

## Article

# The Effect of Different Modifying Methods on Physical, Mechanical and Thermal Performance of Cellular Geopolymers as Thermal Insulation Materials for Building Structures

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**Abstract:** Geopolymers represent a new class of inorganic materials that have great potential for practical application due to the properties of used raw materials, as well as the peculiarities of the cementitious matrix structure formed during the geopolymerization process. Cellular geopolymer specimens were produced in this study using class F fly ash product, which is characterized by low reactivity during geopolymerization. Several standard methods, as well as microstructural studies were applied to evaluate the effect of the following factors on the physical-mechanical and thermophysical characteristics of cellular geopolymers: the use of various mineral modifying components for synthesis of geopolymer systems; high-temperature treatment; the introduction method of alkaline activator. It was observed that “ageing” an aqueous alkali solution for 24 h before mixing with fly ash and foam agent was able to provide a boost of compressive strength of cellular geopolymer specimens up to about 2.5 times, while decreasing the average density by about 28% for all experimental mixes, except for PC-modified mixes. Additionally, high-temperature treatment at 600 °C enables an enhanced strengthening effect of pore structure in cellular geopolymer matrix up to 1.5 times. This phenomenon is especially pronounced for the mixes with 24 h “aged” alkaline solution with exception for PC-modified mixes; for those, high-temperature treatment at 600 °C leads to strength decrease up to 40%. The introduction method of alkaline activator and high-temperature treatment showed a controversial effect on thermal conductivity coefficient depending on the mineral modifying component used for the synthesis of cellular geopolymers. The proposed method for calculation of total porosity of cellular structure of geopolymers as a polycomponent material demonstrated a high degree of correlation with the  $R^2$  value of at least 0.96 between the average density and the calculated total porosity. However, a low degree of correlation with  $R^2$  not exceeding 0.29 was observed for the measured nanoporosity, regardless of the introduction method of alkaline activator and high-temperature treatment.

**Keywords:** cellular geopolymer; mineral modifying component; high-temperature treatment; porous structure; thermal resistance; class F fly ash



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## 1. Introduction

Porous materials are widely used and found in many industrial applications, including the production of membranes and high-performance adsorption materials. In addition, they are commonly used in the construction industry as heat-insulating or sound-insulating

building structures, such as panels and blocks, as well as in the production of lightweight products and structural elements. Polymers, metals or inorganic/mineral components are commonly used as raw materials for the production of various types of cellular composites for building structures. For example, for styrofoam, the polymer-based composites are especially well-suited as insulating materials in civil engineering due to their low density, low thermal conductivity and good sound insulation properties [1]. However, most polymer composites are subject to thermal decomposition at high temperatures, which leads to degradation of their structural integrity and the release of toxic or flammable gases into the atmosphere as a result of polymer decomposition [2]. In this aspect, ceramic materials [3] and their structural analogous produced from aluminosilicate materials [4,5], such as geopolymers [6,7], are considered to be more effective in terms of environmental friendliness and durability.

Additionally, another advantage of geopolymers is the use of a wide range of secondary raw materials, by-products and industrial waste, such as fly ash from coal combustion power plants, blast furnace slag and rice husk ash [8–12]. On the other hand, due to their ceramic nature, geopolymers as elements of building structures have relatively high fragility and low flexural strength, which limits their areas of practical application.

A wide range of aluminosilicate raw materials used for the synthesis of geopolymers vary greatly in the degree of chemical reactivity during geopolymerization processes that makes it challenging to achieve consistency in strength performance and structure formation stability of the final product. To a greater extent, the above-noted challenge for geopolymers appear in their cellular structure because an increase in porosity of the matrix inevitably comes with a decrease in compressive strength.

