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PRELIMINARY RESULTS OF CONSTRUCTION AND DEMOLITION WASTE (CDW) AND FLY ASH (FA) BASED GEOPOLYMER COMPOSITE MATERIALS

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Abstract

There is need to research and develop green construction materials, to counter increasing infrastructural demand and environmental challenges. Preliminary investigation of construction and demolition waste (CDW) and fly ash (FA) based geopolymer composite materials is presented in this paper. The average compressive strength of mechanically activated materials based on ground fly ash (GFA) were 53.8% higher compared to the raw fly ash (RFA) based specimen. Investigation on CDW and GFA based geopolymer composite materials, show that 50% CDW precursor content had the highest strength. Cracks were observed on the GFA composite specimen surface, attributed to surface moisture loss. The optimum compressive strength, 47 ±9.13 MPa can be considered for structural application. Mitigation measures for adverse cracks should be applied.

Keywords: *construction and demolition waste, fly ash, compressive strength, geopolymer composite, mechanically activated materials*

1. Introduction

There is need to research and develop green construction materials, to counter challenges due to increase in infrastructural demand due to rapid population growth and urbanization, also environmental and climatic change issues including circular economy, carbon footprint and reduction of solid waste challenges. The processing and use of construction and industrial waste for geopolymer composites could promote reduction of carbon dioxide emission, from cement production, diversion of solid waste from waste streams, up-cycling and value addition for circular economy. Geopolymer cement has been applied for building pavements, retaining walls and water tanks and with rising popularity as an eco-friendly solution, it is expected that the same will be utilized for other buildings (Hanson-Heidelberg cement group, 2021). Though geopolymer concrete can be beneficial, both economically and sustainably, public authories and sceintist have raised concerns (European commission, 2018-last updated). According to Contreras-Llanes *et al.* (2021), CDW was among the largest waste streams globally hence within the circular economy concept, resulted to increased interest in re-use. Study by Udvardi *et al.* (2019), the highest compressive strength for CDW based geopolymer concrete was at 8M NaOH as compared to the 4M and 12M of NaOH. Komnitsas *et al.* (2015) study on CDW from tiles, concrete and bricks, indicated that for finer particles sizes <150 µm (<D_{50} µm), there was substantial increase in resultant geopolymer compressive strength. In Ojha & Gupta (2020) study, conventional concrete had strength of 30.3 MPa and class-F FA based geopolymer concrete strength was 28.8 MPa, the reduced strength was attributed to weaker inter-transitional zone (ITZ) for geopolymer concrete. In Ahmari *et al.* (2012) study on geopolymer concrete, the addition of grounded waste concrete increased upto 50%, attributed to Ca and Si that resulted to CSH formation and geopolymer gels. In Bassani *et al.* (2019) study, alkaline solution significantly impacted on the hardened CDW-based material, with higher concentration resulted to significantly higher compressive strengths. The mineralogy of CDW can affect the reactivity and fine construction material properties. In Frías *et al.* (2020) study, calcareous based CDW waste reacted less intensely with lime compared to the siliceous based CDW. In Gao *et al.* (2021) study, the addition of concrete slurry waste to ordinary portland cement for geopolymer composites resulted to reduced initial and final setting times. In Allahverdi & Kani (2009) study on waste bricks and concrete based geopolymer binder, samples with higher proportions of waste bricks had higher compressive strengths, attributed to the brick's calcinated alminosilicate content. According to Tan *et al.* (2022), wastes from red clay brick, waste glass and waste tiles could be utilized as a aluminosilicate source, while the unbound aggregates and waste concrete can be a silica and calcium source.

Mechanical activation increases the reactivity hence produce materials with improved properties. According to Kumar S. & Kumar R. (2011), the merits of mechanical activation (MA) for improvement of bulk and surface reactivity is well accepted, grinding duration impacted the intensity of quartz (Q) & mullite (M) peaks of FA, compactness, compressive strength of geopolymer composites improved. In Kumar *et al.* (2017) study, the 120 minutes milling of FA resulted to quartz peak intensity reduction & broadening. Hounsi *et al.* (2013) study showed reduced kaolinite after milling for kaolin based geopolymer composite, attributed to partial amorphization of milling, hence promoting geopolymerization. In Tan *et al.* (2022) study, milling reduced the insoluble residue for CDW. However, smaller size particles makes grinding significantly harder to reduce individual sizes further, according to Ulugöl *et al.* (2021), glass in particular may require longer milling times, resulting to energy-ineffective, time consuming and costly. It is important to determine optimum conditions for grinding that will achieve sufficient material properties, while optimizing the inputs.

