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Research paper

The effect of adding nano-silica on the ultrasonic pulse velocity of geopolymer concrete

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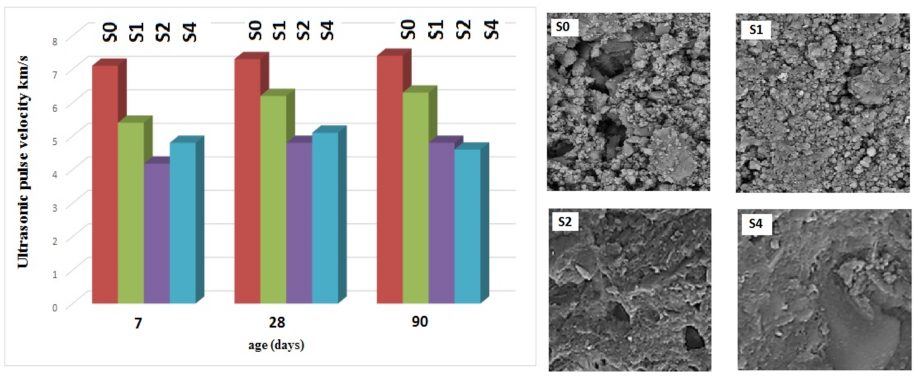
HIGHLIGHTS

- Compressive strength increased with time in all samples. However, the rate of increase varied in different samples and alkalinity.
- The sample with 4% weight of nano-silica provided the maximum strength after 90 days.
- The increase of the nano-silica additive in geopolymer concrete improved the mechanical strength of concrete due to strengthening the concrete bonds.
- The reduction of concrete porosity according to SEM images, it was seen that the ultrasonic pulse velocity decreased.
- The minor increase of the amount of nano-silica improved the efficiency of the concrete proposed in this study.

GRAPHICAL ABSTRACT



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ABSTRACT

Nanotechnology plays an important role in the current construction industry. It has been observed that several properties of cement-based concrete are affected by different nano materials. In this study, different weight percentages of nano-silica (0, 1, 2, and 4 wt%) were used to build geopolymer specimens. The mechanical properties of the specimens were then measured, including the compressive and flexural strength. Results demonstrated that increasing the percentage of nano-silica from 0 to 4 wt% increases the compressive strength of the specimens from 50.4 to 75.2 MPa and the flexural strength from 11.4 to 26.2 MPa. Furthermore, it was observed that the ultrasonic pulse velocity was altered by increasing SiO_2 (nano-silica), decreasing the ultrasonic pulse rate from 7.0 to 4.5 $km.s^{-1}$.

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1. Introduction

Alkaline-activated aluminosilicate, known as Geopolymers, can be used as alternatives to conventional Portland cement to reduce the high emissions from the industrial and commercial use of cement [1]. Nowadays, concrete plays an unavoidable role in civil engineering and construction [2]. However, the high consumption of concrete and huge demand for cement production increases the harmful effects of CO_2 emissions in the atmosphere. To avoid these circumstances, the utilization of non-renewable energy, such as electricity and fossil fuels, is inevitable. Therefore, one of the approaches compatible with an eco-friendly environment is to reduce OPC (Ordinary Portland Cement) or conventional pozzolanic materials consumption [3,4].

In an effort to reduce the use of OPC, different types of concrete have been studied. Zeolites are minerals that contain mainly aluminum and silicon compounds. Although zeolites are a family of minerals, i.e., their chemical composition is similar to zeolite materials, their microscopic structure is amorphous instead of crystalline. Geopolymer is a cohesive material that can be used to produce geopolymer concrete. Supplementing zeolites in this type of concrete is a practical approach to address the main disadvantages of using conventional Portland cement [5].