To address this issue, there are various ways to improve the strength performance of geopolymer materials (Figure 1). For example, to increase tensile strength, some authors [13,14] proposed to use fiber elements of different shape, length and chemical compositions to provide reinforcement to a fragile matrix. Another way to strengthen structural geopolymer frameworks is the application of a reinforcing net using a polymer compound, such as epoxy resin, which is introduced into the geopolymer matrix. Such structures are often referred to as hybrid materials [15–18]. Roviello, G et al. [19] demonstrated a method for the production of hybrid composites based on geopolymer and epoxy-melamine resins with a high degree of compatibility between the organic and inorganic constituents. This was realized using a method based on the synthesis of a joint network of polymer and geopolymer phases in a liquid state due to the interpenetration of the resin into the inorganic matrix at the nanoscale level. Through the use of a hybrid framework, the resulting geopolymer composites had higher mechanical characteristics vs. unmodified composites. At the same time, heat resistance and fire resistance performance remained the same as for geopolymers without modification.

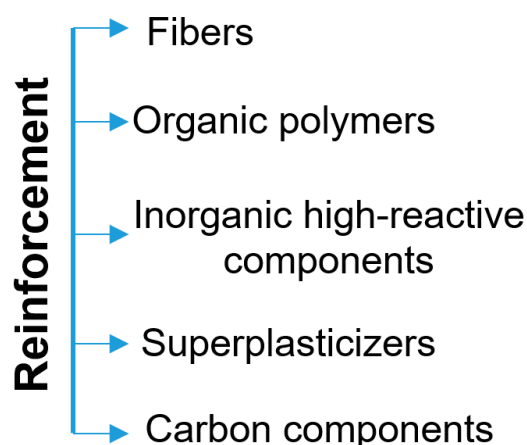


Figure 1. Reinforcement methods of cellular geopolymers.

Similar studies were carried out for cellular composites based on organic–inorganic geopolymers [20], where a mixture of dialkylsiloxane oligomers was used as the organic component. As a result, the produced hybrid cellular geopolymers showed higher fire resistance, a density ranging from 0.25 to 0.85 g/cm<sup>3</sup> as well as compressive strength reaching 5 MPa. The thermal conductivity of these composites was considerably improved in comparison to unmodified cellular geopolymers and other cellular composites with similar density and demonstrated an average value of  $\approx 0.145$  W/m<sup>2</sup>·K.

The introduction of more reactive mineral additives as modifying components as an alternative way to improve strength performance of geopolymers was demonstrated by Dali Bondar et al. [21], who studied the effect of the introduction of mineral additives as pozzolanic components, such as kaolinite, lime and other calcined pozzolans on the compressive strength of the geopolymer under ambient conditions and autoclave curing. The use of these additives in a concentration of 15–20% allows for increasing the strength of the geopolymer up to 13% during curing both in ambient conditions (25 °C) and in an autoclave.

The authors of the study [22] demonstrated the positive effect of combined application of carbon nanotubes and nano-Al<sub>2</sub>O<sub>3</sub> powder in geopolymer composites designed for cementing wells. The introduction of these nano-additives promotes the increases of 28 days compressive strength of the geopolymer composites by 50% vs. unmodified geopolymers.

An example of using carbon textile as a textile reinforcing element in a metakaolin-based geopolymer is presented in [23]. The research results showed a weak adhesion force between carbon textile and geopolymer matrix, which leads to an undesirable effect: a layering of the geopolymer composite.

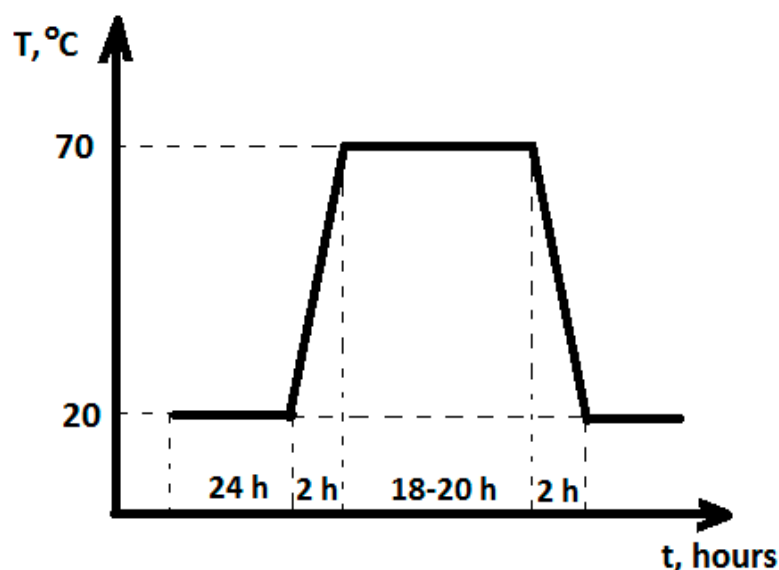
In fact, the use of superplasticizers makes it possible to reduce water demand in cementitious systems and thus strengthen the matrix structure. M. Fadhil Nuruddin et al. [24] used various dosages of superplasticizer up to 7% in the compositions of geopolymer concrete and demonstrated an increase in compressive strength performance.

