

Effective Use of Waste Plastic As Sand in Metakaolin/Brick-Powder Geopolymer Concrete

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ABSTRACT

This study proposes recycling waste clay brick and waste polyethylene terephthalate (PET) bottles as substitution materials in geopolymer concrete. To accomplish this goal, the control mix of geopolymer concrete was prepared based on blended metakaolin and waste clay brick powder (CBP) at a 1:1 mixture by weight. To evaluate the use of shredded PET particles as fine aggregate, three mixtures were made by replacing sand with PET aggregate at volumetric percentages (10%, 15% and 20%). The specimens containing PET aggregate were tested and compared against the control mix (0% PET), with emphasis on the fresh and dry densities, mechanical performance, water absorption and microstructure characteristics. The results indicated the inclusion of PET aggregate to slightly reduce density and improve mechanical properties. When compared to the control mix, the compressive strength of the 20% PET replacement increased to 28.1 MPa after 28 days. Moreover, the concrete with 20% PET obtained the lowest water-absorption rate. The scanning electron microscopy images revealed that the inclusion of waste PET as sand had a significant effect on the microstructure of Mk-CBP geopolymer concrete. When compared to the control mix, the matrix containing 20% PET had a denser microstructure, as well as fewer holes and microcracks, in addition to the packing of paste at the interfacial transition zone.

KEYWORDS: Brick powder, Fine aggregate, Geopolymer concrete, Metakaolin, Polyethylene terephthalate (PET).

INTRODUCTION

Recently, global warming, environmental pollution and large-scale consumption of natural resources have become major crises threatening the world and life on earth (Wang et al., 2021). Having been associated with rapid population growth and urbanization, the vast amount of concrete produced as the key building material of most infrastructures and construction applications has caused significant environmental impacts across the globe (Jain et al., 2023; Rama and Shanthi, 2023). The main eco-drawbacks of concrete come from the energy intensive nature of manufacturing ordinary Portland cement (OPC), which is the main binder in concrete, the volume of raw materials consumed in the process and the contribution of 7-8% of

the overall worldwide carbon dioxide (CO₂) emissions (Ahmed, 2021). Thus, much effort has been spent to develop an alternative to conventional cement concrete. One such method is the use of geopolymer binder to produce sustainable concrete. Moreover, the mineral aggregate of gravel and sand makes up 70%-80% of the total concrete volume and global aggregate consumption is expected to reach 47 billion tons in 2023 (Ahmad et al., 2022). Therefore, it is urgent to alleviate this massive depletion of non-renewable resources and provide sustainable alternative materials as new aggregates in the production of concrete. Meanwhile, waste clay bricks are also an inevitable by-product of the construction industry as something usually generated during construction, renovation and demolition activities. In the same context, huge quantities of water-bottle waste made of polyethylene terephthalate plastic are generated every year. In 2021, more than 580 billion

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(PET) bottles were manufactured and consumed worldwide (Tamburini et al., 2021). Most of these bottles are non-biodegradable and will remain in the aquatic and terrestrial ecosystems for hundreds - if not thousands- of years. This will result in a global pollution crisis. (al-Hadithi & Alani, 2018). One of the most efficient solutions for overcoming these drawbacks involves reusing and recycling brick waste and PET plastic to partially replace the raw materials in concrete manufacture. Moreover, the chemical composition of waste clay brick powder contains sufficient levels of SiO_2 , Al_2O_3 and Fe_2O_3 . Thus, many researchers have encouraged the use of brick powder as a precursor in the synthesis of geopolymer concrete. Tuyen et al. (2018) investigated the effect of alkaline solution concentrations and curing conditions on the strength of geopolymer mortar based on waste clay brick powder and determined a maximum compressive strength of 36.2 MPa to be obtained from a 10% Na_2O -concentration cured at 90°C for 5 days. Meanwhile, Ahmed et al. (2022) replaced metakaolin with waste clay brick powder at weight levels of 10%, 15% and 20% to produce geopolymer concrete. The results revealed the inclusion of brick powder to decrease the static modulus of elasticity by up to 47% and the shrinkage from drying for the samples containing brick powder to be considerably lower than that of the reference mixture at day 28 and later.

A few attempts have been made to recycle waste PET in alkali-activated materials. Akçaözoglu and Ulu (2014) utilized waste PET aggregate in amounts of 20% up to 100% by volume for fine aggregate in alkali-activated mortar based on a blend of blast furnace slag and metakaolin. The authors found the strength values of the alkali-activated composite to decrease as the PET aggregate content increased. In the study of Shaikh (2020), waste PET was reused as a fiber at ratios of 1% and 1.5% by volume for the reinforced ambient curing of a geopolymer composite. The results showed the compressive strength of geopolymer composite with PET-fiber to be higher than that of the cement-based mixture for the same PET content, with all strength values decreasing as the PET-fiber content increased from 1% to 1.5%. Despite the published studies and increased attention on using waste PET in concrete, limited data is available regarding the inclusion of shredded PET as a fine aggregate in geopolymer

concrete based on blended metakaolin and waste brick powder.

Accordingly, this study aims to investigate the synergistic effect of utilizing waste PET aggregate as a fine-aggregate replacement, as well as clay brick powder as a sustainable precursor on the physical, mechanical and microstructural properties of metakaolin-based geopolymer concrete. The study addresses the compressive, flexural and splitting tensile strengths, the density of fresh and hardened states and scanning electronic microscopy (SEM) features. The results show that a highly sustainable (zero-cement) concrete can be produced for use in a wide range of civil-engineering applications.

