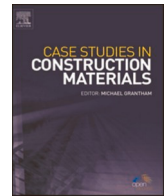




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## Case study

## Strength and microstructural properties of binary and ternary blends in fly ash-based geopolymer concrete

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## ABSTRACT

Carbon dioxide emissions and the consumption of natural resources related to the cement manufacturing have prompted the need to develop more sustainable and environmentally friendly types of concrete. Geopolymer concrete is considered eco-friendly concrete because it is free of cement. Otherwise, nanomaterials have been introduced into geopolymer concrete in previous works with the aim of improving its properties. However, very restricted studies have focused on the combined utilizing of nano-clay and nano-titanium in geopolymer concrete. Therefore, in the current research, geopolymer concrete was developed from industrial wastes (fly ash; FA) by using a novel mixture of different nanomaterials: nano-clay (NC) and nano-TiO<sub>2</sub> (NT). Mixtures with constant water to FA (12 %), and different alkaline contents: (40 %, 45 %, and 50 %) by FA, were performed and divided into three groups. In the first group, only FA was used as a binder, meanwhile, a combination of (FA+NC) and (FA+NT) was used separately in group two (binary). In the third group, (FA+NC+NT) were mixed together (ternary). Several hardened tests have been investigated: compressive, tensile strengths and density. Also, microstructural characteristics were monitored using XRD and SEM tests. The findings revealed that the addition of nanomaterials obviously enhanced the density of the microstructure, reducing the pores of the produced geopolymer concrete. Moreover, the compressive strength was enhanced up to 38 % for NC, and 24 % for NT in the binary blends while the improvement reached 55 % in the ternary blends.

## 1. Introduction

Geopolymer is a type of inorganic polymer created by chemical reactions between aluminosilicate cementitious powder and alkaline activators. Geopolymer combines the properties of both: polymer and cement [1]. The fly ash and metakaolin are commonly representing source of aluminosilicate in geopolymers [2–4]. Fly ash is an industrial waste resulting from burning coal in electric power stations [5]. The use of fly ash in geopolymer concrete can reduce the carbon dioxide emissions by a percentage within the range 25–45 % due to the use of coal combustion waste [6,7]. In other words, the use of such waste in concrete contributes to reducing its

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harmful impact on the environment [8,9]. The utilization of fly ash in geopolymer concrete has many benefits, including improving workability, reducing dryness shrinkage, increasing resistance to chemical attack and corrosion of steel reinforcement, in addition to reducing the heat of hydration [10,11].

There are many uses for geopolymer, such as airport floor, railway sleepers and it may be utilized in rigid pavement repairs [12]. Quite recently, considerable attention has been paid to the inclusion of nanomaterials to geopolymers. Selection of nanomaterials has been recognized as an important strategy to obtain novel properties and maintain sustainability [13–16]. Among the nanomaterials that have been incorporated in the manufacture of geopolymer concrete are nano-clay (NC) and nano-titanium dioxide, TiO<sub>2</sub>, (NT).

Nano clays are silicate particles with a nano-size scale that have nanopores. It can be classified into four fundamental collections: the montmorillonite/smectite, the kaolinite, the chlorite, and the illite groups [17]. Nano clays had been used in geopolymers in many previous studies to examine their impact on different characteristics of geopolymers. For instance, Khater [18] work revealed that incorporating NC in alkali-activated geopolymer by 1 % led to improve the compressive strength up to 90 days. Beyond that ratio, the mechanical strength was decreased.

On the other hand, Assaedi et al. [19] researched the influence of nano-clay (NC) on mechanical, thermal and microstructural characteristics of fly ash geopolymer concrete. They used NC in proportions of 1 %, 2 % and 3 % by weight. Results indicated that NC enhanced the compressive and flexural strength, hardness and flexural modulus of geopolymer concrete with an optimum additional ratio of 2 %. Moreover, NC reduced the porosity of the microstructure. Furthermore, they found that the NC acted as an activator for geopolymer reaction as well as a filler. The same findings were also recorded by Assaedi et al. [20] for low calcium fly geopolymer composites made with nano clay and flax fabric reinforcement. Moreover, Ravitheja and Kumar [21] investigated the impact of adding NC on geopolymer concrete made from fly ash. Results indicated that the presence of NC in the proportion of 6 % led to improve the compressive, splitting tensile and flexural strengths by 25.09, 29.02 % and 36.80 % respectively compared to the reference sample.

Otherwise, nano-TiO<sub>2</sub> is a successful material having a few of a kind of benefits, like photocatalytic characteristics [22]. Yang et al. 2015 [23] investigated the influence of 0.5 % NT on characteristics of geopolymer pastes. Results showed that the compressive strength values were improved by 10 %, 15 % and 9 % correlated to the reference mixture at 3, 7 and 28 days, respectively. Microstructure findings had proved that NT addition accelerated the reaction of alkali-activated slag paste and densified the structure. Moreover, the porosity was reduced and the pore structure was changed.

