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## Sustainable utilization of waste carbon black in alkali-activated mortar production

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## ABSTRACT

This article investigates the potential utilization of waste carbon black (WCB) resulting from the aluminum industry as a by-product material in the fly ash-based geopolymer composites production. Experimental study was conducted to evaluate the effect of WCB on the performance of the geopolymer. Different contents of WCB including 5%, 10%, 15%, 20%, 30%, and 40%, by weight of the fly ash, have been incorporated in the geopolymer mix as either additives or fly ash replacement. Life cycle assessment (LCA) has also been conducted to evaluate the landfills utilization and the environmental impact of the WCB incorporation. The experimental results reflected that the WCB could be used as additives in small quantities (5% of fly ash weight) to the geopolymer mix without negatively affecting its performance. Adding 5% of WCB insignificantly enhanced the compressive strength of the geopolymer by 5%, increased its workability and density by 3% and 4%, respectively, and did not affect its excellent thermal stability. Scanning electron microscopic (SEM) imaging showed more unreacted fly ash particles combined with more voids and cracks within the microstructure of the geopolymer with high WCB content. Finally, incorporating WCB in the geopolymer production improved the utilization of landfills use and reduced the global warming potential, acidification potential, eutrophication potential and abiotic depletion potential.

## 1. Introduction

Concrete is the most popular and widely used material in construction industry. In which, ordinary Portland cement is used as a binder. Unfortunately, the production of cement releases about 5% of carbon dioxide emission to the atmosphere [1], which makes it the second largest contributor to global warming [2]. Therefore, coming up with a new binding material to substitute ordinary Portland cement in concrete represents a priority for the researchers in the field of construction industry. In this regards, geopolymers or alkali-activated materials showed the ability to play a vital role. Geopolymers are ceramic-like materials with amorphous or semi crystalline 3D silica aluminate structures, form due to the activation of source materials that are rich of silica and alumina by alkaline solutions. Geopolymer composites not only own lower carbon dioxide footprint than the traditional cement-based materials [3,4], but also have comparable or even better mechanical and durability properties [5–7]. That make the geopolymers attractive to be utilized in many construction applications such as heavy metal immobilization [8,9], coating applications [10], and fire resistance applications [11].

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The performance of geopolymers highly depends on the type of source materials [12–14]. The research in the field of geopolymers was initially limited to use naturally available source materials such as clay and kaolin [15]. The research was then expanded to include the waste by-products as source materials in geopolymer production [16]. The wastes could be agricultural waste (rice husk ash, palm oil fuel ash, corncob ash) [17,18], municipal waste (paper waste, glass wool fibers, plastic and rubber waste) [19–21], or industrial waste (fly ash, silica fume, slag) [22–25]. This trend made the geopolymer production more sustainable, green and eco-friendly, and economical [26,27]. Recent research showed the ability of geopolymer composites to valorize and immobilize various industrial waste materials [16]. Rashad et al. [16] investigated the effect of using carbonation beet residue, resulting from the sugar beet industry as a by-product material, as an additive or accelerating agent on the properties of fly ash geopolymer cured at room temperature. Their results indicated that using 10–25% of carbonation beet residue gave good strength and a reasonable setting rate. Kakria et al. [28] studied the feasibility of incorporating the non-metallic fractions (NMF) of waste printed circuit boards (WPCB) in geopolymer mortar as fly ash and metakaolin replacement. Their results showed that replaced fly ash with NMF of WPCB increased the compressive strength of geopolymer mortar and reduced its water absorption. Gholampour et al. [29] investigated the properties of ambient cured geopolymer concrete made of fly ash and ground granulated blast furnace slag as source materials and lead smelter slag and glass sand. Their results showed that incorporated the GGBS enhanced the mechanical strengths and reduced the water absorption of geopolymer mortar.

Waste carbon black (WCB) is unwanted and difficult to dispose material resulting from many industries as a by-product material. It is very fine powder with high carbon content. This waste is usually decomposed into landfills causing soil contamination and water pollution [30]. Therefore, the researchers tried to recycle this waste in various applications. Many previous studies investigated the ability of recycling WCB in concrete production. The results reported in [31,32] showed the ability of the WCB to enhance the compressive strength of concrete. Another study [30] showed that adding WCB combined with other fillers caused a reduction in the electrical resistance of concrete. Another source of WCB is the aluminum industry. The WCB is generated as a byproduct in the aluminum factories, and it is usually dumped in landfills. The authors investigated the feasibility of utilizing this waste in cement mortar production [33]. The results showed that replacing up to 13% of cement content with WCB resulted in enhancing the compressive strength of cement mortar.

The above-mentioned literatures showed the ability of geopolymer composites to immobilize and valorize various industrial waste materials. In addition, incorporating the WCB in Portland cement mortar showed positive impact on its compressive strength. However, there was no published work, as per the authors' knowledge, investigated the feasibility of recycling this waste material in geopolymer composites production. The novelty of this work comes from the fact that this study tried to fill the gap and investigated the environmental effects and mechanical properties of geopolymer composites with WCB. In this research, comprehensive study was firstly conducted to identify the optimum geopolymer mix design. The optimum mix design was used to investigate the performance of geopolymer mortar with WCB as additives or replacement to the fly ash. After that, the environmental impact of using WCB in geopolymer composites production was investigated by conducting life cycle assessment.

## 2. Experimental program

### 2.1. Materials

The geopolymer mortar was prepared using Class F fly ash as a source material, silica sand, and sodium hydroxide (NaOH) and sodium silicate ( $\text{Na}_2\text{SiO}_3$ ) as alkaline solutions. Commercially available superplasticizer (PC 485, EPSILONE) was used to enhance the workability of the mix. The fly ash had particle size distribution as shown in Fig. 1. It was used as received from SMEET Qatar with chemical composition complying with ASTM C618–12a standard as shown in Table 1. The moisture content and the density of the fly ash were equal to 0.5% and  $2.23 \text{ g/cm}^3$ , respectively. Locally available silica sand with fines modulus of 2.31, specific gravity of 2.56, and water absorption of 1.87% was used. The sand conforms ASTM C778 standard. The sodium silicate solution was received from

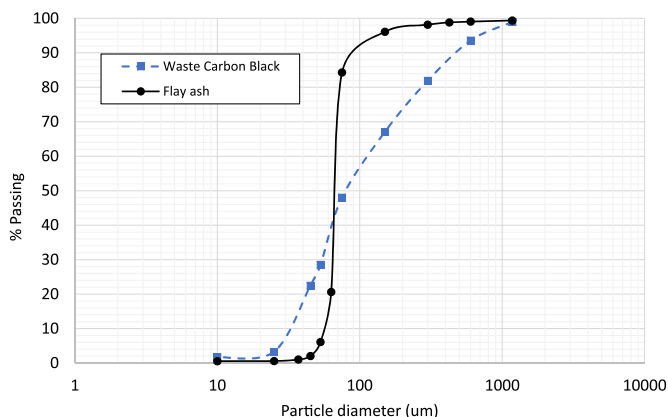


Fig. 1. Particle size distribution of fly ash and waste carbon black.