Experimental Program

Materials

Metakaolin (MK)

This study utilizes Iraqi metakaolin (MK; see Figure 1) as a binding material for synthesized geopolymer concrete. MK was obtained by burning ground kaolinite clay extracted from the Anbar province in western Iraq at 700°C for 2 hours. Table 1 presents the chemical analysis and physical properties and shows the MK to be in compliance with ASTM C618-19 specification as a natural pozzolan.

Waste Clay Brick Powder (CBP)

The CBP is the waste material generated by grinding the hollow clay bricks collected from certain construction sites in the city of Baghdad. The residual pieces of the non-used yellow clay brick were first crushed into small particles and then ground using a cyclone extractor machine to obtain a powder form. The final state of the CBP is shown in Figure 1, with Table 1 illustrating its chemical composition and physical properties.

Alkali Activator Solution

The alkaline activator was a solution composed of sodium hydroxide (NaOH) and sodium silicate (Na_2SiO_3). Firstly, NaOH pellets (98% purity) were dissolved in distilled water to make a concentrated solution of 14M. Then, a heavy liquid of Na_2SiO_3 containing 13.5% Na_2O , 32.5% SiO_2 and 54% H_2O was

mixed with the NaOH solution at a 2:1 weight ratio. This process was exothermal and therefore, the final solution

was left for 24 hours to cool at room temperature before use.



Figure (1): Precursor (binder) materials

Table 1. Chemical analysis and physical properties of MK and CBP

Chemical Composition % by Weight	MK	CBP	ASTM C 618 Limits
SiO ₂	54.2	56.8	SiO ₂ + Al ₂ O ₃ + Fe ₂ O ₃ ≥ 70%
Al ₂ O ₃	39	11.5	
Fe ₂ O ₃	0.92	2.4	
CaO	1.37	20.2	/
MgO	0.15	3.02	/
SO ₃	0.45	0.83	≤ 4.0%
LOI*	0.71	1.19	≤ 10%
Physical Properties			
Specific surface area (kg/m ²)	14.300	462	/
Amount (%) retained on sieve no. 325 (45 μm)	18.2	32.0	≤ 34%
Pozzolanic-activity index on day 7 (%)	113	89.2	≥ 75%

* LOI = Loss on Ignition.

Natural Coarse Aggregate

Natural crushed gravel (max. diam. 14 mm) brought from the al-Nebai region (northern Baghdad) was used as coarse aggregate in all mixes. The gradation and physical properties for the coarse aggregate were satisfied per Iraqi standard IQ. S No. 45-1984 as presented in Figure 2 and Table 2, respectively.

Natural Fine Aggregate

Local natural sand up to 4.75 mm in diameter was extracted from Karbala province and employed as fine aggregate. The gradation curve for the sand lays within Zone 2 (see Figure 2) and its physical properties conform to Iraqi specification standard IQ. S No. 45-1984 (see Table 2).

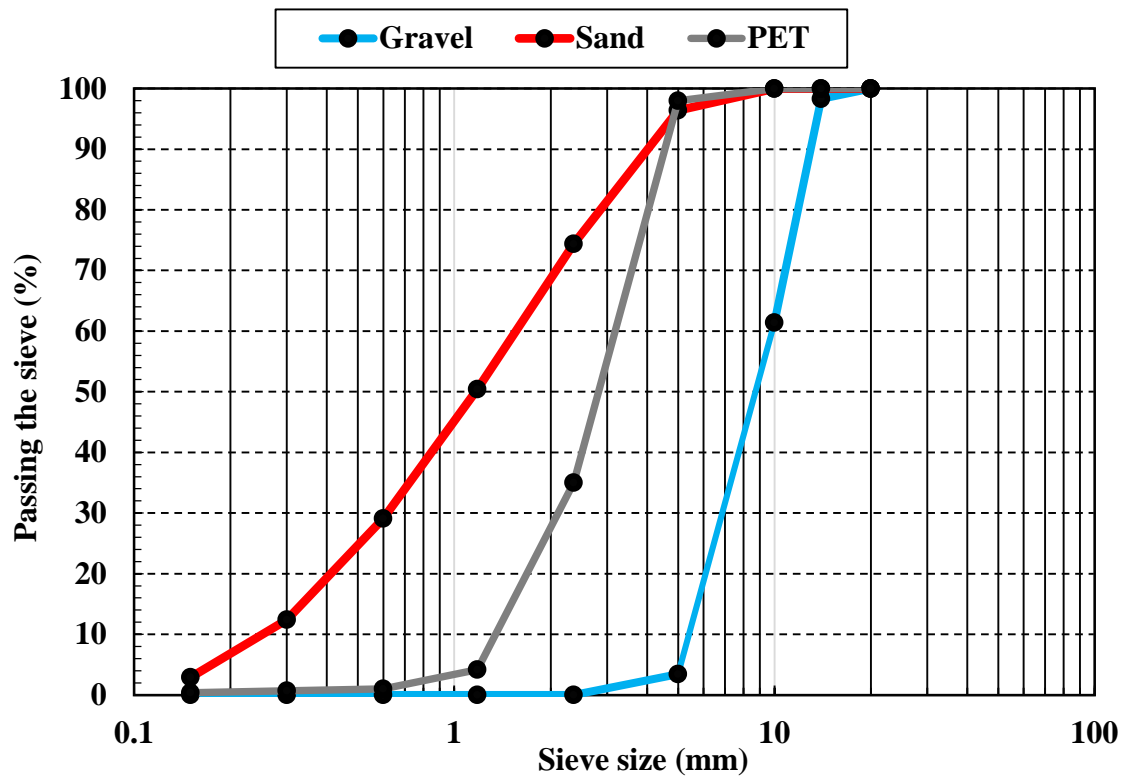


Figure (2): Gradation curve for natural aggregates and PET

Table 2. Properties of natural aggregates and PET

Property	Results		
	Gravel	Sand	PET-aggregate
Dry-rodded density (kg/m ³)	1,595	1,787	447
Specific gravity	2.60	2.58	1.28
Water absorption (%)	1.3	1.6	0.00
Sulfate content (%)	0.034	0.063	/



