Effect of nano-silica on microstructure formation of low-cost geopolymer binder

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Abstract Influence of Nano-SiO₂ (NS) addition on properties of Geopolymer materials has been studied through measurement of physicomechanical and morphological characteristics of the hardened specimens. Alumino–silicate materials are water-cooled slag, albite, kaolin, and metakaoline. Materials were prepared at water/binder ratios in a range of 0.244:0.320 for water-cooled slag based materials, while it increased to 0.46 for albite-based mixes, whereas the used activator sodium hydroxide is 10 wt.%. The control geopolymer mix has been composed of water-cooled slag, metakaoline, and kaolin in the ratio of (4:2:1). Albite used for comparison with slag to demonstrates the possibility of producing lightweight geopolymer binder. Nano-silica was added in the range from 0.3% from the total weight with 0.5% increment. Results indicated that, compressive strengths of geopolymer mixes incorporating NS were obviously higher than those of control one, specially at early ages as well as up on using 2.5% NS with the lowest percentage of water absorption; also uses of albite instead of slag results in formation of light binder with promising mechanical characteristics.

Keywords Nanostructures, lightweight, dense, geopolymer, environment, microstructure

Introduction

Over recent years, there has been a rapid increase in the range of available nanomaterials and the number of companies that supply them. Different production techniques are used, each with benefits and drawbacks, but these are being improved all the time. In building industry, nanotechnology has known by its self-cleaning windows, smog-eating concrete, and many other advances. However, these advances products are minor compared to those developed in the world’s nanotech labs today. Nano era advent in building could not have come at a better time, as the building industry moves aggressively toward sustainability.¹,²

Construction industry is under increasing pressure for adopting green practices in the delivery of built infrastructure. CO₂ is major concern after it has reached critical level of 390 ppm in atmosphere.³ Major CO₂ producing sectors, such as power generation, transportation, oil refining, and manufacturing of steel and concrete are obligated to reduce CO₂ global emission rate by 2030. Within the concrete industry, cement manufacturing is the main culprit.⁴ The use of blended cements is one of the avenue that is expected to significantly reduce the concrete industry’s carbon footprint.⁴,⁵ In addition to being more sustainable, blended cements are also less susceptible to cracking and more durable. Geopolymer binders are examples of non-traditional materials used in blended cements include those derived from pozzolans. Such use of geopolymer binders for building application is not a new concept – the use of pozzolans in construction had precedence in ancient civilization, also less vulnerable to cracking. Following industrialization, there was a shift in the market from pozzolans, triggered by the emergence of Portland cement which is now indisputably the most widely used binder in construction applications.⁶ The renewed interest in new classes of binders for built environment applications has been largely fueled by: (1) concerns over greenhouse gas emissions associated with the cement production process; (2) the need for binders with enhanced durability performance.⁷ The scope of this paper is restricted to construction applications of geopolymer binders incorporating secondary input materials (volcanic ash, fly ash, and slags). The deployment of such products is particularly advantageous in buildings, where durability, strength, and fired resistance are of primary concern.
Despite the existence of strong economic and environmental drivers, geopolymers are still not widely used throughout the world. Alkali-activated binders represent an attractive alternative for the partial or complete substitution of Portland cement in the production of mortars and concretes, offering comparable performance and cost while reducing greenhouse gas emissions.11

Many researchers have applied industrial wastes as water-cooled slag and study its thermal and acid resistance;12 also the crystalline electric arc furnace slag can be activated for the production of geopolymer materials using cement kiln dust as an activator as it has high alkali content capable of dissolving aluminosilicate precursors,13,14 geopolymer produced from construction and demolition wastes were also enhanced by calcium hydroxide producing a more valuable products with high mechanical and morphological properties as investigated by Khater15, where the addition of calcium compound improved the mechanical and microstructural properties with the increase in hydrated lime up to 10 wt.% for mix specimens that were water cured under ambient temperature, while it slightly decreases for those cured in 100% R.H at 40 °C. The influence of silica fume (SF) addition on the properties of geopolymer materials produced from alkali activation of aluminosilicates, metakaolin, and waste concrete produced from demolition works has been studied by Khater15, and results indicated that compressive strengths of geopolymer mixes incorporating SF increases up to 7% substitution and then decreases up to 10%.