**Table 1**  
Chemical composition of Fly ash and WCB used in this study.

Fly ash		Waste carbon black	
Oxide	(%)	Element	(%)
SiO <sub>2</sub>	49.90	Carbon	85
Al <sub>2</sub> O <sub>3</sub>	17.10	Iron	7
CaO	11.80	Fluoride	4
Fe <sub>2</sub> O <sub>3</sub>	7.83	Sodium	2.5
MgO	4.90	Sulphur	1
SO <sub>3</sub>	0.42	Silicon	0.5
K <sub>2</sub> O	0.28		
Na <sub>2</sub> O	0.14		
Cl	0.01		
LOI	7.62		

Qatar Detergent Company in Qatar. The chemical composition of the solution is as follow: 13% Na<sub>2</sub>O, 27% SiO<sub>3</sub>, and 60% water. The NaOH solution was prepared using NaOH pellets with purity of 98% that was acquired from a local supplier. The NaOH solution with concentrations of 4, 6, 8, and 10 Molar were prepared by mixing 160 g, 240 g, 320 g and 400 g of NaOH pellets, respectively, in one liter of distilled water. The NaOH pellets were dissolved gradually in distilled water until reaching the desired concentration of the solution. Each solution was prepared in a big plastic container immersed in a water bath to accommodate the heat of the reaction between the NaOH and water as the reaction is exothermic reaction. The solution was left at room temperature for 30 min to lower its temperature. The solution was then poured in an airtight glass jar to prevent the reaction with air and to be used later to prepare the different mix designs of geopolymers mortar.

The carbon black used in this study was a waste material with a density of 1.9 g/cm<sup>3</sup> generated in large quantities during the production process of Aluminum in Qatar Aluminum Company (Qatalum). After receiving the WCB from Qatalum, it was characterized to determine its chemical composition using X-ray fluorescence (XRF) and X-ray diffraction (XRD) tests, particle size distribution (PSD) using sieve analysis, morphology and particle shapes using scanning electron microscopic (SEM) imaging. The PSD of WCB is plotted in Fig. 1. The figure indicated that the WCB had continuous graded PSDs with some particles finer than the fly ash particles. According to the SEM imaging shown in Fig. 2, the particles of the WCB owned angular and irregular shapes. According to the XRF and XRD analysis, 85% of the total mass of the WCB is carbon (Table 1) with very wide peak at  $2\theta = 25^\circ$  as shown in Fig. 3.

## 2.2. Geopolymer mix design and specimens preparation

The experimental program was divided into two stages as shown in Fig. 4. In the first stage, sixteen geopolymer mixes were prepared with four different molarities of NaOH solution (4 M, 6 M, 8 M, and 16 M), two fluid to binder ratios (0.6 and 0.65), and two sodium silicates to sodium hydroxide ratios (1.0 and 1.5). The optimum mix design from stage one was determined based on the compressive strength values, and used in stage two. In the second stage, geopolymers with WCB were prepared. The WCB was incorporated into the mixes by either partially replaced the fly ash or as an additives. The optimum mix design that extracted from stage one was used with four different WCB contents 5%, 10%, 20%, and 30% by weight of fly ash to prepare twelve more mixes. Detailed mix design is listed in Table 2.

The geopolymer mortar was prepared by mixing the NaOH and Na<sub>2</sub>SiO<sub>3</sub> solutions for two minutes in a glass measuring jar. Then, the activator solutions were placed in the mixing bowl, and fly ash and WCB were added and mixed in the mixing bowl for 30 s at low speed. After that, the sand was added to the mix and mixes for 30 s at low speed. The mixing speed increased to high speed and the mixing continues for 120 s. At this point, the flow table test was conducted, followed by 15 s mixing at high speed. The prepared mortar was used to cast specimens with dimensions of 50 mm × 50 mm × 50 mm and 40 mm × 40 mm × 160 mm in order to perform the compressive and flexural strength tests, respectively. All geopolymer mortar specimens were cured at a fixed temperature of 80 °C and for a duration of 24 h.

## 2.3. Test procedures

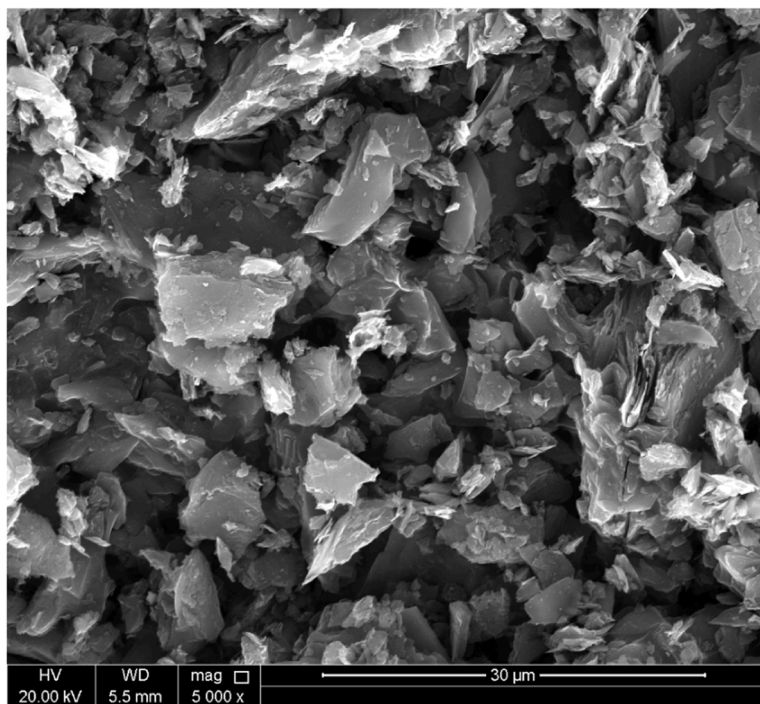
### 2.3.1. Mechanical strengths

The mechanical behavior of geopolymer mortar with WCB was studied through compressive and flexural strengths. Compressive and flexural strength tests of geopolymer mortar were performed using universal testing machine according to the ASTM C109 and ASTM C348 standards, respectively. Geopolymer mortar specimens were loaded until failure with loading rate of 1.3 kN/s and 0.044 kN/s for compression and flexural tests, respectively. The average strength value of three tested specimens for each mix was reported. Fig. 5 shows samples preparation and tests procedures.



(a)

(b)



(c)

Fig. 2. WCB used in this study (a) packed for landfill (b) powder (c) SEM image.

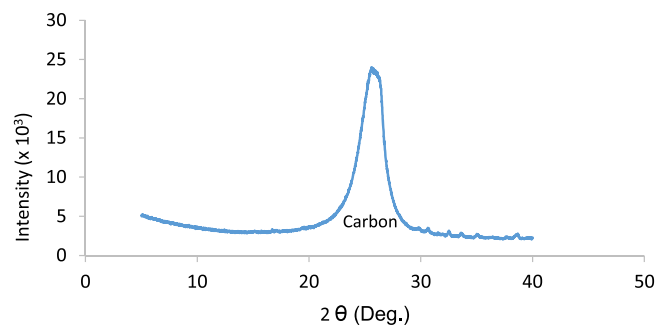


Fig. 3. XRD pattern of WCB.

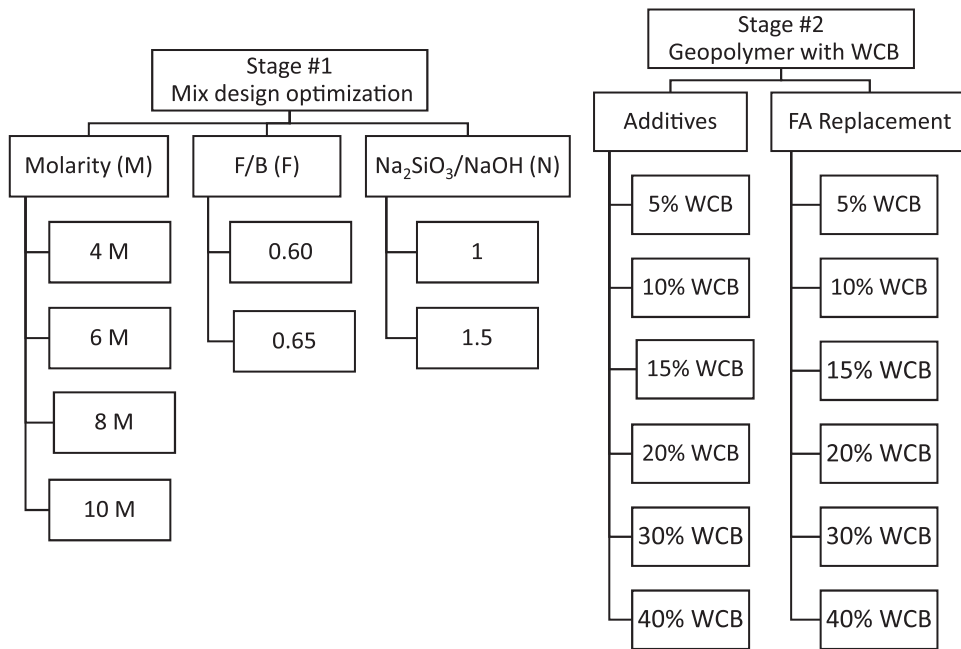


Fig. 4. Experiments flowchart.

