

# THE INFLUENCE OF GRAPHITE ADDITIONS ON THE PROPERTIES OF FOAMED GEOPOLYMERS BASED ON GLASS WASTE

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*Foaming geopolymers (light-weight porous inorganic materials) were successfully synthesized by alkali activator of mixed color waste glass powder without/with pure graphite powder (0.1 wt%) and dry intercalated graphite powder(0.1 wt%) as foaming agents; the specimens were thermally treated at 500,600,700 and 800°C for 1hour. An important volume increase was recorded (-11.8-240.68%), (68.25-146.64%) and (43.79-968.54%) for MG-N5, MGG-N5 and MGIG-N5 respectively after thermal treatment. A sharp increase in volume (important swelling) and shape deformation was recorded for the specimens with intercalated graphite (MGIG-N5 pastes) at temperature 600°C. The microstructure and mechanical properties of these materials were also studied. The foaming geopolymers containing glass and graphite powder (G or IG) which were obtained from this work are strong candidates mostly for the applications demanding thermal insulation with favorable combination of light weight, porosity, mechanical strength and performance to produce construction materials.*

**Keywords:** Glass waste; graphite; foam geopolymers; thermal treatment

## 1. Introduction

Geopolymers (inorganic polymers) considered as relatively new materials which has possess a special properties and it can be used widely in various industry applications. Currently, these new materials are mainly applied in the construction and in the future may be replace the conventional concrete based on Portland cement. Actually, geopolymers are more desirable especially may find these new materials at anywhere which have applications used in the liners of cars and airplanes or used as foaming panels for the thermal insulation in shields for aircraft or in the building materials industry and as a composites materials for fireproof properties[1,2].

In 1970, geopolymers have been used as fireproof materials-a core (made of wood) covered with two layers of geopolymers [3].

Foamed geopolymers (inorganic polymers) are light materials which can be applied in numerous areas of industry, particularly in building materials [4,5]. It is possible to obtain inorganic polymers as well as foams (porous materials) from glass precursors [5-8]. The phenomenon of intumescent can be obtained by the thermal treatment of inorganic polymers based on glass waste [7,8]. Also, the low durability in humid medium of inorganic polymers based on glass waste powder can be settled by a minor heating processing [9,10].

The glass foams processing at low temperature is favorable methods which reuse glass waste [11]. Moreover, the reuse of the glass waste as solid materials considered as one of the most attractive and effective choices [12-14]. In addition, the recycling of glass used as raw materials minimizes the power consumption, extraction, processing and transportation of minerals [6].

Foaming materials based on glass were synthesized by thermal treatment of different types of glass powders or other sources of aluminosilicate minerals, in combination with swelling agents [15]; in other works others minerals like (metakaolin, blast furnace slag and fly ash) were added in order to achieve a high ratio of silica content[16,17]. The materials which was used as blowing agents includes aluminum nitride [17], magnesium carbonate, calcium carbonate, eggshells, calcite and dolomite [18], manganese dioxide and silicon carbide (SiC) [19], carbon and graphite [5].

Chemical intumescent systems have been used as flame retardants for nearly 50 years. The effectiveness of these flame retardants depends on the heat-induced decomposition of the organic components, and the creation of a char layer that insulates the substrate from the heat source. However, as intumescent materials are required to address more severe and diverse applications, new approaches are needed that provide improved performance over conventional systems [20].

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Table 1

The composition of studied geopolymers				
Sample	Solid component		Liquid component	Liquid/Solid Ratio
MG-N5	Mixed color glass powder (MG)	Without additive	NaOH solution (N5)	0.3
MGG-N5	Mixed color glass powder (MG)	Graphite powder(G) 0.1wt%		
MGIG-N5	Mixed color glass powder (MG)	Intercalated graphite (IG) 0.1wt%		

Intumescent materials which swells under firing were produced by the alkaline activation of glass waste powder with borax additions [8,21] or of a mixture of glass waste powder and fly ash [22] or slag [23].

A relatively new intumescent admixture is expandable graphite (EG) that can be produced by the treatment of flake graphite and mixture of acids and some oxidizing agents [24-27]. Expandable graphite can also improve many properties (barrier properties, resistance to aggressive mediums and resistance to weathering) of coating under normal operation.

Expandable graphite EG, is prepared by inserting a certain reactant into the layers of graphite through chemical or electrochemical reaction (intercalating graphite-IG). The intercalation compound is also known as expandable graphite, since it can be exfoliated upon heating [28]. When this material (IG) is heated, it will expand. EG is a good intumescent flame – retardant. When EG is exposes to flame, it will expand, and its cellular structure acts as physical barrier for mass and heat transfer [29].

This work aims to investigate the reuse of glass waste in the manufacture of intumescent materials with graphite content. The glass foams obtained in the research can be potentially used as thermal and acoustic insulations.

## 2. Experimental

### 2.1. Materials

In this work, glass cullet from bottles with different colors (transparent, green and brown) was grinded in order to obtain a mixed glass waste powder (MG); MG was used as a main solid component.

The oxide compositions glass waste powder (MG) is: silica (76.71 wt%), two alkalis ( $\text{Na}_2\text{O} + \text{K}_2\text{O}$ ) (15.87 wt%), calcium oxide (4.96 wt%), aluminum oxide (0.83 wt%), iron oxide (0.55 wt%) and MgO (0.58 wt%). The Blaine specific surface area of glass waste powder (MG) was 3035  $\text{cm}^2/\text{g}$  and the average particle size ( $D_{0.5}$ ) is 41.778  $\mu\text{m}$  [30].

Graphite powder (99%, Sigma Aldrich, Germany) was used as pore forming agent; two types of graphite powders were used:

1. Pure graphite powder (G)
  2. Intercalated Graphite (IG)
- IG was prepared by mixing graphite powder(G)

with Nitric Acid ( $\text{HNO}_3$ , 69%, 14.6 M, CDH, India) and Potassium permanganate ( $\text{KMnO}_4$ , 99%, Merck, Germany) in weight ratio (1:2:1) [31]. The components were mixed for 3 minutes and the resulted mixture was washed several times with distilled water using vacuum filtration until the pH reading was 7; the resulted material was dried at 110°C for 6 h to obtain intercalation graphite (IG) [32]. The resulting product stored maximum 24 hours in the covered polystyrene container before its use.

