Civil and Environmental Engineering
Vol. 20, Issue 2, 1120-1128, DOI: 10.2478/cee-2024-0081

PRODUCING SUSTAINABLE LIGHTWEIGHT GEOPOLYMER CONCRETE USING WASTE MATERIALS

Ameer A. HILAL[1,*], Awaz S. NADER[2], Badoor M. KURJI[3]

[1] Department of Civil Engineering, College of Engineering, University of Anbar, Al-Ramadi, Iraq
[2] Kalar Technical Institute/ Garmian polytechnic University/ Kurdistan Region/ Iraq
awaz.nader@gpu.edu.iq
[3] Department of Chemical and Petrochemical Engineering, College of Engineering, University of Anbar, Al-Ramadi, Iraq
bdoorm.kurji@uoanbar.edu.iq
[*] corresponding author: ameer.hilal@uoanbar.edu.iq, https://orcid.org/0000-0002-3070-1284

Abstract

A research has been undertaken to investigate the possibility of producing sustainable lightweight geopolymer material by using waste zeolite (zeolite molecular sieve) as aluminates and silicates source and at the same time as lightweight medium. This was done by totally or partially replacement of fly ash by waste zeolite particles. Six geopolymer mixes were designed and produced using different source materials (fly ash F, fly ash C and zeolite) and alkaline solution of sodium silicate and sodium hydroxide. From the results, it was found that using fly ash C resulted in producing geopolymer mix with higher density than those made with fly ash F (Gf mix). However, replacing the fly ash totally with the zeolite particles helped in producing lightweight geopolymer concrete (1600 kg/m^3) by achieving about 27% weight reduction and gaining about 5.1 MPa compressive strength. Not only reducing CO_2 emissions by adopting zeolite- based geopolymer binder (instead of cement paste) but also achieving operational energy saving by producing lightweight concrete was obtained from producing lightweight geopolymer concrete as constructional material.

Keywords:

Lightweight concrete;
geopolymer;
zeolite;
fly ash;
strength.

1. Introduction

In concrete industry, by incorporating different types of waste instead of concrete raw materials (cement, sand and gravel), sustainability in concrete production process is integrated [1-2]. Cement was partially or totally replaced by some industrial waste materials [3]. This encourages researchers to replace cement, the main concrete raw material, with more environmentally friendly pozzolanic wastes and then to improve the performance of concrete [4].

Geopolymer concrete can be considered as an innovative construction material with zero cement as a binding medium. This type of concrete produces by polymerizing the materials having high contents of aluminates and silicates in their chemical compositions such as fly ash, slag, metakaolin, high calcium wood ash, rice husk ash and zeolite. In addition to the source material, production of geopolymer concrete is dependent on the activator solutions [5]. This geopolymer material is normally produced with natural materials or/ and industrial wastes having rich amounts of silicates and aluminates [6].

Generally, geopolymer is an inorganic material which compared to pozzolanic materials can be considered as an amorphous rather than crystalline. It has very good properties such as high strength, low permeability, excellent fire resistance, great chemical resistance, good erosion resistance [7].

With its development since 1970's, geopolymer has been considered as the future cement [6]. It depends also on thermally activated natural materials such as metakaolinite or/ and industrial byproducts as sources of aluminum (Al) and silicon (Si). To represent the mineral polymers which are generating from geochemistry, a chemical reaction under highly alkaline conditions on Si-Al minerals can be processed, and subsequently polymerized into Si-O-Al-O bonds in amorphous form to finally produce the binder. It has been stated that, even with high alkalinity, the geopolymer materials do not suffer from alkali-aggregate reaction (AAR), and they possess excellent fire resistant [8]. The geopolymerization process occurs according to the Eq. (1) and (2) [9].

$$(Si_2O_5.Al_2O_2)_n + H_2O + OH^- \rightarrow Si(OH)_4 + Al(OH)^{4-} \quad (1)$$

$$Si(OH)_4 + Al(OH)^{4-} \rightarrow (-Si-O-Al-O-)_n + 4H_2O \quad (2)$$

Water releases during the chemical reaction of geopolymers formation. Later, during the curing and further drying periods; this water expelled from the geopolymer matrix leaving nano-pores in the matrix, which responsible on the geopolymers performance. In contrast to the chemical reaction of water in a Portland cement concrete during the hydration process, water has no role in the chemical reaction in geopolymer mixture. It just provides the workability to the geopolymer mixture during handling.