Khater et al.17 also studied the effect of nano-silica in the ratio from 0 to 1.5% on alkali-activated slag geopolymer, and results showed that increasing the percentage of nano-silica results in an enhancement in the mechanical properties as compared with the control mix up to 90 days. The study of thermal properties took place for the different ratios by experimental and mathematical evaluation. The study shown that the thermal properties as well as thermal insulation property are improved with the increase in the ratio of nano-silica. Kuo et al., shown that the compressive and flexural strengths of cement mortars enhanced with SiO₂ and Fe₂O₃ nanoparticles.18 It was found that the nanoparticles which dispersed uniformly in a cement paste will accelerate cement hydration due to their high activity.19 Additionally, the nanoparticles will fill pores leading to increase in strength and improvement in the microstructure of cement and the interface between the cement paste and aggregates in concrete. It was also found that nano-Fe₂O₃ exhibits a self-sensing of strain capability which can be useful for structural health monitoring.20

Effects of nano-silica (NS) and silica fume (SF) on rheology, spread on flow table, compressive strength, water absorption, apparent porosity, unrestrained shrinkage, and weight loss of mortars up to 28 days were also studied by Senff et al.21 Samples with NS (0–7, wt.%), SF (0–20, wt.%) and water/binder ratio (0.35–0.59), were investigated through factorial design experiments. Nano-silica with 7 wt.% showed a faster formation of structures during the rheological measurements. Maximum unrestrained shrinkage increased 80% for NS mortars (7 days) and 54% (28 days) when compared to SF mortars in the same periods.

Use of an alternative activator based on nano-silica/NaOH (M = K⁺ or Na⁺) blended solutions on the performance of alkali-activated fly ash binders was assessed by Erich et al.22 Binders produced with commercial silicate activators display a greater degree of reaction, associated with increased contents of geopolymer gel; however, mortars produced with the alternative nano-silica-based activators exhibited lower water demand and reduced permeability, independent of the alkali cation used. Results suggest that there are variations in the availability of Si in the system, and consequently in the alkalinity, depending on the silicate source in the activator, which is important in determining the nanostructure of the geopolymer gel.

The main goal of the current paper is to study effect of nano-silica addition on the physicochemical and microstructural features of the resulting geopolymer materials produced from various aluminosilicate precursors producing nontraditional cementing materials that can be applied in many building applications. Another target is to compare the physical and morphological properties between the albite-based lightweight and other formed geopolymer structure. Testing of the resulting geopolymer product is studied with X-ray diffraction (XRD), FTIR, SEM, compressive strength testing, water absorption and bulk density that are conducted on pastes of Geopolymer-based sample, in order to generate a better understanding of the effect of the nano-silica on the behavior of Geopolymerization reaction.

**Experimental procedures**

**Materials**

Materials used in this investigation are Water-cooled slag as a type from ground granulate blast furnace slag (GGBFS) supplied by the Iron and Steel Factory – Helwan, Egypt; kaolin collected from El-Dehesa, South Sinai, Egypt as well as albite collected from Aswan governorate, Egypt. Sodium hydroxide (NaOH) with purity 99% in the form of pellets used as alkali activators, obtained from SHIDO Co., Egypt.

The chemical compositions of the starting raw materials are given in Table (1). Mineralogical characterization of the raw materials was done using X-ray diffraction analysis in powder form as represented in Figure (1). Water-cooled slag is a rich aluminosilicate material and composed of SiO₂, CaO, Al₂O₃, Fe₂O₃, and MnO as illustrated in Table (1), while its mineralogical content is composed of amorphous materials.