The liquid component was sodium hydroxide NaOH 5M (N5). The alkali activator solution 5M NaOH as liquid component was prepared at least 2 h prior to its use, in order cool down at room temperature.

Three types of alkali activated materials (geopolymers) were prepared (see Table1).

The NaOH solution was added to the mixture (MG without/with G or IG) corresponding to a liquid to solid ratio equal 0.3 (Table1); the workability of pastes was good. The resulted pastes were casted in a cuboids mold (20x20x20mm) and vibrated for 2-3 minutes. The molds with the paste were wrapped with cling film and were cured in the electric oven at 60 °C for 1 day, then de-molded and subsequently cured at ambient temperature ( $20 \pm 2$  °C) up to 7 days.

After this initial curing (1 day at 60°C + 6 days at 20°C), the specimens were thermally treated for 1 h at various temperatures (500, 600,700 and 800 °C) in the furnace (heating rate was 10°C/minute). The thermally treated specimens were cooled slowly in the furnace for 24 h. These experiments (thermal treatments) were done twice for each temperature.

### 2.2. Methods

The volume and mass variation of the pastes specimens were estimated from the equations (1 and 2) respectively as previous works [7,8 and 30]:

$$\Delta V = [(V_a - V_b)/V_b] \times 100 (\%) \quad (1)$$

where:  $V_b$  and  $V_a$ ; volume of specimens before and after thermally treated respectively.

$$\Delta M = [(M_a - M_b)/M_b] \times 100 (\%) \quad (2)$$

where:  $M_b$  and  $M_a$ ; mass of specimens before and after thermally treated respectively.

X-ray fluorescence spectrometry (SHIMADZU XRF-1800) was used to assess the chemical (oxide) composition of mixed color glass waste

powder (MG).

X-ray diffraction analysis of graphite powder (G and IG) were assessed with Model Mini Flex II, Rigaku Co., Japan, using with Cu target operating at 30 Kv and 15 mA with standard wave length 1.54 Å for the XRD patterns were recorded in the range of  $2\theta = 5 - 80$  degrees.

Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray spectroscopy (EDS) analyses were performed on paste specimens, coated with Au (30nm), using a TESCAN- Vega III microscope.

The compressive strength of hardened pastes before and after thermal treatment was assessed with a material testing machine (SEWHA SI 4010, Korea).

### 3. Results and discussion

The XRD patterns of pure dry graphite powder (G) and dry intercalated graphite (IG) are presented in Figs.1 and 2. From Fig.(1), one can assess the graphite powder characteristic peak at  $2\theta = 26.3^\circ$ ; for the characteristic peak for intercalated graphite this shifts at  $2\theta=24.8^\circ$ , a clear indication of the expansion of graphite network. The decrease in the value of  $2\theta$  and the increase in d-spacing, indicate the occurrence of intercalation. After intercalation, the intensity of XRD peak is reduced, which shows that the number of crystal defects increases and crystallinity degree decreases.

Figures (3-5), present the visual aspect images of geopolymer pastes specimens; (MG-N5, MGG-N5 and MGIG-N5) before and after thermal treatment at temperatures (500, 600, 700 and 800°C).

From Fig. 3, it can be observed the MG-N5 specimens thermally treated at 500 and 600°C have a regular structure with slightly decrease in volume (contraction) as illustrated in Fig. 6. A local swelling with significant expansion at 700°C with small open porosity and partial melting can be noticed particularly in the upper surface of the specimen. At 800°C, the specimen exhibited swelling and volume increase (see Fig. 6) with self-glazing phenomenon which determines a small open porosity.

Figure 4 presents the MGG-N5 specimens before and after thermal treatment. After the thermal treatment at temperatures 500 and 600°C, the specimens retains the cubic shape but have with some cracks; small open porosity and melting in the upper surface of the specimen with a significant expansion (local swelling) was recorded when the thermal treatment was performed at 600°C and 800°C (see Figs. 4&6). The partial melting and shape deformation are noticed at temperature 800°C (see Fig.6).

The specimens with intercalated graphite addition (MGIG-N5) before and after thermal treatment at temperatures (500, 600, 700 and 800°C) for 1h are presented in Fig. 5. It can be

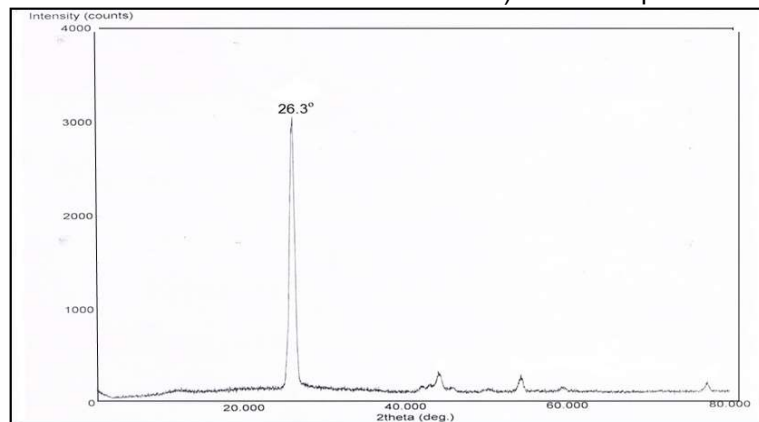


Fig.1 - XRD of graphite powder (G) [32].

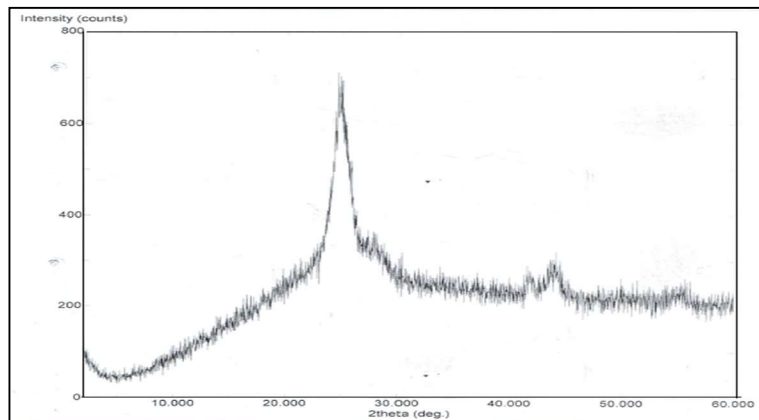


Fig.2 - XRD of dried intercalated graphite powder (IG) [32].