Fly ash, byproduct material, has been widely used as an additive in concrete production. According to ASTM standard and based on its main constituents, fly ash can be classified into three types as class F, class C and class N, [10]. The class F fly ash has less than 10% of calcium. The issue of using class F fly ash is the need for using high temperatures in order to accelerate the geopolymer reaction during hardening process to complete its structural integrity [11, 12]. Similar to the reaction of Portland cement, presence of large content of Calcium in fly ash class C helps also in forming calcium silicate hydrate [4, 13].

In addition to being a natural pozzolan, zeolite could be used as a geopolymer binding material [7]. Zeolite as molecular-sieve has unique microporous structure, morphology and physicochemical characteristics [14]. It often uses as cement substitution due to having large amounts of active Al_2O_3 and SiO_2 with similar chemical composition [4]. Zeolite is crystalline aluminosilicates material with open 3D frame-work structures built of $(Al,Si)O_4$ tetrahedra and it can be classified according to the ratio of SiO_2:Al_2O_3. The main characteristics of zeolite is its large specific surface area (about 600–800 m^2/g), regular structure, uniform size of air pores and good thermal stability [15]. In addition, zeolites has molecular sieves properties, low relationship with temperature (range of 10–150 °C) and high absorption capacity at low pressures [15]. It was reported that addition of zeolite enables to manufacture lighter structural elements without decreasing the strength.

The aim of this research is to produce sustainable lightweight geopolymer material by using waste zeolite (zeolite molecular sieve) as aluminates and silicates source and at the same time as lightweight medium. This will be done by totally or partially replacement of fly ash by waste zeolite particles. Then the effect of this replacement on the density and strength of the geopolymer mix will be investigated.

2. Experimental work

2.1 Materials

As a geopolymer binder, low calcium fly ash type F and high calcium fly as (Type C) which are conformed to ASTM C 618 [16], were used as waste aluminates and silicates sources. In addition, waste zeolite molecular sieve (aluninosilicate microporous crystal) as sphereical particles with an avergae diameter of 2 mm was used as source for aluninates and silicates which are necessary for polymerization process, see Fig. 1. Table 1 presents the chemical compositions of the source materials used in this research. It can be seen from Table 1 that the total of SiO_2, Al_2O_3 and Fe_2O_3 contents in the zeolite is more than 70% indicating that it is typical of fly ash [6].

As activation liquid, sodium silicate (SS) and sodium hydroxide (SH) solutions were adopted. SS (viscosity of 1100 CPS at 20 ^{0}C) was used with concentration of 45.9% (54.1% water) and the weight proportion of SiO_2 to Na_2O equals to 2.4, (32.5% of SiO_2 and 13.4% of Na_2O). From the other hand, preparing of SH was by dissolving 99% purity of NaOH in water [17]. For all investigated geopolymer mortars, natural fine aggregate with maximum size of 4.75 mm was used as filler material.

Fig.1. Waste zeolite molecular sieve used

Table 1. Chemical composition type F and C fly ashes and zeolite determined by (XRF) (mass %).

Oxides	CaO	SiO2	Al2O3	Fe2O3	K2O	MgO	SO3	L.O.I
Type F fly ash, [%]	5.22	46.88	28.43	17.92	1.86	2.27	0.37	3.21
Type C fly ash, [%]	28.18	31.32	6.3	28.62	2.48	0.26	0.89	-
Zeolite [%]	3.34	67.76	13.11	0.872	1.423	1.2	-	9.9

2.2 Mix Design and Production

An absolute volumes method [18] was adopted to design all investigated geopolymer mixes since the zeolite can be considered as lightweight material with density of 780 kg/m^3. Firstly, 500 kg/m^3 for fly ash was chosen and then for alkaline liquid (AA) to solid equals to 1, the amount of AA to start with was 500 kg/m^3. For mixes with zeolite, the volume of fly ash (its specific gravity is 2.2) was replaced totally or partially by the zeolite particles as 177 kg/m^3.

The proportions of the investigated geopolymer concrete mixes are summarized in Table 2. Noted that, G denoted to geopolymer mix. The small letters f, c and z denoted to the source as fly ash F, fly ash C and zeolite, respectively. In mixes Gfz, Gzz and Gcz the 25% of the zeolite weight (177 kg/m^3) was replaced by fly ash F, fly ash C and zeolite powder, respectively.