Duan et al. [24] investigated the impact of nano titanium dioxide (NT) on the properties of fly ash geopolymer pastes. NT additions were 1 %, 3 % and 5 % by mass. Results were in comparison to reference mix showed that the higher the NT content the higher is the early and later age compressive strength of geopolymer. The strength improvement was more obvious at 5 % of NT at 28 days. Moreover, the microstructure of geopolymer-based NT is becoming less cracked and more compacted. Hemalatha and Ramjee [25] researched the influence of using NT (in proportions of 2 %, 4 % and 6 %) with and without wollastonite on self-compacting geopolymer concrete. The fresh and mechanical properties were observed. Results indicated that the optimum percentage of NT was 4 % which improved the fresh, compressive and splitting tensile strengths of geopolymer concrete in the presence or absence of wollastonite.

The microstructure and micromechanical characteristics of fly ash geopolymer concrete with 2 % NT and nano SiO<sub>2</sub> (NS) were studied by Luo et al. [26]. Results indicated that the micromechanical and compressive strength properties were enhanced after the addition of both nanomaterials. NT was a less reinforcing effect than that for NS, however, micromechanical properties were enhanced importantly in the presence of NT. The strength of geopolymer, at 28 days, was increased by 10.49 % for NT and 17.38 % for NS compared to the reference sample. It was also found that the nanoparticles are integrated with the gel to make a compound of better strength. Moreover, Safiuddin et al. [27] stated that the application of nanoparticles in concrete enhances the performance of concrete and can be considered cost-effective on the long term as a result of reducing the cost of the rehabilitation and maintenance, despite their high (initial) cost.

Based on the reviewed literature, the combined effect of nano-clay and nano-TiO<sub>2</sub> particles on geopolymer has been limited described in the published literature. For example, according to Luo et al. [26] research, NS and NT were used in the geopolymer separately (binary blend). However, in this study, NC (not NS) and NT were used separately (binary blend) and combined (ternary blend) with the fly ash. Additionally, according to the authors' knowledge, there is very little research that came up with blending fly ash (FA) with nano clay or nano titanium as suggested in the current study. Moreover, Jindal and Sharma [28] reported that, in geopolymer products, a sufficient amount of literature dealt with the addition of nano-titanium dioxide is still not available. Further, in this study, a new technique that improves strength is suggested using a novel combination of binary and ternary blends of

**Table 1**  
Chemical composition of fly ash (FA) and nano-clay (NC).

Component, %	FA	NC	ASTM C-618 limits [29]
SiO <sub>2</sub>	49.6	54.2	≥ 70
Al <sub>2</sub> O <sub>3</sub>	45.8	36.4	
Fe <sub>2</sub> O <sub>3</sub>	4.53	1.92	
CaO	/	1.74	/
MgO	/	1.68	/
SO <sub>3</sub>	0.033	< 0.07	≤ 4
Loss of Ignition	/	2.07	≤ 10
Insoluble Residue	/	0.95	/
Fineness	20	= 60 nm	≤ 35

(FA+NC+TiO<sub>2</sub>) with a curing method of a low-pressure steam curing system. So, the outcomes for this research will be valuable for better performance of the resulted geopolymer concrete on one line with industrial wastes consumption.

## 2. Materials and methods

### 2.1. Materials characteristics

The raw materials included: fly ash powder, fine aggregate, coarse aggregate, tap water, superplasticizer and alkaline activator (Sodium Silicate + Sodium Hydroxide). The chemical analysis of FA and NC is shown in Table 1. It is compliant with ASTM C-618 [29] Class F and N requirements. The used nano clay was brought from (Nanocor® Inc., USA). This montmorillonite clay was calcined at 750 °C for 2 h following [30–32]. The superplasticizer, (known as Master RHEOBULD SP1), was imported from Sika company in Egypt according to ASTM C 494 Type F [33]. The fine and coarse aggregates are conformed to ASTM C 33 specification [34]. The grading of the fine and coarse aggregates is presented in Tables 2 and 3. The nano TiO<sub>2</sub> was provided by Nanoshel Company (USA), and Table 4 shows its characteristics. Sodium silicate (Na<sub>2</sub>SiO<sub>3</sub>) produced in the U.A.E. was utilized to make the alkalis liquid. Table 5 illustrates the properties of the Na<sub>2</sub>SiO<sub>3</sub>. Sodium hydroxide (NaOH) (Table 6), of 99 % purity in flake shape was utilized. Dissolving solid particles and flakes in distilled water is the first thing that should happen in order to prepare an activator with the specified molarity (10 M). There are three common methods for dispersion of nano materials: mechanical agitation, ultrasonic vibration and chemical dispersion following the procedure outlined by Yu et al. [35]. Different ratios of Na<sub>2</sub>SiO<sub>3</sub>/NaOH were used to examine the effectiveness of alkalis activators on the geopolymers. Extra potable water used in the mix design of geopolymer is free from organic substances and suspended particles. Extra water, as 10 % by weight of FA, is used for enhancing the workability and homogeneity of the geopolymer end product.