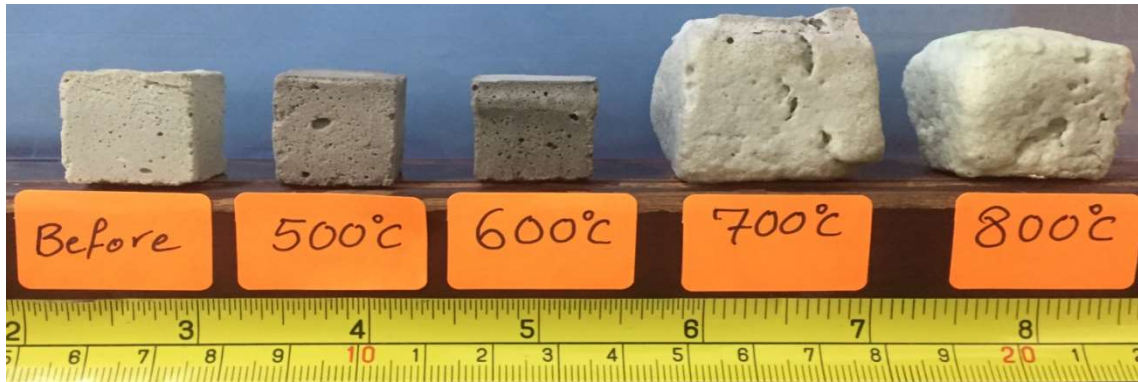


Fig.3 - MG-N5 paste specimen's before and after thermal treatment at 500, 600, 700 and 800°C /1h.



Fig.4 - MGG-N5 paste specimen's before and after thermal treatment at 500, 600, 700 and 800 °C /1h.



Fig.5 - MGIG-N5 paste specimen's before and after thermal treatment at 500, 600, 700 and 800°C /1h.

seen clearly the change in color (black color) for all specimens after thermal treatment at temperatures 500-800°C, this is most probably due to some reactions of the intercalate materials like nitric acid and potassium permanganate. The expansion (swelling) and deformation started at temperatures 600°C and 700°C; this phenomenon is associated with cracks and increase of open porosity (see Figs.5&6). The paste specimens presented a partial melting; the open porosity increases when the temperature rises to 800°C (Fig.5).

The volume changes of MGIG-N5 specimens are significantly higher as compared with the MGG-N5 and MG-N5 specimens after thermal treatment, especially for 600, 700 and 800°C (see Fig.6). The volume changes values ( $\Delta V\%$ ) calculated from equation (1), were (-11.4-240.68%), (68.25-146.64%) and (43.79-968.54%)

for MG-N5, MGG-N5 and MGIG-N5 pastes respectively as recorded in Fig. 6. Due to a foaming process, the percentage of volume changes were increased with increases the temperatures of thermal treatment in the range from 500-800°C (see Figs.3-5). A sharp increase MGIG-N5 volume was recorded when the thermal treatment was performed at 700°C, much higher as compared with the volume increase recorded for MG-N5 (as a reference formulation) and MGG-N5 (see Fig.6).

For the composition with intercalated graphite (MGIG-N5), paste specimens started to swell at lower temperatures i.e. 600°C. The higher values for the volume increases (intumescent behavior) are recorded at temperatures comprised between 600°C and 700°C. This is due to the spontaneous expansion by the intercalated  $\text{KMnO}_4$

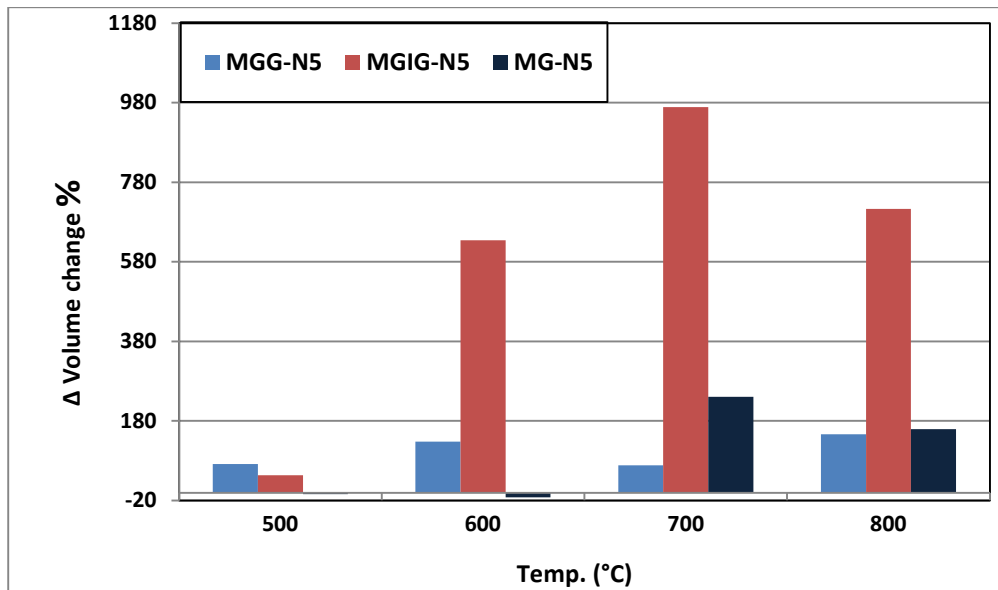


Fig.6 - Volume changes of MG-N5, MGG-N5 and MGIG-N5 paste after thermal treatment at 500, 600, 700 and 800 °C/1 h.