The molarity and the alkalie liguid were chosen to be 8M and 500 kg/m^3, respectivelly. However, the alkaine liguid was corrected acoording to the source type (fly ash F or C or zeolite) in order to prevent the floatting up of the lightweight zeolite (balls) particles.

Geopolymer mixes investigated in this research were produced by mixing the source material and fine sand (dry materials) and then the alkaline solution was added gradually. The geopolymer slurry was then poured into the molds. The geopolymer concrete specimens were cured in an oven at 60 ^{0}C for 24 hours. Finally, the samples were removed from the oven and kept for 5-6 hours in room temperature until the test age [17]. The steps of production and curing are ilustrated in Fig. 2.

Table 2. Proportions of investigated geopolymer mixes.

Consists	Mixes					
	Gf	Gc	Gz	Gfz	Gcz	Gzz
Sand [kg/m^3]	1036	1036	1036	1036	1036	1036
Fly ash (F) [kg/m^3]	500	-	-	125	-	-
Fly ash (C) [kg/m^3]	-	500	-	-	125	-
Zeolite [kg/m^3]	-	-	177	123.75	123.75	123.75
Alkaline liquid (A) [kg/m^3]	500	500	500	500	500	500
Extra solution [kg/m^3])	-75	-150	-	-25	-75	+20
NaOH/ Na_2SiO_3	1	1	1	1	1	1
Molarity	8	8	8	8	8	8

(a) (b) (c)

(d) (e) (f)

Fig. 2. Steps of production and curing of investigated geopolymer mixes (a) dry materials mixing (b) solution incorporation (c) Mixing (d) molding (e) heat curing (f) final products

3. Tests

At ages of 7 and 28 days, densities of investigated geopolymer mixes were measured by dividing the specimen weight (kg) over its volume (m^3). Moreover, compressive strength test was determined according to ASTM C513 [19] on cubes of 70 mm^3. In addition, flexural strength test was done according to ASTM C78 [20] using 40mm× 40mm× 160mm prisms. For each property, the average of three specimens of each mix was taken. To investigate the distribution of zeolite particles and air pores, slices were cut from the centers of 50 mm^3 cubes and polished with 400#, 800#, 1200# and 4000# silicon carbide abrasive (35, 21.8, 15.3 and 5 microns, respectively) using a rotating grinder. Then, after oven drying; images were captured for the specimens surfaces using a suitable camera [18].

4. Results and discussion

It is well known that the geopolymerization process is started with the dissolve of the aluminosilicate solids (from the source material) into the solution of SS and SH (alkaline liquid) [21]. This chemical reaction controls by the source types due to the differences in aluminate and silicate amounts in one type to another as illustrated in Table 1 and Fig. 3. It can be noticed from Fig. 3 that Si/Al ratio differs among the source types [22]. In this research, two fly ash types (Type F and Type C) were used in addition to the zeolite as source materials. Furthermore, replacing 25% of the zeolite volume by fly ash resulted in producing lightweight geopolymer mixes with density around 1850 kg/m^3 as shown in Fig. 4.

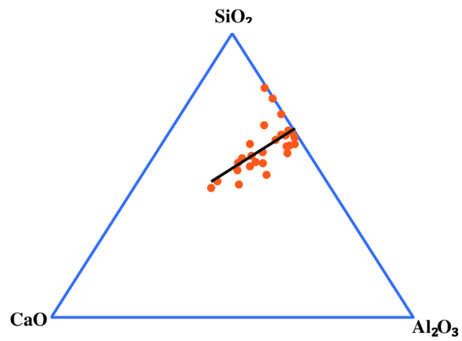

Fig. 3. Ternary diagram of calcium, silica and alumina contents [18]

In terms of density, Fig. 4 illustrates the average density results of the investigated mixes. The results show that the dry densities of all investigated geopolymer mixes at 7 days was slightly higher than those at 28 days. Whereas, the zeolite- based geopolymer mix (Gz mix) gave the lower density (about 1600 kg/m^3) among the all investigated mixes. In other words, replacing the fly ash totally with the zeolite particles helped in reducing the mix density by about 27% and producing lightweight material. In addition, it was noticed that using the zeolite particles helped in generating some entrapped air pores (pores creating during mixing process), see Fig. 5, since the vibration method did not adopt in order to avoid the floating up of the lightweight zeolite particles.