Table 2  
Geopolymer mortar mix design.

	Mix No.	Specimen ID	NaOH Concentration (M)	F/B	Na <sub>2</sub> SiO <sub>3</sub> /NaOH	Fly ash (Kg/m <sup>3</sup> )	Carbon Dust (Kg/m <sup>3</sup> )	Sand (Kg/m <sup>3</sup> )	NaOH solution (Kg/m <sup>3</sup> )	Na <sub>2</sub> SiO <sub>3</sub> solution (Kg/m <sup>3</sup> )	SP (Kg/m <sup>3</sup> )
Stage 1	M1	4 M-F1-N1	4	0.6	1	711	–	1956	213	213	7
	M2	4 M-F1-N2			1.5	711	–	1956	171	256	7
	M3	4 M-F2-N1		0.65	1	711	–	1956	231	231	7
	M4	4 M-F2-N2			1.5	711	–	1956	185	277	7
	M5	6 M-F1-N1	6	0.6	1	711	–	1956	213	213	7
	M6	6 M-F1-N2			1.5	711	–	1956	171	256	7
	M7	6 M-F2-N1		0.65	1	711	–	1956	231	231	7
	M8	6 M-F2-N2			1.5	711	–	1956	185	277	7
	M9	8 M-F1-N1	8	0.6	1	711	–	1956	213	213	7
	M10	8 M-F1-N2			1.5	711	–	1956	171	256	7
	M11	8 M-F2-N1		0.65	1	711	–	1956	231	231	7
	M12	8 M-F2-N2			1.5	711	–	1956	185	277	7
	M13	10 M-F1-N1	10	0.6	1	711	–	1956	213	213	7
	M14	10 M-F1-N2			1.5	711	–	1956	171	256	7
	M15	10 M-F2-N1		0.65	1	711	–	1956	231	231	7
	M16	10 M-F2-N2			1.5	711	–	1956	185	277	7
Stage 2	M17	5AWCB	10	0.6	1.5	711	35.55	1956	171	256	7
	M18	5RWCB				675	35.55	1956	171	256	7
	M19	10AWCB	10	0.6	1.5	711	71.1	1956	171	256	7
	M20	10RWCB				640	71.1	1956	171	256	7
	M21	15AWCB	10	0.6	1.5	711	106.65	1956	171	256	7
	M22	15RWCB				604	106.65	1956	171	256	7
	M23	20AWCB	10	0.6	1.5	711	142.2	1956	171	256	7
	M24	20RWCB				569	142.2	1956	171	256	7
	M25	30AWCB	10	0.6	1.5	711	213.3	1956	171	256	7
	M26	30RWCB				498	213.3	1956	171	256	7
	M27	40AWCB	10	0.6	1.5	711	284.4	1956	171	256	7
	M28	40RWCB				427	284.4	1956	171	256	7

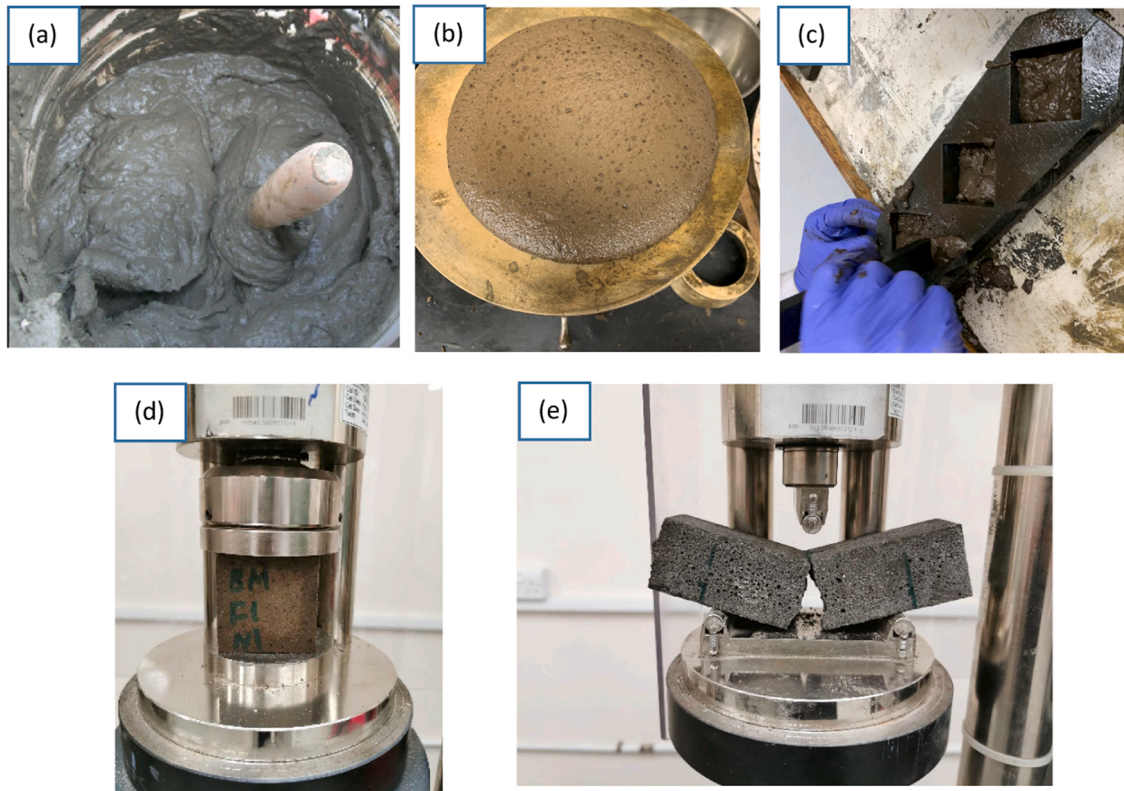


Fig. 5. Samples preparation and testing procedure (a) Mixing (b) flow table test (c) casting (d) compression test (e) flexural test.

### 2.3.2. Workability

The workability of geopolymer mortar with WCB was investigated using flow table test according to the ASTM C1437 standard. The average of four readings for each mix was calculated and reported.

### 2.3.3. Density

The effect of WCB on the density of geopolymer mortar were determined according to the ASTM C642 standard. After the flexural test, selected specimens were used to measure the density. The specimens were weighted and then placed in an oven at  $110 \pm 5$  °C for 24 h. After that, the specimens were removed from the oven, left in the ambient temperature to cool and then weighted. The procedure was repeated until the weight loss did not exceed 1% of the original weight of the specimen. Finally, the dry density was calculated as the weight of the specimen divided by its volume.

### 2.3.4. Microstructural investigation

SEM imaging was performed according to the ASTM C1723 to explore the microstructure of geopolymer mortar with WCB. After the compression test, small fragments were extracted from different locations of the specimens, coated with gold to enhance the conductivity, and tested using NOVA NanoSEM 450 device. Energy dispersive X-ray spectroscopy (EDS) analysis was conducted using same device according to the ASTM E1508 standard to locate the WCB particles within the geopolymer products.