Figure (3): Shredded PET aggregate

Waste PET Aggregate

This study uses waste polyethylene terephthalate

particles (PET-aggregate) obtained by shredding PET bottles to replace a volume fraction of sand in geopolymer mixtures. The process of the PET-aggregate production is based on previous research (see al-Hadithi & Alani, 2018). The PET particles have an irregular flaky shape (see Figure 3) and the PET aggregate was passed through a 10-mm sieve and tested to determine the physical properties, as summarized in Table 2.

Water and Superplasticizer

Extra tap water and high-range water-reducing admixture (superplasticizer- type Conplast SP2000) in conformance to ASTM C 494-17 type G were added to all mixes to improve workability and enhance the mixing process.

Mixture Proportions

Four geopolymer concrete mixes were prepared by replacing the natural fine aggregate with PET-aggregate at levels of 0%, 10%, 15% and 20% by volume. The precursors consist of the MK and CBP at a 1:1 ratio by weight. The mix design of the reference mix was adopted from a previous study (Ahmed et al., 2019). For

all mixtures, the ratio of alkali solution to binder was fixed at 65%, while the dosages of SP and extra tap water were taken respectively at 2% and 5% of the binder weight. Table 3 illustrates the weights of the ingredients and the percentage of PET-aggregate for each mix.

Table 3. Mixture details and proportions (kg/m³)

Items	Concrete mixture ID			
	Ref.	M10PET	M15PET	M20PET
Replacement percent of PET	0%	10%	15%	20%
MK	207.5	207.5	207.5	207.5
CBP	207.5	207.5	207.5	207.5
Gravel	1240	1240	1240	1240
Sand	475	427.47	403.7	380
PET-aggregate	0	11.9	17.8	23.8
Alkaline liquid	270	270	270	270
SP	8.3	8.3	8.3	8.3

Preparing the MK-CBP Geopolymer Concrete Specimens

The current study adopted a mixing process for producing the geopolymer specimens similar to the one used by Ahmed et al. (2019). The natural coarse and fine aggregates were prepared for a saturation surface dry (SSD) condition by being immersed in water for 48 hours then left to lose their surface moisture. Before adding the ingredients in an electrical pan mixer, the PET-aggregate was mixed with the sand manually. The MK was also thoroughly blended with the CBP to obtain the precursor binder.

Meanwhile, the fresh geopolymer concrete's stickiness might cause it to adhere to the molds. Therefore, all molds were cleaned very well and polished with a thin layer of polymeric wax before casting. The fresh concrete was cast immediately in layers into the required test molds. Each layer was compacted manually by tamping (30 blows) with a steel rod, followed by 20-25 sec. on an electrical vibrating table. The top surface of the specimens was leveled by trowel and then covered with a polyethylene sheet to minimize plastic shrinkage. After 24 hours, the hardened specimens were removed from the molds, sealed with thick nylon bags and then placed in an electrical oven at 80°C for 24 hours as a curing regime. All specimens were left exposed to the air outside the laboratory in shade until test

time.

Testing

This study implemented several tests to assess the PET-aggregate's effect on the characteristics of the blended MK-CBP geopolymer concrete. For each mixture, the physical and mechanical properties were identified by casting and testing six cubed specimens with dimensions of 100×100×100 mm, six prismatic specimens of 100×100×400 mm, two prisms with the dimensions of 75×75×300 mm and six cylindrical samples of 100-mm diameter and 200-mm height. The details of the tests are shown in Table 4. The microstructure analysis was carried out using the scanning electron microscopy (SEM) model (TESCAN-VEGA III system) at the Department of Materials Development – Ministry of Sciences and Technology in Iraq. The study investigated the main features of the interfacial transition zone (ITZ) between waste PET-aggregate and geopolymer paste for each mixture. The SEM specimens have approximately dimensions of (10×10×10) mm. These specimens were obtained by cutting the samples of dynamic-modulus elasticity after testing them using a concrete-cutting machine. All samples were dried inside an electrical-oven dryer at 60°C for 6 days, after which one surface was polished and coated with gold as a pre-treatment for SEM imaging.