According to the results obtained, the geopolymer concrete mix obtained using fly ash C has higher density than that obtained using fly ash F, because fly ash C has higher Ca and Fe amounts in its composition [23]. Additionally, because of its light weight; using of zeolite as source in producing geopolymer concrete helped in achieving the lowest density. Noting that zeolites have pores smaller than 2 nm in diameter making it light and leading to reduce the concrete density [14].

With regard to strength, it was stated that fly ash (Type F) is preferred in the producing geopolymer concrete than fly ash Type C. This is because of the presence of high calcium in Type C which interfered with geopolymer binder sitting time and making it faster. Therefore, the strength (compressive and flexural) of mix made with fly ash Type C was higher than that made with Type F as shown in Fig. 6 and Fig. 7.

It was found that using the zeolite particles as source material instead of fly ash (Gz mix) caused reduction in strength by about 80% of that of geopolymer mix made with fly ash F (Gf). However, with reducing its weight, this substantiable lightweight materials can be used in insulation applications such as partitions. Some trials were carried out to enhance the strength of geopolymer mix with zeolite (Gz) by using fly ash instead of 25% of the zeolite. It was found that adding fly ash (Type F) instead of part of zeolite (Gf mix) helped in enhancing the strength by about 13% of Gz mix. However, this replacement resulted in increasing the density to about 1850 kg/m^3. It was noticed that adding zeolite powder (Gzz mix) had a negative effect on the strength by reducing it by about 58% of that of Gz mix. The reason for this strength reduction may be the fineness of the zeolite powder was not enough for polymerization process.

It was reported in the literature that the strength of a geopolymer material improves with increasing of alkaline liquid (AA) to solid ratio. This may be due to bringing more (Si) with increasing the added AA leading to the more formation of Si-O-Si bonds [24]. In addition, the role of silicates in geopolymerization process and the importance of presence of silicates in producing geopolymer materials were discussed [25]. Therefore, without increasing its density; future attempts will be carried out in future to enhance the strength of the zeolite- based geopolymer concrete by taken the above considerations into account.