In earlier work, the authors of this study determined the influence of the introduction method of alkaline activator into class F fly ash-based geopolymer binders, which were modified with various mineral components, such as kaolin, metakaolin and portland cement [25]. Geopolymer specimens were prepared using two curing approaches:

- (1) Natural curing in laboratory conditions at temperature  $22 \pm 3$  °C and relative humidity  $\approx 16\%$  before testing;
- (2) Natural curing in laboratory conditions at temperature  $22 \pm 3$  °C and relative humidity  $\approx 16\%$  for 24 h, followed by heat treatment in an oven according to the following temperature regime: heating to 70 °C—2 h → isothermal curing at 70 °C—18–20 h → cooling to a temperature of  $22 \pm 3$  °C—2 h → natural laboratory conditions until testing (Figure 2).

The research results showed that the aging of the aqueous NaOH solution for 24 h before use can increase the strength indicators by up to 84 %. At the same time, the average density remains practically unchanged. However, systems with a similar chemical composition, yet different matrix with compact and cellular structures, usually reveal various quantitative and qualitative changes in performance. For example, the compressive strength of compacted cement paste exceeds that for cellular concrete based on the same binder by a factor of 10 or higher [26–28].

Similar to work of [25], current research studies the effect of the alkaline activator introduction method and mineral modifying components on physical-mechanical, thermal-physical and heat-resistant characteristics of cellular geopolymers.



**Figure 2.** Scheme of the curing approach 2.

In the above-studied literature sources, experimental results are presented on the influence of a small number of external factors (one or two) on one or two parameters of the final geopolymer. However, given the good prospective for the use of cellular geopolymer products in buildings, it is relevant to evaluate the response when three or more external factors simultaneously affect important characteristics of cellular geopolymer composites.

The goal of this article is to establish the complex effect of different external factors, such as the nature of the mineral modifier, the method of using the alkaline activator and high-temperature treatment on the basic performance characteristics of geopolymer foams.

## 2. Materials and Methods

### 2.1. Materials

Class F fly ash product produced in Russia was used as the main aluminosilicate component. The Type F fly ash used in this study is a by-product of the combustion of bituminous coal in the form of a wet furnace mass in a circulating fluidized bed. Furnace mass consists of crushed coal particles up to 30 mm. Portland cement CEM I 42.5 (Russia), kaolinite (Russia) as well as metakaolin were used as mineral modifying components (MMC) for synthesis of cellular geopolymers (CGP). Metakaolin was synthesized in laboratory conditions by dihydroxylation of natural kaolin in a muffle furnace according to the following regime: elevation of temperature up to 700 °C for 2 h → curing at 700 °C for 2 h → slow cooling down to  $22 \pm 3$  °C for 8–12 h (Figure 3).

Caustic soda or sodium hydroxide NaOH with 98% purity was used as an activating alkaline agent.

The component BioFoam produced in Russia was used as a forming agent in this study to produce CGP. BioFoam is a protein compound based on polymeric ampholytic surfactants. It is produced in the form of a homogeneous dark brown water solution liquid with a specific gravity 1.1 g/cm<sup>3</sup>.

