Preparation and characterization of lightweight geopolymer composites using different aluminium precursors

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Abstract

Production of lightweight building materials attract attention of the scientists worldwide with the need for reducing the structures dead-weight, provide better thermal insulation for buildings, and cost less to transport. The current work focuses on production of lightweight geopolymer composites by the incorporation of aluminium powder and aluminium slag in various ratios for water cooled slag/ kaolinite sand composite, the activators used were 6% of equal ratio from sodium hydroxide and sodium silicate. The properties of the produced lightweight geopolymer composites have been studied through measurement of compressive strength, bulk density, water absorption, FTIR, XRD and SEM imaging. Results demonstrate modification for both physico-mechanical and microstructural characteristics by using aluminium powder and aluminium slag forming lightweight composites with density below 2.15 g/cm³ depending on the studied mix composition.

Keywords: lightweight geopolymer, activation, aluminium, slag
Kulcsszavak: könnyű geopolimer, aktiválás, alumínium, kohósalak

1. Introduction

Today, the need for production of new ecofriendly building materials from country's natural resources and waste by-products has been increased vastly with an increase of general awareness about environmental issues which led to a growing concern over waste generation and the realization that such materials must be treated, eliminated or re-used. Rising consumption and the concomitant increase in industrial production has occasioned both a rapid decline in natural resources and generation of large volumes of waste or by-products [1]. Recent researches into the economically viable re-use of wastes have given rise to different proposals, including the possibility of its inclusion in building materials [2,3].

These materials categorized under the alkali activated materials and commonly known as geopolymer if their resources are from nature geological origins rich in silicon and aluminium, whilst industrial by-products rich in alumina and silica oxides such as coal fly-ash, silica fume and granulated blast furnace slag considered as alkali activated materials which is the major category, while the geopolymers can be considered as subset from this mother set as coincide with the pioneer scientists Davidovits in 1970s [4,5], who discovered and established of the research in geopolymer binders.

The basic binding phase in geopolymers is an amorphous aluminosilicate gel that consists of a three dimensional framework of SiO₄ and AlO₄ tetrahedra linked by corner-shared O atoms [6-8]. Geopolymer binders hold many advanced properties over traditional ones although the most important is their low manufacturing energy consumption and low CO₂ emission [9,10], which can be sorted as a *Green Material* [11]. The important of green materials increased also vastly with the increased ability for producing lightweight building materials in construction sector that can insulate heat and sound, where traditionally, ordinary Portland cement (OPC) has been used as the binder for concrete

[12-14], exhibits high thermal transmission and said to be responsible for about 7% of the CO₂ emission worldwide.

The development of lightweight building materials with good mechanical properties can reduce the deadweight of structures, provide better thermal insulation for buildings, and cost less to transport and erect when prefabricated structural components are made in factory, whereas using lightweight aggregates is one of the most common known ways for making lightweight building materials [15,16].

The use of lower densities concrete is beneficial in terms of structural load-bearing, and as acoustic and thermal insulation. The density can be reduced by replacing part of the solids by air voids or lightweight aggregates, several approaches exist for this objective. In the method known as no-fines concrete, the fine aggregate is omitted; whereas in the called lightweight concretes, the normal aggregates are replaced by lightweight to reduce the density. If air voids are introduced into the cement paste to reduce its density, the concrete is known as cellular, aerated, or foamed concrete [17,18], which can reduce the quantity of raw material (sand, cement and lime) of up to 30%, which knocks down the cost of building materials.

There are many types of materials that can be suitable for lightweight thermal insulation and fall under the following basic materials and composites [19, 20]:

1. Inorganic materials
 - a. Fibrous materials such as glass, rock, and slag wool, fly ash.
 - b. Cellular materials such as calcium silicate, bonded perlite, vermiculite, ceramic products and geopolymer.
2. Organic materials
 - a. Fibrous materials such as cellulose, cotton, wood, pulp, cane, or synthetic fibers.
 - b. Cellular materials such as cork, foamed rubber, polystyrene, polyethylene, polyurethane, poly-isocyanurate and other polymers.