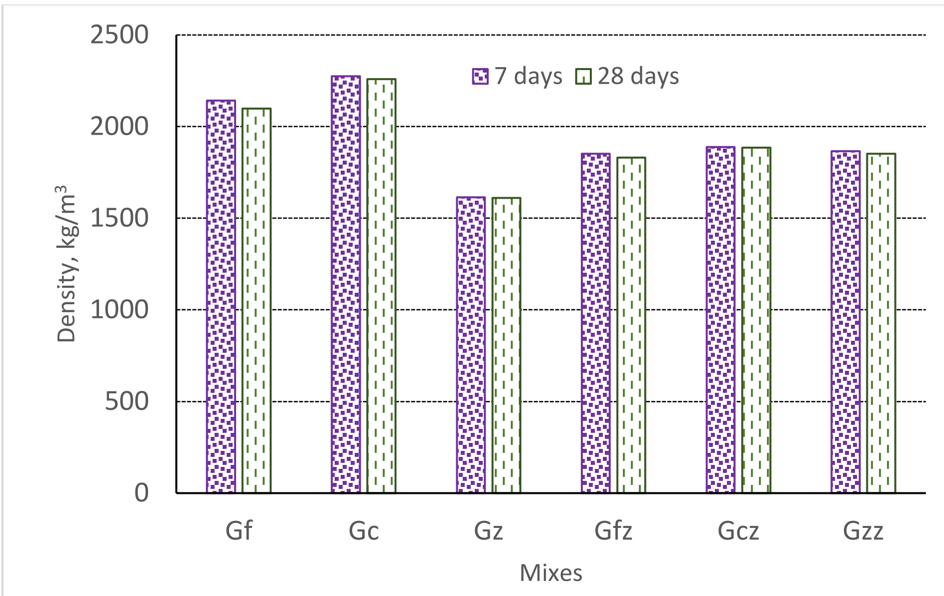

Fig. 4. Densities values of investigated geopolymer mixes

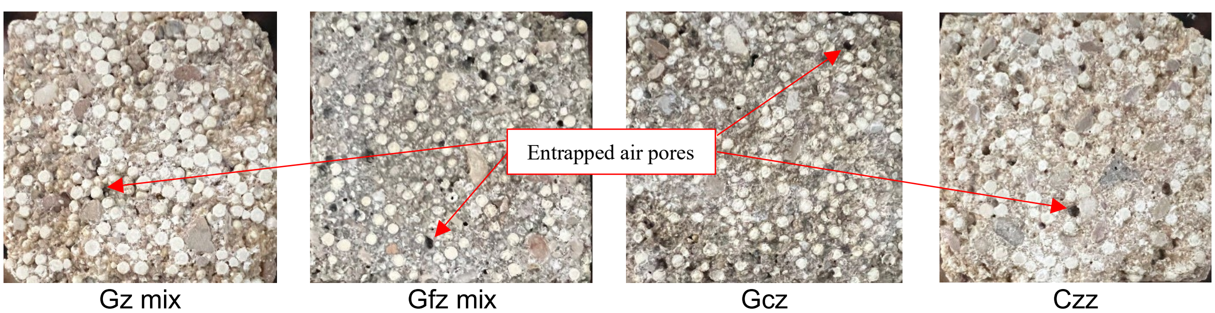

Fig. 5. Polished surfaces of geopolymer mixes made with zeolite [scale: 50mm× 50mm].