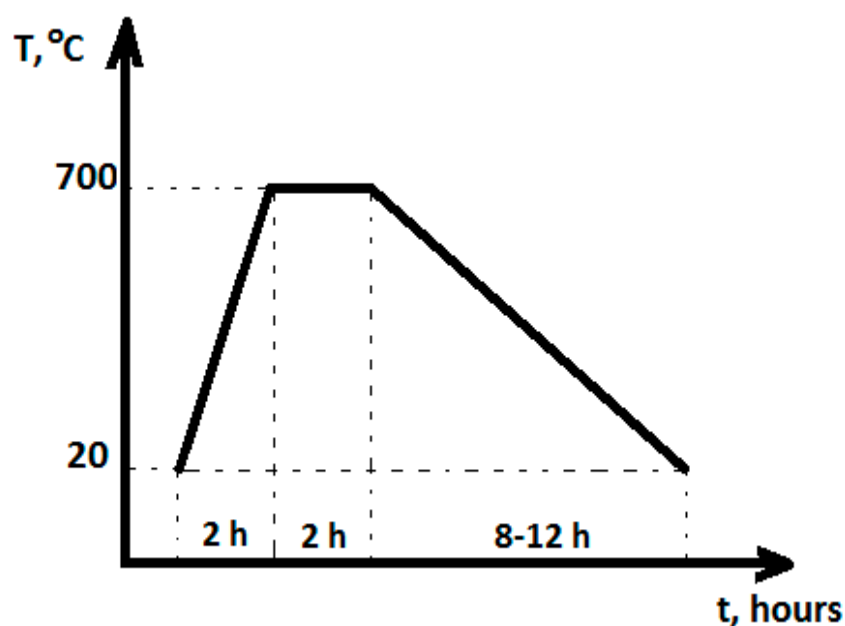
### 2.2. Testing Methods and Procedures

#### 2.2.1. Chemical and Microstructural Characterization

Chemical and mineral compositions of class F fly ash (FA), portland cement (PC), kaolinite (K) and metakaolin (MK) were determined by X-ray fluorescence analysis (XRF) using an X-ray WorkStation ARL 9900 (Thermo Scientific, USA). A Co-anode was used as a radiation source. The measurement of the specific surface area (SSA) and the average particle size of the powder solid-phase components FA, PC, K and MK was carried out



using a multifunctional machine PSH-12 (SP) (Russia) operating on the principle of an automated Blaine method.



**Figure 3.** Scheme of temperature regime for metakaolin synthesis.

Analytical porosity measurement of the experimental CGP was carried out using a Tristar II 3020 Micromeritics specific surface area and porosity analyzer (USA).

Chemical composition and physical characteristics of solid-phase FA, PC, K and MK are presented in Table 1.

**Table 1.** Chemical composition and physical characteristics of solid-phase mineral components.

MMC	Oxides Content, wt.%												Real Density, g/cm <sup>3</sup>	SSA, m <sup>2</sup> /kg
	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	K <sub>2</sub> O	MgO	CaO	P <sub>2</sub> O <sub>5</sub>	SO <sub>3</sub>	N <sub>2</sub> O	LOI	SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub>		
FA	58.9	28.3	4.63	0.97	0.65	1	3.74	0.36	-	0.63	6.07	2.08	1.87	290
PC	19.1	5.21	3.58	0.32	0.6	1.28	65.4	-	3.47	-	0.23	3.67	3.05	320
K	53.8	43.4	1.02	0.58	0.56	0.21	0.01	0.06	-	0.03	4.2	1.23	2.61	2610
MK	53.1	42.8	0.7	0.3	0.9	-	0.15	-	-	0.02	0.4	1.24	2.52	2520

The microstructure of the cellular geopolymer specimens was studied using scanning electron microscopy (SEM). A scanning electron microscope Mira 3 FesSem (Tescan, Czech Republic) operating in a high vacuum mode (InBeam) with a high brightness Schottky cathode was used. Before the analysis, all samples were preliminarily covered with a thin layer of chromium (Cr) for a better conductivity.