On the other hand, chemical composition of kaolinite material showed that it contains about 56% alumina oxide and 27% of silica oxide, in addition to small content of calcium and magnesium oxides as presented in Table (1), however the mineralogical composition shows that it composed of kaolinite mineral with minor amount of quartz minerals as shown in Figure (1). This kaolinite material was thermally treated at 800 °C for 2 h with a heating rate of 5 °C/min., to produce metakaolin (Mk). This temperature was chosen on the basis of an earlier research works, where calcinations below 700 °C results in a less reactive metakaolinite with more residual kaolinite, above 850 °C crystallization occurs and reactivity declines.18,16,23,24 Albite’s mineralogical composition, on the other
hand, is composed of montomorillonite, kaolinite, calcite, quartz and albite, while its composition is composed mainly of SiO₂ as major constituent and Al₂O₃, Fe₂O₃, and CaO as minor constituents.

Nano-silica used in this investigation is a synthetic product with spherical particles in the range of 8–18 nm and 60 m²/g blain fineness imported from Sigma–Aldrich (Germany). The chemical analysis showed that it consists mainly of pure silica, 99%, while its X-ray diffraction (XRD) pattern (Figure 1) shows that nanosilica is mostly composed of amorphous quartz. The physical properties of nano-silica are given in Table (2). Laser particle size distribution pattern of nano-silica indicate that the crystallinity of NS is of an average particle size of 60 nm.

Transmission Electronic Microscopic (TEM) was carried out using type JEOL – JEM – 1230 of magnification up to 60000 to test the particle size of the used nano-silica powders. Figure (2) shows the morphologies of NS, where its particles are represented by highly agglomerated clusters with size of (40–65 nm).

**Dispersion of nano silica**

Nano silica material was first subjected for sonication using a Fritish 450 Sonifier Analog Cell Distributor for 15 min. Glenium Ace 30-poly-carboxylate based superplasticizer used for nano-silica dispersion as this polycarboxylate-based superplasticizer has been proven to be effective for nano-silica dispersion. Solutions with concentration of 0–3 wt.% of the total dry weight and replaced the used kaolinite content were used to identify the effect of nano-silica concentrations for the evaluation of its threshold ratio.

**Geopolymerization and curing**

Nanoparticles are not easy to disperse uniformly due to their high surface energy. Accordingly, mixing was performed as follows:

1. Nano-SiO₂ particles were stirred with 50% of the mixing water using high-speed magnetic stirrer for 15 min.
2. Geopolymer materials passing a sieve of 90 μm as represented in Table (3) were hand mixed with the alkaline activator solution as well as with the other part of mixing water for 10 min followed by a further 5 min using rotary mixer and mixed at medium speed (80 rpm) for another 30 s.
3. Glenium Ace-30 superplasticizer (polycarboxylate based) was added and stirred with the mixture at high speed for additional 30 s, followed by the previously stirred nano-silica.
4. The mixture allowed to rest for 90 s and then mixed for 1 min at high speed.

All investigations involved the addition of 10 wt.% NaOH of dry mixes. Water–binder material ratio (w/b) increases depending on the added nano content as indicated in the table (3). The paste mixture was casted into 25 × 25 × 25 mm cubic-shaped moulds, vibrated for compaction and sealed with a lid to minimize any loss of evaporable water.

Specimens were left to cure undisturbed under ambient temperature (23 °C) for 24 h, demolded and then some specimens were left to be water cured under room temperature.
collected from 0°to 50° 2θ (step size 0.02°2θ and speed 0.4°/min). Silica was used as an internal standard. Data were identified according to the XRD software (pdf-2: database on CD-Release 2005). Particle size analysis was done using a laser scattering particle size distribution analyzer (Horiba

**Methods of investigation**

Chemical analysis was carried out using Axios (PW4400) WD-XRF Sequential Spectrometer (Panalytical, Netherland). Compressive strength tests were carried out using five tones German Brüf pressing machine with a loading rate of 100 kg/min determined according to ASTM C109 M. XRD analysis was recorded on a Philips PW 1050/70 Diffractometer using a Cu-Kα source with a post sample Kα filter. XRD patterns were

| Description       | Results          |
|-------------------|------------------|
| Diameter (nm)     | 8–18             |
| Purity (%)        | 98               |
| Surface area (m²/g) | 240             |
| Density (g/cm³)   | 0.5              |
| Molecular weight  | 60.08            |

At the end of the curing regime, the specimens were removed from their curing condition, dried well at 80 °C for 24 h then exposed to the compressive strength measurements, while the other parameters estimated by using the resulted crushed specimens.
Stopping of hydration was performed on crushed specimens by subjecting to stopping alcohol/acetone solution (1:1), followed by washing with acetone as recommended by different investigators for preventing further hydration.