### 2.2. Experimental work

Two kinds of nanomaterials were utilized; Titanium dioxide and calcined nano-clay. According to review, alkaline doses and two systems of nano blends (binary and ternary) were assessed. The used mix proportion which is presented in Table 7 and the mixing steps of the geopolymer concrete mixes are following the procedure outlined by Ali et al. [36]. Several experiments were carried out to verify the validity of using binary and ternary blends in geopolymer concrete. An optimization process has been carried out to specify an optimal percentage for both nanomaterials. After 28 d, tests of compressive strength, density and splitting tensile strength were examined. 10 cm cubes were chosen to perform the compressive strength (see Fig. 1) and bulk density tests following BS 1881: Part 116 [37] and BS 1881: Part 114 [38] specifications, respectively. While (10 \* 20) cm cylindrical specimens were utilized to test the splitting tensile strength following to BS 1881: Part 117 [39] standard (see Fig. 1). After molding is completed, an autoclave was applied for 3 h according to the ASTM C 151 [40] specification. After the autoclave curing (for 3 h), the samples were kept in mold, wrapped by a polythene cover for about 21 h to prevent moisture loss (the evaporation of the extra water), then lifted from the molds. ELE apparatus (as it is shown in Plate 1) were used associated with a bursting pressure of 295 ± 10 psi (2 ± 0.07 MPa), which meets to a temperature of 421 ± 2°F (215 ± 3 °C) for 3 h. This process will ensure the speed up of pozzolanic reactions. Additionally, X-ray diffraction (XRD) and SEM microscope tests were performed using samples taken from 3 various composites. Thereafter NaOH has been prepared, it should be mixed together with Na<sub>2</sub>SiO<sub>3</sub> to make the alkaline solution at most one day before utilizing.

## 3. Results and discussion

### 3.1. Hardened properties

First of all, in the preliminary stage of this research, a lot of trial mixes were performed in order to choose the optimal doses of NC and nano TiO<sub>2</sub> that maximize the strength of the resulting concrete. So, different addition percentages were assessed in this stage based on the data presented in the introduction. For this reason, SigmaXL Version 6.1 program was used depending on the analysis of response surface method (RSM), using the design of experiments (DOE) principles. The following quadratic model is used:

$$y = b_0 + b_1X_1 + b_2X_2 + b_{12}X_1X_2 - b_{11}X_1^2 + b_{22}X_2^2 \quad (1)$$

**Table 2**  
Grading of fine aggregates.

Sieve size (mm)	Cumulative percentage passing	Limits of ASTM C33
9.50	100	100
4.75	90	95–100
2.36	85	80–100
1.18	55	50–85
0.60	26	25–60
0.30	14	5–30
0.15	6	0–10

**Table 3**  
Grading of coarse aggregate.

Sieve size, mm	Cumulative percentage passing	Limits of ASTM C33 4.75–19 mm
25	100	100
19	95	90–100
12.5	–	–
9.5	45	20–55
4.75	7	0–10
2.36	3	0–5

**Table 4**  
The properties of Titanium dioxide according to manufacturer.

Property	Titanium dioxide
Chemical composition	TiO <sub>2</sub>
Purity (%)	99
particle size (nm)	49
Specific surface area (m <sup>2</sup> /gm)	37
Color	Off white
Phase	Rutile
Density (g/cm <sup>3</sup> )	4.1

**Table 5**  
Properties of sodium silicate.

Description	Value
SiO <sub>2</sub> / Na <sub>2</sub> O	2.45
Na <sub>2</sub> O % by weight	13.30
SiO <sub>2</sub> % by weight	32.55
Viscosity (CPS) 20 °C	750
Specific Gravity	1.540
Density - 20 °C	51
Appearance	Hazy

**Table 6**  
Properties sodium hydroxide.

Appearance	Unit	Specification ASTM E291-09	Results
Sodium hydroxide (NaOH)	%	≥ 97.5	98
Sodium carbonate (Na <sub>2</sub> CO <sub>3</sub> )	%	≤ 0.40	0.25
Sodium chloride (NaCl)	%	≤ 0.15	0.09
Iron oxides (Fe <sub>2</sub> O <sub>3</sub> )	%	≤ 0.01	0.006
Sulfate as Na <sub>2</sub> SO <sub>4</sub>	Ppm	≤ 200	90
Copper as Cu <sup>+2</sup>	Ppm	≤ 4.0	0.5
Nickel as Ni <sup>+2</sup>	Ppm	≤ 5.0	2.5
Manganese as Mn	Ppm	≤ 4.0	0.07
Silicate as SiO <sub>2</sub>	Ppm	≤ 20	12
Water Insoluble	Ppm	≤ 200	74

where:

y = Dependent variable,

X<sub>1</sub>, X<sub>2</sub>, X<sub>3</sub>... X<sub>n</sub> =Independent variables,