#### 2.2.2. Porosity and Density Characteristics

Porosity is one of the fundamental parameters that is directly associated with the quality of a material, as well as its functionality and area of application. Usually, in case of cellular materials, porosity as a parameter on its own can define thermal-and-physical characteristics. The issue of evaluating the porosity of cellular composites is usually addressed by using standard method, for example, Russian Standard GOST 12730.4-78 “Concretes. Methods of determination of porosity parameters” or using analytical instruments, such as the BET method and mercury porosimetry. For example, in the study [29], the evaluation of general or cumulative porosity was conducted by the combination of data from mercury porosimetry, BET method and SEM analysis. However, there is reason to believe that all of the above methods can give errors either due to the human factor or due to the instrument error.

Additionally, errors can be caused by the characteristics of the studied material, or by the specifics of the used measurement method.

The most common factors that can affect the accuracy of a porosity value are the following:

1. Destruction of samples integrity, which takes place during sample preparation or directly during analysis. This issue is typical, for example, for the BET method, where the crushing/breaking the test sample into pieces of 4–7 mm is a required preparation protocol. This leads to a violation of the integrity of the internal pores. In addition, the study of porosity on samples of such small dimensions does not provide a complete picture of the porosity cellular material in bulk;
2. Testing limitations related to the type and origin of the studied materials. These limitations apply to the subject of testing. For example, resistance to external conditions, such as heating, immersion in a liquid medium, degassing and other conditions are important to consider for each specific material tested. For example, the standard method for determining porosity by immersing a sample in water is not suitable for gypsum materials that are not water-resistant and easily degrade in water;
3. Features of pore structure of the tested material. The impossibility of determining the total porosity if there are closed pores in the structure is one common drawbacks for existing analytical methods for porosity evaluation. Below provides the definitions used in this article:

True or total porosity is the total volume of all pores in the material, open and closed, expressed as a percentage of the total volume of the material. This value is numerically equal to the sum of the closed and open porosity.

The apparent or open porosity is the ratio of the volume of all open pores in the material to the entire volume of the material, including the volume of all pores.

Closed porosity is the ratio of the volume of all closed pores in the material to its total volume, including the volume of all pores. Normally, concrete materials, regardless of their composition, are characterized by the presence of closed pores. Commonly, the closed porosity  $P_{closed}$  is determined only experimentally or by calculation using a standard technique (for example, the Russian standard GOST 12730.4-78 “Concretes. Methods of determination of porosity parameters”):

$$P_{closed} = P_{total} - P_{open} \quad (1)$$

where  $P_{total}$  is the total porosity, %;  $P_{open}$  is the open porosity, %.

In turn, open porosity is also a calculated value, where it is necessary to determine water absorption  $W$  (%) of the tested material:

$$P_{open} = W \cdot \rho_{average} \quad (2)$$

where  $\rho_{average}$  is the average density, g/cm<sup>3</sup>.

However, due to instability in water, not all materials can withstand the water absorption test. The cellular geopolymer concrete specimens produced in this study are characterized by low water resistance (especially, without high-temperature treatment). In this regard, the determination of total porosity for those specimens was carried out using the following calculation method that is based on determining the total porosity  $P_{total}$  as a result of taking into account the difference in true density (excluding pores) and average density (including pores) for the tested material (see Equation (3)):

$$P_{total} = \left(1 - \frac{\rho_{average}}{\rho_{true}}\right) \cdot 100\% \quad (3)$$

where  $\rho_{average}$  is the average density, g/cm<sup>3</sup>;  $\rho_{true}$  is the true density, g/cm<sup>3</sup>.

To determine these parameters, the classical formula can also be used, which is expressed as the ratio of the mass of the material to the volume it occupies:

$$\rho_{true} = \frac{m}{V_{real}} \quad (4)$$

$$\rho_{average} = \frac{m}{V_{average}} \quad (5)$$

where  $V_{real}$  is the volume of the specimen without pores,  $\text{cm}^3$ ;  $V_{average}$  is the volume of the specimen, taking into account the pores,  $\text{cm}^3$ .  $m$  is the mass of the specimen, g.