Water absorption measurements of the bricks were carried out according to ASTM C140. The percentage absorption was calculated using the equation:

\[
\text{Absorption} \% = \left( \frac{W_2 - W_1}{W_1} \right) \times 100
\]

where \( W_1 \) = weight of specimen after complete drying at 105 °C, \( W_2 \) = final weight of surface dry sample after immersion in water for at least 24 h.

Bulk density was calculated using the formulae:

\[
\text{Bulk Density} = \frac{D}{(W - S)} \text{ (g/cm}^3\text{)}
\]

where \( D \) = weight of specimen, \( S \) = weight of suspended specimen in water, and \( W \) = weight of soaked specimen suspended in air.

Transmission Electronic Microscopic (TEM) (type JEOL – JEM – 1230) of magnification up to 60000 was to measure the particle size of the nano materials. Bonding characteristics of the alkali-activated specimens were analyzed using a Jasco-6100 Fourier transform infrared spectrometer (FTIR). Test sample was ground and uniformly mixed with KBr at a weight ratio KBr: specimen = 200:1. The mixture, 0.20 g was pressed to a disk of 13 mm in diameter for analysis at 8 t/cm². The wave number was ranging from 400 to 4000 cm⁻¹.

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**Results and discussion**

**FTIR investigations**

FTIR spectra of 28 days hardened geopolymer specimens having various NS content are shown in Figure (3). Bands description are as follows: stretching vibration of O–H bond at about 3455 cm⁻¹, bending vibration of H–O–H bond at about 1630 cm⁻¹, stretching vibration of CO₂ located at about 1430 cm⁻¹, asymmetric stretching vibration (Si–O–Si) at about 1060 cm⁻¹ for non-solubilized silica, asymmetric stretching vibration (T–O–Si) at about 980 cm⁻¹ where T = Si or Al, symmetric stretching vibration of CO₂ at about 870 cm⁻¹, symmetric stretching vibration (Si–O–Si) at 665 and 535 cm⁻¹ and bending vibration (Si–O–Si and O–Si–O) at about 455 cm⁻¹.

FTIR spectra of control mix without nano-silica indicated the formation of broad asymmetric T–O–Si band for three-dimensional geopolymer network (N-A-S-H gel) at about 980 cm⁻¹ increased in its intensity up to 2.5% NS reflecting the increased content of the formed amorphous geopolymer structure as related to NS enhancement effect on geopolymerization reaction, as well as offering more nucleation sites for geopolymer gel growth and accumulation; however, further increase in NS beyond 2.5% results in a decrease in the broadness of amorphous geopolymer band with the formation of shoulder at about 1050 cm⁻¹ for non-solubilized silica as resulted from the agglomeration of nano-silica, where the difficulty in dispersing of NS uniformly is a common feature, so that hindering the interaction and solubilization of reacting materials negatively decrease amorphous structure, therefore a weak zone in the form of voids is formed, consequently the
Formation, while increase again with 3% NS as a result of increased porosity that will be more prone to air entering and carbonation increase. It can be seen that intensity of the carbonate at about 1430–1450 cm\(^{-1}\) (\(\nu\) C–O) subjected to be splitted into two peaks indicates the distorted nature of CO\(_3\) mineral.\(^3\)\(^8\),\(^3\)\(^9\) This could be attributed to partial carbonation of C-S-H gel in air atmosphere.