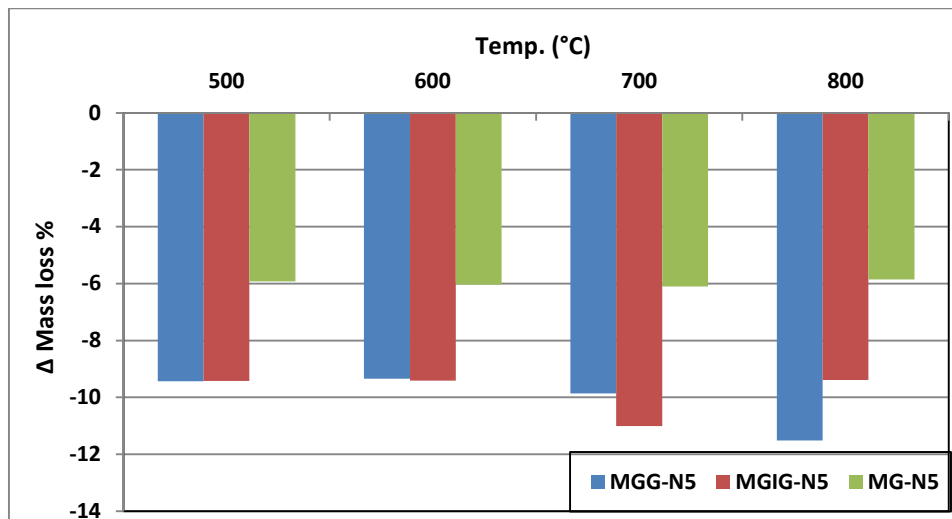


Fig.7 -Mass loss of MG-N5, MGG-N5 and MGIG-N5 specimen's after thermal treatment at 500, 600, 700 and 800 °C/1 h.

between graphite layers [33]; this behavior is different from the specimens without graphite additives (MG-N5) for which the highest swelling is recorded at 700°C (see Figs.3 & 6). The initiation temperature of intumescent process decreases with approximately 100°C after IG powder is added to the raw mix. These results are in a good agreement with that obtained by borax addition [8, 21 and 34] i.e. the swelling of the specimens containing waste with IG powder additives at temperatures comprised between 600°C-700°C.

The swelling of studied compositions during the thermal treatment are due to several phenomena: i) the dehydration and sodium (aluminate) silicate hydrates formed by the alkali activation of glass waste [7,8] correlated with the softening/partial melting of the material; ii) important increase of IG volume due to the presence of intercalated species.

The mass changes vs. temperatures of thermal treatment ( $\Delta M$  %) are presented in Fig. 7.

It can be seen that the mass losses comprised between 5.86-15.1% are recorded for all geopolymer pastes. For the specimens with graphite addition (MGG-N5 and MGIG-N5), the mass losses are higher as compared with reference MG-N5. The mass loss in MG-N5 is determined mainly by the water loss from sodium silicate hydrates; for MGG-N5 and MGIG-N5 pastes at higher temperatures (over 600°C) is possible to have also a burning of graphite which determines the increase of weight loss [5,35].

### 3.1 SEM and EDS analysis

Figures 8-11 present the SEM and EDS images for MG-N5, MGG-N5 and MGIG-N5 pastes. Fig.8-a & b, Fig. 8- c & d and Fig. 8-e&f show the SEM images for MG-N5, MGG-N5 and MGIG-N5 pastes respectively, cured 24 h at 60°C and up to 7 days at 20 °C (i.e before thermal treatment). One can assess glass grains included in a continuous matrix formed most probably from sodium silicate