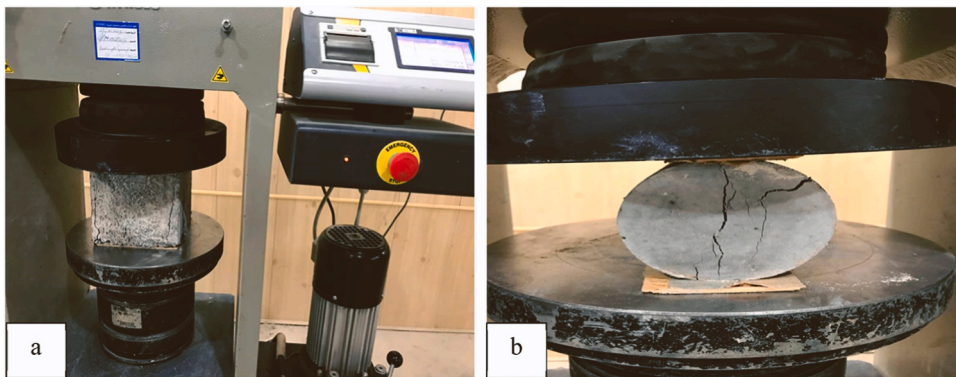
b<sub>0</sub>, b<sub>1</sub>, b<sub>2</sub>, b<sub>3</sub>...b<sub>ij</sub>= vector of main effect coefficients.

The variables are different nano-clay and nano-titanium dioxide percentages (0.25 %, 0.5 %, 0.75 %, 1.0 %, 1.25 % and 1.5 %) that will be added separately to geopolymer concrete using different alkaline doses to investigate composite compressive strength. After that in stage two, the optimal doses were used to make all binary and ternary blends. A similar procedure has been adopted by previous researchers as well [41,42]. Fig. 2 presents the maximum compressive response surface graphs that show the trends in effects of alkaline doses of (40 %, 45 % and 50 %), and nanoparticles levels (0, 0.25 %, 0.5 %, 0.75 %, 1.0 %, 1.25 % and 1.5 %) by weight of FA. Each response surface plot is a particular response as a function of two variables (alkaline and nanoparticles). Each black-shaded area in plots corresponds to the optimum condition of manufacturing parameters of geopolymer under autoclave curing. This optimum

**Table 7**  
Mix proportions.

Group No.	Mix <sup>a</sup> No.	Fly ash (kg/m <sup>3</sup> )	Fine aggregate (kg/m <sup>3</sup> )	Super-plasticizer (kg/m <sup>3</sup> )	Coarse aggregate (kg/m <sup>3</sup> )	Na <sub>2</sub> SiO <sub>3</sub> / NaOH (%)	NC (%)	TiO <sub>2</sub> (%)
1	MC	400	720	12	1100	40	0	0
	MBC						1	0
	MBT						0	1.25
	MR						1	1.25
2	MC	400	720	12	1100	45	0	0
	MBC						1	0
	MBT						0	1.25
	MR						1	1.25
3	MC	400	720	12	1100	50	0	0
	MBC						1	0
	MBT						0	1.25
	MR						1	1.25

<sup>a</sup> MC: refers to the control mix (without nano powders); MBC: refers to the mix of binary blends with nano clay; MBT: refers to the mix of binary blends with nano titanium; MR: refers to the mix of ternary blends with nano clay and nano titanium.



**Fig. 1.** Experimental tests: (a) compressive strength and (b) splitting tensile strength.



**Plate 1.** ELE apparatus for autoclave curing.

percentage enhanced the studied features, whereas improper addition may result in agglomeration and adversely influenced the geopolymer. The R-Square value is given as 92 %. This is very desirable for a Designed Experiment. The plots reveal a quadratic relation (initially increased thereafter declined and then increased again for all investigated parameters). Therefore; the optimum level of the manufacturing parameters derived from the response surface method for all investigated samples are as follows: nano clay percentage equal to 1.0 %; and nano TiO<sub>2</sub> equal to 1.25 % added by weight of FA.

Generally, the addition of nanoparticles to the concrete maximizes the surface area of powders and therefore, workability loss occur. Nevertheless, all mixtures having nano powders shown slightly lower workability in comparison with reference mix. Fig. 3 shows the compressive strengths for all the examined specimens after 28 d. As shown, all binary and ternary samples exhibited higher