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Various routes are available to suit lightweight production, one of such chemicals to generate gas within the mixture is metallic aluminium powder, which is very reactive in alkaline environments, such as calcium hydroxide or alkaline hydroxides; the aluminium dissolves as aluminate Al_2O and H_2 gas is liberated and trapped within the cementitious paste or mortar, expanding and increasing the volume. In order to prevent the escape of the gas, the paste must have a proper consistency and fast setting times according to the following reaction [21,22]:

Another low cost material for lightweight production and available in greater quantities is aluminium slag (dross) acts which incorporates air into the mixture and can be used in the manufacturing of building blocks (bricks), pre-molded panels, subfloors and other kinds of surfaces. Approximately, up to 4 million tons of white dross and more than a million tons of black dross are reported throughout the world each year, and around 95% of this material is landfilled [23, 24].

This material also can be used raw materials in cement production industry as proved by Puertas et al. [25] as well as Pereira et al. [26] who explored partial replacement of aluminium dross to cement mortar in certain limits to obtain environmental and economic advantages. Others recommended the usage of aluminium dross as filler in concrete bricks and non-aerated concrete [27].

The objective of this contribution is to investigate the effect of aluminium powder and aluminium slag into the geopolymer composites as gas liberating agent and producing lightweight geopolymer structures. The other target is to study the effect of added materials on the hardened composites by monitoring their impacts on physico-mechanical and microstructural characteristics. X-ray diffraction and FTIR are used for investigation of the resulted composite structure.

2. Experimental procedures

2.1. Materials

Materials used were ground granulated blast furnace slag (GGBFS) sourced from Iron and Steel Factory - Helwan, Egypt, and kaolinite sand material brought from Middle East for Mining Investment Co., Egypt. Materials used for production of lightweight composites were aluminium slag procured from Nagh Hammadi Factory for aluminium production, Egypt, where it produced from recovery of aluminium through scrap recycling, where a great quantity of oxide layer formed on the surface of molten metal and has to be removed from the melt to refine the quality of the final product. This material has dual effect in geopolymer composite formation; one is the participation in the geopolymer formation as it contains

both alumina and silicon oxides, the second is the very high active alumina content which acts as initiator for lightweight products.

Aluminium powder (Adwic chemicals, Egypt, 94% of purity) used as hydrogen gas liberating materials for geopolymer composites. Sodium hydroxide (NaOH) with purity 99 % in the form of pellets used as alkali activators, obtained from Piochem Co., Egypt, while liquid sodium silicate (LSS, $\text{Na}_2\text{SiO}_3 \cdot 9\text{H}_2\text{O}$) from Fisher company consists of 32% SiO_2 and 17% Na_2O with Silica modulus $\text{SiO}_2/\text{Na}_2\text{O}$ equal 1.88 and its density is 1.46 g/cm^3 .

Chemical compositions of the starting raw materials are illustrated in Table 1, whereas the mineralogical characterization was done using X-ray diffraction analysis as represented in (Fig. 1). Blast furnace slag is a rich aluminosilicate material and composed from the dominant content of SiO_2 , CaO, Al_2O_3 , Fe_2O_3 , and MnO as illustrated from Table 1, while its mineralogical structure revealed the amorphous nature of this material.

Fig. 1. X-Ray diffraction pattern of the starting raw materials [K=Kaolinite; Q=Quartz; D=Diaoyudaoite, $\text{syn}(\text{Na}_{1.22}\text{Al}_{11}\text{O}_{17.11})$; S=Spinel, $\text{syn}(\text{Mg}_{0.35}\text{Al}_{0.65})(\text{Al}_{1.79}\text{O}_4)$; C=Corundum (Al_2O_3); L=Lime (CaO); F=Fayalite magnesian ($\text{Mg}_{0.75}\text{Fe}_{1.10}\text{Mn}_{0.15}\text{SiO}_4$); H=Halite (NaCl)]

1. ábra Alapanyagok röntgendiffraktogramjai [K=Kaolinit; Q=Kvarc; D=Diaoyudaoit ($\text{Na}_{1.22}\text{Al}_{11}\text{O}_{17.11}$); S=Spinell, ($\text{Mg}_{0.35}\text{Al}_{0.65})(\text{Al}_{1.79}\text{O}_4$); C=Korund (Al_2O_3); L=Egetett mész (CaO); F=Fayalit magnézia ($\text{Mg}_{0.75}\text{Fe}_{1.10}\text{Mn}_{0.15}\text{SiO}_4$); H=Halit (NaCl)]

Kaolinite sand showed that it has about 27% aluminium oxide as well as 64.5% of silicon oxide, in addition to little amount of calcium and magnesium oxides as presented in Table 1, however the mineralogical composition showed that kaolinite sand composed of about 75% quartz in addition to 25% kaolinite as shown in Fig. 1, the figure reflected also the crystalline nature of aluminium slag where most of its minerals are reach in alumina as Diaoyudaoite, Fayalite, spinel and corundum.