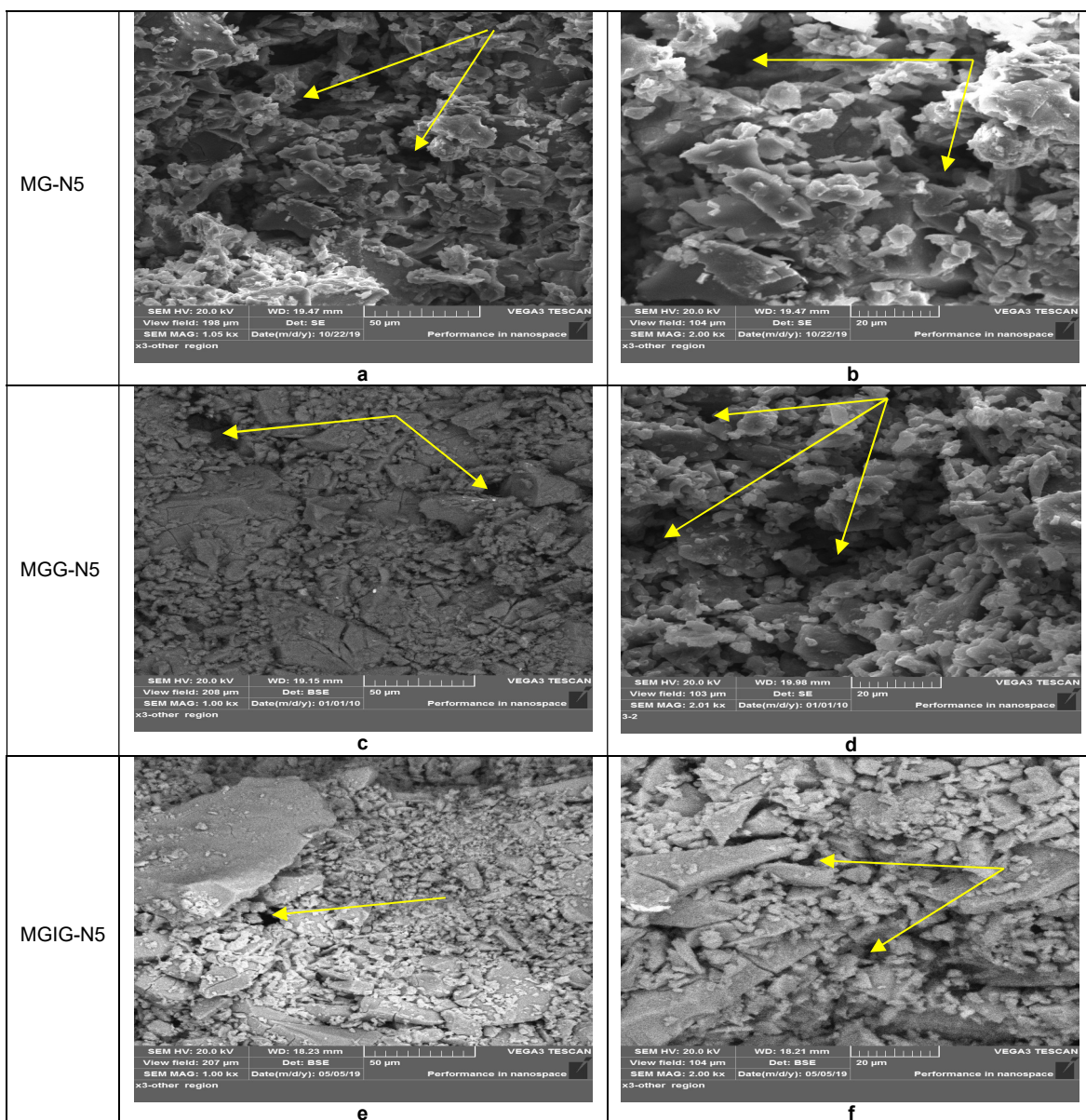


Fig.8 - SEM micrograph for MG-N5 (a&b), MGG-N5 (c&d) and MGIG-N5 (e&f) pastes curing 24 h at 60°C and 6 days at 20°C.

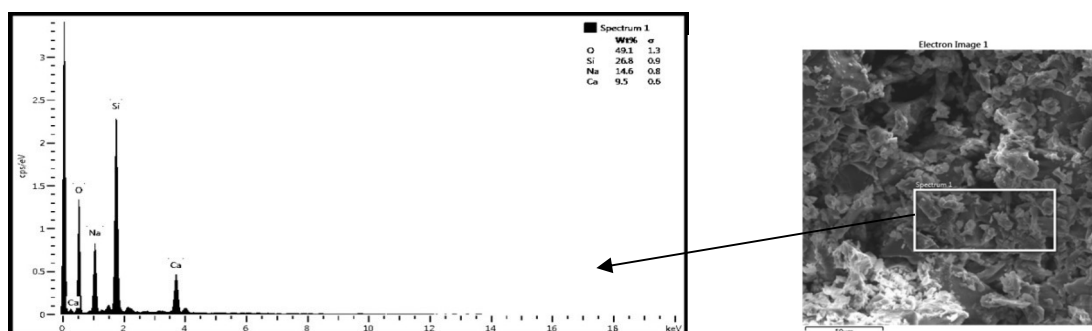


Fig.9 - SEM micrograph and corresponding EDS for MG-N5 paste curing 24 h at 60°C and 6 days at 20°C.

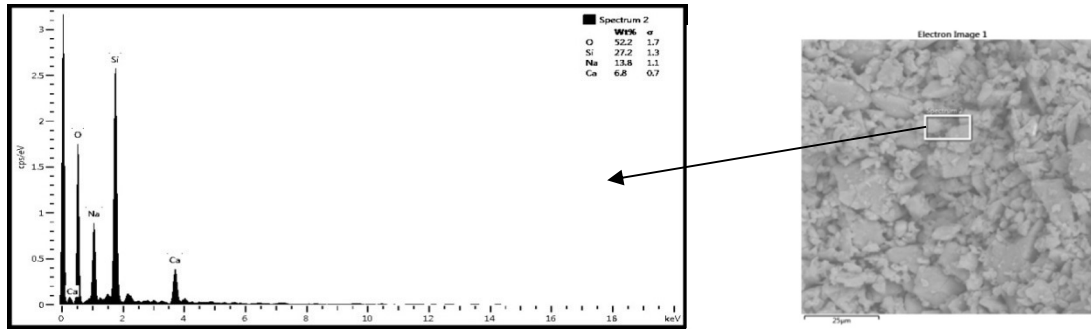


Fig.10 - SEM micrograph and corresponding EDS for MGG-N5 paste curing 24 h at 60°C and 6 days at 20°C.

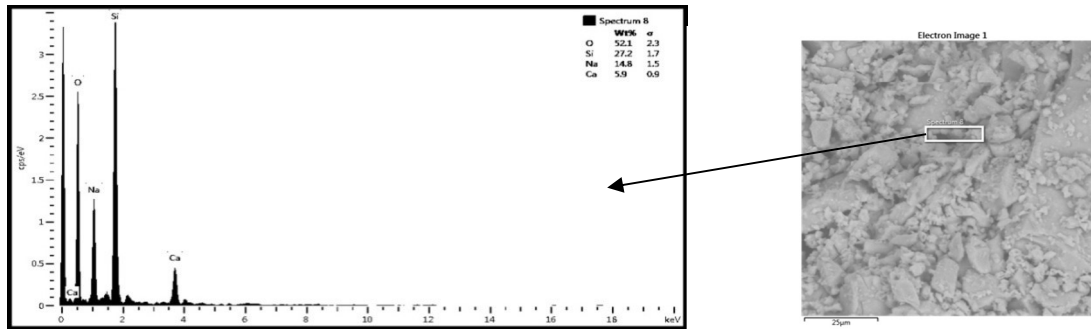


Fig.11 - SEM micrograph and corresponding EDS for MGIG-N5 paste curing 24 h at 60°C and 6 days at 20°C.

(aluminate) hydrates – resulted by the alkaline activation of waste glass powder [6]. Pores with different sizes are present on SEM images (see arrows in Fig.7). These round shape pores result mainly due to the entraining of air during the mixing of pastes. EDS analyses (Figs.9,10 and 11) confirm the presence of Si, Na, Ca in the studied pastes.

The microstructure of MG-N5, MGG-N5 and MGIG-N5 specimens after thermal treatment at 500, 600, 700 and 800°C was examined by SEM - Figs. 12,13 and 14. One can assess the increase of the porosities as a result of the presence of sodium silicate hydrates and graphite- G and IG (foaming agents). One can notice the presence of spherical shape pores because the sodium silicate hydrates tends to decompose and generate a balanced pressure on the surrounding walls, so the constant pressure gives the spherical form [36].