The cellular geopolymer concrete specimens investigated in the article are in the form of cubes with a size of 70 mm. Therefore, the determination of the average volume in this case was carried out by a simple measurement of their geometric dimensions (width, length, height) with subsequent calculations according to Equation (6):

$$V_{average} = a \cdot b \cdot h \quad (6)$$

where parameters  $a$ ,  $b$  and  $h$  are the length, width and height of the experimental cellular geopolymer concrete specimen, respectively, mm.

The values of the true density for simple components, which are the constituent of the studied CGP, are known and contained in the corresponding guidance books. In the case of CGP, it is a multi-component material. Therefore, its true density can be taken as a sum of the true densities of all the constituents of the composite, taking into account their ratio in the total volume:

$$\rho_{true.composite} = x \cdot \rho_1 + y \cdot \rho_2 + \dots + z \cdot \rho_n \quad (7)$$

where  $\rho_1, \rho_2, \rho_n$  are the true densities of the constituents 1, 2, ...,  $n$ ;  $x, y, z$  are the proportions of the constituents in the total volume of the multi-component composite, taken as 1.

True density values for class F fly ash and mineral modifying components PC, K and MK were determined using a Pycnomatic ATC helium pycnometer (Thermo Fisher Scientific, Italy). The true density value for the alkaline component NaOH is  $2.13 \text{ g/cm}^3$ . It was taken from the Chemical Abstracts Service (CAS Number: 1310-73-2). The porosity of experimental CGP was determined with a specific surface area and porosity analyzer Tristar II 3020 using the BET multipoint method.

### 2.2.3. Compressive Strength Testing

The compressive strength test was carried out according to the standard procedure applicable for cellular concrete following Russian Standard GOST 10180-2012 "Concretes. Methods for strength determination using reference specimens". The tested samples were cube of  $70 \times 70 \times 70 \text{ mm}$ . The compressive strength was determined as the arithmetical average of three cubes.

### 2.2.4. Thermal Conductivity Testing

The measurement of thermal conductivity for cellular geopolymer concrete was carried out with a heat probe using the ITP-MG4 "ZOND" equipment setup. For the testing, holes were made in the center of CGP cube specimens corresponding to the probe dimensions with diameter of 4 mm and hole depth of 45 mm. The duration of one measurement lasted 8 min.

This work presents the averaged data based on the results of three tested specimens of each mix to determine physical, mechanical and thermophysical characteristics of CGP specimens.

## 2.3. Sample Preparation

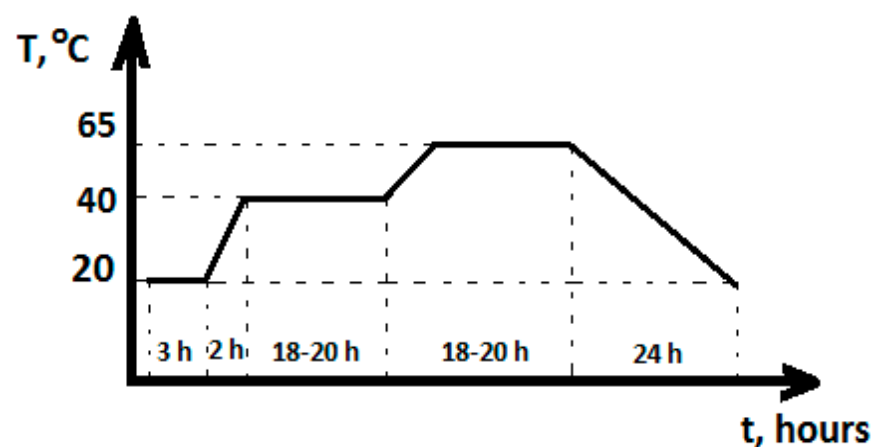
In this experimental program, two series of CGP specimens were produced. For the first series, a freshly prepared aqueous NaOH solution was used. For the second series of specimens, an aqueous NaOH solution was prepared and "aged" for 24 h before use in

the cellular geopolymer mixes preparation. Each series were represented by specimens with four different component compositions. Experimental mixes of CGP are presented in Table 2.

