

An Investigation into Durability Aspects of Geopolymer Concretes Based Fully on Construction and Demolition Waste

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Abstract

The focus of the construction industry has shifted towards the development of alternative, eco-friendly and green construction materials due to the energy-inefficient and carbon-intensive nature of Portland cement (PC) production and aggregate quarrying. Meanwhile, increased number of repetitive repair/renovation/maintenance activities and demolition operations for the end-of-life buildings generate significant amounts of construction and demolition waste (CDW). For the purposes of sustainability and upcycling wastes into high-value-added materials with improved greenness, components from CDW streams can be used in producing geopolymer concretes without using PC and natural aggregates, given the rich aluminosiliceous nature of CDW components. The focus of current work is therefore on the analysis of durability of aspects (*i.e.*, drying shrinkage and resistance against sulfate attack, cyclic freezing-thawing, and chloride penetration) of geopolymer concretes made entirely of CDW. Different types of bricks, tile, concrete, and glass were used in mixed form as precursors for geopolymerization while different-size grains of waste concrete were used as recycled aggregates. As alkali activators, sodium hydroxide, calcium hydroxide and sodium silicate were used. In a companion mixture, CDW-based precursors were replaced with slag and class-F fly ash. Results showed that sulfate and cyclic freeze-thaw exposure did not cause any noticeable weight and compressive strength loss in CDW-based geopolymer concretes, while chloride penetration was found comparable to PC-based concrete. While drying shrinkage was found high in entirely CDW-based geopolymer concrete and resulted in surface microcracks, it was possible to lower the drying shrinkage substantially via substitution of CDW-based precursors with fly ash and slag.

Keywords: Construction and demolition waste, chloride penetration, drying shrinkage, freezing-thawing, sulfate resistance, geopolymer concrete.

1 Introduction

The construction industry has been forced to seek alternative building materials mostly due to the high energy requirements and irreparable harm caused by carbon emissions of PC production to the environment [1]. In recent decades, the concept of geopolymers has attracted a great deal of attention as an eco-friendly alternative to traditional PC concrete. Geopolymers are synthesized by activating aluminosilicate materials with alkali silicates and/or hydroxides and subsequently gain strength by the formation of geopolymeric gels via series of chemical reactions [2]. Heretofore, cement-free geopolymers that exhibit superior mechanical properties have successfully been developed by the use of mainstream industrial wastes (*i.e.*, fly ash, ground granulated blast furnace slag, silica fume) thanks to aluminosilicate-rich content and reactivity of these materials [3]. However, due to increased attention by the cement industry, limited supply, and cost-related concerns of these wastes, an increasing effort is being made lately on the discovery and utilization of new aluminosilicate resources in the development of geopolymers.

The construction industry generates around 10 billion tons of CDW annually worldwide due to repetitive maintenance, repair, and demolition activities causing CDW to be responsible for about 30-40% of total solid waste [4]. The recovery/recycling activities of CDW are still limited and the majority of such activities is restricted to downcycling practices such as landfilling [4]. CDW with their aluminosilicate-rich nature has recently become popular as a possible resource for the development of geopolymers. Recently, increasing emphasis is being placed on development of geopolymers out of CDW-based components such as brick, ceramic, and concrete wastes. However, the subject is still in its infancy considering the fact that the published studies have mainly focused on the development of geopolymer pastes and mortars only detailing fundamental properties (*i.e.*, strength measurements and microstructural characterization) [5,6]. To the authors' best knowledge however, studies trying to scale up CDW-based geopolymers and mortars to the level of structural concrete mixtures and evaluating their long-term durability aspects is almost non-existent. Therefore, the main focus of the current study is on the assessment of durability properties (*i.e.*, drying shrinkage and resistance against sulfate attack, cyclic freezing-thawing, and chloride penetration) of structural geopolymer concretes made entirely of CDW-based components. Different types of bricks, tile, concrete, and glass were used in mixed form as precursors for geopolymerization while different-size waste concrete grains were used as recycled aggregates in the development of concrete mixtures. For comparison, a mixture where CDW-based precursors were replaced with the mixture of slag and class-F fly ash (15% and 5%) was also developed.