Before geopolymer technology can be accepted and further developed, the control mechanisms and general alkali activation must be better understood [6]. Geopolymers are formed after being hardened from 3D networks, including tetrahedral AlO_4 and SiO_4, that are connected by an oxygen element. The geopolymerization process involves the dissolution of aluminosilicates, which generates a gel that eventually converts into a rigid body [7,8]. Metakaolin is a hyperactive reactive aluminosilicate with mechanical properties and durability similar to micro-silica [9]. However, unlike fly ash, blast furnace slag, and micro-silica (by-products of the other substances), metakaolin is produced exclusively for cementitious additives. Therefore, metakaolin, a Class N (natural) pozzolan, is required to comply with the requirements of AStM C618. The raw material needed for the production of metakaolin is kaolin soil, also known as Chinese soil [10].

Research has been conducted on incorporating nanoparticles, i.e., zinc oxide (ZnO) nanoparticles, copper oxide (CuO) nanoparticles, nano-silica (SiO_2), iron oxide, alumina (Al_2O_3), nanoclays, etc., into the geopolymer matrix or concrete to enhance physical and mechanical properties. For example, Zidi *et al.* studied the influence of zinc oxide nanoparticles on the mechanical and thermal properties of the geopolymer. Their results showed that the addition of ZnO nanoparticles could increase the compressive strength to its maximum value, depending on the ZnO content [11,12]. Furthermore, the acid resistance of geopolymer concrete can be improved by using 20% granite debris as natural, fine aggregates. While the use of granite waste in geopolymer concrete reduces its workability, workability may be effectively enhanced by using a variety of super plasticizers [13].

In its purest form, kaolin soil is a white mineral composed mainly of hydrated aluminum di-silicate $Al_2Si_2O_5(OH)_4$ [14]. The temperature at which kaolin becomes the crystalline structure of metakaolin is about 600 to 1000 ^{o}C. If the material is heated less during cooking, the amorphous mineral phase conversion will not occur, leading to an incomplete pozzolanize procedure. On the other hand, if the material is overcooked, it will clump [1] and form a hard dead material [15] and non-reactive mullite [2] ($Al_2O_3.2SiO_2$). Therefore, temperature plays an essential role in converting kaolin soils into meta-kaolin super reactive pozzolans [16]. From a civil engineering point of view, meta-kaolin increases the quality of cement additives and provides higher performance compared to micro-silica [17]. Nowadays, there is no doubt that nanotechnology is an inevitable part of many industries. Recent studies have shown that nanoparticles (e.g., nano-silicates) provide a high surface-to-volume ratio that can increase chemical reactivity.

Many studies have been conducted on the influence of adding nano-silica to concrete and reinforcement, i.e., to improve workability [14]; however, only a few have been done on the effect of adding nanoparticles to concrete's water bodies to enhance its physical and mechanical properties. Naniz and Mazloom studied the influence of different levels of nano-silica on fresh and hardened properties of self-compacting concrete with different water-to-binder ratios. According to their experimental results, replacing 3% of the cement with nano-silica produced satisfactory self-compacting concrete [18,19]. Tarangini *et al.* proved that nano-silica could increase the micro voids of pervious concrete more than several other mineral admixtures, thus significantly increasing the freeze-thaw resistance of concrete [20,21].

Since the velocity of the ultrasonic pulse is influenced by the mechanical properties of the concrete, it can be used to measure these properties. Variation in the pulse velocity along different paths in the structure is a sign of variation in the quality of the concrete. In cases where parts of the tested concrete have low density, high porosity, and injury, the pulse velocity is reduced, in which case one can determine the amount of possible failure. Moreover, changes in the structure of concrete resulting from strength or damage affect the pulse rate and may appear as increases or decreases in its velocity. Therefore, it is possible to detect changes in concrete structures by measuring the pulse velocity at appropriate intervals [22].