Increasing curing time up to 180 days for the control of slag-based geopolymer mix (Figure 4) results in an increased enhancement in the broadness of the main T–O–Si asymmetric stretch vibration absorption peak.\(^3\)\(^6\) The spectra showed also a decrease in carbonate content at about 1430, 870 cm\(^{-1}\) with NS as a results of increased geopolymer formation, while increase again with 3% NS as a results of increased porosity that will be more prone to air entering and carbonation increase. It can be seen that intensity of the carbonate at about 1430–1450 cm\(^{-1}\) (\(\nu\) C–O) subjected to be splitted into two peaks indicates the distorted nature of CO\(_3\) mineral.\(^3\)\(^8\),\(^3\)\(^9\) This could be attributed to partial carbonation of C-S-H gel in air atmosphere.

Also, there is a broad band for hydrating materials (C-SH & CASH) and combined water within the matrix at 3455 cm\(^{-1}\), this band seems to be increased with NS addition as a results of interaction of free dissolved Ca species from slag precursor forming CSH that will offers an additional binding materials as well as nucleation sites for geopolymer gel growth.\(^3\)\(^7\) The spectra showed also a decrease in carbonate content at about 1430, 870 cm\(^{-1}\) with NS as a results of increased geopolymer formation, while increase again with 3% NS as a results of increased porosity that will be more prone to air entering and carbonation increase. It can be seen that intensity of the carbonate at about 1430–1450 cm\(^{-1}\) (\(\nu\) C–O) subjected to be splitted into two peaks indicates the distorted nature of CO\(_3\) mineral.\(^3\)\(^8\),\(^3\)\(^9\) This could be attributed to partial carbonation of C-S-H gel in air atmosphere.

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Effect of nano-silica on geopolymer nanocomposites

The present unreacted kaolinite material that decreases with curing time as a result of increased kaolinite dissolution. Further increase in curing time up to 180 days for the albite control-based geopolymer mix (Figure 5) results in a decrease in non-solubilized silica (Si–O–Si) at 1100 cm\(^{-1}\) as a result of increased dissolution with time and incorporation in the formation of geopolymer chains as is in accordance with the increased intensity of T–O–Si band with time. Also, an increased intensity of the hydration and combined water at 3400 cm\(^{-1}\) as compared with slag-based mix favoring increased porosity of the first one, also the kaolinite bands beyond 3600 cm\(^{-1}\) indicate the lower dissolution efficiency of unreacted kaolinite in that mix as compared with slag-based one.

**Figure 5** FTIR spectra of control albite-based geopolymer specimens cured up to 180 days. [1: Stretching vibration of O–H bond, 2: Bending vibrations of (HOH), 3: Stretching vibration of CO\(_2\), 4: Asymmetric stretching vibration (Si–O–Si), 5: Asymmetric stretching vibration (T–O–Si), 6: Symmetric stretching vibration of CO\(_2\), 7: Symmetric stretching vibration (Al–O–Si), 8: Symmetric stretching vibration (Si–O–Si), 9: Bending vibration (Si–O–Si and O–Si–O)]

**Figure 6** XRD pattern of 28 days alkali-activated geopolymer having various NS content. [Q: Quartz, Fj: Faujasite, CSH: Calcium silicate hydrate, R: Reversedite, HS: Hydroxysodalite, A: Zeolite A, C: Calcite, K: Kaolin]
NS up to 2.5%, whilst an increased NS over this ratio results in the formation of weak zone of agglomerated nanomaterials that are susceptible to carbonation as reflected on the increased intensity of calcite peaks that coincide with FTIR illustrations. On the other hand, the zeolite phases decrease with nano-silica up to 2.5%, followed by an increase as a result of hindering the intact between the geopolymer chains as well as hindering the formation of more branched chains as a result of crystallization.

Studying effect of time on the control slag-based geopolymer mixes up to 180 days (Figure 7) results in decrease in intensity of the kaolinite content, whilst the CSH phases increase with time as results of increased interaction of dissolved materials.