### 2.3.5. Phase composition and mineralogical analysis

Selected specimens were grinded to powder with a size of less than 150  $\mu\text{m}$ . The grinded powder was used to conduct the XRD test using JSX 3201 M (Jeol) spectroscopy device in order to investigate their mineralogical content and phase composition.

### 2.3.6. Thermal stability analysis

Thermal characteristics of geopolymer mortar with WCB was examined by conducting thermogravimetric analysis (TGA) tests. The test gave an indication about the stability of the geopolymer when exposed to elevated temperatures in the range of 30 °C and 750 °C. Selected specimens were grinded to powder with a size of less than 45  $\mu\text{m}$ . The grinded powder was used to conduct the TGA test using

TGA 4000 PerkinElmer device.

### 3. Life cycle assessment (LCA)

The LCA process usually consists four components: goal and scope definition, inventory phase, impact assessment phase, interpretation phase [26,27,34], as shown in Fig. 4. The LCA has been employed in this research to estimate the environmental impact of incorporating the WCB in geopolymer mortar production. The scope was to investigate the effect of incorporating WCB as an additive or fly ash substitution in the geopolymer mix through the four processes of cradle-to-gate. The Centrum voor Milieukunde Leiden (CML) 2016 method [35] was followed to perform the LCA. In addition to the land use, the assessment focused on the impact of four categories: acidification potential (AP), global warming potential (GWP), abiotic depletion potential (ADP fossil), and eutrophication potential (EP). Finally, the LCA was simulating using GaBi software.

GaBi database was used to source the unit processes of fly ash, NaOH, Na<sub>2</sub>SiO<sub>3</sub>, carbon black, sand, water and electricity. The WCB was used as a waste material with proper properties to mimic the real life situation [36].

## 4. Experimental results and discussions

### 4.1. Geopolymer mix design optimization

Optimum mix design of geopolymer mortar was determined based on the compressive strength values of the sixteen different mixes tested in this study. The results are summarized in Table 3. According to the results, the molarity of the NaOH solution greatly affected the strength of geopolymer mortar for all F/B and Na<sub>2</sub>SiO<sub>3</sub>/ NaOH ratios. In general, increasing the molarity enhanced the compressive strength. The improvement in the compressive strength due to the increase in the molarity was more pronounced for specimens prepared with Na<sub>2</sub>SiO<sub>3</sub>/ NaOH of 1.5 compared to Na<sub>2</sub>SiO<sub>3</sub>/ NaOH of 1.0, and for specimens prepared with F/B ratio of 0.6 compared to F/B ratio of 0.65 as shown in Fig. 5. The enhancement in the compressive strength of geopolymer mortar with increasing the NaOH molarity could be attributed to the more dissolution of silica and alumina in the mix [15]. Moreover, increasing the Na<sub>2</sub>SiO<sub>3</sub>/ NaOH ratio from 1 to 1.5 increased the rate of the polymerization reaction due to the present of more soluble silicates [37].

Finally, it is clear that mix 10 M-F1-N2 owned the highest compressive strength among the others. Based on that, it was selected to be used in the next stage to study the behavior of geopolymer mortar with WCB.

### 4.2. Performance evaluation of geopolymer mortar with WCB

#### 4.2.1. Mechanical strengths

The target of this part of the study was to determine the optimum amount of WCB that can be incorporated into the geopolymer mix without negatively affecting its mechanical properties. The WCB either was added to the mix or replaced the fly ash. Six WCB contents (as fly ash weight percentages) were considered herein including 5%, 10%, 15%, 20%, 30% and 40%. The results are presented in Fig. 6 and Table 4. The compressive strength of control specimen was equal to 44.8 MPa. Adding different dosages of WCB to the geopolymer mix up to 40% of its fly ash content insignificantly affected its compressive strength. On contrary, substituting the fly ash by WCB clearly reduced the compressive strength of the geopolymer mortar regardless of the replacement ratio. The reduction in the compressive strength started with 5% in the case of 5% replacement ratio and reached up to 44% in the case of 40% replacement ratio. The reduction in the compressive strength due to the substitution of fly ash with WCB could be attributed to the reduction in the amount of silica and alumina in the mix. The Si content is responsible for determining whether the condensation occurs between Al-Si or Si-Si, whereas the Al content governs the type of chemical structure and network formation [38]. In general, high content of reactive

**Table 3**  
Compressive strength results for mix design optimization.

Mix design ID	Compressive strength (MPa)	Standard deviation
4 M-F1-N1	12.33	0.1
6 M-F1-N1	33.72	0.15
8 M-F1-N1	48.83	0.7
10 M-F1-N1	45.59	0.6
4 M-F1-N2	19.07	0.14
6 M-F1-N2	37.52	0.24
8 M-F1-N2	43.46	0.75
10 M-F1-N2	51.1	0.9
4 M-F2-N1	13.09	0.11
6 M-F2-N1	32.91	0.14
8 M-F2-N1	40.41	0.6
10 M-F2-N1	39.14	0.56
4 M-F2-N2	21.26	0.15
6 M-F2-N2	31.46	0.2
8 M-F2-N2	37.68	0.5
10 M-F2-N2	47.50	0.7

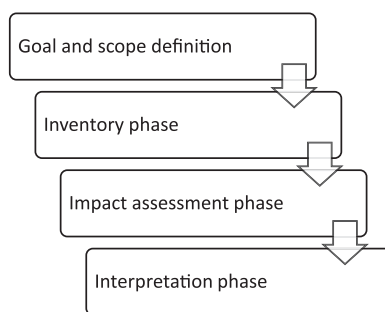


Fig. 6. LCA components.

Table 4

Experimental results of geopolymer with WCB.

Sample ID	Compressive strength			Flexural strength			Spread diameter	
	Value (MPa)	Change (%)	Std.	Value (kN)	Change (%)	Std.	Value (mm)	Change (%)
control	44.79	–	0.77	2.86	–	0.14	231	–
5 A	46.89	5	0.56	2.68	-6	0.07	239	3
5 R	42.45	-5	0.39	2.64	-8	0.21	211	-9
10 A	39.72	-11	0.91	3.03	6	0.20	209	-9
10 R	34.73	-22	1.16	2.38	-17	0.39	218	-6
15 A	41.41	-8	1.19	2.68	-6	0.10	184	-20
15 R	35.23	-21	0.96	2.47	-14	0.16	188	-19
20 A	45.90	2	1.57	2.64	-8	0.06	168	-27
20 R	27.92	-38	0.91	2.32	-19	0.20	163	-29
30 A	43.28	-3	3.95	2.55	-11	0.19	167	-28
30 R	26.58	-41	0.60	2.07	-28	0.09	164	-29
40 A	41.63	-7	1.03	2.63	-8	0.08	167	-28
40 R	25.27	-44	0.66	2.09	-27	0.09	164	-29

silica significantly influences the formation of high amount of alkaline aluminosilicate gel [15]. Moreover, the reduction in the compressive strength due to fly ash replacement with WCB could be attributed to the increase in the water to fly ash ratio [39].

The flexural strength of control geopolymer mortar was equal to 2.86 kN. Adding up to 10% WCB to the mix insignificantly enhanced the flexural strength of the geopolymer mortar by 6%, whereas adding 15% or more WCB reduced the strength. Maximum reduction of 11% was noticed in the case of adding 30% of WCB into the mix. On contrary, replacing fly ash by WCB in the mix reduced the flexural strength of the geopolymer mortar regardless the replacement ratio. The reduction in the strength could reach up to 28% if 30% of the fly ash replace by WCB.