In this regard, pulse velocity measurements are used for concrete quality control. For this purpose, one should note that the mechanical test conducted on concrete cubes and control cylinders for measuring the pulse velocity pertains only to the concrete. This is because the cubic and concentric cylinders may not represent the actual concrete of a structure. Therefore, the magnitude of the pulse velocity in comparison with the mechanical test is preferred over the specimens tests if the relationship between ultrasonic pulse velocity of the concrete properties in the tested structure is obtained and the results of this experiment can be achieved. Although it is possible to derive empirical relationships between the pulse velocity and static/dynamic modulus of elasticity as well as the concrete strength, these relationships are uninfluenced by other factors, including the type, content, additives of cement, the type and size of aggregates, operating conditions, and the type of concrete. In investigating the results of pulse velocity in the determination of elastic properties and resistance based on the results of pulse velocity, the influence of other factors should be considered, especially in cases where concrete resistance is more than 60 Mpa [17]. One of the important applications of ultrasonic pulse velocity (PUV) in evaluating the mechanical properties of concrete is to estimate the compressive strength of concrete. Karimaei used ultrasonic pulse velocity technology as a nondestructive testing method to estimate the compressive strength of 11 groups of concrete samples containing coal gangue [23]. The compressive strength and UPV parameters of concrete of different ages and different proportions of coarse and fine aggregate replaced by coal gangue were studied, and the exponential relationship between compressive strength and UPV was obtained [12].

Due to its mechanical properties, Geopolymers concrete has the potential to be used in fire-resistant materials, thermal insulation, construction materials, cement, and concrete. Nano-silica is one of the materials that can change the mechanical, microstructural, and structural properties of concrete. This study demonstrates how UPV may be used to investigate the trend of strength evolution in geopolymer concrete.

2. Experimental Procedure

2.1. Basis of ultrasonic pulse velocity device operation

An electro-acoustic generator produces pulses of longitudinal vibrations. This generator is tested on a concrete surface. After passing the pulses through a certain length of the concrete, the pulse vibrations are converted into electrical signals by an additional generator (i.e., receiver) [4]. The electronic circuit of the device is able to measure the passage time of the pulse in microseconds. The pulse rate V in the unit of $km.s^{-1}$ and $m.s^{-1}$ is obtained from the Eq. (1).

$$V = L / T \tag{1}$$

Where L and T denote the pulse path length and the pulse passage time (i.e., the length of time it takes the pulse to pass through the length L), and the ultrasonic pulse vibration is represented in the unit of frequency. This is because the pulse is transmitted precisely and fully in the concrete, and the maximum energy is created along the pulse development. On the other hand, concrete is composed of different phases, which means that when the pulse is applied to concrete, it is exposed to different exposures [5].

2.1.1. Effective factors for pulse velocity

Basically, the amount of the pulse velocity depends on the properties of the concrete being tested. Therefore, it is necessary to consider the different factors that affect the pulse rate and pulse velocity, such as moisture content, concrete temperature, path length, and sample size and shape. These factors are briefly described below.

2.1.2. Moisture content

The moisture content has two effects, physical and chemical, on the pulse rate. These are important when estimating the concrete resistance with pulse speed. The pulse rate in a standard sample may differ significantly with structural organs made from other concrete types. This difference is mainly due to different treatment conditions and hydration degrees, as well as the existence of free water in the pores. Hence, these factors must be considered when estimating the resistance [5].

2.1.3. Concrete temperature

The variation in concrete temperatures has no significant effect on the pulse velocity.

2.1.4. Path length

The path length where the pulse velocity is measured should be long enough so that the pulse is unaffected by the heterogeneity of the concrete.

2.1.5. Sample size and shape

The velocity of the short pulse is independent of the size and shape of the specimen it pulses through unless the minimum lateral dimension is greater than the minimum value. If the length of the specimen is sufficiently clear, the pulse velocity may be reduced significantly. The value of this reduction greatly depends on the ratio of the pulse-wave length to the minimum lateral dimension of the sample. However, if the ratio is greater than unity, the reduction in the velocity rate should be considered. In other words, the minimum lateral dimension of the sample should be more than the pulse-wave length. In the present study, ultrasonic pulse velocity was used to investigate the effect adding nano-silica powder to SiO_2 has on the mechanical properties of geopolymer specimens [5,6].

2.2. Preparation

Metakaolin has been used as the main material. A coarse aggregate of different sizes of 20, 16, 12.5, 10, and 4.75 mm and locally available sand were mixed in different percentages (Table 1). The chemical composition of metakaolin is listed in Table 2.

Table 1. Concrete mixing plan.