Mucsi *et al.* (2015) study, the lignite (Matra power station – Visonta) and brown coal fly ashes (Tiszaújváros) had amorphous contents of 52% and 70% respecively, with both type containing quartz, mullite, cristoballite, maghemite, anhydrite and albite phases. Mádai *et al.* (2015) study, the class F, coal fired unitreated fly ash phases including amorphous – 80%, quartz – 3.48, mullite 2 : 1 – 12.45% and cristobalite high – 3.98%, with the treated/geopolymerised fly ash had higher contents of amorphous and lower contents of quartz phases. Study by Paaver *et al.* (2021), mechanical activation did not have a significant impact on mineral composition.

Preliminary investigation of CDW and FA based geopolymer composite materials was conducted. This investigation were in 2 series of geopolymer materials. In the 1st series, the compressive strengths for GFA and RFA-based geopolymer composite materials were investigated. The focus was on the impact of mechanical activation of FA precursor on geopolymer composite materials. Based on the results of the 1st series, the FA precursor with better results was adopted for the 2nd series of tests. In the 2nd series, compressive strengths for varying amounts of CDW and GFA precursors, based on mass were investigated. The CDW was milled in a ball mill, and the fraction finer than 106 μm size used. The main aim for the 2nd series was to investigate the impact of varying proportions of FA and CDW on the geopolymer composite material. The second series utilizes 2 aluminosilicate precursors, for geopolymer composite materials i.e. FA having a higher SiO_2 content and CDW, with higher CaO content. Blending 2 waste materials, can improve the properties of resultant construction eco-material, hence promoting feasibility.

2. Methods and materials

2.1. Materials

Alkaline solution included: 8M sodium hydroxide (8M NaOH), 10M sodium hydroxide (10M NaOH), Sodium silicate (SS), GFA, ground for 120 minutes in ball mill, raw brown coal FA, sourced from

Tiszaújváros, CDW material mainly aggregate material waste with some attached binder/cement material.

The particle size distribution for FA and CDW powder are presented in *Figure 1.*, showing a significant reduction of particle size for GFA samples.

Chemical composition based on X-ray flourescense (XRF) analysis results for FA and CDW are shown in *Table 1*. The CDW powder had lower SiO_2 and Al_2O_3, and higher CaO content as compared to FA sample. According to the X-ray diffraction (XRD) results shown in *Table 2* the ground CDW had 36.4% and 26.3% quartz and amorphous amounts respectively.

The specific gravities for CDW and GFA were almost similar, with average values of 2.5 and 2.4 respectively.

Table 1. *Chemical composition for GFA and CDW powders*

Composition	**CDW Powder (%)**	**GFA (%)**
SiO_2	56.7	60.6
Al_2O_3	7.6	25.7
MgO	1.42	1.18
CaO	15.8	1.84
Na_2O	0.74	0.98
K_2O	1.69	1.48
Fe_2O_3	2.46	5.6
MnO	0.123	0.034
TiO_2	0.285	0.573
P_2O_5	0.138	0.047
S	0.82	0.08
F	<0.3	<0.3

Table 2. *CDW sample composition based on XRD analysis*

Names	**Quantity (%)**
Quartz	36.4
Calcite	13.3
Albite	6.6
Vaterite	7.8
Muscolite 2MI	6.5
Chlorite IIb	0.5
Microcline max	2.2
Portlandite	0
Actinolite	0.5
Amorphous	26.3