#### 4.2.2. Workability

The results of the workability test are summarized in Table 4 in terms of spread diameter. Except for the mix with 5% WCB dosage, incorporating WCB into the geopolymer mix clearly reduced the flowability of the mortar. The reduction reached 29% in the case of 20% WCB dosage as shown in Fig. 7. After that, the flowability of the mix did not affect by the addition of more WCB. The reduction in the flowability in the case of WCB addition could be credited to the decreasing in the water to solid ratio. While the reduction in the case of fly ash substitution with WCB could be attributed to the fact that WCB owned more fine particles than fly ash.

#### 4.2.3. Thermal stability

Thermogravimetric profiles of geopolymer mortar with WCB are presented in Fig. 8 to show the weight loss of the specimens when exposed to elevated temperature up to 750 °C. It is clear in Fig. 8 that the total weight loss of the fly ash-based geopolymers did not exceed 9%, which agreed with the literatures [40,41]. The weight loss of specimens with WCB substitution was higher than that of specimens with WCB addition for WCB dosages up to 10% as shown in Fig. 8a and b. In the case of using 15% WCB and more, the situation was reversed as shown in Fig. 8c-f. In general, all thermogravimetric curves could be divided into four stages as shown in Fig. 8. The first stage (S1) was located between 0 °C and 120 °C. The weight loss in this stage was associated with the evaporation of the free or evaporable water presented on the surface and porosity or cavity of the specimens [42,43]. The second stage (S2) was located between 120 °C and 230 °C. The weight loss in this stage could be attributed to the evaporation of the chemically bound water [42,44],



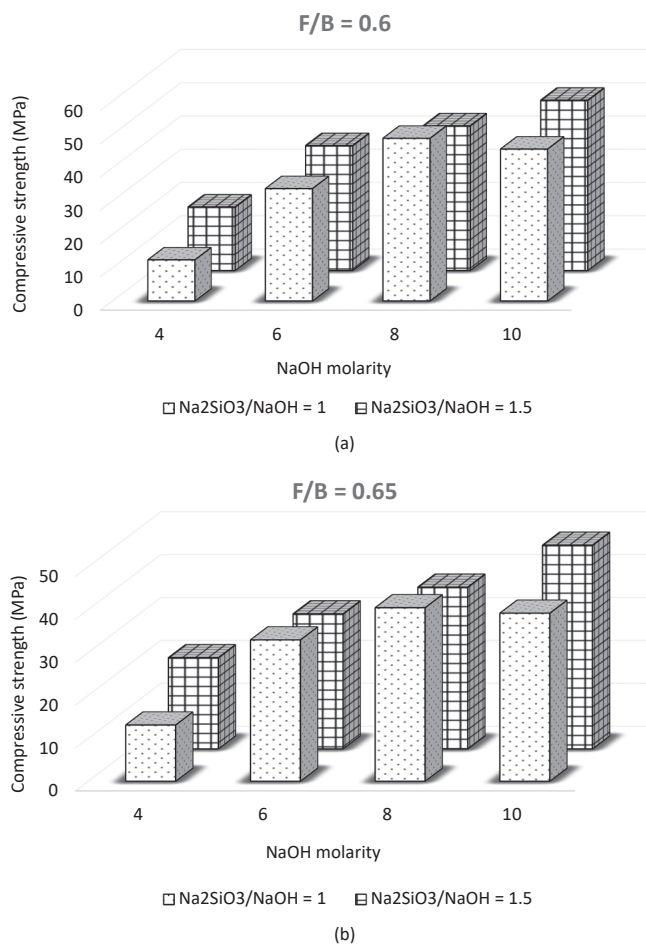


Fig. 7. Effect of synthesis parameters on compressive strength of geopolymer mortar.

or was associated with the presence of NASH gel [45]. After the second stage, the rate of the weight loss stabilized between 230°C to 630°C (S3). Another weight loss was captured between 630 °C and 750 °C (S4). This weight loss could be attributed to the dehydroxylation and recrystallization of geopolymer specimens [43].

Extracted from the TGA results, the differential Thermogravimetric (DTG) profiles of geopolymers with different WCB contents are presented in Fig. 9. It is clear from the figure that presence of WCB (either addition or substitution) caused a clear shift in the temperature corresponding to the first peak for all WCB content. In the case of specimens in which the WCB substituted the FA, the shift in the peak value could be attributed to the fact that the geopolymers with high FA content contain more calcium (Ca) atoms; therefore, they can hold pore water more strongly than geopolymers with lower FA content [42]. Knowing that presence of Ca in the geopolymer gel is helpful in holding the pore water through the formation of new phases such as N-(C)-A-S-H (calcium substitute sodium in sodium aluminosilicate hydrate) [46]. In the case of specimens in which the WCB added to the mix, the shift in the peak value could be attributed to the fact that these specimens owned more voids with larger size compared to control specimens (as shown later in the SEM section). Thus, the evaporation of the free water will be easier which caused an increase in the mass loss at lower temperatures [42].

It can be concluded from the above discussion that the geopolymers own excellent thermal stability, and the WCB does not affect this stability.

#### 4.3. Effect of WCB on the density and microstructure of geopolymer mortar

The density of geopolymer mortar with WCB was measured according to the ASTM C642 standard. The results are presented in Fig. 10. The density of the control specimen was equal to 1.97 g/cm<sup>3</sup>. Incorporating WCB insignificantly affected the density of the geopolymer mortar as clear in the figure. The maximum changes in the density due to WCB addition and substitution were equal to 6% and 7%, which happened in the case of adding 20% or substituting 5% of the fly ash content.

On the other hand, the effect of fly ash substitution with WCB on the microstructure of geopolymer mortar was investigated using SEM imaging. Fig. 11a shows the SEM image of control geopolymer specimen. The figure reflects the existence of amorphous geopolymerization products with reacted and unreacted FA particles. Substitution 5% of the fly ash content with WCB caused limited

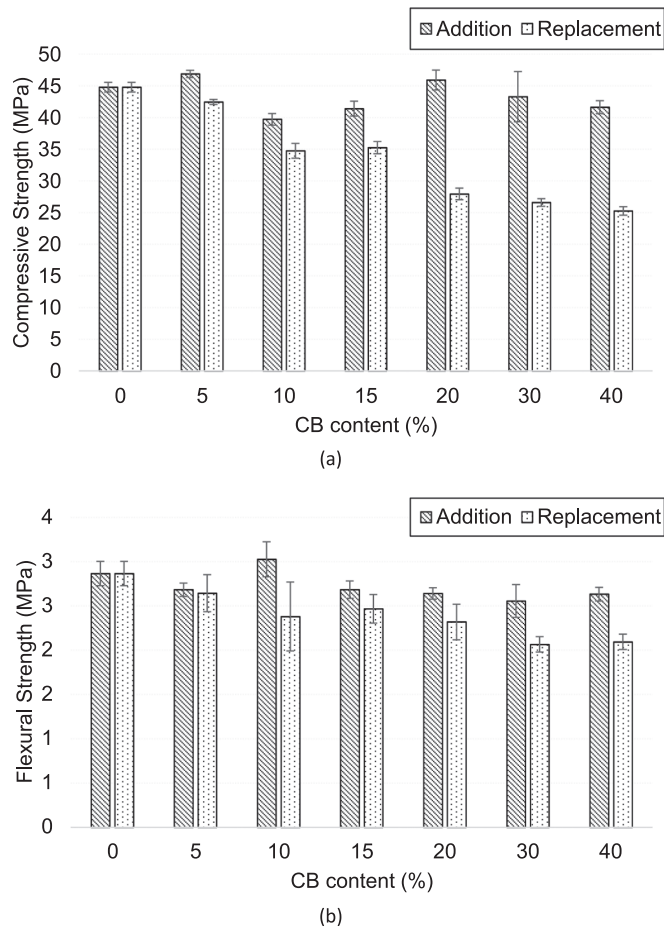


Fig. 8. Effect of waste carbon black on mechanical strengths of geopolymer mortar.