Fine aggregate ($kg.m^{-3}$)	Coarse aggregate ($kg.m^{-3}$)	Metakaolin ($kg.m^{-3}$)	Fly ash ($kg.m^{-3}$)
850	850	200	400

Table 2. Chemical composition of Metakaolin.

wt%	Composition
54.1	SiO_2
43.5	Al_2O_3
0.2	Fe_2O_3
0.1	CaO
0.12	Na_2O
0.02	K_2O
0.08	MgO
1.3	TiO_2
1.3	L.O.I.

The coarse and fine aggregates used in the experiment were in a saturated surface dry condition. A combination of sodium hydroxide and sodium silicate was considered the alkaline liquid. Commercial-grade sodium hydroxide was mixed with distilled water to produce a solution of sodium hydroxide with 16 molar concentrations. The characteristics of this solution are presented in Table 3.

The sodium hydroxide to sodium silicate ratio was kept at 1:2.5 (by volume). Colloidal nano-silica with 0% (S0), 1% (S1), 2% (S2), and 4% (S4) fly ash (wt%) was added to the alkaline liquid. The nano-silica powder, with a particle size of about 14 nm, was produced by the Xunyu Henan Chemical Company, China. The nano-silica was initially mixed dry with the methacholine for 4 hr to distribute it in the geopolymer samples uniformly. The alkaline solution was obtained by calculating the proportion of distilled water, sodium silicate solution, and sodium hydroxide pellets. The geopolymer paste was prepared by gradually adding

Table 3. Specifications of the sodium silicate solution.

Value	Specifications
8.2%	Na_2O
27%	SiO_2
64.8%	H_2O
1.34 $g.ml^{-1}$	Density
< 0.005	Fe
< 0.005	Heavy metals (such as Pb)

an alkaline silicate solution to a mixture of nano-silica and metakaolin. The mixing procedure was continued until complete uniformity, approximately 30 min. The geopolymer paste was poured into polyethylene column molds and shaken for several minutes to expel air bubbles. The samples were then cured for 24 h at 67 °C. Closing the mold lids during the curing process prevented the water from escaping from the samples during geopolymerization.

2.3. Mechanical and non-destructive test

The compressive and flexural strength of the specimens were provided by AStM C39 and AStM C1161 standards, respectively. Microstructural images were obtained with an electron scanning electron microscopy 2300 Scan Cam. These images were taken from the failure level of the samples. The surface of the samples was covered with a thin layer of gold to prevent electronic charging. The size of the standard cube specimens was 150×150×150 mm, and samples with 40×40 mm section and 160 mm length were cast to determine the compressive and flexural strength of the geopolymer concrete with different mixtures of nano materials and a control sample without them. All specimens were tested for 7, 28 and 90 days after casting to determine the compressive strength at different ages.

3. Results and discussion

3.1. Mechanical strength

The variations in the amount of the compressive and flexural strength (in the unit of MPa) at different ages and nano-silica additives (S0-S4) are shown in Fig. 1. Non-destructive tests via ultrasonic pulse velocity were also performed on these samples to determine the concrete quality as per IS code. As can be observed in Fig. 1, both compressive and flexural strength values increase as the age of the specimens increases. Moreover, one can discern that the strength amounts present upward trends. The highest strength level belongs to sample S4, which included 4% weight of the nano-silica. More precisely, the compressive and flexural strength values varied from 50.4 to 75.2 MPa and 11.4 to 26.2 MPa from S0-S4. Moreover, this increasing trend in the two graphs is such that we can conclude that as the weight percentage of silica nanoparticles increases to values above 4% by weight, the strength will continue to increase.

Furthermore, in the unit of $km.s^{-1}$, the ultrasonic pulse velocities from samples S0-S4 at 7, 28 and 90 days are shown in Fig. 2. The Fig. 2 shows that in samples with nano-silica (S1, S2, and S4), the ultrasonic pulse velocity increases as the age of the specimens increases from 7 to 28 days, but the ultrasonic pulse velocity changes very little after that, up to 90 days (greatest time analyzed in this study). However, for samples without nano-silica (S0), the ultrasonic pulse velocity decreased from day 28 to 90. The highest level of the ultrasonic pulse velocity belongs to sample S4, with a 4% weight of the nano-silica.