Material	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	K_2O	Na_2O	TiO_2	MnO_2	P_2O_5	Cl-	L.O.I.	SrO	BaO	Total
Kaolinitic sand	64.5	26.90	0.28	1.28	0.37	0.34	0.07	0.11	1.11	0.01	0.22	0.07	4.34	-	-	99.60
Ground granulated blast furnace slag (GGBFS)	36.67	10.31	0.50	38.82	1.70	2.17	1.03	0.48	0.57	4.04	0.04	0.05	0.12	0.18	3.28	99.96
Alumina slag	14.52	76.15	0.60	2.39	3.15	0.19	0.36	2.37	0.02	0.00	0.01	-	-	-	-	99.75

Table 1. Chemical composition of starting materials (m%)

1. táblázat Alapanyagok kémiai összetétele (m%)

Mix no.	Water cooled slag (WCS), %	Kaolinite sand, %	Al-slag, %	Al-powder, %	NaOH, %	Sodium silicate, %	Water/binder
A0	50	50	-	0.0000	3	3	0.25
A1	50	50	-	0.0125	3	3	0.25
A2	50	50	-	0.0250	3	3	0.25
A3	50	50	-	0.0375	3	3	0.25
A4	50	50	-	0.0500	3	3	0.25
A5	50	50	-	0.0625	3	3	0.25
A6	50	50	-	0.0750	3	3	0.25
AS1	50	50	1	-	3	3	0.225
AS2	50	50	2	-	3	3	0.25
AS3	50	50	3	-	3	3	0.275
AS4	50	50	4	-	3	3	0.288

Table 2. Composition of the geopolymers mixes
2. táblázat Geopolimer keverékek összetétele

2.3. Geopolymerization and curing

Geopolymer were set by mixing raw materials (passing 90 µm sieve) of each mixture with the alkaline solution as represented in Table 2 for 15 min with an electronic mixer. Aluminium powder was added to the binding material in small ratios from 0.0125 up to 0.075% with the increment of 0.0125% from the total weight; whereas aluminium slag was added in the ratio from 1 up to 4% from the total weight. The used aluminium materials mixed well with the total water incorporated the added activators for better production of foaming materials. All mixes were left to cure undisturbed under ambient temperature for 24 hrs, and then subjected to curing temperature of 40 °C with a 100 % relative humidity. At the end of curing regime, specimens were removed from their curing condition, dried well at 80 °C for 24 hrs then subjected to compressive strength measurements whereas the resulted crushed specimens were subjected to stopping of the hydration process by methyl alcohol/acetone method [28, 29] in order to prevent further hydration and then preserved in a well tight container until the time of testing.

2.4. Methods of investigation

Chemical composition of the started raw materials were performed using Axios, WD-XRF Sequential Spectrometer (Panalytical, Netherland, 2009). Compressive strength tests were performed according to ASTM-C109 [30] using German Brűf pressing machine. XRD analysis was carried out using Philips PW3050/60 Diffractometer. Microstructure of hardened materials was examined using FEI-SEM Inspect S equipped with an energy dispersive X-ray analyzer (EDX). Amorphous structure was traced using FTIR using potassium bromide binder, whereas their range from 400 to 4000 cm⁻¹ [31, 32].

Water absorption measurements of the bricks were carried out according to ASTM C140 [33]. The percentage absorption was calculated using the equation:

$$\text{Absorption (\%)} = [(W2 - W1) / W1] \times 100$$

where W1 = weight of specimen after complete drying at 105°C, W2 = final weight of surface dry sample after immersion in water for at least 24 hours.