Figure 12 presents the SEM images of MG-N5 pastes after thermal treatment at temperatures 500-800°C. Pores with various shapes and sizes are present in these materials. One can also notice the presence of closed small round pores in the walls of big pores at temperatures (see Fig. 12 e, f, g&h).

The SEM images of MGG-N5 pastes showed an increase of the internal porosity after the thermal treatment at 500°C (Fig. 13 a & b); pores with various shapes and sizes (0.63- 10.22 µm) appeared in this specimen. This significant rise of the porosity is mostly due to the dehydration and transformation of sodium silicate (aluminate)

hydrates which resulted by the alkaline activation of waste glass powder. The pores size increase (1.84-134.56 µm) in the paste thermally treated at 600°C (Fig.13 c & d). Larger pores (13.52-106.25 µm) can be noticed when the thermal treatment is performed at 700°C; small round pores are preset in the walls of the large pores (Fig.13 e & f). The water molecules from sodium silicate hydrate structure commence to be released at temperature range 600-620°C [36]. The glass particles start to soften, and the melt will prevent the releasing of water vapors. This mechanism will create a remarkable closed porosity for paste specimens at thermally treated at 700°C (see Fig. 13 e,f). The increase in temperature of thermal treatment from 700°C to 800°C has an important effect on the microstructure of MGG-N5 foams; the size of pores increases up to 55.69-218.08 µm - Fig. 13 g, h.

Figure 14 presents the SEM micrographs of the MGIG-N5 specimens after thermal treatment at 500-800°C. The size of pores is in the range of 1.87-4.58 µm, 72.5-288.42 µm, 21.73-221.42 µm and 318.49-1512.93 µm respectively. The increase of temperature from 500°C to 800°C has a significant effect on the foams microstructure. The further increase in size of pores with irregular shapes indicates activation of the foaming process by the thermal treatment. The use of intercalated graphite (IG) as pore forming agent gives the best result i.e. the higher porosity is achieved for MGIG-N5 pastes as compared with MG-N5 and MGG-N5 pastes- Fig. 14 d, f, h.

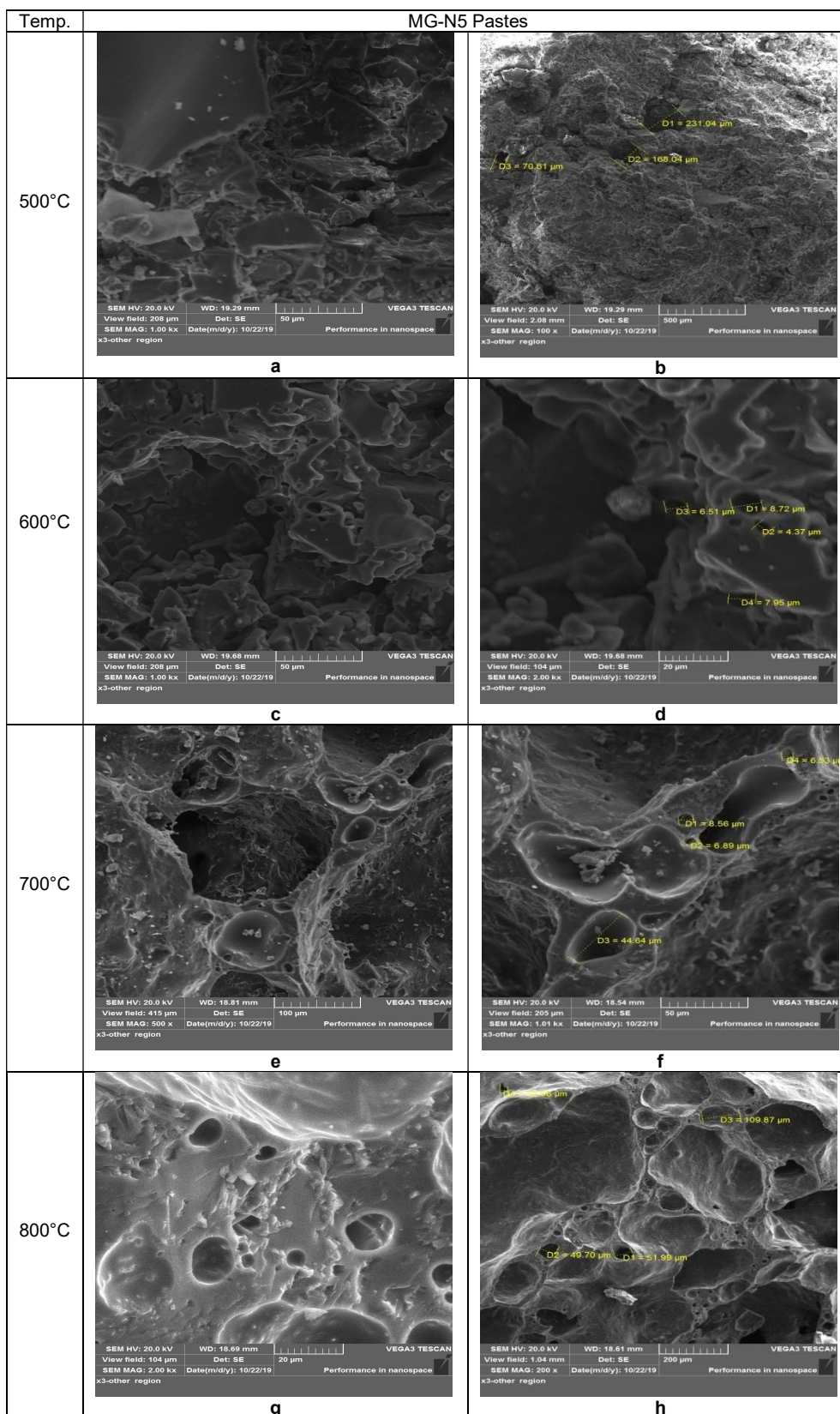


Fig.12 - SEM images of MG-N5 paste specimens after thermally treated at temperatures 500, 600, 700 and 800 °C (fracture).



