **(a)** low calcium, **(b)** medium calcium, and **(c)** high calcium regions.

with high Si content in the M3 mixture, the formation of Ca-based gel structures is also expected in the presence of materials such as S and CH in the mixture. In this context, figure 4b demonstrates the different gel structures formed through the increased Ca content. In this region, NASH gels started disappearing, and C(N)ASH gels became more prominent than in the low-Ca region (Figure 4a). The presence of hybrid gel formation with varying structural arrangements and random distribution caused the appearance of microcracks and voids found in low-Ca regions. The process of geopolymerization takes place quickly when Na_2SiO_3 is present, along with the presence of Ca^{+2} ions that act as additional sites for nucleation. These factors enhance the polymerization process and result in the formation of more compact matrices. In figure 4c, Ca was the most distinct element in this region, and it also provided insights into the formation of CASH and C(N)ASH gels.

Conclusion

This study focused on investigating the compressive strength and microstructure of mixed CDW-based geopolymer mortars containing untreated FRCA. Experimental findings demonstrated that depending on the composition, compressive strength exceeding 30 and 50 MPa can be obtained from geopolymer mortar mixtures based on CDW entirely and CDW + S, respectively, which were found promising for achieving completely CDW-based structural concretes and/or structures in future. Utilization of Na_2SiO_3 and S had a positive impact on the mechanical performance. At later ages, a similar finding was reported for combinations including CH as part of the alkaline activators. According to SEM/EDX analysis, formation of NASH gels was clearer in areas with low Ca. In high-Ca regions, the microstructure was found to be denser mostly due to the presence of hybrid NASH gels together with Ca-based gels including CASH and C(N)ASH.

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Conflict of Interest

None.

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