2 Experimental Program

2.1 Materials and Mixture Preparation

In the current research, unknown-origin CDW-based materials including different masonry elements (*e.g.*, hollow brick [HB], red clay brick [RCB] and roof tile [RT]), concrete (C) and glass (G) were utilized in mixed form as the precursor in the production of geopolymer concrete. These materials were taken individually from a demolition site and then subjected to identical two-step crushing-grinding operation to obtain adequate fineness for the geopolymerization reactions. In addition to CDW-based materials, ground granulated blast-furnace slag (GGBS) and fly ash (FA) were used as co-binders in the precursor design of selected mixtures. Images of the precursors taken in the bulk form and after crushing-grinding were presented in Fig. 1. Chemical composition as obtained by the X-ray fluorescence analysis of the precursors was shown in Table 1. Untreated recycled concrete was used as aggregates. Recycled concrete aggregates (RCA) in different sizes were obtained by simple crushing and sieving of waste concrete. The particle size range was chosen as 0.1-4.75 mm for fine RCA (FRCA) and 4.75-16 mm for coarse RCA (CRCA). The particle size distributions of the CDW-based materials and RCA were presented in Fig. 2. Various concentrations of sodium hydroxide (NaOH), sodium silicate (Na_2SiO_3) and calcium hydroxide ($\text{Ca}[\text{OH}]_2$) were used as alkali activators in the production of mixtures. Mixtures and alkali solutions were prepared with tap water.

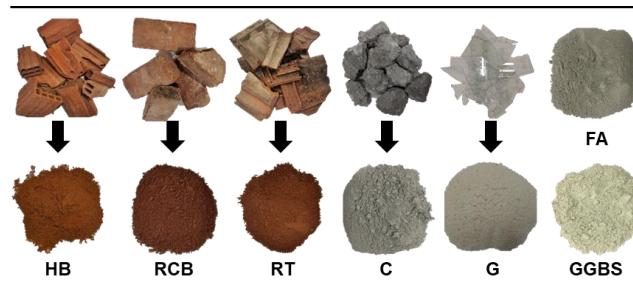


Fig. 1. Images of the precursors

Table 1. Chemical compositions of the precursors (units are in %).

Chemical Composition	HB	RCB	RT	C	G	FA	GGBS
SiO ₂	39.7	41.7	42.6	31.6	66.5	60.1	32.1
Al ₂ O ₃	13.8	17.3	15.0	4.76	0.93	21.4	11.2
Fe ₂ O ₃	11.8	11.3	11.6	3.50	0.25	7.41	0.62
CaO	11.6	7.69	10.7	31.3	10.0	0.99	36.1
MgO	6.45	6.49	6.26	5.12	3.93	1.82	5.64
SO ₃	3.40	1.41	0.71	0.92	0.24	0.22	1.21
Na ₂ O	1.45	1.15	1.60	0.45	13.6	0.99	0.31
K ₂ O	1.55	2.66	1.60	0.71	0.20	2.91	0.83
Loss on ignition	7.80	7.96	7.49	20.9	4.15	2.61	9.09

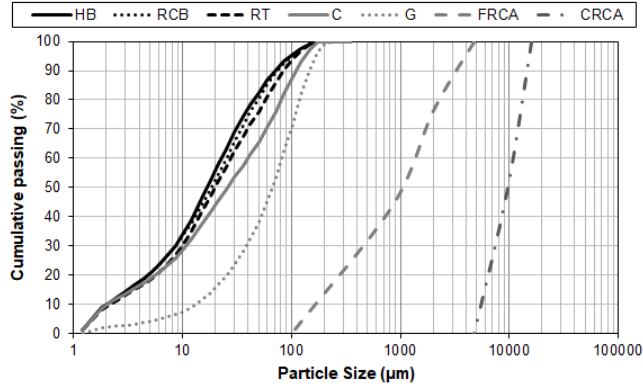


Fig. 2. Particle size distributions of the materials

Two different mixture designs were made. In the first design, both binder and aggregate phases were from CDW, while in the second design, the binder phase was comprised of 80% CDW + 15% GBFS + 5% FA and aggregates were from RCA. Mixture designs, details of which can be found in Table 2, were decided according to the preliminary laboratory works performed. In Table 2, CDW refers to different types of materials in mixed form with proportions of 26.7% HB, 26.7% RCB, 26.6% RT, 10% C and 10% G, by weight. While preparing the mixtures, first NaOH was dissolved in tap water and left to cool in a laboratory room conditions for 24 h. Then precursors and RCA were mixed for 60 s at 100 rpm to obtain homogeneous dry mixture. Alkali solutions were then added to the mixer by the following order of NaOH solution, Na_2SiO_3 and $\text{Ca}(\text{OH})_2$. After adding each alkali activator, mixing at 100 rpm was performed for 60 s before the addition of next one.