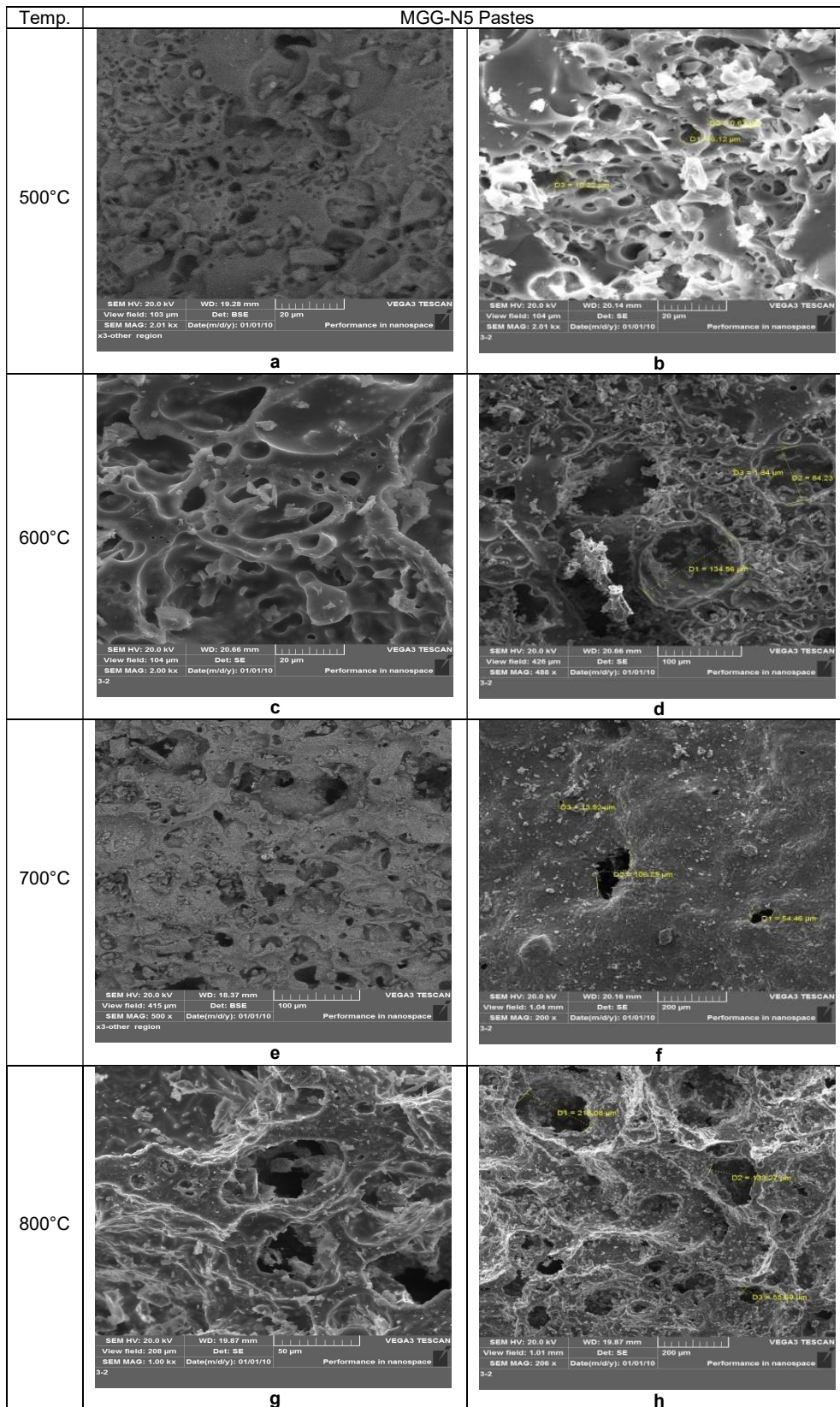


Fig.13 - SEM images of MGG-N5 paste specimens after thermally treated at temperatures 500, 600, 700and 800 °C (fracture).