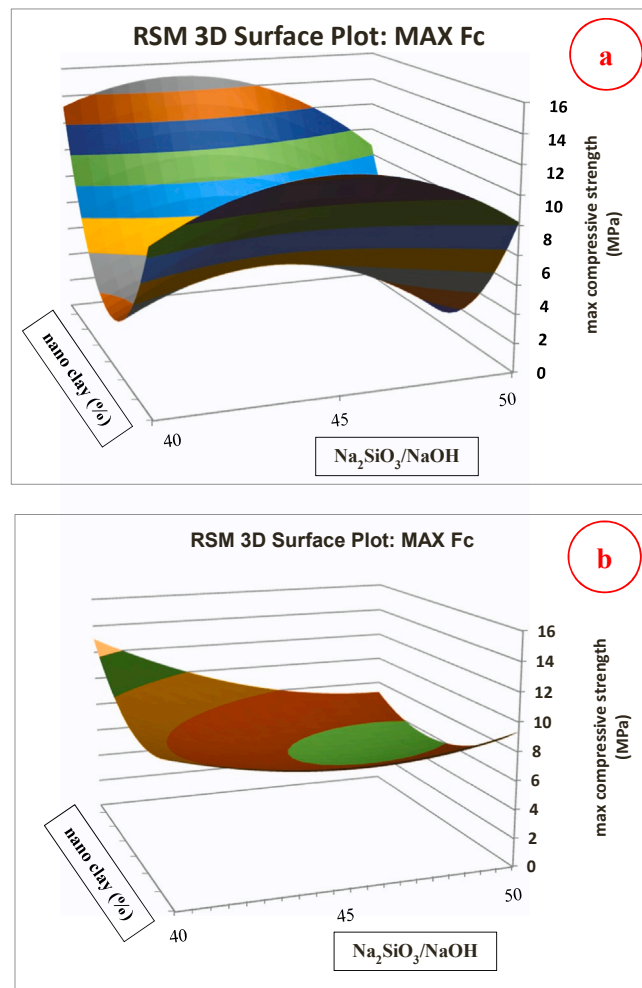


Fig. 2. Maximum compressive response surfaces plots for samples with different alkaline and nanoparticles doses using: (a) NC and (b) TiO<sub>2</sub>.

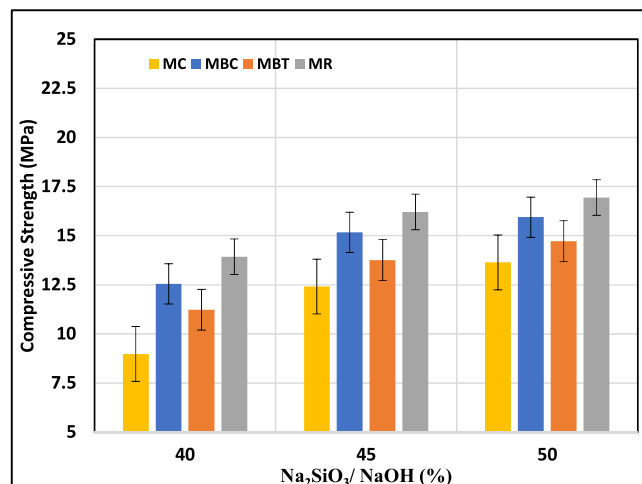


Fig. 3. Effect of nano TiO<sub>2</sub> and nano clay on the compressive strength at different percentages of alkaline.

compressive strengths in comparison with reference mix (MC). Besides; binary blend results demonstrated that the addition of nano materials was remarkably affected through increasing the compressive strength by (38 %, 24 % and 20 %) for mix of binary blends with nano clay (MBC), and by (24 %, 12 % and 9 %) for mix of binary blends with nano titanium (MBT) at alkaline doses of (40 %, 45 % and 50 %) respectively. Furthermore, in the ternary system, it is clear that the addition of two types of cementitious materials in the nano-scale could be helpful in geopolymer performance. As compared to MC, results of mix with ternary blends (MR) demonstrated that the addition of nano materials was remarkably affected through increasing the compressive strength by (55 %, 30 % and 24 %) at alkaline doses of (40 %, 45 % and 50 %). These improvements may be due to the pozzolanic action which consuming calcium hydroxide and making of more stable hydrated calcium silicate that fills the black holes and minimize the interfacial transition zone width [21], as well as a result of the filling ability of nanoparticles that minimize the size and sum of pores as proved later in the SEM micrographs. Nonetheless, NT shows lower compressive increments in comparison with NC. This may occur according to the higher pozzolanic reactivity for NC with calcium hydroxide. Similar observation was found by Mohan et al. [43].

Moreover, the compressive strength for all the investigated geopolymer mixes enhanced as the dosage of alkaline solution increased. This enhancement at high alkalis resulted in greater reaction with FA, leading to a denser microstructure. Further, the compressive strength was raised by 64 %, 78 % and 89 % for MBC, MBT and MR respectively in the case 50 % of  $\text{Na}_2\text{SiO}_3/\text{NaOH}$ , in comparison with reference mix.

The splitting tensile strength results of geopolymer concrete are depicted in Fig. 4. The splitting behavior is comparable to that in compression, but with a lower rate of increase. Moreover; all binary blend mixes incorporating nanoparticles yielded better splitting strength in comparison with reference mix (MC) by (36 %, 28 % and 20 %) for MBC, and by (29 %, 17 % and 18 %) for MBT at alkaline doses of (40 %, 45 % and 50 %) respectively. Moreover, in comparison with reference mix (MC), results of ternary blends MR demonstrated that the addition of nano materials was remarkably affected through increasing the splitting tensile strength by (50 %, 34 % and 33 %) at alkaline doses of (40 %, 45 % and 50 %) respectively. According to these results, higher strength improvements were gained in mix MR (ternary) as compared to mix MC, MBC and MBT respectively. The pozzolana reaction of nanoparticles may importantly enhance the microstructure and improving the studied properties of resulting geopolymer. This pozzolanic reactivity is related to the chemical composition and in particular, its high content of silicon dioxide ( $\text{SiO}_2$ ), which can improve C-S-H gel in concrete and affect the hardening properties of concrete. However, the results are agreeable with those obtained by Kumar et al. [44].