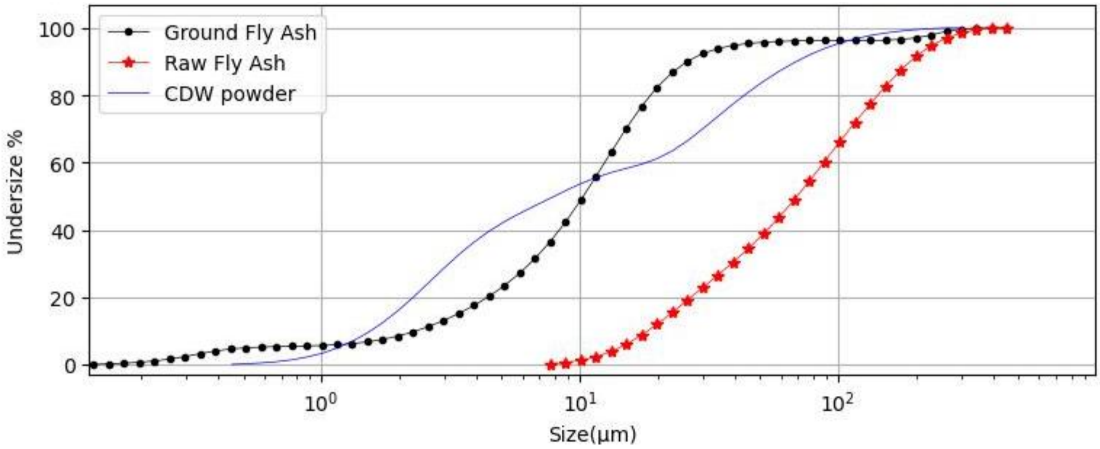


Figure 1. *Particle size distribution for FA and CDW powders measured by laser scattering*

2.2. Method

2.2.1. RFA and GFA based geopolymer composites

FA were individually mixed with alkaline solution. The ratios were based on % mass (w/w). The ratio of 8M NaOH: SS alkaline solution was 1 : 3 w/w. The alkaline to FA precursor was 0.82 w/w. The geopolymer paste were placed in cylinder mould of ø35 mm diameter and 65 mm height and demoulded the next day. Samples were demoulded and cured at 60 °C for 6 hours, subsequently cured under ambient conditions.

Analysis and tests were as follows: dimensions, masses, uniaxial compressive strength test at the rock mechanics laboratory, particle size distribution for FA samples, geopolymer composites were analyzed by FTIR spectrometry.

2.2.2. CDW powder and GFA based geopolymer composites

Assorted sizes of CDW was milled using ball mill, and fines <106 μm were used. Experience from Institute of Raw Material Preparation and Environmental Technology, University of Miskolc indicated that, higher CDW sizes have significant amounts of unreactive quartz. In Szabó *et al.* (2023) study, CDW had 54.4% quartz content, hindering milling efficiency and influencing reactivity. The ratios were based on % mass (w/w). Varying proportions of FA and CDW precursors were mixed with alkaline solution. The percentages of CDW precursor were 100%, 75%, 50%, 25% and 0%. The ratio of 10M NaOH: SS alkaline solution was 1 : 3 w/w. The alkaline to FA precursor was 0.82 w/w. 20 mm cube moulds were used to place the paste. Samples were demoulded after 2 days and cured at 60 °C for 6 hours, subsequently cured under ambient temperature.

Analysis and tests include: dimensions, mass, uniaxial compressive strength test was conducted in the rock mechanics laboratory. The CDW powder was tested for particle size distribution, using laser scattering analyzer Horiba LA-950, X-ray diffraction (XRD).

The 2nd series of experiment used 10M NaOH, different from the 1st series – 8M NaOH. Based on previous researches. 10M and 8M NaOH are expected to be within the optimum molarity range. Based on previous researches this are optimum ranges for geopolymer composition. Application of different molarity for the two series did not affect the research objectives. 20 mm cubes moulds were adopted for the 2nd series because constraint amount of ground CDW material (<106 μm). Szabó *et al.* (2023) study, high content of quartz CDW hindered milling efficiency.

3. Results and discussion

Densities and uniaxial compressive strength measurements of cylinder specimen are shown in *Table 3*. The average compressive strength of GFA geopolymer composite specimen was 15.52 ±0.41 MPa higher that for RFA geopolymer composite specimen, with 10.09 ±0.79 MPa. GFA based geopolymer composite materials achieved 53.8% higher compressive strength compared to the RFA based geopolymer composite specimen. The average density of GFA based geopolymer composites was higher compared to that of RFA based geopolymer composites. The higher compressive strengths for denser specimen could be attributed to the compactness of particles. Study by Sanjaya (2020), the compressive strength and density of concrete increased with increasing sand proportion, while the porosity reduced. There is a significant reduction of particle size between the RFA and GFA samples, with the ground samples resulted to increase strength for geopolymer composites, as compared to the RFA, this can be attributed to increase in specific surface area for reaction. The particle sizes for CDW, RFA and GFA are shown in *Table 4*. There was not much difference between GFA and CDW powder median sizes (D_{50}), at D_{25}, the CDW were finer compared to GFA while the GFA were finer at D_{75} compared with CDW.