The bulk density is calculated according to Archimedes, principle as in the following equation [34,35]:

$$\text{Bulk density} = D / (W - S) \text{ g.cm}^{-3}$$

Where;

D = weight of dried specimens

W = weight of soaked specimen suspended in air

S = weight of suspended specimens in water

3. Results and discussion

3.1. Effect of aluminium powder

FTIR spectra of neat geopolymer composite without aluminium powder cured up to 90 days are shown in Fig. 2. It can be noticed a decrease in kaolinite peaks beyond 3600 cm⁻¹ with time as a results of continuous dissolution and polymerization of residual kaolinite forming additional geopolymer constituents leading to an increased reinforcement of the matrix, this confirmed by an increased intensity of the asymmetric stretching vibration (Ti-O-Si) at about 1000 cm⁻¹ where T=Si or Al, as resulted from the increased geopolymerization and precipitation, this occur in conjunction with an increased intensity of symmetric stretching vibration (Al-O-Si) at about 780 cm⁻¹ as well as symmetric stretching vibration (Si-O-Si) at about 670-685 cm⁻¹. Also, regions close to 460 cm⁻¹ attributed to angular deformations of SiO₄ tetrahedrons (O-Si-O), as well as asymmetric stretching vibration for Si-O-Si (for non-solubilized particles) probably associated with quartz [36], these results are similar to those obtained by Palomo et al. [37]. Stretching vibration for Al-OH at about 910 cm⁻¹ resulted from free unreacted aluminium species decreased also with time as unreacted free alumina exposed to dissolution and polymerization with time [38].

It can be noticed a shift in the wave number of main asymmetric band to right with an increased intensity up on time reflecting the increase of vitreous content which agrees with the observation that the incorporation of kaolinite within the sand retards polymerization reaction in the very early period of reaction [39, 40], while at later ages an increased dissolution of Al from kaolinite sand leading to an enhancing polymerization and modifying the chemistry of the Si-O-T bonds (giving a decrease in the wavenumber) [39, 40]

Fig. 2. FTIR spectra of control geopolymer cured up to 90 days in 40 °C with 100% R.H.) [1,2,3: Stretching vibration of O-H bond for kaolin; 4: Bending vibrations of (HOH); 5: Stretching vibration of CO₂; 6: Asymmetric stretching vibration (Si-O-Si); 7: Asymmetric stretching vibration (T-O-Si); 8: Symmetric stretching vibration of Al-OH; 9: Symmetric stretching vibration of CO₂; 10: Symmetric stretching vibration (Al-O-Si); 11: Symmetric stretching vibration (Si-O-Si); 12,13: Bending vibration (Si-O-Si and O-Si-O)]

2. ábra 90 napos korú (40 °C, 100% RH) geopolimer keverékek FTIR spektrumai [1,2,3: Vegyértékrengés O-H kötés (kaolin); 4: Deformációs rezgés (HOH); 5: Vegyértékrengés (CO₂); 6: Aszimmetrikus vegyértékrengés (Si-O-Si); 7: Aszimmetrikus vegyértékrengés (T-O-Si); 8: Szimmetrikus vegyértékrengés (Al-OH); 9: Szimmetrikus vegyértékrengés (CO₂); 10: Szimmetrikus vegyértékrengés (Al-O-Si); 11: Szimmetrikus vegyértékrengés (Si-O-Si); 12,13: Deformációs rezgés (Si-O-Si és O-Si-O)]

Fig. 3. FTIR spectra of 28 days lightweight geopolymer composite having various doses of aluminium powder [1,2,3: Stretching vibration of O-H bond for kaolin; 4: Bending vibrations of (HOH); 5: Stretching vibration of CO₂; 6: Asymmetric stretching vibration (Si-O-Si); 7: Asymmetric stretching vibration (T-O-Si); 8: Symmetric stretching vibration of Al-OH; 9: Symmetric stretching vibration of CO₂; 10: Symmetric stretching vibration (Al-O-Si); 11: Symmetric stretching vibration (Si-O-Si); 12,13: Bending vibration (Si-O-Si and O-Si-O)]

3. ábra 28 napos korú könnyű geopolimer kompozitok FTIR spektrumai különböző mennyiségű alumínium por adagolás mellett [1,2,3: Vegyértékrengés O-H kötés (kaolin); 4: Deformációs rezgés (HOH); 5: Vegyértékrengés (CO₂); 6: Aszimmetrikus vegyértékrengés (Si-O-Si); 7: Aszimmetrikus vegyértékrengés (T-O-Si); 8: Szimmetrikus vegyértékrengés (Al-OH); 9: Szimmetrikus vegyértékrengés (CO₂); 10: Szimmetrikus vegyértékrengés (Al-O-Si); 11: Szimmetrikus vegyértékrengés (Si-O-Si); 12,13: Deformációs rezgés (Si-O-Si és O-Si-O)]