According to studies, the process of geopolymerization includes four stages that occur simultaneously [3]. These four steps are briefly related to the dissolution of Si and Al from aluminosilicate in a strong alkaline aqueous solution. Moreover, the formation of Si-O-Si and O-Al-Si in the aqueous phase compacts the polymerization to create a three-dimensional aluminosilicate structure, the bonding of solid particles in the geopolymer structure, and the capture of the whole system as the final polymer structure.

As a source of Si required for the formation of geopolymer reactions, the presence of nano-silica improves the reactivity and dissolution of the raw

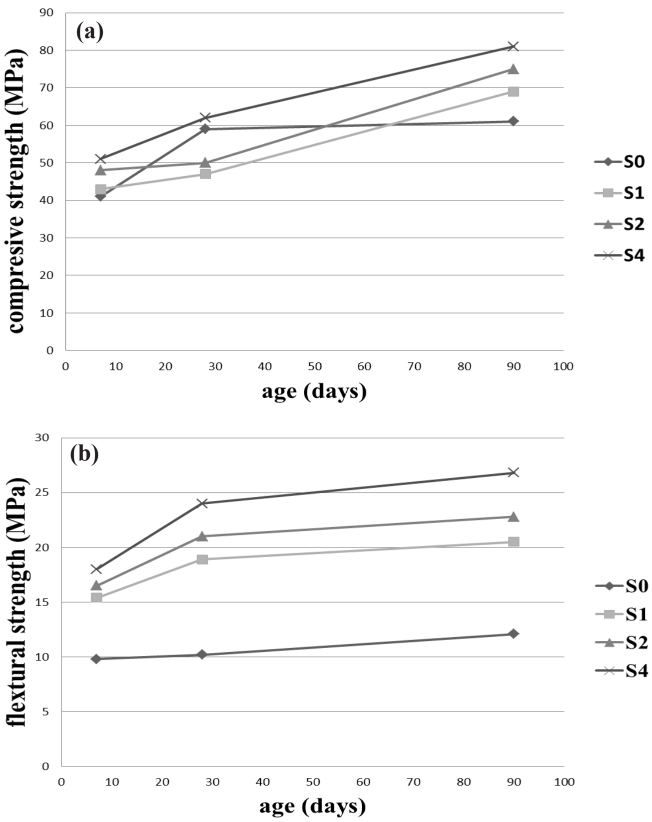

Fig. 1. Variations in the strength values of samples S0-S4 on different days and nano-silica additives. (a) the compressive strength and (b) the flexural strength.

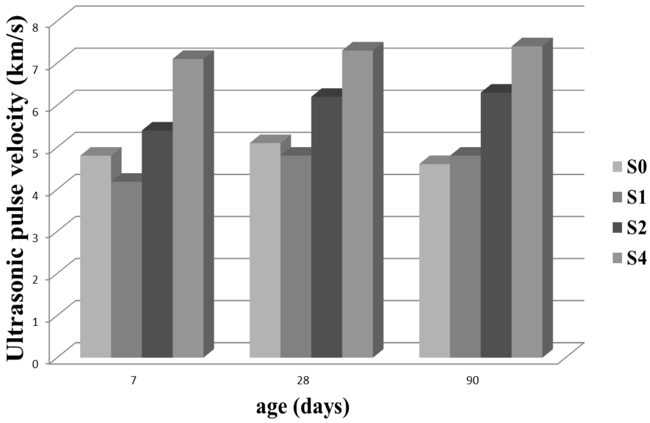

Fig. 2. The ultrasonic pulse velocities of samples S0-S4 under different ages of 7, 28, and 90 days and different amounts of nano-silica additives.

material due to the amorphous, nano-presence of Si particles. Subsequently, the dissolved Si in the softened augmentation system increases. Hence, the Si-O-Si bonded reactions, which are stronger than the other present bonds, increase, improving the strength of the geopolymer samples [6-9]. In other words, the use of amorphous silica at a nanoscale particle size results in more system uniformity owing to higher reactivity and the absence of unreacted particles left over from the raw materials in concrete [8]. The presence of unreacted particles in the final body causes the uniformity of the microstructure to be lost, and the weak bond in the joint between these particles and the background, due to non-homogeneity, will be a place for stress concentration and a consequent reduction in the strength. As can be seen in Fig. 3, the geopolymer body containing 4% nano-silica (the upper right plot) roughly provides the uniformity characteristic and better cohesion than the other samples, confirming the above-mentioned descriptions.