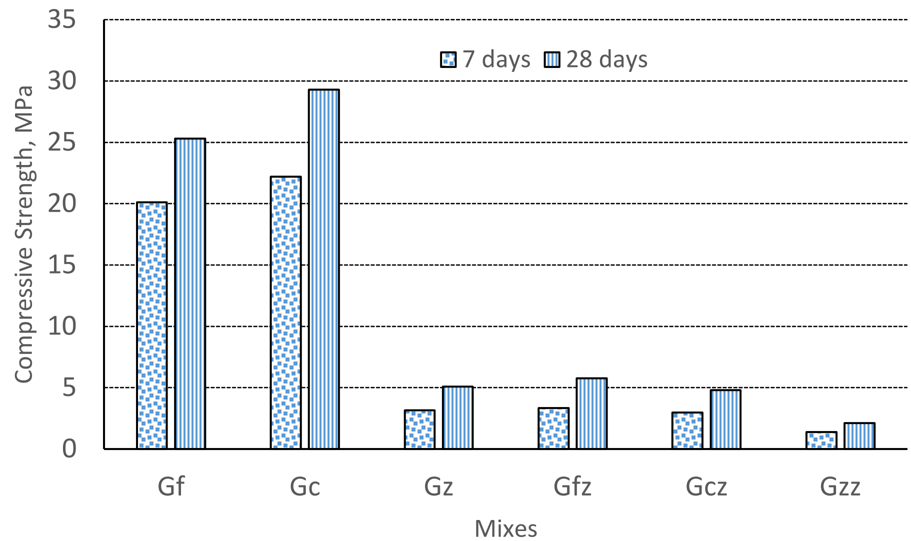

Fig. 6. Compressive strength values of investigated geopolymer mixes

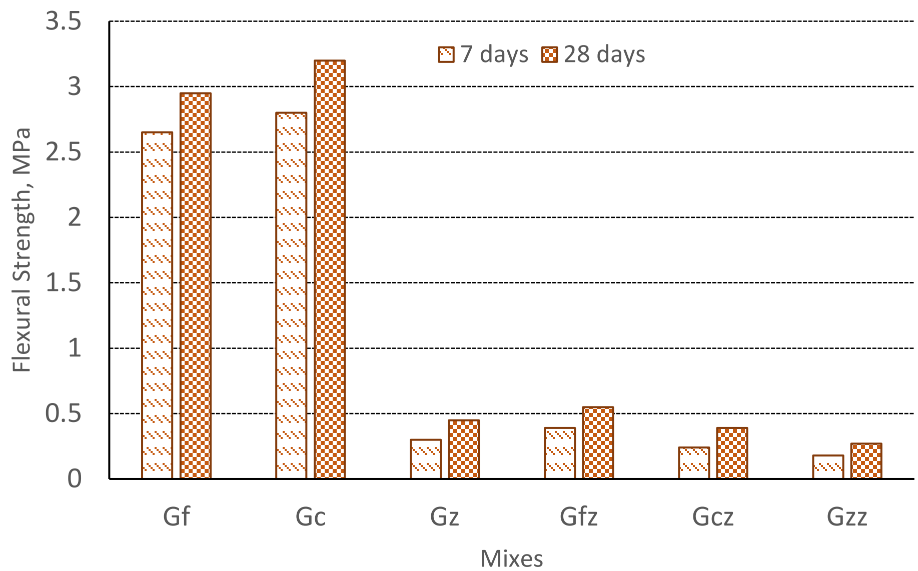

Fig. 7: Flexural strength values of investigated geopolymer mixes

5. Conclusion

The best finding from this research is producing lightweight geopolymer concrete from waste material, zeolite. By producing such a sustainable constructional material, not only reducing CO_2 emissions by adopting zeolite- based geopolymer binder (instead of cement paste) but also achieving operational energy saving by producing lightweight concrete. From the experimental work, it was found that, due to their different chemical compositions, fly ash (Type C)- based geopolymer mix gained higher density and strength than that made with Type F. Lightweight geopolymer with 27% weight reduction (about 1600 kg/m^3) was successfully produced by using zeolite particles as source medium instead of fly ash. With strength of 5.1 MPa, it is suitable for insulation applications such as partitions.

This study has suggested a number of avenues for future work including enhancing the strength and durability of the zeolite- based geopolymer concrete by taken into account some considerations regarding to increasing of alkaline liquid (AA) to solid ratio leading to more formation of Si-O-Si bonds and the altering the amount of silicates in the zeolite. In addition, the effect of zeolite particles roughness on strength and permeation properties [26] needs to be investigated.

References

[1] AL-HADITHI, A. - ABBAS, M.: The effects of adding waste plastic fibers on the mechanical properties and shear strength of reinforced concrete beams, Iraqi Journal of Civil Engineering, 2018, vol. 12, no. 1, pp. 110–124.
[2] HILAL, A.: Effect of crumb tyres rubber on some properties of foamed concrete, Anbar Journal for Engineering Sciences, 2011, vol. 4, no. 2, pp. 1–17.
[3] BHAVANI, S. - NAGESH, K. G. - REDDY, M. - SANJEEVA, R. E.: Experimental Study on Geopolymer Rubberized concrete using natural Zeolite, IOP Conf. Ser. Mater. Sci. Eng., 2021, vol. 1132, no. 1, p. 012038, doi: 10.1088/1757-899x/1132/1/012038.
[4] AMINI, I. - KRISNAMURTI, W. W.: Compressive strength of geopolymer concrete with fly-ash from Paiton Steam Power Plant and variations of substitution sodium silicate (Na2SiO3) with natural zeolite, IOP Conf. Ser. Mater. Sci. Eng., 2020, vol. 930, no. 1, doi: 10.1088/1757-899X/930/1/012025.
[5] MA, C. - ZAWAWI, A. - OMAR, W.: Structural and material performance of geopolymer concrete: A review, Constr. Build. Mater., 2018, vol. 186, pp. 90–102, 2018, doi: 10.1016/j.conbuildmat.2018.07.111.
[6] VILLA, C. - PECINA, E. T. - TORRES, R. - GÓMEZ, L.: Geopolymer synthesis using alkaline activation of natural zeolite, Constr. Build. Mater., 2010, vol. 24, no. 11, pp. 2084–2090, doi: 10.1016/j.conbuildmat.2010.04.052.
[7] LIBAN, R. I. - KESKIN, Ü. S. - ÖZTÜRK, O.: Mechanical strength variation of zeolite-fly ash geopolymer mortars with different activator concentrations, Chall. J. Res. Lett., 2021, vol. 12, no. 3, pp. 96–103, doi: 10.20528/cjcrl.2021.03.003.
[8] ALEEM, M. I. A. - ARUMAIRAJ, P. D.: Geopolymer concrete – A review, Int. J. Eng. Sci. Emerg. Technol., 2012, vol. 1, no. 2, pp. 118–122, doi: 10.7323/ijeset/v1.
[9] SINGH, B. - ISHWARYA, G. - GUPTA, M. - BHATTACHARYYA, S. K.: Geopolymer concrete: A review of some recent developments, Constr. Build. Mater., 2015, vol. 85, pp. 78–90, doi: 10.1016/j.conbuildmat.2015.03.036.
[10] BAKHAREV, T.: Geopolymeric materials prepared using Class F fly ash and elevated temperature curing, Cem. Concr. Res., 2005, vol. 35, pp. 1224–1232, doi: 10.1016/j.cemconres.2004.06.031.
[11] KOZHUKHOVA, N. I. - TESLYA, A. Y.: Comparison study of Class F and Class C fly ashes as cement replacement material on strength development of non-cement mortar, Appl. Sci. Eng. Conf., 2018, vol. 288, no. 012019, doi: 10.1088/1757-899X/288/1/012019.
[12] NEMEC, J. - GANDEL, R. - JERABEK, J. - SUCHARDA, O. - BILEK, V.: Properties of selected alkali-activated materials for sustainable development, Civil and Environmental Engineering, vol. 20, no. 1, 2024, pp. 307–318, https://doi.org/10.2478/cee-2024-0024.
[13] MARK, O. - EDE, A. - ARUM, C. - JOLAYEMI, K.: Empirical modeling of high-performance self-compacting concrete with induction-furnace slag, Civil and Environmental Engineering, vol. 20, no. 1, 2024, pp. 440–460, https://doi.org/10.2478/cee-2024-0034.
[14] KURJI, B. M. - MUJTABA, I. M. - ABBAS, A. S.: Synthesis, characterizations, and recent applications of the silica-based Mobil Composition of Mesoporous Material: A review, Iraqi J. Chem. Pet. Eng., 2023, vol. 24, no. 3, pp. 1–12.
[15] NAGROCKIENE, D. - GIRSKAS, G.: Research into the properties of concrete modified with natural zeolite addition, Constr. Build. Mater., 2016, vol. 113, pp. 964–969, doi: 10.1016/j.conbuildmat.2016.03.133.
[16] ASTM C618: Standard specification for coal fly ash or calcined natural pozzolan for use in concrete, Am. Soc. Test. Mater, 2005.