Mineralogical investigation

XRD pattern of 28 days hardened alkali-activated geopolymer mix admixed with various ratios of nano-silica is shown in Figure (6). The pattern illustrates a broad band in the region of 20° to 35° 2θ characterizing glassy phase of geopolymer constituent, this region exposed to increase with nano-silica addition up to 2.5% as a result of increased NS enhancement and possessing of nucleation centers for geopolymer growth; however control mix without NS shows the presence of kaolinite peaks as a result of incomplete dissolution of source materials, which will diminish with NS as mentioned before. An increase in the CSH phases (CSH & reversedite C$_2$S$_2$H$_3$) with NS up to 2.5%, whilst an increased NS over this ratio results in the formation of weak zone of agglomerated nanomaterials that are susceptible to carbonation as reflected on the increased intensity of calcite peaks that coincide with FTIR illustrations. On the other hand, the zeolite phases decrease with nano-silica up to 2.5%, followed by an increase as a result of hindering the intact between the geopolymer chains as well as hindering the formation of more branched chains as a result of crystallization.

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intensity of the kaolinite content whilst its intensity higher than slag-based mix, as results from decreased amorphous content of albite mix so that lower dissolution ability will be resulted. Whilst CSH phases increase with time as results from increased interaction of dissolved calcium and silica species forming more binding materials, however the glassy geopolymer band looks smaller as compared with slag mix one where the amorphous constituents of slag form more geopolymer chains and reflected positively on the hump in the previous region.

calcium and silica species forming the binding materials, this coexist with the increased broadness of glassy geopolymer band; reflecting an increased formation and growth of the geopolymer structures with time forming a dense structure, this is in alignment with the decreased Faujasite with time. The previous notice confirmed by lowering carbonate content with curing time.

Investigating the effect of time on the albite control-based geopolymer mixes up to 180 days(Figure 8) results in decreased intensity of the kaolinite content whilst its intensity higher than slag-based mix, as results from decreased amorphous content of albite mix so that lower dissolution ability will be resulted. Whilst CSH phases increase with time as results from increased interaction of dissolved calcium and silica species forming more binding materials, however the glassy geopolymer band looks smaller as compared with slag mix one where the amorphous constituents of slag form more geopolymer chains and reflected positively on the hump in the previous region.

Figure 9 SEM micrographs of control water-cooled slag-based geopolymer specimens cured at; (A) 90 days, (B) 6 months

Figure 10 SEM micrographs of 28 days alkali-activated geopolymer specimens having various doses of NS. (A) 0%, (B) 0.5%, (C) 2.5%, and (D) 3%
Microstructural investigation

Microstructure of the control slag-based geopolymer specimens without NS and cured at 90 and 180 days are shown in Figure (9). It can be noticed that morphology of geopolymer specimens cured up to 90 days (Figure 9(a)) composed of small plates spread within the matrix in addition to formed CSH, which fill mostly all the voids within the matrix, however increasing curing time to 180 days (Figure 9(b)) results in an increased densification of geopolymer plates into big ones that enhance the formation of more homogenic morphology.

Replacing kaolinite constituents by NS from 0 to 3% for hardened specimens at 28 days Figure (10), where the control mix without NS (Figure 10(a)) depicts the coexistence of CSH along with aluminosilicate gel, whilst heterogeneity of the matrix is predominant with the presence of pores structure. Adding NS in the ratio of 0.5% results in the enhancement in the morphological texture where the voids in the plain mix almost diminishes, however the intensity of the formed geopolymer structures is still not in dense morphological shape (Figure 10(b)). Increasing NS to 2.5% (Figure 10(c)) results in the formation of dense homogeneous structure with little evidence of the presence of micropores, this can be related to the enhanced effect of NS in accelerating the hydration reaction as well as offering more nucleation sites for geopolymer growth and accumulation. Further increase in NS to 3% (Figure 10(d)) results in the formation of wide pores with the matrix in addition to the formed binding materials, these pores aroused from the NS agglomeration leading to difficulty in dispersing uniformly, therefore a weak zone in the form of voids is formed, consequently the homogeneous hydrated microstructure cannot be formed.