On investigating the effect of addition of various ratios of aluminium powder up to 0.075% cured at 28 days (Fig. 3), first there is a slight decrease in the main asymmetric band up on adding 0.025% aluminium as a results of alteration in the Si/Al ratio, however a gradual increase in the band intensity with shifting to right can be recognized with increasing the aluminium powder up to 0.0625% where the added alumina results in enhancing the degree of polymerization by the effect of seeded nucleation results in the formation of nucleation sites for geopolymer formation and precipitation which positively enhance the resulted geopolymer structure [41], also this shift

indicating the formation of a new product (the amorphous aluminosilicate gel phase) [31]. However, further increase in the aluminium powder results in an increase of liberated hydrogen gas as mentioned previously up on reacting with alkali, this gas results in an increased porosity as well as hinder interaction between binding materials leading to a lowering in intensity of the main asymmetric band with increased intensity of kaolinite bands reflecting low dissolution rate of the kaolinite precursor as compared with the lower aluminium powder ratio, additionally carbonate bands at about 1440 and 870 cm⁻¹, these bands exposed to an increased intensity with aluminium powder up to 0.0625 % then decreased up on using 0.075% aluminium powders which can be linked by the behaviour of asymmetric vibration of T-O-Si as hydrogen gas which hinder intact between reacting particle leads to lowering in carbonation rate of geopolymer composites at higher aluminium powder ratio.

Fig. 4 shows the XRD pattern of 28 days alkali activated geopolymer composites incorporating various ratios of aluminium powder, where a gradual increase in the amorphous geopolymer constituents with aluminium powder up to 0.0625% as represented by increased broadness with peak distortion between 20-35° 2θ characterizing glassy phase of geopolymer constituents, however this broadness decreased with further aluminium increase as well as up on using lower doses as come in coincide with FTIR. The increased intensity of amorphous constituents by formation of new phases come in alignment with the increased broadness in the range from 6-10° 2θ for aluminosilicate gel, where this increase reflects the increased dissolution of geopolymer precursor [42].

Fig. 4. XRD pattern of 28 days lightweight geopolymer composite having various aluminium powder ratios [K=Kaolinite; Q=Quartz; Fj: Faujasite; CSH: Calcium silicate hydrate; ZF: Zeolite Na-F]

4. ábra 28 napos korú könnyű geopolimer kompozitok röntgendiffraktogramjai különböző mennyiségű alumínium por adagolás mellett [K=Kaolinit; Q=Kvarc; Fj: Faujasit; CSH: Kalcium szilikát hidrát; ZF: Zeolit Na-F]

Another more likely reason is that the unique conditions in the early stages of seeded geopolymer formation by Al substitutes that may have created an environment where zeolite Na-F was able to be directly synthesized by hydrothermal treatment of kaolinite or metakaolin at relatively low temperatures (~80 °C) [43, 44]. These phases his Al-rich gel, which does not occur in the unseeded system, also explains the formation of Na-F [45]. On further increase in alumina powder; there is an intense peak for Faujasite of zeolite favoured the lower geopolymer characteristic as known by their lower branching ability, the

previous notice comes in accordance with Zuhua et al. [46], where the crystalline zeolite phases are destructive to the consistent distribution of geopolymers.

Fig. 5. Compressive strength of lightweight geopolymer composite having various aluminum powder ratios, cured up to 90 days

5. ábra 90 napos korú könnyű geopolimer kompozitok nyomószilárdsága különböző mennyiségű alumínium por adagolás mellett

The results of compressive strength and absorption as well as bulk density for hardened geopolymer composite incorporated various ratio of aluminium powder, cured in 100 % relative humidity at 40°C up to 90 days are shown in Figs. 5-6. The results showed an increase in strength for all mixes along with hydration age as attributed to the continuing pozzolanic reaction as well as continuous growth of geopolymer chains forming tightly bound structure. Dissolving aluminium powder with its high reactivity in alkaline environment transforming into aluminate species and giving off free hydrogen gas. The aluminate species can promote the reaction by the effect of seeded nucleation as mentioned latter and enhance propagation as well as precipitation of geopolymer chains, while the liberated gas can facilitate the formation of lightweight composites with lower density as compared with common building materials units.