Table 2. Material proportions used in mixtures (units are in kg/m^3)

Mix ID	Precursors			Water	Alkali Activator			Aggregates	
	CDW	GGBS	FA		NaOH	Na_2SiO_3	$\text{Ca}(\text{OH})_2$	FRCA	CRCA
M1	800	150	50	202	112	224	50	500	500
M2	1000	-	-	202	112	224	50	500	500

2.2 Curing and Testing

After the mixing operation, mixtures were cast into different molds and kept in their molds for 24 h at an average temperature of 23 ± 2 °C and a relative humidity of $50 \pm 5\%$. Three cubic specimens with the dimensions of 100 mm were produced to determine the 7- and 28-day compressive strength for each mixture in accordance with ASTM C109 standard. After removed from their molds, specimens were kept in sealed condition at an average temperature of 23 ± 2 °C and relative humidity of $95 \pm 5\%$ until the testing age. The final compressive strength value was determined by averaging the test results of three different specimens.

Drying shrinkage was determined in accordance with the ASTM C157 standard. For each mixture, three prismatic specimens with the dimensions of 75×75×285 mm (height×width×length) were produced and cured under ambient conditions at the average temperature of 23±2 °C and relative humidity of 50±5%. After, specimens were measured for the changes in their length with time at the end of 7 and 28 days.

Three separate cubic specimens measuring 100 mm were produced for each mixture to be used in sulfate resistance tests. Specimens were first cured in sealed conditions at average temperature of 23±2°C and relative humidity of 95±5% for 7 days and then immersed into sulfate solution (5% Na₂SO₄) following the ASTM C1012 standard. Changes in the weight and compressive strength of the specimens were recorded after immersion in the sulfate solution for further 7 and 21 days.

Resistance against freeze-thaw cycles was measured by using three 100 mm-cubic specimens. Before the start of the test, all specimens were cured in a sealed condition at average temperature of 23±2°C and relative humidity of 95±5% for 14 days to gain adequate strength for the test. Changes in the weight and compressive strength of the specimens were recorded after exposure to 25, 50 and 100 freeze-thaw cycles. Each cycle adjusted in the freeze-thaw cabinet was composed of 6 hours at -18 °C and 6 hours at +5 °C, and all specimens were kept in water.

To have an idea about the resistance of specimens against chloride ion penetration, rapid chloride permeability test (RCPT) was performed in accordance with the ASTM C1202 standard. Four cylindrical specimens (Ø100×50 mm) were produced for each mixture. Specimens were first cured for 14 days under sealed conditions at an average temperature of 23±2°C and relative humidity of 95±5% and their chloride ion permeability was recorded in Coulomb after 7 and 14 days from the completion of first curing process.

3 Results and Discussion

3.1 Compressive Strength

Average 7- and 28-day compressive strength results of geopolymer concretes were given in Fig. 3. Results for 100% CDW-based M2 mixture were 15.9 and 30.2 MPa after 7 and 28 days, respectively. This suggests that geopolymer concretes can be successfully manufactured from entirely CDW-based materials allowing wastes to be used as valuable secondary raw materials and are likely to be used for structural purposes. On the other hand, at the end of 7 and 28 days, M1 mixture produced by replacing CDW with the GGBS+FA (15%+5%) achieved a compressive strength of 22.6 and 39.4 MPa, respectively. The increase in the compressive strength as a result of GGBS+FA replacement can be explained as follow: (i) introduction of GGBS with high pozzolanic and self-cementing capability leads to the formation of extra Ca-based gel types resulting in improvements in the microstructure and compressive strength of geopolymers ultimately [7], (ii) introduction of FA with suitable chemical composition in terms of SiO₂+Al₂O₃+Fe₂O₃ content and spherical morphology leads to an improved rheological and mechanical properties of alkali-activated systems [8].