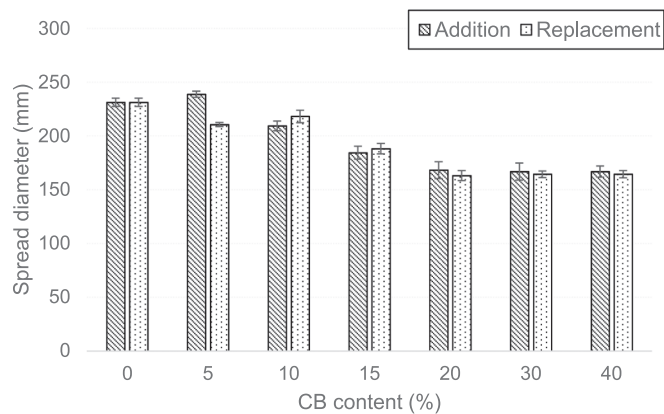


Fig. 9. Flow table results of geopolymer mortar with carbon black.

changes to the microstructure of the specimen as clear in Fig. 11b. On contrary, substitution 10% or 15% of the fly ash content with WCB significantly affected the microstructure of the geopolymer. More unreacted fly ash particles were noticed within the specimen. In addition, fragmented matrix combined with more voids and cracks were captured as shown in Fig. 11c. Finally, substitution of more than 20% of the fly ash content with WCB resulted in crumbled matrix and more wider cracks and voids distributed all over the specimen as shown in Fig. 11d. The fragmented matrix that captured in the SEM images of specimens with high substitution dosage could be attributed to the reduction in the amount of silica and alumina in the mix. The low content of reactive silica negatively

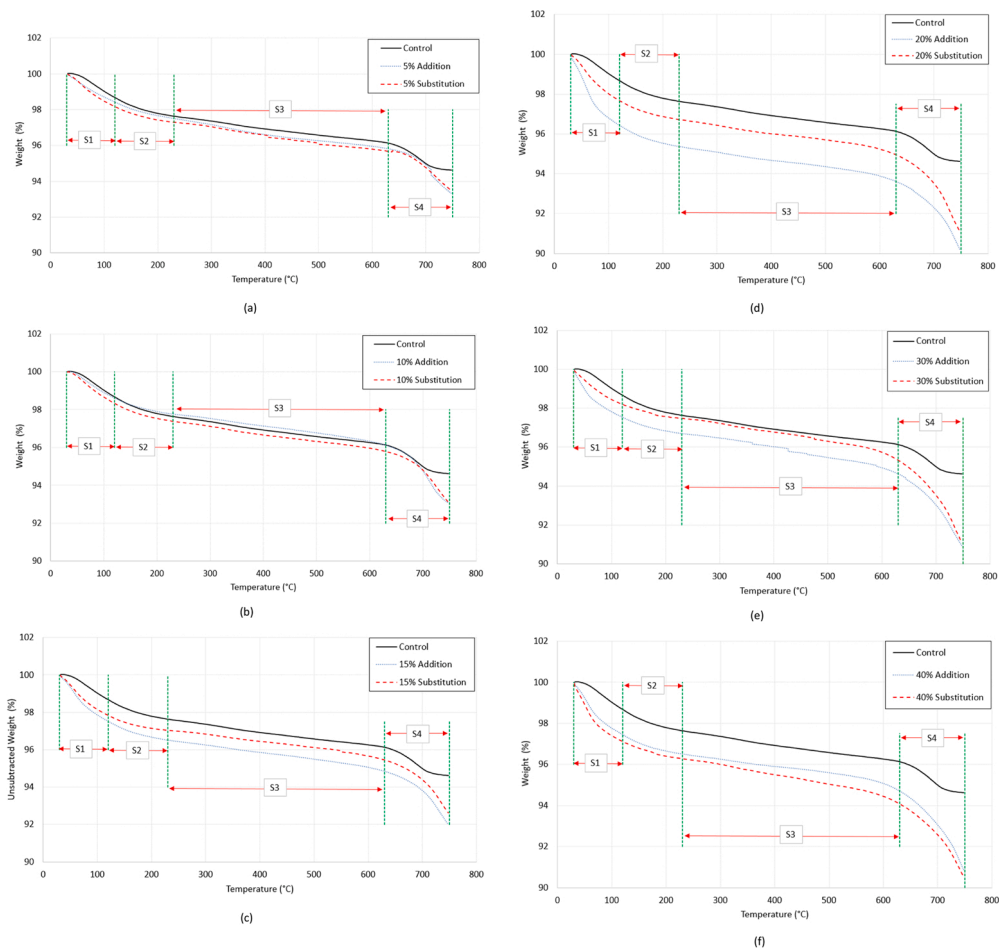


Fig. 10. TGA curves for geopolymers with different WCB contents.

influenced the formation of alkaline aluminosilicate gel [15].

#### 4.4. Phase composition and mineralogical analysis

Fig. 12 shows the XRD patterns of geopolymer specimens with and without WCB. It was clear in the figure that the geopolymer mortar consisted of main crystalline phases of quartz and mullite. The presence of these two minerals in the geopolymer composites reflected that not all silica and alumina originally existed in the fly ash were fully used in the geopolymerization process [47,48]. The diffuse halo located at  $2\theta$  values between  $20^\circ$  and  $40^\circ$  present in all of the XRD figures indicated to the typical geopolymer gels [48]. Even though no other crystalline phases were monitored due to the presence of the WCB in the mix, it was noticed that the intensity of the peaks of the main crystalline phases (quartz, mullite) decreased with the addition of the WCB. These results could be attributed to the fact that in the presence of the WCB, the silica and alumina originally presented in the fly ash were not totally dissolved and participated in the geopolymerization reaction [49] which may affect the mechanical strengths of the geopolymer.

### 5. Life cycle assessment

The results of the LCA for the production of  $1.0 \text{ m}^3$  of geopolymer mortar (GPM) with different addition and substitution percentages of WCB are presented in Figs. 13 and 14. Fig. 13 reveals that among all materials used in the fly ash-based geopolymer mix design, production of the alkaline solution was the main contributor to all impact categories in the production of GPM. The contribution of the alkaline solution production in the GWP, AP, EP and ADP categories were accounted for 80%, 68%, 69% and 91%, respectively. This trend might be attributed to the fact that the production of the alkaline solution was associated with high amount of energy consumption [50].

The effect of WCB incorporation in each category of the LCA is presented in Fig. 14. Similar trend was captured for the categories of GWP, AP, EP, and ADP due to the waste carbon black incorporation. Substitution the fly ash with WCB resulted in positive environmental impact on all four categories as shown in Fig. 14. The reduction in the GWP, AP, EP, and ADP increased with increasing the