Table 3. *Average densities & compressive strengths for specimen for RFA and GFA geopolymer composite material*

Specimen	**Density (kg/m³) (Avg.)**	**Uniaxial Compressive strength (MPa) @ failure (Avg.)**
RFA geopolymers composite	1170 ±12.41	10.09 ±0.79
GFA geopolymers composite	1392 ±5.15	15.52 ±0.41

Table 4. *Particle sizes for CDW and FA*

	D_{25} (mm)	**D_{50} (mm)**	**D_{75} (mm)**
RFA	32.4	69.5	125.4
GFA	5.4	10.3	16.8
CDW	2.7	8	35.9

Since the GFA geopolymers composites had higher compressive strength compared to RFA based geopolymer composites, the GFA was selected for the 2nd series of test. The compressive test and densities for the GFA and CDW based geopolymer composites are as shown in *Figure 2,* for varying % of CDW and FA. The highest compressive strength of the geopolymer composite was at 50% CDW & FA, averaging 47 ±9.13 MPa, with the lowest being for 100% CDW precursor, averaging 18 ±1.50 MPa. The highest average density was for 100% CDW geopolymer based composite with 1785 ±42.38 kg/m³, with the lowest being the 25% CDW, at 1696 ±29.98 kg/m³.

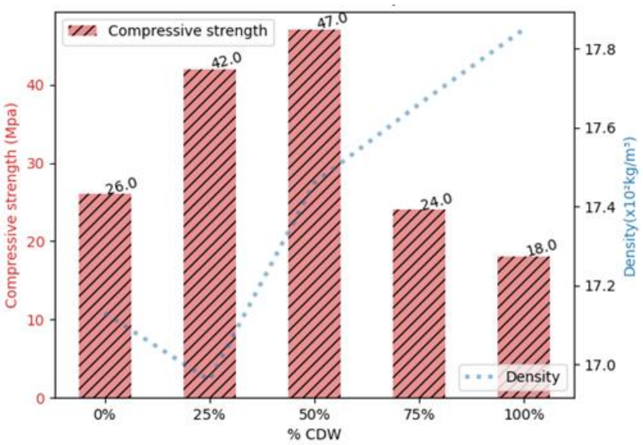


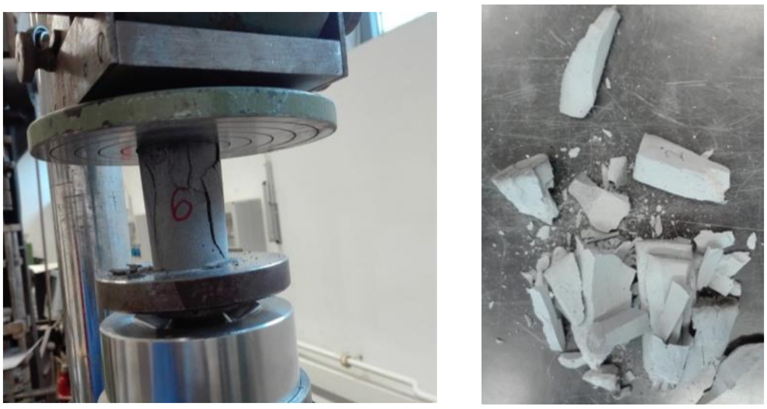
Figure 2. *Compressive strengths and Densities for CDW & FA based geopolymer composites*

The 100% FA geopolymer composite had better workability compared to those composites containing CDW, while the 100% CDW based geopolymer fresh paste had relatively lower workability. Generally, the average densities were higher with increasing amount of CDW powder proportions.