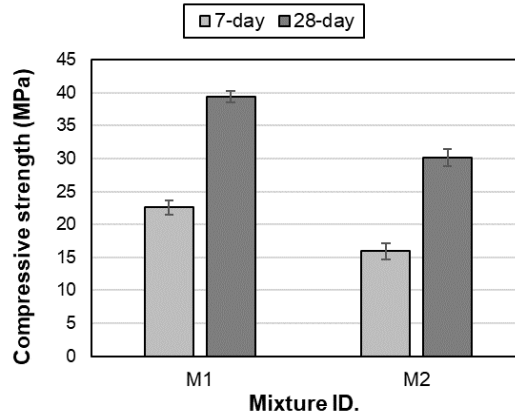


Fig. 3. Average compressive strength results

3.2 Drying Shrinkage

7- and 28-day drying shrinkage results of geopolymer concretes were shown in Fig. 4. After 7 days, drying shrinkage strains of M1 and M2 mixtures were calculated as 2632 and 4129 $\mu\epsilon$, respectively. After 28 days, drying shrinkage strains for both mixtures increased and were recorded as 3622 $\mu\epsilon$ for the M1 mixture and 5841 $\mu\epsilon$ for the M2 mixture. As clear from the results, the inclusion of GGBS+FA caused a reduction in drying shrinkage of approximately 37% at both ages. This was associated with the effect of GGBS+FA addition boosting the compressive strength, which reduces the transport of water via limiting the number of interconnected capillary networks and reducing the total evaporation from specimens' surface by using some of the free water in the mixtures, particularly due to self-cementing capability of GGBS [9]. In contrast to cementitious systems, water does not directly participate in gel development in alkali-activated materials; rather, it tends to evaporate over time. Therefore, substitution of CDW precursors, especially with GGBS herein, might have the ability to significantly reduce drying shrinkage in geopolymeric systems where free water can directly participate in gel formation. Free water availability often causes geopolymers to exhibit 3 to 6 times greater drying shrinkage than PC systems [10]. ACI committee 555 further states that concretes having 100% RCAs show 70% to 100% greater drying shrinkage compared to those produced with normal aggregates [11]. Based on these findings, it can be claimed that the drying shrinkage values found in this study can still be regarded quite acceptable compared to PC-based concretes with clean aggregates which are generally reported to exhibit drying shrinkage in the range of 500-1000 $\mu\epsilon$ at 28 days.

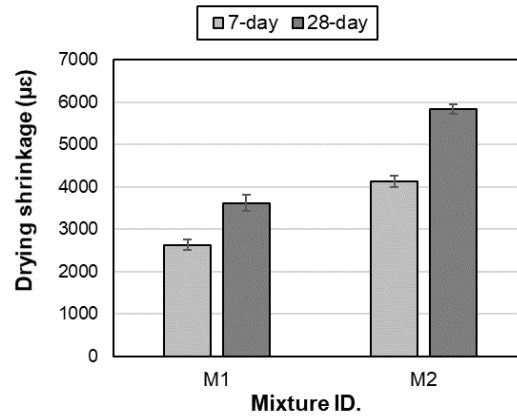


Fig. 4. Average drying shrinkage results

3.3 Sulfate Resistance

In Fig 5, changes in the compressive strength and weight of mixtures were shown after exposure to Na_2SO_4 solution for different further curing periods. Control mixtures were also produced to compare the compressive strength and weights of the mixtures immersed in the sulfate solution. Results showed that the compressive strength of the 7-day-old specimens of M1 mixture decreased by 9.63% and 9.13% after being kept in Na_2SO_4 solution for 7 and 21 days, respectively. A similar decrease in the same-age specimens of M2 mixture was also noted as 8.29% and 6.29% for 7+7 and 7+21 days, respectively. All samples immersed in sulfate solution experienced weight losses in the range of 1.3-2.6%. The reason behind these decreases is likely to be the decrease in the overall alkalinity of the geopolymer systems due to dissolution of alkalis in the sulfate solution exposed to a sulfate-rich environment. Thereafter, the geopolymeric gel structure starts to deteriorate depending on the exposure conditions leading to an increase in porosity and loss in compressive strength. Slightly better sulfate resistance of M2 mixture, which was entirely CDW-based and produced with lower calcium content, can be explained by the less ion exchange between the sulfate solution and network structure which was eventually reflected as higher sulfate resistance in the low calcium alkali-activated systems [12].