[17] HILAL, A. A. - MAHMOOD, F. I.: On production of pre-formed foamed geopolymer concrete, in 2018 11th International Conference on Developments in eSystems Engineering (DeSE), 2018, pp. 290–294.
[18] OBAID, H. A. - HILAL, A. A.: Foam concrete made with micro and nano silica sand: Pore structure and properties, Adv. Concr. Constr., 2021, vol. 12, no. 3, pp. 207–216.
[19] ASTM C513: Standard test method for obtaining and testing specimens of hardened lightweight insulating concrete for compressive strength, Am. Soc. Test. Mater., 2019.
[20] ASTM C78: Standard test method for flexural strength of concrete (using simple beam with third-point loading), Am. Soc. Test. Mater., 2015.
[21] ZHANG, Z. - PROVIS, J. L. - REID, A. - WANG, H.: Geopolymer foam concrete: An emerging material for sustainable construction, Constr. Build. Mater., 2014, vol. 56, pp. 113–127, doi: 10.1016/j.conbuildmat.2014.01.081.
[22] WATTIMENA, O. - ANTONI, A. - HARDJITO, D.: A review on the effect of fly ash characteristics and their variations on the synthesis of fly ash-based geopolymer, in AIP Conference Proceedings, 2017, vol. 1887, p. 20041, doi: 10.1063/1.5003524.
[23] ANTONI, S. - WIJAYA, W. - HARDJITO, D.: Factors affecting the setting time of fly ash-based geopolymer, Mater. Sci. Forum, 2016, vol. 841, pp. 90–97, doi: 10.4028/www.scientific.net/MSF.841.90.
[24] AL BAKRI, A. M. - KAMARUDIN, H. - OMAR, A. - NORAZIAN, M.: The effect of alkaline activator ratio on the compressive strength of fly ash-based geopolymer paste, IOP Conf. Ser. Mater. Sci. Eng., 2011, vol. 5, pp. 1916–1922, doi: 10.1088/1757-899X/209/1/012064.
[25] NADER, A. S. - HILAL, A. A. - ALWAN, G. H.: Investigating the properties of foamed fly ash and metakaolin-based geopolymer concrete, Civil Engineering and Architecture, 2022, vol. 10, no. 7, pp. 2820–2832.
[26] HILAL, A. A.: Effect of aggregate roughness on strength and permeation characteristics of lightweight aggregate concrete, Journal of Engineering, vol. 2021, Article ID 9505625, 8 pages.