Fig. 5 shows the effects of the binary and ternary blends on the bulk density of blended geopolymer concrete. It is clear from the results that there is a direct relationship between strength and density, when the density value of a particular mixture increases, the resistance value of the same mixture increases accordingly. Additionally, as compared to (MC), there is an enhancement in density by (1.5 %, 0.7 % and 1.4 %) for MBC, and by (2.4 %, 1.6 % and 1.8 %) for MBT at alkaline doses of (40 %, 45 % and 50 %). It is clear that the amount of improvement in NT density is higher for the same percentage of addition, due to the higher unit weight of NT. These findings agree with what found by Irshidat and Al-Saleh [45]. Moreover, as compared to control (MC), results of ternary blends (MR) demonstrated that the addition of nano materials was remarkably affected through increasing the bulk density by (3.6 %, 2.4 % and 2.3 %) at alkaline doses of (40 %, 45 % and 50 %) respectively. This happens as a result of the filling ability of nanoparticles that minimize the size and sum of pores that lie in the microstructure. More alkalis doses in the mix produce higher reactivity with FA. Resulting in a denser microstructure. This would increase the thickness of the ITZ, so the bonding forces between the matrix and the aggregate particles would increase [26].

### 3.2. Microstructure

The results shown in Fig. 6 represent data from all mixes (MC, MBC, MBT and MR) subjected to autoclaving and constant alkalis by

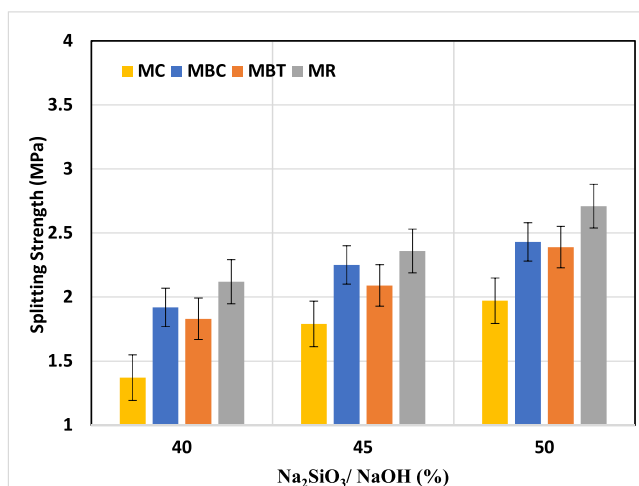


Fig. 4. Effect of nano  $\text{TiO}_2$  and nano clay on the splitting tensile strength at different percentages of alkaline.

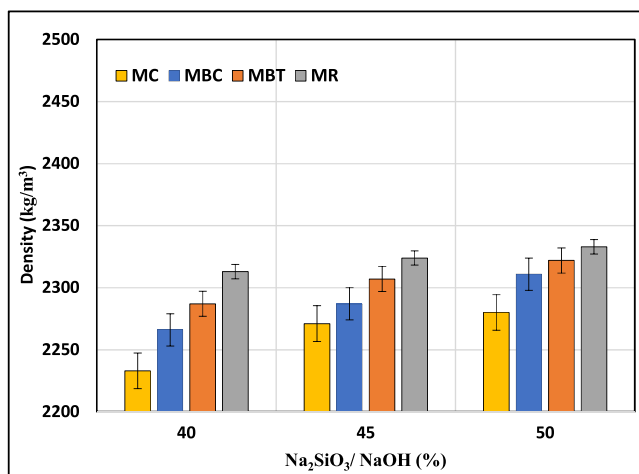


Fig. 5. Effect of nano TiO<sub>2</sub> and nano clay on the dry density at different percentages of alkaline.

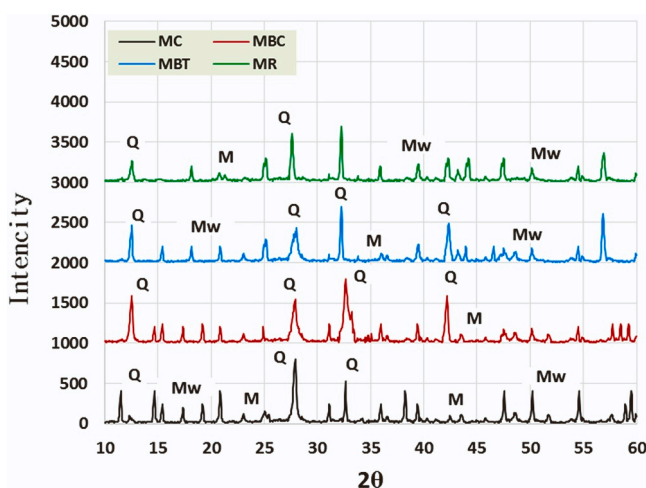


Fig. 6. X-ray diffraction for specimens: (a) control MC; (b) MBC with 1 % NC; (c) MBT with 1.25 % TiO<sub>2</sub>; and (d) MR with (1 % NC and 1.25 % NT).