Table 4. Details on the experimental test methods

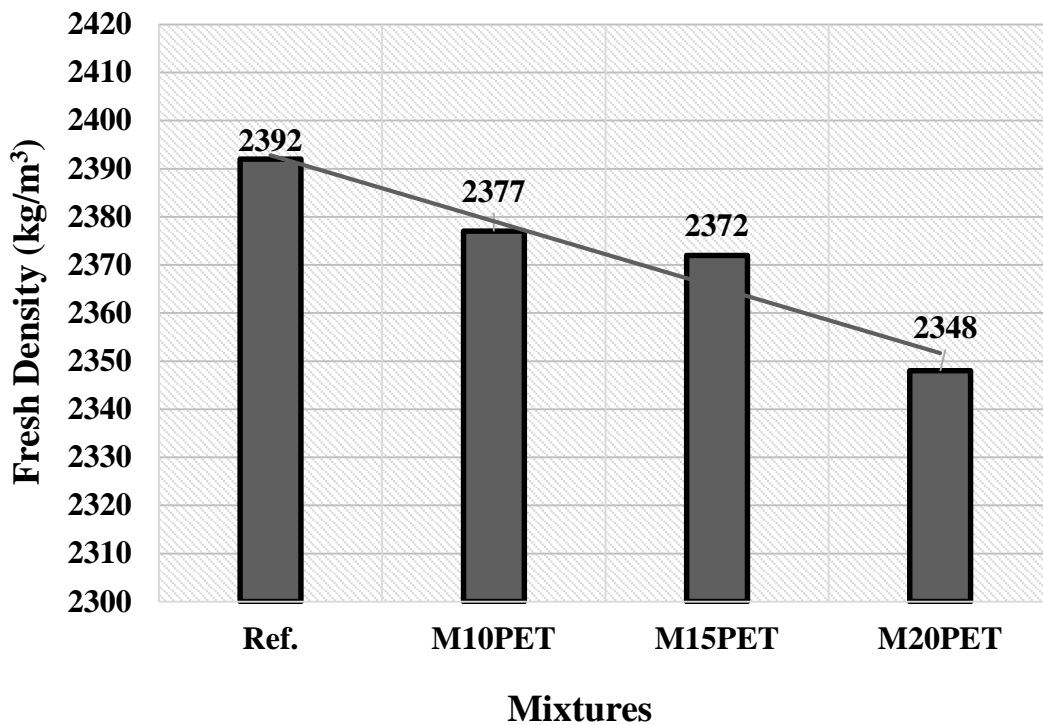
Targeted Properties	Standard	Specimens	Time of Test
Fresh density	ASTM C 138-2017	3 cubes	immediately after casting
Compressive strength	BS 1881:116 -2003	6 cubes	day 7 and day 28
Flexural strength	ASTM C 78-2018	6 prisms	day 7 and day 28
Splitting tensile strength	ASTM C 496-2017	6 cylinders	day 7 and day 28
Bulk density	ASTM C 642-2013	6 fractured pieces	day 7 and day 28
Water absorption			day 28

RESULTS AND DISCUSSION

Fresh Density

Figure 4 presents the fresh unit weights for the mixes with different percentages of PET aggregate. One can clearly note how the fresh density decreases as the PET-

aggregate ratio increases. The fresh density for the mixtures containing PET particles at 10%, 15% and 20% dropped respectively by 0.62%, 0.83% and 1.84% when compared with the control mix. This reduction in fresh weight is attributed to the lower density of PET particles compared to the density of natural sand.

**Figure (4): Fresh density of mixtures containing PET aggregate**

Dry Density

Figure 5 summarizes the hardened bulk densities for all mixes on day 7 and day 28. The results indicate the dry weight of concrete to decrease slightly for each test day as the ratio of PET-aggregate increases. Increasing the amount of PET particles in the mixture by 10%, 15% and 20% decreased the bulk density of the control mix on day 28 by 0.22%, 0.27% and 0.40%, respectively.

The reason for this slight decrement is attributed to the lower density of the PET-aggregate (four times) compared to the fine natural aggregate. This observation agrees well with other research on shredded PET particles being incorporated in concrete (Rahmani et al., 2013; al-Hadithi & Alani, 2018; Lee et al., 2019).

Meanwhile, the results show the bulk density of all mixes to decline over time, whereas the dry density of

the PET specimens at 0%, 10%, 15% and 20% volumes on day 28 reduced respectively by 1.72%, 1.55%, 1.24% and 0.67% compared to the values from day 7. This reduction in bulk density includes the voids between

particles and is related to the loss of free water within the composite, with a geopolymer gel developing with a lower unit weight.