**Table 2.** Mix design of the cellular geopolymer concrete.

Mix ID	Component Composition,% wt.						Foaming Agent
	FA	MMC			NaOH	Water	
		PC	MK	K			
NaOH freshly prepared aqueous NaOH solution							
R	53.5	-	-	-	8.02	38.3	0.17
GPC	32.09	21.4	-	-	8.02	38.3	0.17
GMK	32.09	-	21.4	-	8.02	38.3	0.17
GK	32.09	-	-	21.4	8.02	38.3	0.17
24-h aged aqueous NaOH solution							
R-24	53.5	-	-	-	8.02	38.3	0.17
GPC-24	32.09	21.4	-	-	8.02	38.3	0.17
GMK-24	32.09	-	21.4	-	8.02	38.3	0.17
GK-24	32.09	-	-	21.4	8.02	38.3	0.17

In this research, for preparation of CGP mixes, a more-gentle heat treatment was applied vs. compact geopolymers. For instance, in the work of [30] it was revealed that a more-effective “compressive strength/average density” ratio is provided for geopolymer foams at a slightly elevated temperature up to 40 °C vs. 20 °C. The use of more-gentle heat treatment mode for geopolymer foams is preferable because the foam, normally, contains about 50% more water than compact geopolymers. Under the influence of high temperature, the pressure in the pore structure rises due to the intensive removal of moisture from the specimen, which leads to its structure destruction. Thus, based on the known regularities of effective consolidation of geopolymers based on class F fly ash at high temperatures [31,32], the CGP specimens were subject to temperature treatment in a drying oven according to the following regime: curing at a temperature of  $22 \pm 3$  °C and relative humidity 14–18% for 3 h → heating to 40 °C for 2 h → isothermal holding at 40 °C for 18–20 h → rise of temperature up to 65 °C for 2 h → isothermal curing at 65 °C for 18–20 h → cooling to  $22 \pm 3$  °C for 2–4 h (Figure 4).



**Figure 4.** Temperature treatment regime for the CGPs.

Before testing, experimental specimens were stored in laboratory conditions at a temperature of  $22 \pm 3$  °C and a relative humidity of 14–18%.

### 3. Results

#### 3.1. Effect of High-Temperature Treatment on the Properties of Cellular Geopolymer Concrete

To expand the application areas for geopolymers, there is a large amount of research devoted to the study of their resistance to high temperatures. In work [33], it was found that the chemical and mineral compositions of fly ash have a significant effect on the temperature threshold resistance for geopolymers in the temperature range from 200 °C to 600 °C. This threshold was determined by the change in compressive strength values.