On performing a close comparison between 6 months hardened slag-based geopolymer specimen and albite one without NS as illustrated in Figure (11), a clear difference between both morphological shapes appeared, where slag-based one enriched with Ca-source results in both dense geopolymer plates (Figure 11(a)); however albite-based geopolymer specimens suffers from deficiency in calcium species as reflected in its morphological shape as CSH results in both binding structure, as well as offers nucleation sites for geopolymer gel growth. The previous findings emphasized by weak cohesion between the reacting materials as well as the presence

Figure 11  SEM micrographs of 6 months alkali activated geopolymer specimens having 0% NS. (A) slag based, (B) albite based

Figure 12  Compressive strength of alkali-activated geopolymer specimens having various Nano silica content

CompressiveStrength, MPA

3days 7days 14days 60days 90days 180days

Nanosilica, %

0 0.5 1 1.5 2 2.5 3 3.5

0 10 20 30 40 50 60 70 80

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The reaction process between SiO2 and Ca(OH)2 may be as follows:

\[ =\text{Si} - \text{O} - + \text{H} - \text{OH} \rightarrow =\text{Si} - \text{OH} - \text{react quickly} \]

\[ =\text{Si} - +\text{H} - \text{OH} \rightarrow =\text{Si} - \text{OH} - \text{react quickly} \]

\[ =\text{Si} - \text{OH} + \text{Ca} (\text{OH})_2 \rightarrow \text{C-S-H} \]

The-formed CSH binding phases possess nucleation sites for geopolymer accumulation and growth in addition to seeding effect of nanomaterials, both shared positively in enhancement of the mineralogical and morphological shape of the resulted hardened specimens.

The increase in compressive strength was noticed with NS addition up to 2.5%, while decrease with further NS increase, the increase in strength as mentioned latter linked to seeding nucleation of nanomaterials, both shared positively in enhancement of the mineralogical and morphological shape of the resulted hardened specimens.

The increase in compressive strength was noticed with NS addition up to 2.5%, while decrease with further NS increase, the increase in strength as mentioned latter linked to seeding nucleation of nanomaterials, whilst the decrease related mainly to insufficient wetting and dispersion of the added nanomaterials in spite of using superplasticizer, as a results of agglomeration, forming weak zone with many points of defects that can be facilitate air carbonation of geopolymer matrix. Furthermore, increasing NS content up to 2.5%, hardened geopolymer paste strength as compared with control sample increased by 75.65, 65.20, 52.04, and 33.96% at 7, 28, 90, and 180 days, respectively, while using 3% NS leads to

**Compressive strength**

Compressive strength of hardened geopolymer mixes having various NS ratios from 0 up to 3% from the total weight with increment of 0.5%, and cured up to 180 days is shown in Figure (12). The strength pattern illustrates an increase in strength of all mixes with curing time as a result of hydration progress forming CSH in addition to geopolymer constituents leading to production of dense and homogeneous structure; where Nano-silica has a specific surface of 160 m²/g, so the reaction between SiO₂ and free Ca(OH)₂ is going in quick pattern rate where NS having many unsaturated bonds Si–O and Si in the surface, the reaction process between SiO₂ and Ca(OH)₂ may be as follows:

\[ =\text{Si} - \text{O} - + \text{H} - \text{OH} \rightarrow =\text{Si} - \text{OH} - \text{react quickly} \]

\[ =\text{Si} - +\text{H} - \text{OH} \rightarrow =\text{Si} - \text{OH} - \text{react quickly} \]

\[ =\text{Si} - \text{OH} + \text{Ca} (\text{OH})_2 \rightarrow \text{C-S-H} \]
Figure (13) shows a comparative diagram between control albite-based and water-cooled slag-based geopolymer hardened specimens, where a wide gap between the two types at all curing times, as a result of deficiency of calcium source in albite-based mix in addition to increased iron content that will precipitate as hydroxide and so lowering the medium alkalinity and so the driving force for geopolymer formation and accumulation.