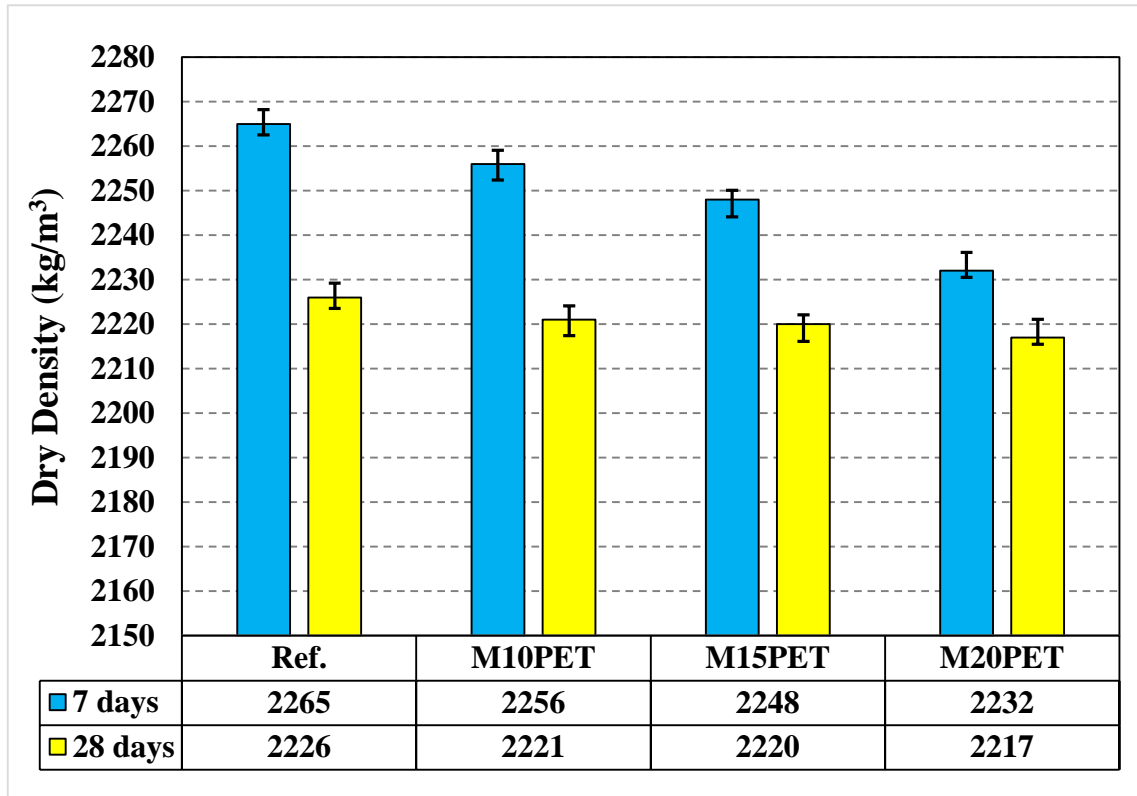


Figure (5): Dry density of mixtures containing the PET aggregate

Compressive Strength

Figure 6 presents the compressive-strength values of the mixes with the different percentages of PET-aggregate after 7 and 28 days. Regardless of the PET content, all mixes exhibited higher strength with aging due to the continuous geopolymerization and formation of a 3D gel network. The results shown in Figure 6 indicate the compressive strength on both day 7 and day 28 to have enhanced systematically with the increased PET ratios. On day 28, the use of 10%, 15% and 20% PET mixtures increased the compressive strength of the control mix by 7.2%, 5.2% and 35.1%, respectively. This positive trend may be ascribed to the chemical interaction between the alkaline solution and PET particles, resulting in the formation of stronger hydrogen bonds on the plastic surface. Thus, the smooth surface of PET will become rougher and lead to improved adhesion between the geopolymer paste and PET

aggregate.

Meanwhile, replacing the natural fine aggregate used in the SSD condition with the dry PET aggregate has led to minimizing the total volume of water in the geopolymer mixture. Thus, the adverse effect of free water on the geopolymerization process will be minimized. Consequently, the geopolymer matrix became denser with fewer defects, especially in the interfacial transition zone (ITZ). These findings agree with previous studies on conventional concrete containing PET aggregate. Lee et al. (2019) found replacing 20% of natural aggregate with PET particles treated with a calcium hypochlorite solution to increase compressive strength by 13.2%. Moreover, Dawood et al. (2021) revealed a 26.8%-43.64% increase in compressive strength for specimens containing 5%-12.5% PET-waste partial sand substitution.

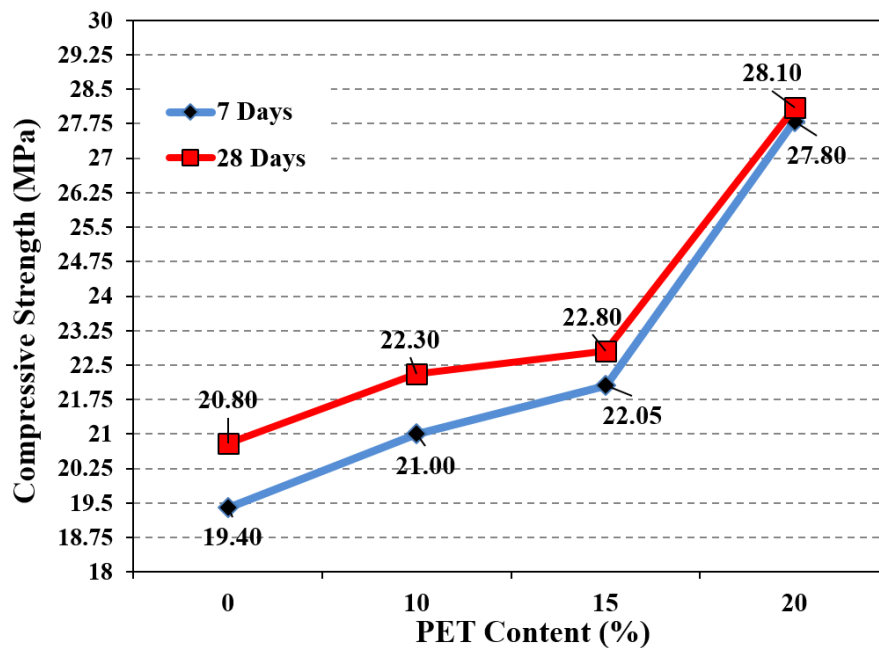


Figure (6): Compressive strength of MK-CBP geopolymer concrete with different PET-aggregate contents