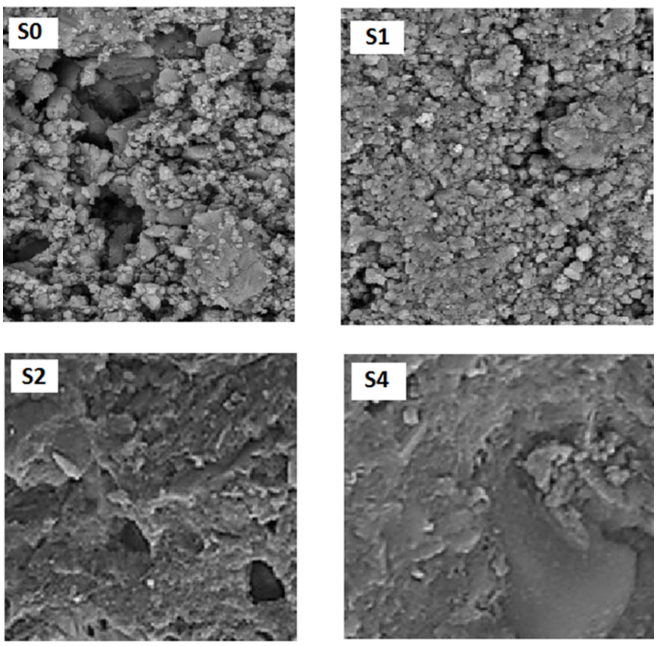

Fig. 3. SEM image of samples with nano-silica additive.

3.2. Ultrasonic pulse velocity (UPV)

Fig. 2 shows the ultrasonic pulse velocity results of geopolymer concrete samples containing nano-silica. As can be seen, the ultrasonic pulse velocity decreases from 7 to 4.5 $km.s^{-1}$ when the amount of nano-silica increases up to 4%. However, the best quality of concrete in all cases is obtained when the pulse velocity values are more than 4.5 $km.s^{-1}$. Under such circumstances, the quality of the proposed concrete proposed is better than concrete samples made with ordinary cement. On the other hand, using nano-silica in a geopolymer concrete reduces the pulse rate by decreasing the porosity and strengthening the Al-Si bonds. In addition, concrete curing time also affects the ultrasonic pulse velocity.

Fig. 3 indicates the scanning electron microscope (SEM) images of the refractive index of the samples containing nano-silica and the control sample without nano-silica. The applicability of standard charts and graphs mainly used for cement-based concrete were also verified.

4. Conclusions

This study demonstrates how UPV may be used to investigate the strength trend evolution in geopolymer concrete. Based on our results, the following conclusions can be drawn:

(1) Compressive strength increased with time in all samples. However, the rate of increase varied in different samples and alkalinity.

(2) The sample with a 4% weight of nano-silica (S4) provided the maximum strength after 90 days.

(3) The increase of the nano-silica additive in geopolymer concrete improved the mechanical strength of the concrete by strengthening the concrete bonds. In addition to this phenomenon and the reduction of concrete porosity (according to SEM images), it was also seen that the ultrasonic pulse velocity decreased.

(4) A minor increase in the amount of nano-silica improved the efficiency of the concrete proposed in this study.

References

[1] S. Naskar, A.K. Chakrabort, Effect of nano materials in geopolymer concrete, Persp. Sci. 8 (2016) 273-275.

[2] J. Davidovits, Properties of geopolymer cements, In Proceedings of the First International Conference on Alkaline Cements and Concretes, Scientific Research Institute on Binders and Materials, Kiev State Technical University, Kiev, Ukraine, 1994, pp. 131-149.

[3] A. Taffe, C. Maierhofer, Guidelines for NDT methods in Civil Engineering, Federal Institute for Materials Research (BAM), Berlin, Germany, 2003.