w.t. of FA of 40 %. These mixes were chosen because they exhibit higher percentages of enhancement in compressive and splitting strengths. The XRD patterns indicate the formation of well-crystallized materials. As shown in Fig. 6, Quartz (Q) and Mullite (M) higher peaks were noticed in the 2θ values of 27–32° for all samples. While MBT still has some of this well-crystallized material but shows the emergence of quartz and mullite crystalline phases which originated from the nano TiO<sub>2</sub>. The presence of glassy phases in the internal structure may lead to such results. Usually, these peaks indicated the formation of the main compound (aluminosilicate hydrate (N-A-S-H)) produced by the geopolymerization process. The wide peaks detected at 2θ = 25°, 38°, and 48°, indicating the formation of geopolymer products. The largest hump was noticed at 2θ = 27.5° as a result of the Quartz appearance. The mix MR features somewhat more pronounced (more highly crystalline, and/or perhaps forming in greater quantity) quartz and mullite peaks, at the expense of the amorphous contribution. These findings agree with other studies [46].

Kanagaraj et al. [47] reported: “when the percentage of NC increased, the amount of Ca(OH)<sub>2</sub> decreased, at the same time the X-ray humps of C-S-H expanded“. Likewise, Kamath et al. [12] recognized a reduction in Ca(OH)<sub>2</sub> from 17 % to 12 %, whilst the amorphous phases improved from 70 % to 75 %.

NT and NC can speed up pozzolanic reaction, giving nucleation sites that gather around it the geopolymerization products. The fundamental hump of geopolymerization occurs earlier with improved peak as mentioned by Kuri et al. [48].

SEM analysis gives a good qualitative sight of the mineralogy that lies inside the geopolymer microstructure. Fig. 7 shows four SEM micrographs of (MC, MBC, MBT and MR) mixes at 28 days. Generally, mix MC has uncomplicated and regular microstructure composed of microcracks which indicating weaker microstructure and besides this gives a reason for the decrease in compressive and splitting tensile strengths. This explains why the fracture surface of the reference mixture has a weak ITZ as pointed by (arrow 1/a). Moreover, these pores are somewhat filled with NC and/or C-S-H from the pozzolanic reaction (arrows 2/b and 3/c). This makes the ITZ thinner and thus, making a better cohesion force in internal structure. So, samples of binary and ternary blends are heavier than the