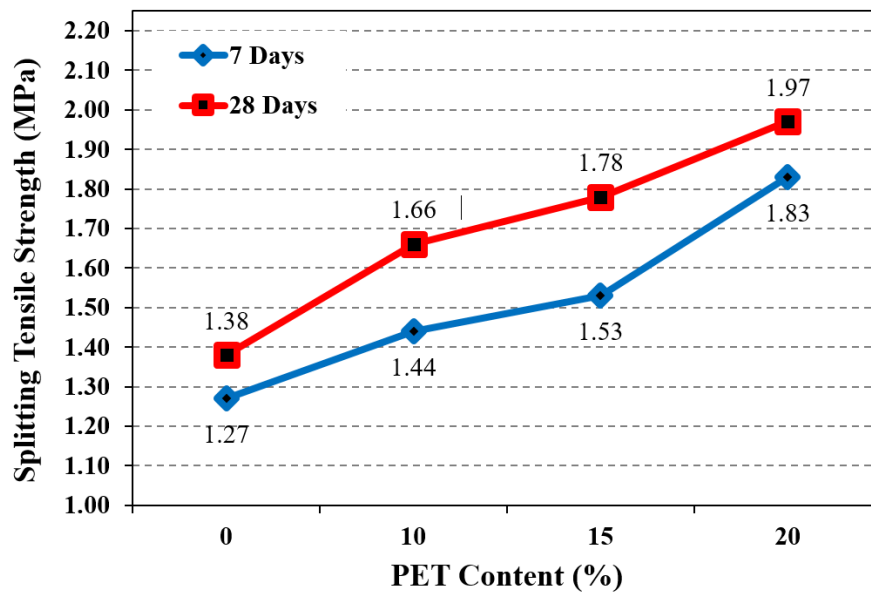


Figure (7): Splitting tensile strength of MK-CBP geopolymer concrete with different PET-aggregate contents

Splitting Tensile Strength

Figure 7 shows the relationship between splitting strength and PET-aggregate content on day 7 and day 28. All geopolymer concrete mixtures exhibited slightly increased splitting strength over time as a result of geopolymerization development. The splitting strengths

of the geopolymer mixes at 10%, 15% and 20% PET-aggregate on day 28 can clearly and respectively be observed to be 20.28%, 28.98% and 42.75% stronger than the control mix. Despite the concrete strength being affected directly by the characteristics of the aggregate, especially hardness, shape and surface texture, the use

of smooth flaky particles of PET as a fine aggregate improves the tensile resistance of the MK-BP geopolymer concrete. This improvement can be related to the positive effect from increasing the reactions in the PET-aggregate surface area compared to those in natural sand, with bond strength consequently being augmented. According to al-Hadithi and Alani (2018), a utilization of 2.5% PET waste as fine aggregate increases the splitting strength by 6% for high-performance concrete specimens. In addition, Dawood et al. (2021) concluded a 26.9% increment to occur in splitting strength when using PET particles that make up 7.5% of the fine aggregate.

Flexural Strength

For all MK-CBP geopolymer mixes, indirect tensile strength was measured using the flexural test over hardened concrete prisms of 100×100×400 mm on day 7 and day 28. As shown in Figure 8, the flexural strength of all specimens ranged from 2.25 to 3.6 MPa over the

28 days of curing. The development and improvement in flexural strength over time resemble the results obtained from the compressive- and splitting-strength tests. As noted in Figure 8, increasing the PET-aggregate percentage resulted in increased flexural strength on both day 7 and day 28. For example, on day 28, the 10%, 15% and 20% PET-aggregate substitutions presented respective increases of 21.57%, 33.33% and 41.17% in flexural strength. This increase in flexural strength is attributed to the dense packing of PET-aggregate and geopolymer gel, especially at the transition zone. Meanwhile, the semi-fibrous form and size of the PET particles can act as fibers within a geopolymer matrix and restrict prolonged cracks at the micro-level and macro-level in composite concrete. This phenomenon matches previous observations from research on the enhanced flexural-strength results for ordinary concrete containing PET as a fine aggregate (Rahmani et al., 2013).

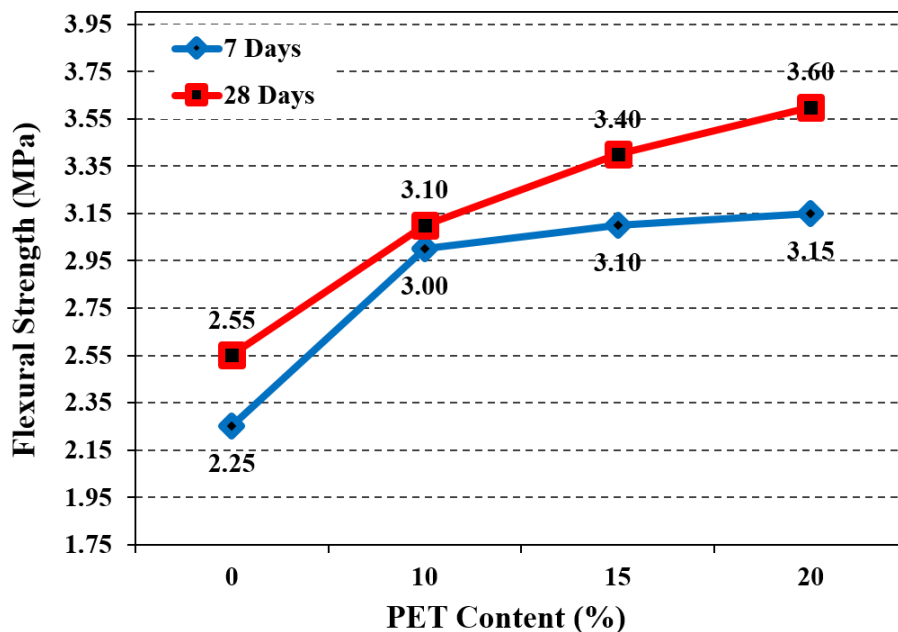


Figure (8): Flexural strength of MK-CBP geopolymer concrete with different PET-aggregate contents

Water Absorption

Water absorption plays an important role in the durability and service life of concrete. Figure 9 shows the water absorption of geopolymer concrete mixes containing various percentages of shredded PET

particles as fine-aggregate replacement over 28 days. The control specimen (0% PET) confirmed a geopolymer concrete with acceptable water absorption at 4.46%. However, the results shown in Fig. 9 reveal incorporating PET-aggregate of 10%, 15% and 20% in

volume to lead to respective decreases of 0.89%, 4.93% and 10.09% in water absorption compared to the reference mixture. The reason for this reduction may involve the chemical interaction between the PET surface and alkali solution that forms a dense compacted cement paste. Thus, less voids and micro-cracks are produced within the geopolymer matrix, especially in

the interfacial transition zone as mentioned previously. In addition, the hydrophobic, non-porous property and flaky-shape of the PET particles restrict water movement into the permeable voids. al-Hadithi and Alani (2018) previously observed including 7.5% PET as a fine-aggregate replacement to decrease water absorption by 14% in high-performance concrete.