The GFA geopolymer composite exhibited cracks as illustrated in *Figure 3*. Cracks were observed on the surface of GFA composite specimen. Those were attributed to loss of moisture at the surface. Rapid moisture loss can result to excessive crack in cement treated materials (Portland cement association (PCA) 2003). In Singh *et al.* (2018) study, cracks were observed in geopolymer composite cubes having pulverized red mud and thermal cured, hence resulted to reduced strength. Kumar S & Kumar R (2011) study for samples cured at 60 ºC, for 4 hours, with unmilled FA had compressive strength of less than 15 MPa while those with milled FA (90 minutes), had strengths in excess of 67 MPa. Hence, with proper mitigation of cracks to GFA geopolymer composite specimen can result to very significant compressive strength increase.

Figure 3. *Cracks in GFA geopolymer composite specimens*

The GFA based geopolymer composite specimen generally shattered when loaded to failure, and indication of higher brittleness as compared with the RFA based specimen, with more tendancy of elastic failure, *Figure 4* shows the shattered specimen and crack on specimen.



(a) *(b)*

Figures 4. *(a) RFA geopolymer composite specimen cracking after load of compressive test machine and (b) shatter for GFA geopolymer composite specimen after loading*

Generally, at/towards failure, the FA based geopolymer composite shattered compared with CDW based geopolymer composite, hence the latter had less brittle tendancy.

Graphs for FTIR, for the RFA and GFA geopolymer composite materials are illustrated in *Figure 5*, with peaks at 3456.78 cm^{-1}, 1648.84 cm^{-1}, 1448.28 cm^{-1}, 1456.96 cm^{-1}, 1001 cm^{-1}, 491 cm^{-1}, 457 cm^{-1}, 431 cm^{-1}, 424 cm^{-1} and 404 cm^{-1}. FTIR, peak of the RFA geopolymer composite material at 3456.78 cm^{-1}, could be due to OH bonds, HOH bond stretching vibration similarly to Mucsi *et al.* (2015) study on FA based geopolymer composite, at 3406.64 cm^{-1} peak. Peak at 1001 cm^{-1}, in both geopolymer composite material specimens and at 491 cm^{-1}, 457 cm^{-1}, 424 cm^{-1} for geopolymer composite with RFA and at 404 cm^{-1}, 431 cm^{-1} for geopolymer composite with GFA could be due to internal vibrations of Si-O-Si, Si-O-Al. The internal vibrations of Si-O-Si, Si-O-Al are found in aluminosilicates, at 950–1250 cm^{-1} and 420–500 cm^{-1} (Davidovits 2008).

Graphs for FTIR, for the 5 samples of GFA and CDW based geopolymer composite materials are illustrated in *Figure 6*. CDW (0%) had a peak at 3648.66 cm^{-1}. All the 5 samples, peaks were recorded between 2364.30 cm^{-1} (25% CDW sample) and 2358.52 cm^{-1} (75% CDW sample). A peak was detected at 1540 cm^{-1} for the 0% CDW sample. All the 5 samples, peaks were recorded between 1456.96 cm^{-1} (50%, 25% and 0% CDW sample) and 1419.36 cm^{-1}, (100% CDW sample), indicating a possible shift, to lower wavenumbers, with increase proportion of CDW, for above 50% CDW sample. All the 5 samples, peaks were recorded between 1008.56 cm^{-1} (75% CDW sample) and 986.41 cm^{-1} (100% CDW sample) also all the 5 samples, peaks were recorded between 484.05 cm^{-1} (50% CDW sample) and 421.37 cm^{-1} (100% CDW sample), and indication of internal vibrations of Si-O-Si, Si-O-Al, according to Davidovits (2008), for aluminosilicates internal vibration, at 950–1250 cm^{-1} and 420–500 cm^{-1}.