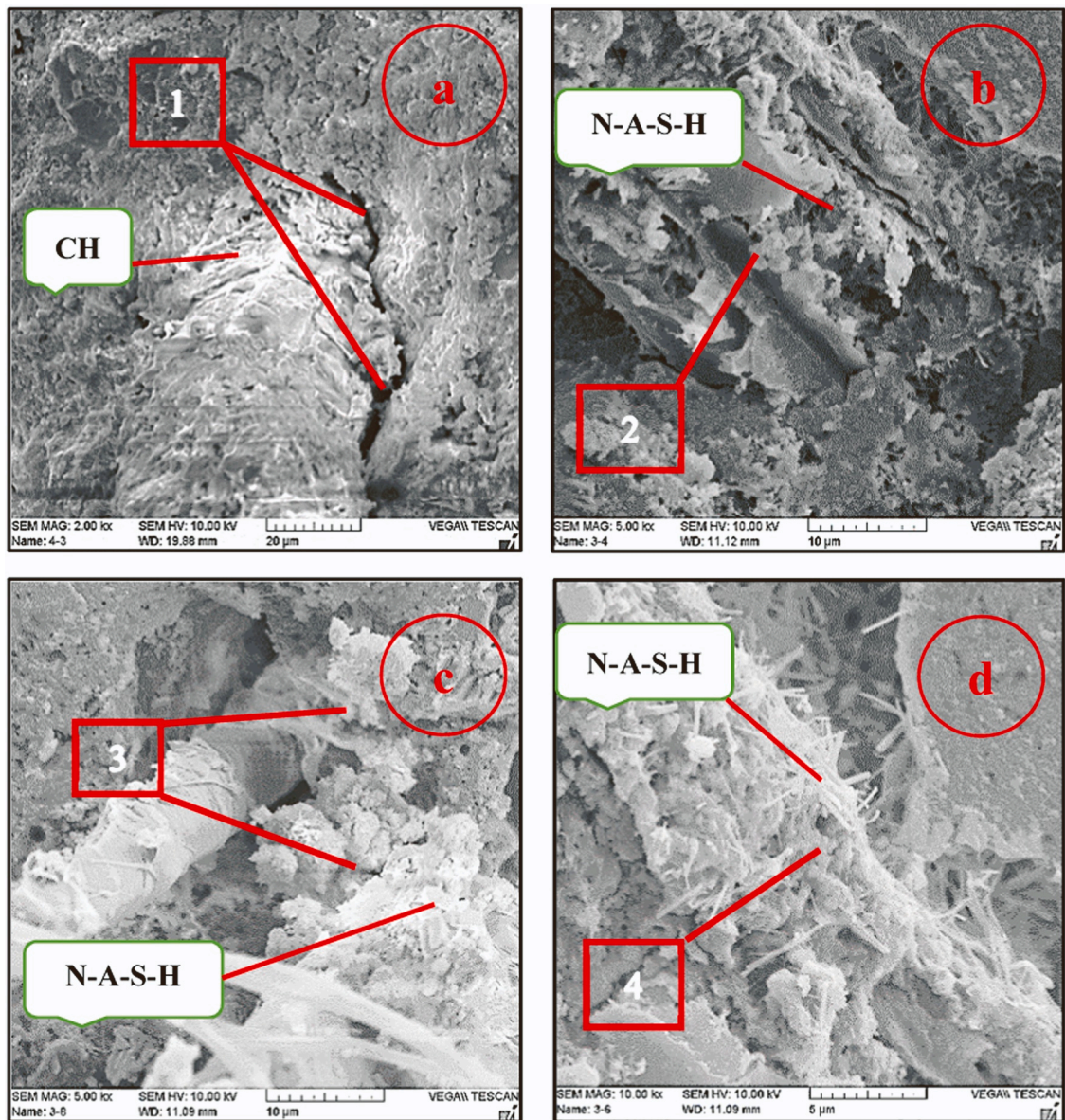


Fig. 7. SEM images of fractured surface for specimens: (a) control MC; (b) MBC with 1 % NC; (c) MBT with 1.25 % TiO<sub>2</sub>; and (d) MR with (1 % NC and 1.25 % TiO<sub>2</sub>).

MC that can improve the ITZ, and developed the strength of resulting concrete. The absence of interfacial voids and cracks is clearly visible for the specimens containing nanomaterials. These findings agree with what found by Ali et al. [49]. The unexpected surprise was the appearance of very thin fibers (arrows 4/d) in the interfacial regions between the paste and the aggregate, which is attributed to the formation of silicate compounds due to the pozzolanic interaction of nanomaterials and the early stimulation of steam accelerated curing. Moreover, from these images, the matrix of the geopolymer is denser (Fig. 7/ b, c and d), improving the adhesion between aggregate and matrix, thus decreasing the thickness of ITZ. The SEM images corroborate his results, and are proof that accelerated pozzolanic reactions improved this interface through better mechanical interlocking effects.

#### 4. Conclusions

##### 4.1. Theoretical and practical implications of findings

From the outcome of this study, it is possible to conclude that:

1. The optimal percentage of addition that maximizes compressive strength for NC is 1 %, and 1.25 % for nano TiO<sub>2</sub> by weight of FA.
2. The addition of nanoparticles improved the compressive strength of produced geopolymer concrete up to 38 % for NC, and 24 % for TiO<sub>2</sub> respectively in the binary blends. Meanwhile, the increase reaches 55 % in the ternary blends.
3. The splitting tensile strength results proved that both: NC and NT are essential elements in developing the strength of geopolymer concrete. Nevertheless; NC shows more strength than NT.
4. Density increased by (0.7–2.4 %) for binary blends and between (2.3–3.6 %) for ternary blends at 28 days respectively.
5. The XRD analysis proved that Quartz (Q) and Mullite (M) amorphous peaks, noticed in the patterns of diffraction of 2θ between 27° and 32°.
6. Electron microscopy images showed the presence of extra-large crystals of hydrates due to the pozzolanic activity of the nano-materials, which precipitated in the empty pores. This densifies the ITZ and that tends to increase the composite strength.
7. By reviewing the results, it is clear that the value of the improvement in the studied properties was low when the percentage of addition of nanomaterials exceeds 1 %. And for economic considerations, this ratio is the best to use for the two types of additives: NC and NT.

#### 4.2. Limitations and recommendations for further studies

1. One of the limitations of this investigation is that the age of the examination was up to 28 days, and therefore further studies should be carried out to study the behavior of these nanomaterials at the later ages (for example 180 and 360 days).
2. The other limitation of this study is the multi-collinearity issue in the optimization problem. The approach that was adopted in this study was represented by optimizing the replacement percentage of the nanomaterials separately and then mixing them together in the geopolymer concrete mixtures. Although this technique has also been used in the literature, the properties of geopolymers containing two nanomaterials together may differ from those containing only one. Therefore, this issue should be addressed extensively in the future works to show their possible effect on the results obtained.
3. A study is recommended to examine the durability characteristics of FA-based geopolymer concrete contained blending of nanomaterials.

#### Declaration of Competing Interest

None.

#### Data availability

No data was used for the research described in the article.

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