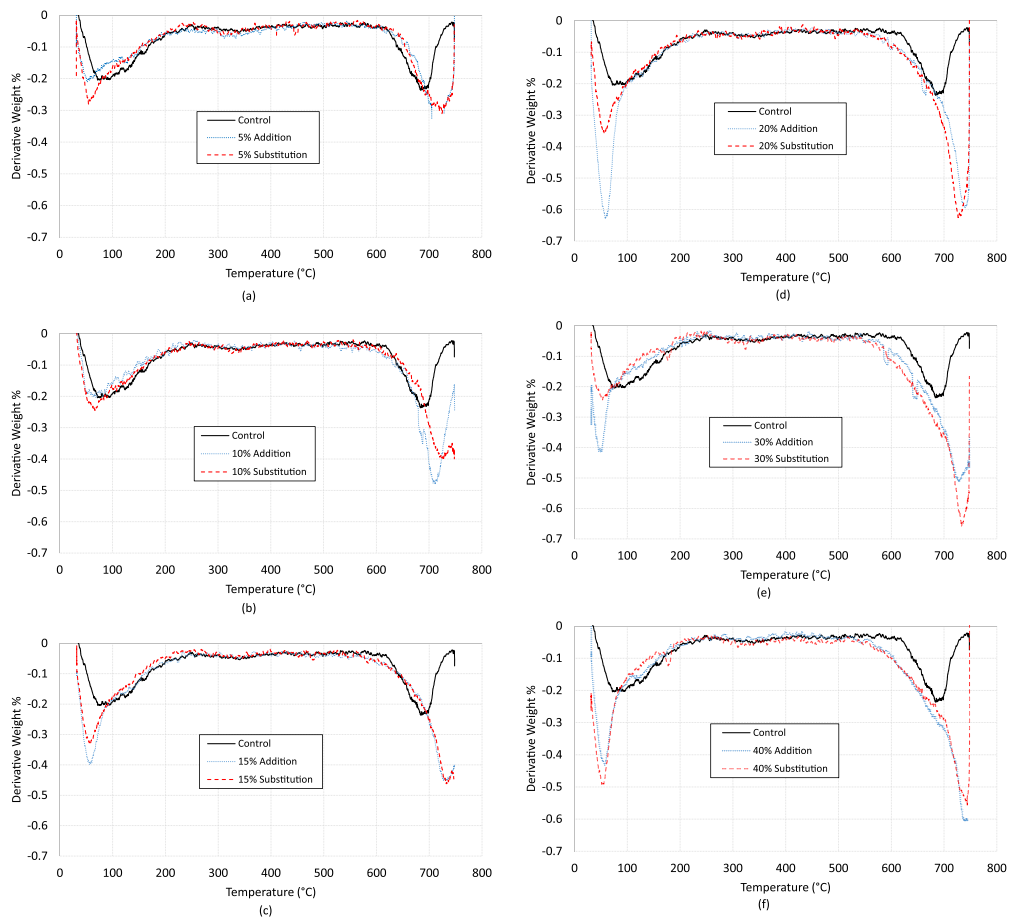


Fig. 11. DTG curves for geopolymers with different WCB contents.

substitution percentages reaching 5%, 6%, 7%, and 5%, respectively, for 40% replacement ratio. On contrary, adding WCB into the geopolymer mix resulted in negative environmental impact. The addition of WCB caused more emissions to the environment in the four impact categories GWP, AP, EP and ADP. The emission increased with increasing the WCB dosage as shown in Fig. 14.

Finally, the significant advantage of using the WCB in the production of GPM is clearly noticed in the land use category as shown in Fig. 14. Including WCB; either as fly ash replacement or addition; in the production of GPM enhanced the land use utilization. The enhancement enlarged to reach up to 131% with increasing the WCB dosage to be 40% of the fly ash content. This results could be justified by the fact of using WCB in geopolymer production instead of disposing it in the landfills reduced the environmental implications by reducing the demand of landfill areas. (Figs. 15 and 16).

## 6. Recommendation and future work

Durability of geopolymer mortar with WCB should be studied before taking a decision to use this waste material in the production of geopolymer composites thus use it in construction applications.

## 7. Conclusions

The potential utilization of WCB in alkali-activated mortar production was covered in this research. Effect of incorporating this waste material on the performance of geopolymer mortar was experimentally studied. After that, life cycle assessment was conducted to explore the landfill utilization and the environmental impact of using the WCB in geopolymer production. The following conclusions could be drawn:

1. The WCB could be used as additives in small quantities (5% of fly ash weight) to the geopolymer mix without negatively affecting its performance. Adding 5% of WCB insignificantly enhanced the compressive strength of the geopolymer by 5%, increased its workability and density by 3% and 4%, respectively, and did not affect its excellent thermal stability. Using large quantity of the WCB (10% and more) insignificantly decreased the mechanical strengths of the geopolymer but clearly reduced its workability.

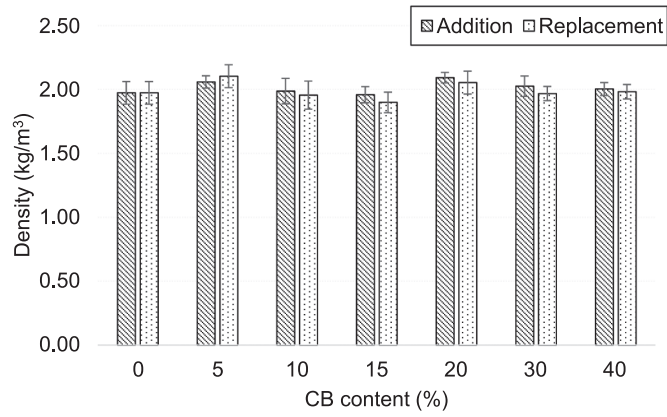


Fig. 12. Effect of WCB on the density of geopolymer mortar.

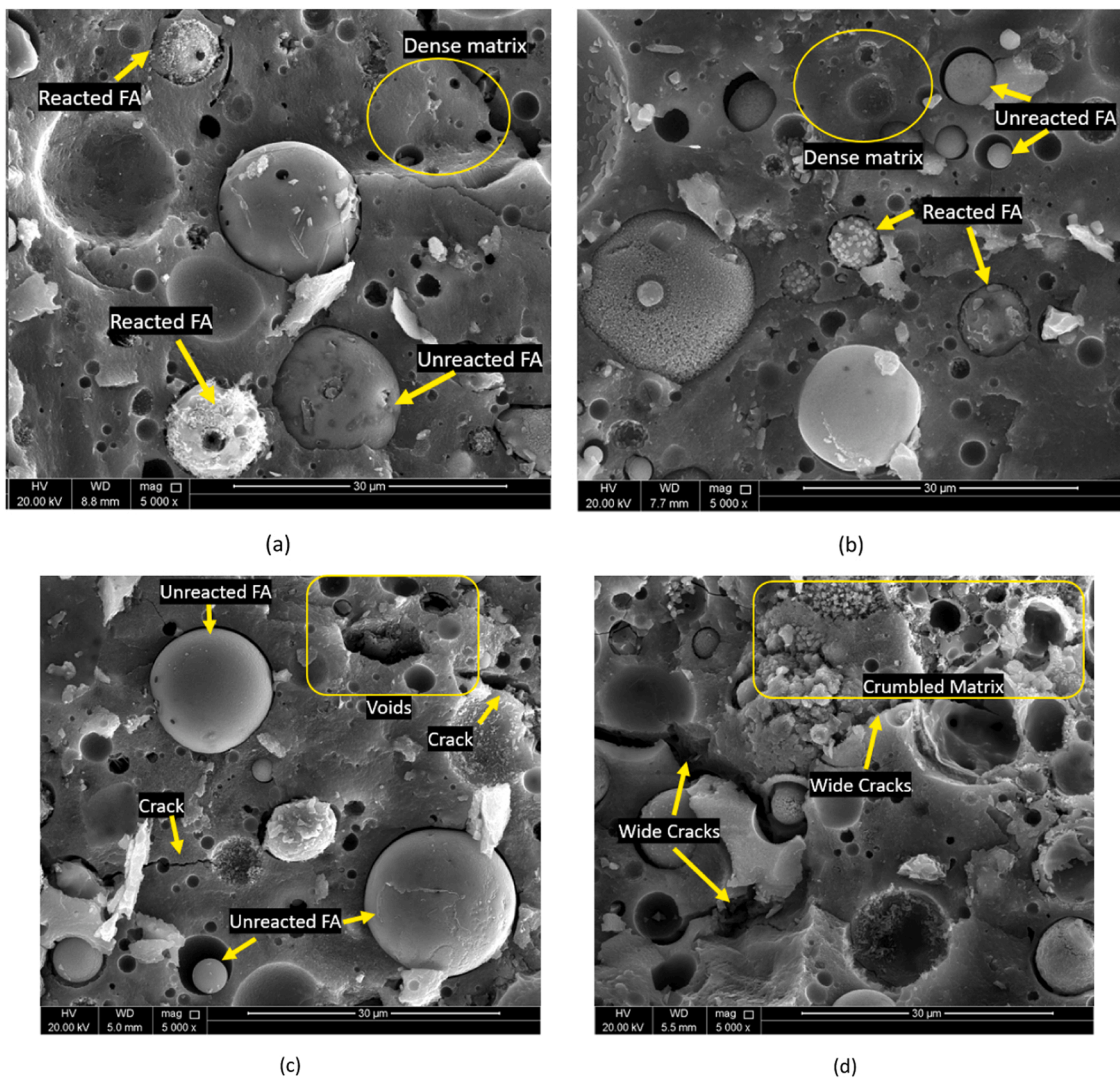


Fig. 13. SEM images of geopolymer mortar with different WCB contents (a) control (b) 5% (c) 10% (d) 20%.