Fig. 6. Bulk density and water absorption of lightweight geopolymer composite having various aluminium powder ratios

6. ábra Könnyű geopolimer kompozitok testsűrűsége és vízfelvevétele különböző mennyiségű alumínium por adagolás mellett

There is a sharp strength decrease can be elucidated from the figure with the addition of 0.0125% aluminium, as explained from both FTIR and XRD as the alteration in Si/Al ratio lead to a decrease in strength. Increasing the ratio of the added alumina, results in a gradual increase in strength,

reaching the maximum values at 0.0625% followed by rapid decrease with further alumina increase. The strength increase can be explained by the increased asymmetric stretching vibration of amorphous glassy geopolymer phases at about 1000 cm⁻¹ where the seeded nucleation effect of the liberated aluminate enhanced the nucleation and geopolymerization of reacting materials, forming zeolite Na-F which known by its ability to strengthen the structure. However, further increase in the aluminium powder results in an extra increase in the liberated hydrogen gas which compensate the activation role of aluminate species by absorption increase and so hinder interaction between the reacting species and so lower the intensity of the formed geopolymer as well as strength

We can notice that the strength values were 214.0, 271.0, 112.4 kg/cm² at 7 days and 239.5, 350.0, 109.1 kg/cm² at 28 days for 0, 0.0625, 0.075 % aluminium powder, respectively, however the density values are 1.874, 2.010, 1.599 g/cm³ at 7 days and 1.886, 2.016, 1.598 g/cm³ at 28 days for the same ratios. Strength pattern also shows that values of the formed green composite incorporating 0.0625 % aluminium powder are much harder than OPC cement according to ASTM C150 [47] where its standard strength values are 200 and 294 kg/cm² at 7 and 28 days, whereas the produced lightweight product with the previously mentioned ratio is much harder in addition to its low production cost as most of the used materials are wastes, however cement uses primary raw materials.

It was recognized that lightweight concrete can be classified according to the purpose of utilizing as: 1) structural lightweight concrete with cylinder compressive strength at 28 days equal or more than 17 MPa and the approximate density range 1400-1800 kg/m³ 2) masonry concrete (structural / insulating lightweight concrete) has a compressive strength between 7-14 MPa and density range 500-800 kg/m³ 3) insulating concrete has a compressive strength between 0.7-7 MPa and density lower than 800 kg/m³ [48, 49].

So the produced product can be used effectively in the production of high-strength lightweight concrete with strength values (20 to 35 MPa). On the other hand lightweight structural concrete is required under European standards to have and density of less than 2000 kg/m³ (2.0 g/cm³), where most of the studied composites have density values ≤ 2.0 g/cm³ at all curing ages which reflects the lightweight characteristics of composites.

Fig. 7. a, shows the typical microstructure of 90 days control geopolymer matrix free from aluminium powder, this matrix exhibits dense morphology with spreading unreacted kaolinite laminar in addition to geopolymer networks which come in accordance with kaolinite bands in FTIR chart. On adding 0.0625% Al powder (Fig. 7. b), an increased dissolution of kaolinite constituent observed as shown from the matrix nearly free from kaolinite laminas which conform the seeding nucleation effect of added alumina leading to modification of the resulted geopolymer by an increased intensity, this can be illustrated clearly from the cohesion of the formed matrix. Another role of the added alumina is to form lightweight composite by releasing free hydrogen gas by alkaline interaction; this confirmed by the micropores within reacted particles. On increasing aluminium powder to 0.075%

an increased liberation of hydrogen gas noticed from the formation of wide pores within the matrix, this will adversely inhibit the intact between formed geopolymer networks and so lower their mechanical properties (Fig. 7. c).

Fig. 7. SEM images of 90 days lightweight geopolymer composite having various aluminium powder ratios [A: 0%; B: 0.0625%; C: 0.075%]

7. ábra 90 napos korú könnyű geopolimer kompozitok elektronmikroszkópos felvételei különböző mennyiségű alumínium por adagolás mellett [A: 0%; B: 0.0625%; C: 0.075%]

3.2. Effect of aluminium slag

On studying the effect of aluminium slag using FTIR for 28 days geopolymer composites incorporating various aluminium slag ratio up to 4% (Fig. 8), there is a marked decrease in the main asymmetric band with the addition of 1% aluminium slag as a results of alteration in the Si/Al ratio, whilst there is a progressing increase in the asymmetric band with slight shifting to right by increasing aluminium slag to 3% where the used slag results in modification of the geopolymer structure and formation of vitreous content [41], also shifting of the band to lower wave number favours the increased amorphous gel phase as stated before [31]. On prolonged investigation of the pattern there is a growth in carbonate band with aluminium slag (3%) which can be considered as beneficial in formation of cellular composite as compared with aluminium powder in spite the strength is lower than that of aluminium powder.