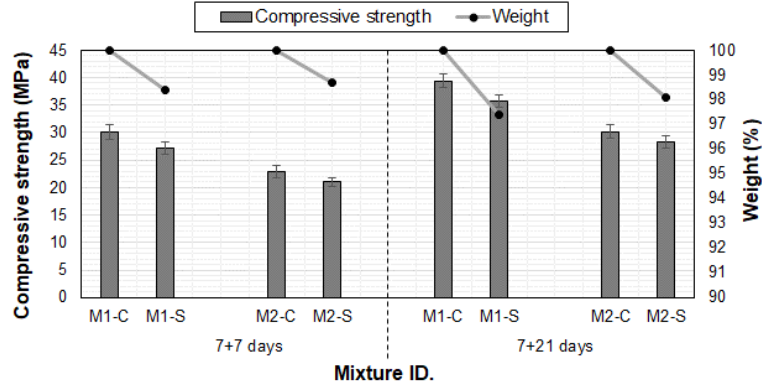


Fig. 5. Changes in the compressive strength and weight of mixtures after Na₂SO₄ exposure (M-C: control mixture; M-S: mixture exposed to sulfate solution)

3.4 Freeze-Thaw Cycles

Changes in the compressive strength and weight of the mixtures after subjected to 0, 25, 50 and 100 freeze-thaw cycles were presented in Fig. 6. Losses in the compressive strength of the M1 mixture after 25, 50, and 100 freeze-thaw cycles were 1.99%, 10.96% and 19.93%, respectively. The same results for M2 mixture were 3.05%, 11.35% and 18.34%, respectively. M1 mixture lost 4.9% of its weight after 100 freeze-thaw cycles, whereas 3.6% weight loss was observed in the M2 mixture. Since the freeze-thaw behavior of geopolymers is controlled by physicochemical processes, similar to PC concretes, air-void distribution is of critical importance [13]. Despite more porous microstructure of M2 mixture, which was mostly reflected by its lower compressive strength, pores seem to be beneficial for freeze-thaw resistance of this mixture. Overall, it can be stated that the results of both mixtures were similar and comparable to PC-based concretes [13].

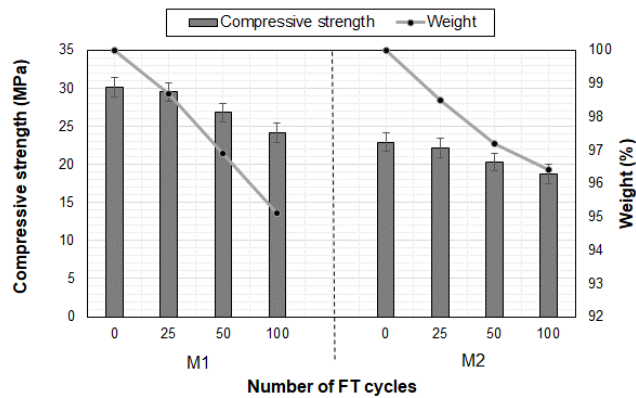


Fig. 6. Changes in the compressive strength and weight of mixtures after cyclic freeze-thaw exposure

3.5 Chloride Ion Penetration

Average rapid chloride permeability test (RCPT) results of the mixtures were given in Fig. 7. RCPT result of the M1 mixture after the 14+7 and 14+14 days were measured as 2900 and 2250, respectively, whereas the same results were 3150 and 2670 for the M2 mixture. The matrices of the geopolymer concretes become denser and more compact with longer curing periods, which decreased RCPT results of the mixtures. There were no big differences in the results for both curing ages and mixtures. According to the ASTM C1202, the chloride ion permeability of both mixtures was found in moderate class. It needs to be stated that these permeability levels were achieved in the presence of porous RCA and are acceptable for numerous applications. Results are also comparable to the traditional PC-based concretes [14].