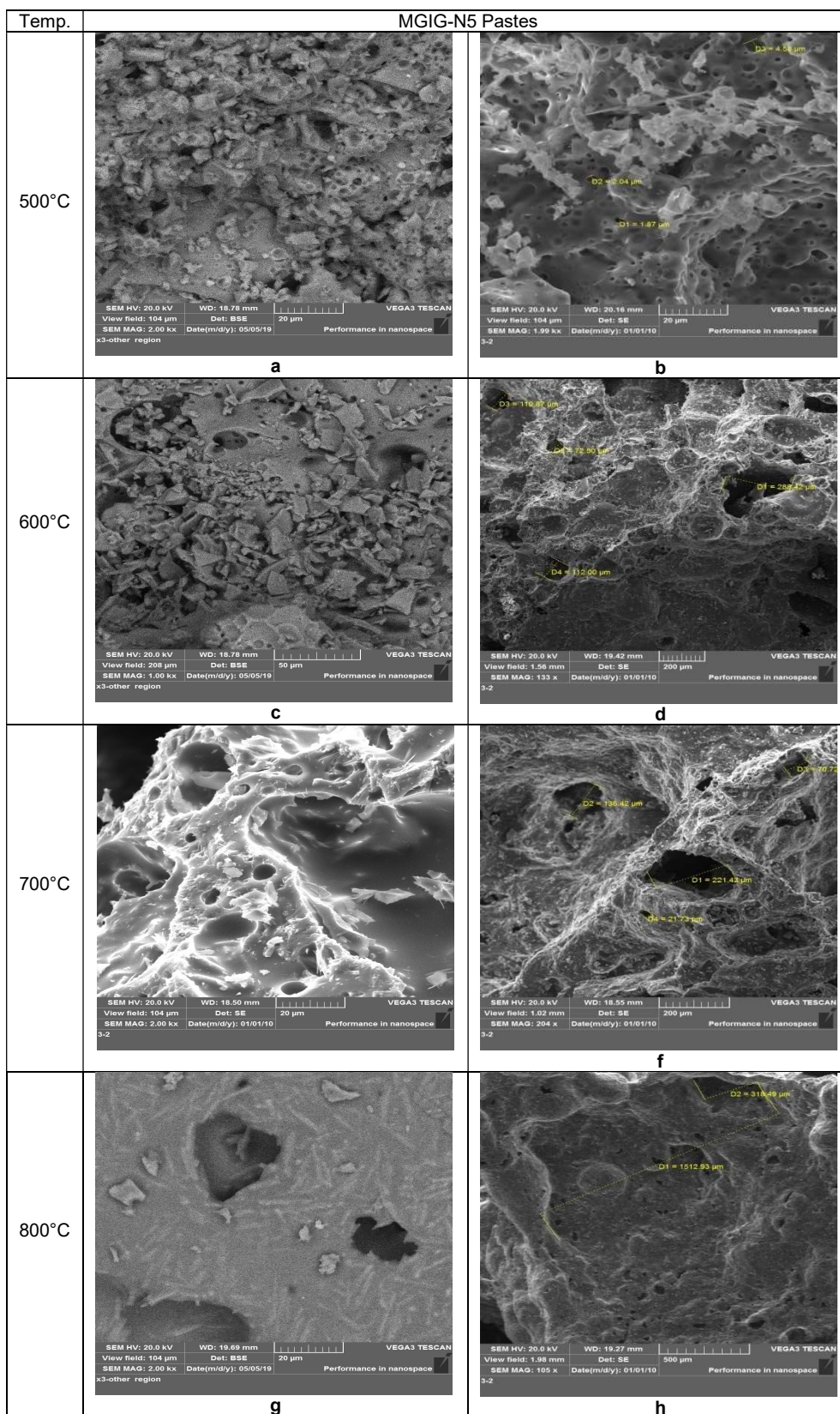


Fig.14 - SEM images of MGIG-N5 paste specimens after thermal treatment at temperatures 500, 600, 700 and 800 °C (fracture).

### 3.2 Mechanical properties

The compressive strength of MG-N5, MGG-N5 and MGIG-N5 after the thermal treatment at 500-800°C is presented in Fig.15.

For the composition without graphite (i.e MG-N5) the increase of thermal treatment temperature from 500 to 700°C determines the decrease of the compressive strength values, due to the increase

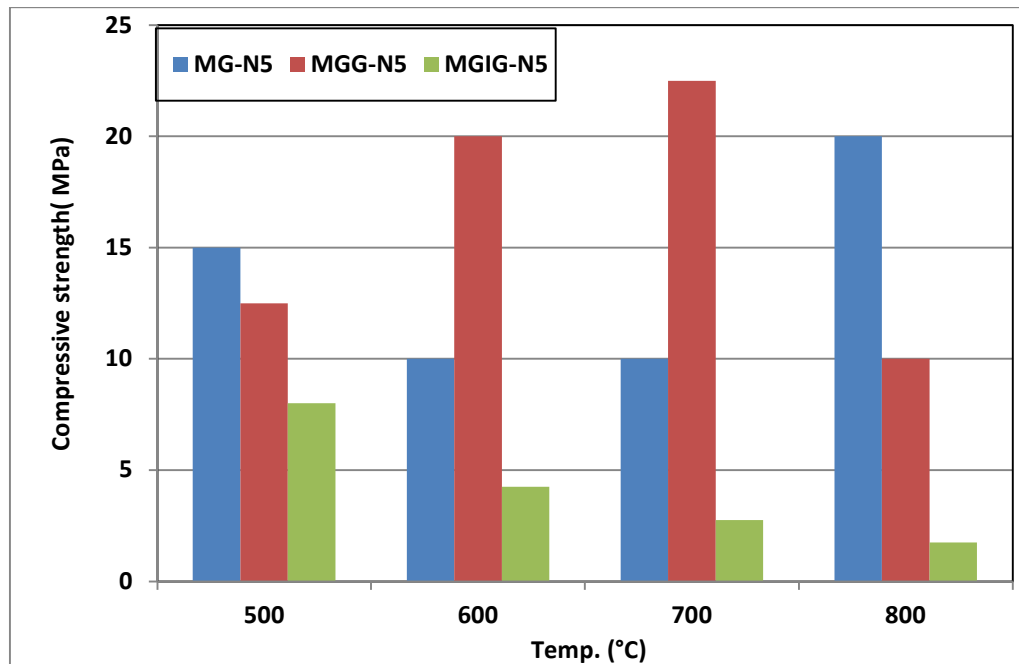


Fig.15 - Compressive strength of MG-N5, MGG-N5 and MGIG-N5 paste specimen's after thermally treated at 500,600,700 and 800 °C /1 h

of the volume and porosity; for the specimen thermally treated at 800°C was recorded an increase in the compressive strength due to a sintering process (reduction of the porosity).

For MGG-N5, the compressive strength values increase after thermal treatment from 500°C and 700°C (see Fig.15). This increase due to the sintering process and reduction of the porosity; a contraction of the specimens due to its densification is noticed for the thermal treatment performed at 700°C (see Figs.4 and 13). The increase of porosity noticed for the specimen thermally treated at 800°C (Fig.13 g,h) determines a decline in the compressive strength values. However, these values are still high- 10MPa (see Fig.15).

For MGIG-N5, the increase of specimens volume and porosity (see figs.6 and 14) determines, as expected the decrease of the compressive strength values, after thermal treatment at 500-800°C (Fig.15). It is well-known that the compressive strength and porosity content of the sample are inversely related [36]. Nevertheless, the compressive strength values between 2–5 MPa represent a perfect result for foaming materials [37].

#### 4. Conclusions

Geopolymer foams were obtained by the alkaline activation with NaOH solution (5M) of mix color glass waste powder (MG) without/with graphite additions (0.1 wt%). The resulted geopolymers were thermally treated at temperatures comprised between 500°C and 800 °C and a significant increase in volume was recorded due to foaming process. For the

geopolymer with intercalated graphite addition (i.e MGIG-N5) was recorded an important increase of volume and porosity when subjected to thermal treatment. The swelling phenomenon is first recorded for the specimens thermally treated at 600°C. A decrease of the temperature at which these specimens started to swell (with approximately 100°C) was recorded when IG powder is added to the raw mix.

The compressive strength values of all specimens decrease after the thermal treatment due to the important increase of porosity.

The resulted foams can be potential used as light thermal and sound insulations; the energy saving materials have a low cost and are environmentally friendly.

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