[4] A.M. Neville, J.J. Brooks, Concrete Technology, 2nd Ed., Prentice Hall, New York, 2010.

[5] E.N. Kani, A. Allahverdi, Effects of curing time and temperature on strength development of inorganic polymeric binder based on natural pozzolan, J. Mater. Sci. 44 (2009) 3088-3097.

[6] Y. Lin, H. Changfan, C. Hsiao, Estimation of high performance concrete strength by pulse velocity, J. Chin. Inst. Eng. 20 (1997) 661-668.

[7] Y. Lin, C.-P. Lai, T. Yen, Prediction of ultrasonic pulse velocity (UPV) in concrete, Aci Mater. J. 100 (2003) 21-28.

[8] E.A. Whitehurst, Evaluation of concrete properties from sonic tests, American Concrete Institute Monograph, 2 (1966) 27.

[9] T.R. Naik, V.M. Malhotra, J.S. Popovics, The ultrasonic pulse velocity method, In: Handbook on Nondestructive Testing of Concrete, 2nd Ed., CRC Press, 2003, pp. 8-19.

[10] B.S. Al-Nu'man, B.R. Aziz, S.A. Abdulla, S.E. Khaleel, Compressive strength formula for concrete using ultrasonic pulse velocity, Int. J. Eng. Trends Technol. 26 (2015) 8-13.

[11] P. Duxson, A. Fernández-Jiménez, J.L. Provis, G.C. Lukey, A. Palomo, J.S. van Deventer, Geopolymer technology: The current state of the art, J. Mater. Sci. 42 (2007) 2917-2933.

[12] Z. Zidi, M. Ltifi, Z. Ben Ayadi, L. El Mir, X. Nóvoa, Effect of nano-ZnO on mechanical and thermal properties of geopolymer, J. Asian Ceram. Soc. 8 (2020) 1-9.

[13] H.M.M. Khater, Physicomechanical properties of nano-silica effect on geopolymer composites, J. Build. Mater. Struct. 3 (2016) 1-14.

[14] P. Deb, P. Nath, P. Sarker, Properties of fly ash and slag blended geopolymer concrete cured at ambient temperature, in S. Yazdani, and A. Singh (ed), The 7th International Structural Engineering and Construction Conference, ISEC-7, Manoa, Honolulu: University of Hawaii, 2013, 571-576.

[15] Y. Tanigawa, K. Baba, H. Mori, Estimation of concrete strength by combined nondestructive testing method, Special Publication, 82 (1984) 57-76.

[16] U. Durak, O. Karahan, B. Uzal, S. İlkentapar, C.D. Atiş, Influence of nano SiO_2 and nano $CaCO_3$ particles on strength, workability, and microstructural properties of fly ash-based geopolymer, Struct. Concr. 22 (2021) E352–E367.

[17] J. Davidovits, Global warming impact on the cement and aggregates industries, World Resour. Rev. 6 (1994) 263-278.

[18] H.M. Khater, Effect of nano-silica on microstructure formation of low-cost geopolymer binder, Nanocomposites, 2 (2016) 84-97.

[19] O.A. Naniz, M. Mazloom, Effects of colloidal nano-silica on fresh and hardened properties of self-compacting lightweight concrete, J. Build. Eng. 20 (2018) 400-410.

[20] D. Adak, M. Sarkar, S. Mandal, Effect of nano-silica on strength and durability of fly ash based geopolymer mortar, Constr. Build. Mater. 70 (2014) 453-459.

[21] D. Tarangini, P. Sravana, P. Srinivasa Rao, Effect of nano silica on frost resistance of pervious concrete, Mater. Today-Proc. 51 (2021) 2185-2189.

[22] J. Davidovits, Geopolymer Chemistry and Applications, 4th Ed., Geopolymer Institute, Saint-Quentin, France, 2015.

[23] M. Karimaei, F. Dabbaghi, M. Dehestani, M. Rashidi, Estimating compressive strength of concrete containing untreated coal waste aggregates using ultrasonic pulse velocity, Materials, 14 (2021) 647.