Increasing aluminium slag to 4%, results in the increase of crystalline minerals associated with aluminium slag which adversely hinder geopolymerization reaction in addition to diverse effect of hydrogen gas in increased porosity and so hinder the interaction between reacting particles as reflected on the decreased intensity of the main asymmetric band.

The pattern clarified the fixed intensity of kaolinite bands beyond 3600 cm^{-1} which may be due to the lower efficiency of aluminium slag in enhancing geopolymerization reaction as compared with aluminium powder. There is an additional notice about carbonate bands at about 1440 and 870 cm^{-1} , these bands decreased up on using 1 % aluminium slag while increased again with 3 % aluminium slag and then exposed to

sharp decrease with further increase in Al-slag. This can be linked by the behaviour of asymmetric vibration of T-O-Si as hydrogen gas which hinder intact between reacting particles leads to lowering in carbonation rate of geopolymer composites at higher aluminium slag (4%).

Fig. 8. FTIR spectra of 28 days lightweight geopolymer composite having various doses of aluminium slag [1,2,3: Stretching vibration of O-H bond for kaolin; 4: Bending vibrations of (HOH); 5: Stretching vibration of CO₂; 6: Asymmetric stretching vibration (Si-O-Si); 7: Asymmetric stretching vibration (T-O-Si); 8: Symmetric stretching vibration of Al-OH; 9: Symmetric stretching vibration of CO₂; 10: Symmetric stretching vibration (Al-O-Si); 11: Symmetric stretching vibration (Si-O-Si); 12, 13: Bending vibration (Si-O-Si and O-Si-O)]

8. ábra 28 napos korú könnyű geopolimer kompozitok FTIR spektrumai különböző mennyiségű alumínium salak adagolás mellett [1,2,3: Vegyértékrengés O-H kötés (kaolin); 4: Deformációs rezgés (HOH); 5: Vegyértékrengés (CO₂); 6: Aszimmetrikus vegyértékrengés (Si-O-Si); 7: Aszimmetrikus vegyértékrengés (T-O-Si); 8: Szimmetrikus vegyértékrengés (Al-OH); 9: Szimmetrikus vegyértékrengés (CO₂); 10: Szimmetrikus vegyértékrengés (Al-O-Si); 11: Szimmetrikus vegyértékrengés (Si-O-Si); 12,13: Deformációs rezgés (Si-O-Si és O-Si-O)]

Fig. 9. XRD pattern of 28 days lightweight geopolymer composite having various aluminium slag ratios [K=Kaolinite; Q=Quartz; CSH: Calcium silicate hydrate; ZF: Zeolite Na-F; C: Calcite]

9. ábra 28 napos korú könnyű geopolimer kompozitok röntgendiffraktogramjai különböző mennyiségű alumínium salak adagolás mellett [K=Kaolinit; Q=Kvarc; CSH: Kalcium szilikát hidrát; ZF: Zeolit Na-F; C: Kalcit]

Fig. 9 shows the XRD pattern of 28 days geopolymer composites incorporating various aluminium slag ratio, where there is a similarity between this pattern and those for aluminium powder pattern as a gradual increase in the amorphous geopolymer constituents with increase of aluminium slag up to 3%, observed as well as an increased broadness in the range between 20-35° 2θ for amorphous geopolymer constituents, while the main difference between them is an increased Faujasite up on using higher aluminium powder. However, this is not the case in aluminium slag mixes having higher ratio (4%) where no Faujasite formed as the

increased crystalline phases in slag constituents hinder the reaction and so a lower intensity of both asymmetric stretching as well as Faujasite, this in coincide with the low dissolution rate of kaolinite constituents.

On investigating compressive strength, density and water adsorption results for hardened geopolymer composite incorporated various ratio of aluminium slag, cured up to 90 days (Figs. 10-11), there is an analogous behaviour to that of aluminium powder where a gradual increase with time as well as with aluminium slag up to 3 % but still lower than that of control mix as a results of lower efficiency of aluminium slag in enhancing the reaction.