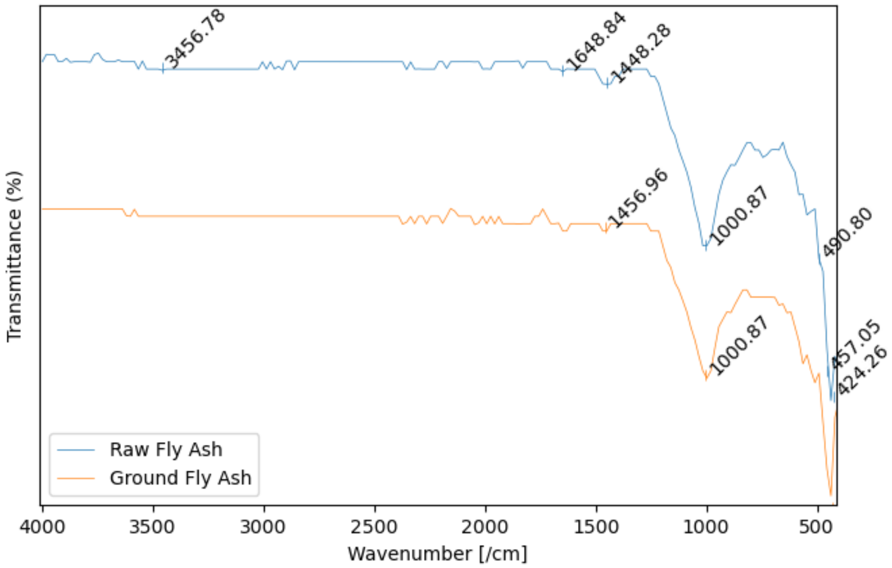


Figure 5. *FTIR for FA based geopolymer composite*

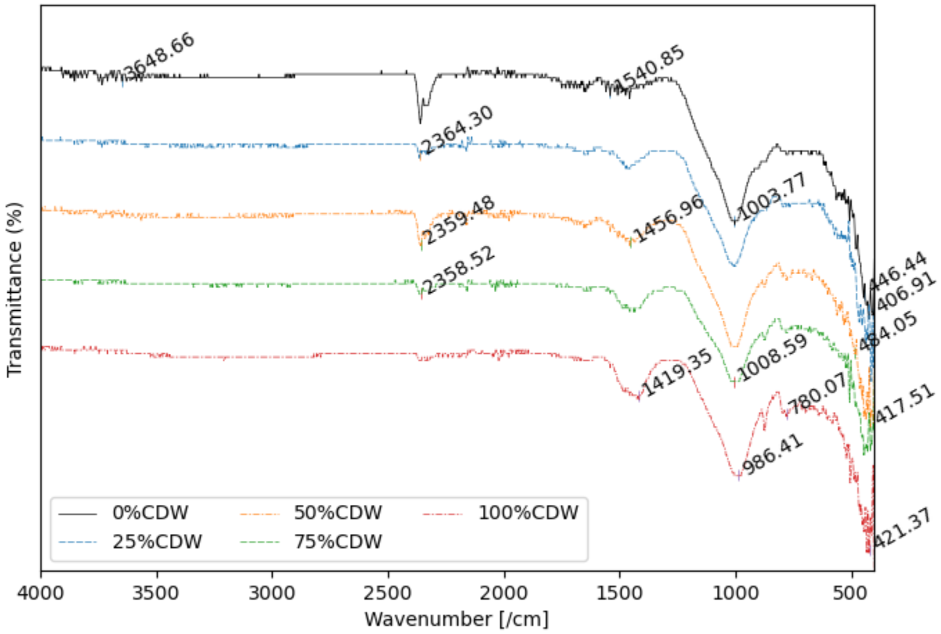


Figure 6. *FTIR for FA and CDW based geopolymer composite*

4. Summary

The GFA based geopolymer composite materials, achieved 53.8% higher strength compared to the RFA based geopolymer composite specimen. The GFA geopolymer composite specimen generally shattered when loaded to failure, indicated higher brittleness tendancy in comparision to RFA specimen, with greater tendancy of elastic failure. There is a need for mitigation of excessive cracking to GFA geopolymer composite specimen, by use of appropriate cover or techniques that would reduce the moisture loss at freshly moulded geopolymer composite material surface, such as adequately covering paste with adequate moisture retaining materials. The FTIR for the RFA and GFA and also FA and CDW based geopolymer composites materials indicated presence of Si-O-Si and Si-O-Al bonds.

The 100% CDW had reduced flowability and workability as compared with other 2nd series materials. Generally, at/towards failure, the GFA based geopolymer composite shattered at higher degrees, an indicator of brittleness compared to the CDW based geopolymer composite. The highest compressive strength for CDW and FA based geopolymer composite was the 50% CDW & FA at 47 ±9.13 MPa, and the lowest was 100% CDW at 18 ±1.50 MPa, this lower strength could be attributed to reduced workability, with increase in CDW. Based on the optimum strength results for CDW and FA based geopolymer composite, the material had compressive strength adequate for structural application consideration.

Further studies should be conducted on optimal CDW particle sizes for crack control.

5. Acknowledgments

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