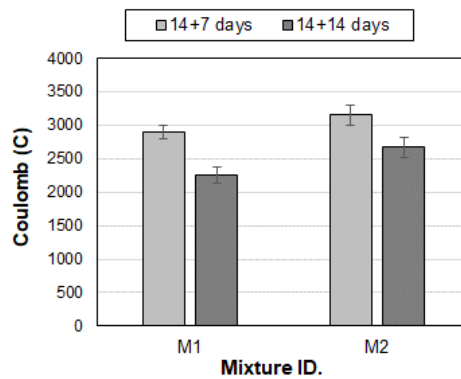


Fig. 7. Average RCPT results

4 Conclusions

Durability aspects of geopolymer concretes based entirely on construction and demolition waste (CDW) were investigated. The findings can be summarized as follows:

- Entirely CDW-based concretes reached 30.2 MPa compressive strength and this value increased to 39.4 MPa when 15% GGBS+ 5% FA substitution was employed.
- Drying shrinkage was found high in entirely CDW-based geopolymer concrete but, it was lowered significantly via GGBS+FA substitution.
- Sulfate and freeze-thaw exposure did not cause any noticeable weight and compressive strength loss in CDW-based geopolymer concretes.
- Chloride ion permeability was found comparable to PC-based concretes.

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References

1. Andrew RM (2018) Global CO₂ emissions from cement production. *Earth System Science Data*, 10(1), 195-217.
2. Provis JL, Lukey GC, van Deventer JS (2005) Do geopolymers actually contain nanocrystalline zeolites? A reexamination of existing results. *Chemistry of materials*, 17(12), 3075-3085.
3. Zhang P, Gao Z, Wang J, Guo J, Hu S, Ling Y (2020) Properties of fresh and hardened fly ash/slag based geopolymer concrete: A review. *Journal of Cleaner Production*, 270, 122389.
4. Zhang C, Hu M, Di Maio F, Sprecher B, Yang X, Tukker A (2022) An overview of the waste hierarchy framework for analyzing the circularity in construction and demolition waste management in Europe. *Science of the Total Environment*, 803, 149892.
5. Yıldırım G, Kul A, Özçelikçi E, Şahmaran M, Aldemir A, Figueira D, Ashour A (2021) Development of alkali-activated binders from recycled mixed masonry-originated waste. *Journal of Building Engineering*, 33, 101690.
6. Tan J, Cai J, Li J (2022) Recycling of unseparated construction and demolition waste (UCDW) through geopolymer technology. *Construction and Building Materials*, 341, 127771.
7. Ulugöl H, Günel MF, Yaman İÖ, Yıldırım G, Şahmaran M (2021) Effects of self-healing on the microstructure, transport, and electrical properties of 100% construction-and demolition-waste-based geopolymer composites. *Cement and Concrete Composites*, 121, 104081.
8. Ye N, Chen Y, Yang J, Liang S, Hu Y, Xiao B, Huang Q, Shi Y, Hu J, Wu X (2016) Co-disposal of MSWI fly ash and Bayer red mud using an one-part geopolymeric system. *Journal of hazardous materials*, 318, 70-78.
9. Deb PS, Nath P, Sarker PK (2015) Drying shrinkage of slag blended fly ash geopolymer concrete cured at room temperature. *Procedia Engineering*, 125, 594-600.
10. Atiş CD., Bilim C, Çelik Ö, Karahan O (2009) Influence of activator on the strength and drying shrinkage of alkali-activated slag mortar. *Construction and building materials*, 23(1), 548-555.
11. Lamond JF, Campbell Sr RL, Giraldi A, Jenkins NJ, Campbell TR, Halczak W, Miller R, Cazares JA, Hale HC, Seabrook PT (2001) Removal and reuse of hardened concrete. American Concrete Institute: Farmington Hills, MI, USA, 26.
12. Chen K, Wu D, Xia L, Cai Q, Zhang Z (2021) Geopolymer concrete durability subjected to aggressive environments—A review of influence factors and comparison with ordinary Portland cement. *Construction and Building Materials*, 279, 122496.
13. Provis JL, Van Deventer JS (2013) Alkali activated materials: state-of-the-art report, RILEM TC 224-AAM (Vol. 13). Springer Science & Business Media.
14. Ramezani-pour AA, Jovein HB (2012) Influence of metakaolin as supplementary cementing material on strength and durability of concretes. *Construction and Building materials*, 30, 470-479.