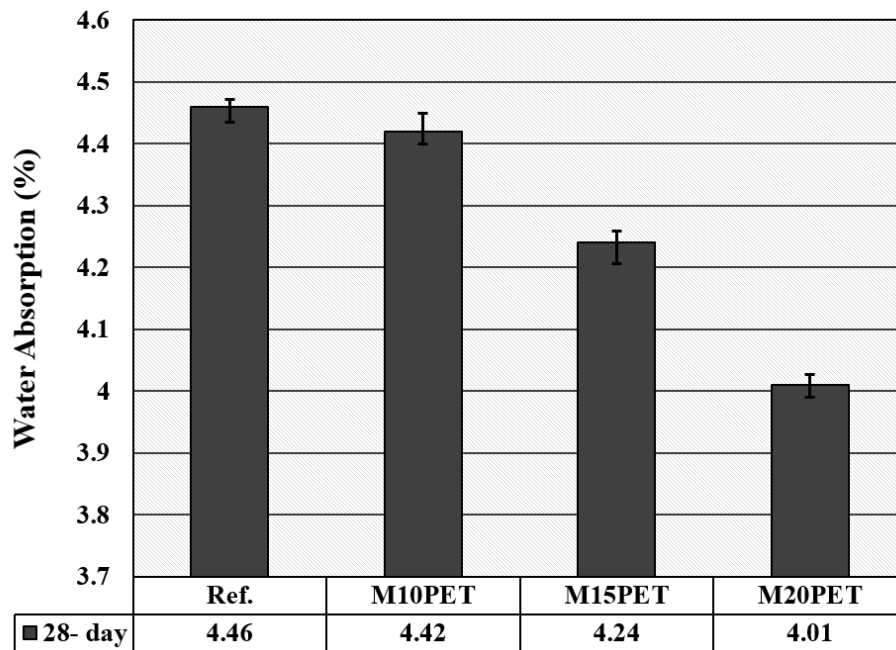


Figure (9): Water absorption of MK-CBP geopolymer concrete mixtures

Scanning Electron Microscopy (SEM) Analysis

Figures 10 and 11 respectively illustrate the SEM figures for the reference mix and M20PET geopolymer concrete mix on day 28. The micrographs of the control mix in Figures 10a-10c show a heterogeneous and porous paste with some unreacted or partially reacted waste brick powder coinciding with the reacted metakaolin and alluding to its strong bond. Furthermore, Figure 10-b shows how the matrix of the reference mix has wide microcracks resulting from drying shrinkage, as well as a significant amount of dendritic shapes produced from the activated geopolymer gel. However, the abundance of CaO (20.2%) in the chemical composition of the waste brick powder will produce more needle-shaped crystalline calcium hydroxide (CaOH) during geopolymerization.

A close look at the micrographs for the 20PET mix in Figures 11a-c show improvements in the microstructure's features by developing a dense gel with

less pores (air voids) and small cracks, especially along the interfacial transition zone (ITZ). Accordingly, the homogeneity of the geopolymer matrix with PET aggregate will be enhanced compared to the reference mix. Figure 11-b clearly shows an intensive geopolymer paste to have bonded strongly with the PET aggregate; therefore, no gaps appear between them along the ITZ. Furthermore, the microstructure image in Figure 11c demonstrates considerable agglomerated geopolymer gel to have set on the PET surface, with strong adhesion between the geopolymer paste and the PET particles. This implies a chemical species to have formed between the surface of the PET aggregate and the alkaline liquid. Hence, a unique composite of geopolymer gel and PET will form within the concrete matrix and fill in all defects (e.g. microcracks, pores) along the ITZ. This may be the reason behind the improvements in the mechanical properties of the mixes containing PET waste.