Fig. 10. Compressive strength of lightweight geopolymer composite having various aluminium slag ratios, cured up to 90 days

10. ábra 90 napos korú könnyű geopolimer kompozitok nyomószilárdsága különböző mennyiségű alumínium salak adagolás mellett

Fig. 11. Bulk density and water absorption of lightweight geopolymer composite having various aluminium slag ratios

11. ábra Könnyű geopolimer kompozitok testsűrűsége és vízfelvétele különböző mennyiségű alumínium salak adagolás mellett

First, strength decreases sharply upon using 1% slag precursor as a results of alteration of silica to alumina ratio, whereas strength increases with further aluminium slag to 3% as illustrated in details before about the efficiency of aluminium species in activating the reaction by seeding nucleation principle, while the liberated gas can facilitate the formation of lightweight composites with lower density as compared with common building materials units. On further increase in aluminium slag to 4%, strength decreases again as the intensity of zeolite F decreased which known by its ability to enhance the formed structure, in addition to decreased intensity of

asymmetric geopolymer band in FTIR pattern, where the increased aluminium slag with its crystalline constituents hinder the formation of geopolymer chains as well as increased liberated gases decreased strength sharply by isolation of the interacting particles.

On comparing with strength values of aluminium powder specimens, a lower strength values of Al-slag species as compared with aluminium powder as related to the lower reactivity of aluminium slag and its crystalline structure. The strength values were 214.0, 91.2, 32.7 kg/cm² at 7 days and 239.5, 133, 63.8 kg/cm² at 28 days for 0, 3, 4 % aluminium slag, respectively, however the density values are 1.874, 1.417, 1.275 g/cm³ at 7 days and 1.886, 1.665, 1.406 g/cm³ at 28 days for the same ratios. Whereas the absorption values are 14.54, 13.19, 18.56 % at 7 days and 11.10, 11.93, 16.05 % at 28 days as illustrated in Fig. 11.

An important conclusion is that the strength values of the formed geopolymer composite incorporating 0.0625% aluminium powder are much harder than aluminium slag, however from the point of view of lightweight principles; the density of the Al-slag species are lower than that of aluminium powder composites while the water absorption values are higher, which can be considered as a very important from the point of view of lightweight composites as the density is lower by about one third and the absorption higher by about the same ratio.

On classifying aluminium slag composites according to previously mentioned classification [48, 49], they can be suitable for masonry concrete (structural / insulating lightweight concrete) which has a compressive strength between 7-14 MPa and density range 500-800 kg/m³, as well as insulating concrete has a compressive strength between 0.7-7 kg/m³ and density lower than 800 kg/m³.

On comparing the morphology of cellular geopolymer composites incorporating aluminium powder and aluminium slag; a clear difference between their microstructure observed where the 3% aluminium slag (Fig. 12.a) enhanced composite shows low cohesion between their matrix as well as spreading of unreacted kaolinite within its morphological structure, this reflecting low enhancing ability of aluminium slag for activation and nucleation of geopolymer precursors as coincided by their low mechanical characteristics as compared with optimum aluminium powder enhanced geopolymer mix (Fig. 12.b) which possess dense structure with little evidence of unreacted kaolinite.

Fig. 12. SEM images of 90 days lightweight geopolymer composite doped with A: 3% Al slag; B: 0.0625% Al powder

12. ábra 90 napos korú könnyű geopolimer kompozitok elektronmikroszkópos felvételei A: 3% Al salak; B: 0.0625% Al por

4. Conclusions

The paper deals with incorporation of aluminium substituents as hydrogen gas emitting materials for lighting the density of the resultant geopolymer composites, while the raw materials for the base geopolymer mixes made from waste materials for lowering the production costs, there are some important findings that can be concluded from the present paper:

Aluminium powder and aluminium slag can be used efficiently in production of lightweight geopolymer materials resulting in low density composites as compared with most common lightweight materials, where density of most cement concrete was 2.2 to 2.60 g/cm³.

Aluminium powder can be used up to 0.075% with acceptable properties and with density values ≤ 2.0 g/cm³ and decreased with aluminium substituents increase, while 0.0625 % can possess both high mechanical and morphological advantages with densities in the same previous range whereas their 28 strength values exceeds 350 kg/cm² which can be used effectively in the production of high-strength lightweight concrete.

Aluminium slag materials can be used without significant loss in the mechanical and physical properties up to 4%, while using 3% possess optimum mechanical characteristics with density values lower than 1.70 g/cm³.

In spite strength values of Al- slag species are lower than aluminium powder, their density values are lower and absorption are higher by about one third, which encourage its beneficial use in lightweight production.

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