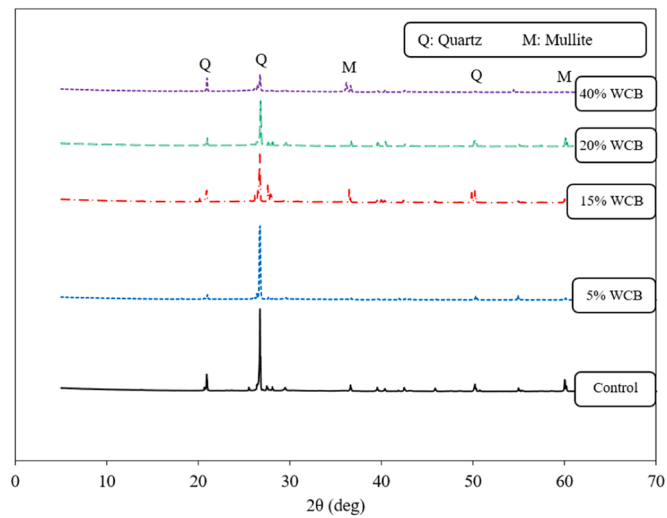


Fig. 14. XRD pattern of geopolymer mortar with different WCB contents.

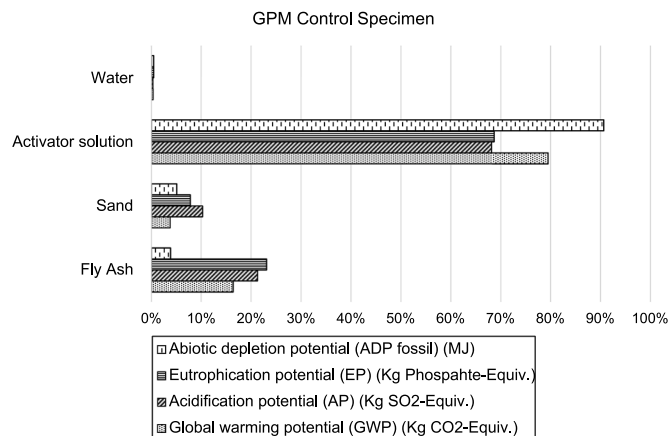


Fig. 15. Contribution of different constitution of geopolymer mortar on the LCA results.

- Substituting fly ash with WCB clearly reduced the mechanical strengths of the geopolymer mortar. The reduction in the compressive and flexural strengths increased from 5% and 8%, respectively, for 5% replacement ratio to be 44% and 28%, respectively, for 40% replacement ratio.
- The microstructure of the geopolymer mortar was affected by substituting the fly ash with WCB especially for high replacement ratio. More unreacted fly ash particles, and fragmented matrix combined with more voids and cracks were noticed within the specimens with high WCB content.
- Incorporating WCB; either as additives or fly ash replacement; in the production of geopolymer composites enhanced the land use utilization. The enhancement increased with increasing the WCB dosage to reach up to 123% and 131% in the case of adding or substituting 40% of the fly ash content with WCB, respectively.
- Among all components materials used to produce the fly ash-based geopolymer composites, production of the alkaline solution was the main contributor to all environmental impact categories used in the life cycle assessment.

### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

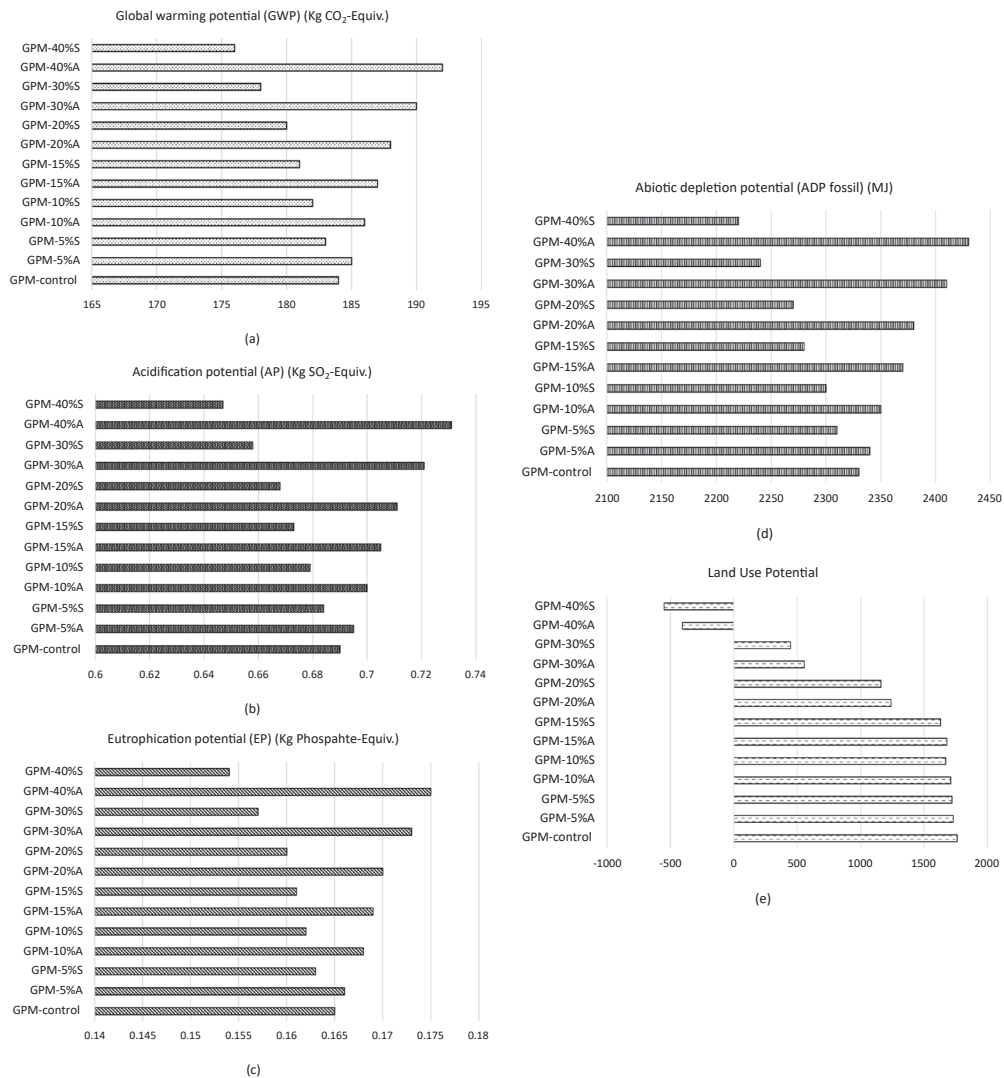


Fig. 16. LCA results effect of geopolymer with WCB.

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