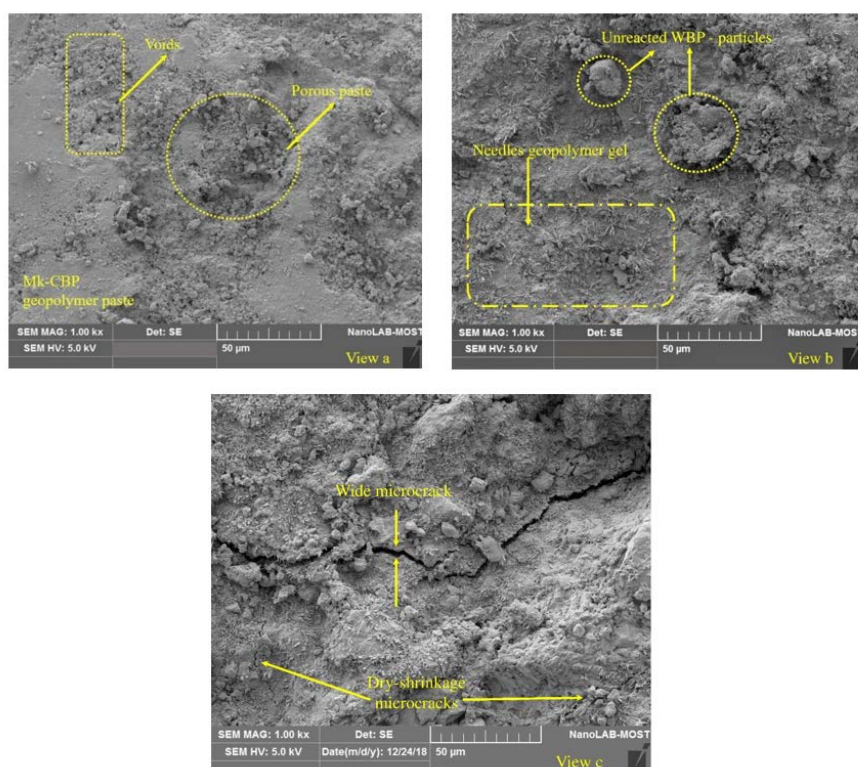


Figure (10): (views a, b and c): SEM micrographs of the reference Mk-CBP geopolymer mixture on day 28

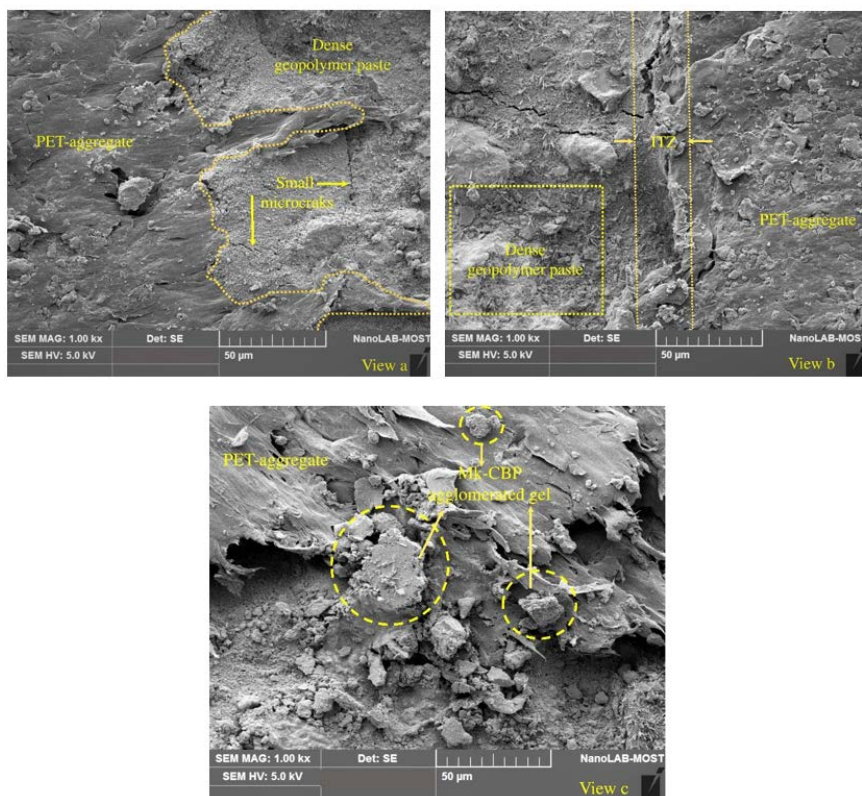


Figure (11): (views a, b and c): SEM micrographs of the 20% PET mixture on day 28

CONCLUSIONS

This study evaluated the effects of incorporating waste PET particles as fine aggregate on various physical, mechanical and microstructure properties of blended MK-CBP-based geopolymer concrete. According to the experimental results, one can draw the following main conclusions:

- The unit weight of MK-CBP geopolymer concrete in its fresh and hardened states reduces slightly when PET particles are used to replace sand. The use of a 20%-PET aggregate will provide the lowest fresh and dry density of concrete to reach 2,348 and 2,217 kg/m³, respectively.
- Including shredded PET waste as fine aggregate strengthens the composite of MK-CBP geopolymer concrete. The compressive strength increases as the PET volume increases, with the 20%-PET mixture gaining a compressive strength of up to 28.1 MPa compared to 20.8 MPa for the reference mix (0% PET).
- The cylinders of the PET-aggregate mixtures present higher splitting strengths compared to the control mixture. The highest tensile strength is 1.97 MPa and occurs in the 20%-PET fine aggregate mixture on day 28.
- The flexural strength of MK-CBP-geopolymer concrete is significantly affected by including PET particles in place of sand, with increases up to

21.57%, 33.33% and 41.17% observed on day 28 respectively for the 10%, 15% and 20% PET mixes.

- The use of shredded PET as a fine aggregate at the three replacement dosages (10%, 15% and 20%) led to a decrease in water absorption for the MK-CBP geopolymer concrete, with the lowest water absorption rate being 4.01% for the 20%-PET mix.
- The microstructure images show non-homogeneous and porous features associated with multi-cracks within the matrix of the reference MK-CBP geopolymer mix.
- The microscopic results substantiate the presence of chemical interactions between the alkaline solution and PET aggregate, accompanied by fewer pores and cracks for the 20%-PET aggregate mixture and more bonding between the geopolymer paste and PET surface being noticed along the ITZ.

In general, substituting natural fine aggregate with waste PET aggregate has been demonstrated to be able to positively affect the properties of blended MK-CBP geopolymer concrete. The incorporation of PET aggregate serves in waste-management development and the preservation of non-renewable resources. Further studies are recommended to examine the effect of high-volume content of PET aggregate (up to 20%) on the geopolymer concrete properties. Additionally, the long-term durability performance and time-dependent properties of blended MK-CBP-based geopolymer concrete should be investigated.

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