Water absorption and bulk density investigation

Water absorption of the slag-based geopolymer specimens (Figure 14) has opposite pattern to compressive strength profile, where the absorption decrease with NS up to 2.5%, then exposed to reincrease with further NS, as well as decrease with curing time up to 90 days. Increased enhancement of NS strength loss by −22.16, −29.84, −20.88, and −22.37% 7, 28, 90, and 180 days, respectively.

The trend of compressive strength variation coincides well with XRD, FTIR observations as well as SEM micrographs Figures (3, 10), where the amorphous content of geopolymer materials increased with increasing NS content up to 2.5% – as indicated from the increased intensity of main asymmetric T–O–Si band as form more precipitation sites for geopolymer accumulation and precipitation upon interacting with free lime, forming CSH. Further increase in NS leads to insufficient wetting of the medium that hinder the propagation of geopolymer chains and so weaken its mechanical properties; as indicated from the increased carbonate content due to the increased weak zone with increased agglomeration of nano-silica and so decrease in intensity of amorphous band N-A-S-H as illustrated previously.

Figure 15 Water absorption comparison diagram between control albite and water-cooled slag-based Geopolymer specimens at different curing times

Figure 16 Bulk density comparison diagram between control albite and water-cooled slag-based Geopolymer specimens at different curing times
results in the formation of well-compacted and dense geopolymer structure, which positively decrease available pores and consequently water absorption as confirmed well from the geopolymer microstructures elucidations represented previously in Figure (10).

A wide variation between water absorption using albite-based geopolymer mix as compared with slag-based one at all curing time, where the increased water absorption in albite mix favors the utilization in formation of lightweight precursors where their strengths are sufficient in formation of lightweight load bearing brick walls after 7 days (Figure 15), as in accordance with their mechanical strength which give values exceeding 9 MPa more than required for load bearing walls.

These previous notices about production of lightweight geopolymer product confirmed by lower bulk density of the albite-based geopolymer mix (Figure 16), whilst slag-based mix confined a higher bulk density giving additional evidence about formation of compacted and dense geopolymer matrix as a result of continuous dissolution and interaction of dissolved Ca and Si species forming CSH in addition to the formed geopolymer chains. The bulk density values of the albite-based mix are lower than those of alkali-activated slag. This may be due to the decrease in Ca\(^{2+}\) in the albite; therefore, lower amounts of CSH formed, which is the main source of mechanical properties of the cement pastes. As the Na\(_2\)O content increases, the bulk density decreases due to the formations of rich CSH with low bulk density and high porosity.\(^{34,42}\)

**Conclusions**

The main findings of this article are summarized below:

1. Addition of NS results in an enhancement in mechanical and morphological characteristics of the formed geopolymer specimens incorporating fired and unfired precursors.
2. The enhancement and modification of the hardened geopolymer specimens achieved when using NS up to 2.5%, however further increase results in agglomeration and decrease in the efficiency of the added nanomaterials.
3. SEM micrographs have proved an increased enhancement in the microstructural properties of the NS-mixes up to formation of dense and homogenous morphological shape when using 2.5% NS, whilst an increase in porosity is a common feature of higher NS-mixes.
4. XRD and FTIR spectra confirm an intense amorphous geopolymer structure using 2.5% NS, while the increased crystalline phases with the formation of zeolite structures along with an increased carbonate bearing compounds are predominant with further NS.
5. The formed geopolymer mixes possess high mechanical properties that exceed 36 MPa after 28 days for control mix and increases with further NS up to 2.5%, giving a value of 60 MPa at 28 days and increased to 71 MPa at 6 months, which can be used in various building applications as infrastructure as well as fire-resistant building materials.
6. Using albite precursor for geopolymer formation results in sharp decrease in mechanical and microstructural properties as compared with slag-based mixes, however the values exceeds 9 MPa after 28 days which exceeds the values required for load bearing brick walls, whilst the absorption and bulk density of the previous mix favours its use in lightweight binding materials.

**Disclosure statement**

No potential conflict of interest was reported by the authors.

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