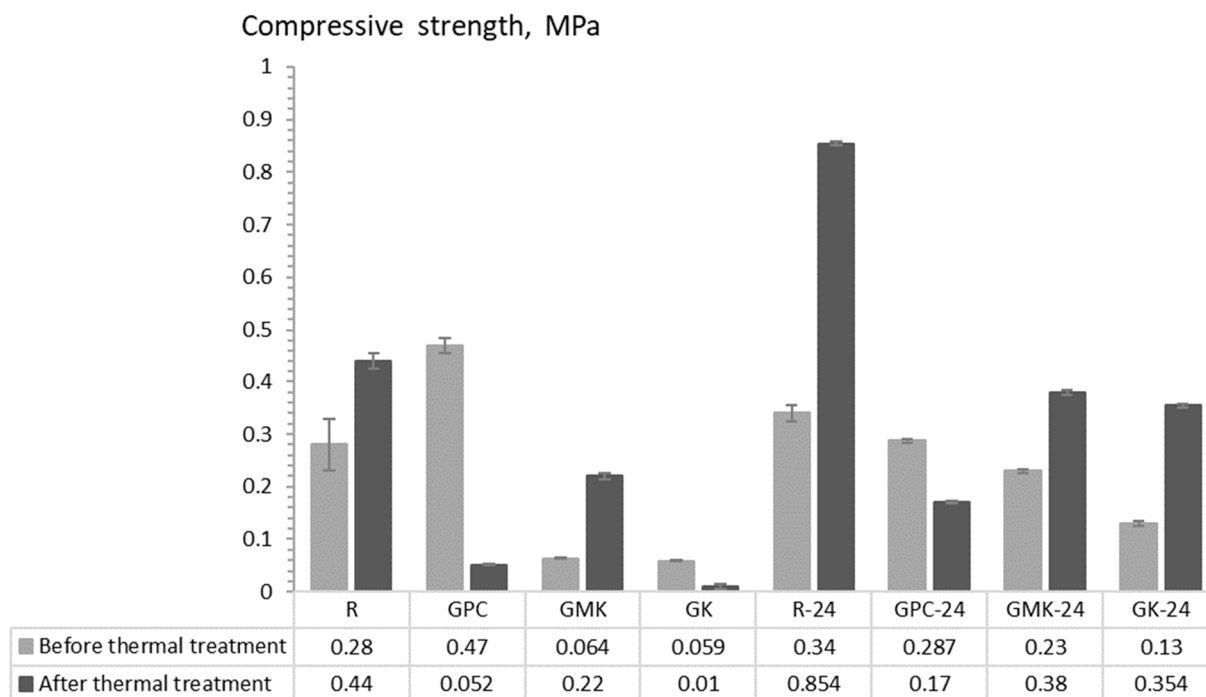
In this article, the influence of such factors was studied as the type of mineral modifying components and the introduction method of the alkaline activator on physical and mechanical properties, such as compressive strength, average density, porosity, thermal conductivity as well as the resistance of the cellular geopolymer structure to high temperatures (Table 3). High-temperature treatment (TT) and tests to determine the above-mentioned characteristics were carried out for CGP at the age of 9 days after demolding.

**Table 3.** Characteristics of cellular geopolymer concrete using different introduction method of the alkaline activator NaOH.

Mix ID	Average Density, g/cm <sup>3</sup>		Compressive Strength, MPa		Thermal Conductivity, W·m <sup>-2</sup> ·K <sup>-1</sup>	
	Before TT	After TT	Before TT	After TT	Before TT	After TT
R	0.732	0.585	0.28	0.44	0.049	0.044
GPC	0.963	0.695	0.67	0.052	0.154	0.071
GMK	0.496	0.564	0.064	0.22	0.043	0.041
GK	0.456	0	0.059	0	0.088	0.001
R-24	0.788	0.726	0.34	0.854	0.047	0.042
GPC-24	0.74	0.682	0.287	0.17	0.096	0.054
GMK-24	0.71	0.63	0.23	0.38	0.045	0.044
GK-24	0.832	0.629	0.13	0.354	0.074	0.055

Based on the obtained results presented in Table 3 and Figure 5, the use of an “aged” alkaline solution initiates the formation of a stronger structure for both before and after heat-temperature treatment. For example, for mixes before high-temperature treatment, more pronounced strength increase is observed for mixes, GMK-24 vs. GMK, GK-24 vs. GK is by more than 2.5 and 1.2 times, respectively, and for the mixes R-24 vs. R the strength increase was only 21 %. The opposite effect is observed for geopolymer foam concrete with PC content (composition GPC-24). A 24 h exposure to an aqueous solution of NaOH leads to the formation of a less-strong structure in the samples of the mix GPC-24. This phenomena is yet to be studied further.

In turn, high-temperature treatment has a favorable effect on the strength formation for all experimental compositions, other than mixes GPC and GPC-24 (with cement content), as well as for the mix GK (containing kaolinite and freshly prepared NaOH solution). The decrease in the compressive strength for the GPC and GPC-24 can be caused by the fact that the hydrated PC under high temperatures dehydrates and breaks down. This destructively affects the cellular geopolymer structure as a whole.



**Figure 5.** Effect of the introduction method of alkaline activator NaOH on compressive strength under different temperature conditions.

However, it should be noted that the modifying additive PC for the freeing of high-temperature treatment mix (mix GPC) provides the highest compressive strength vs. other experimental geopolymer foams. On the other hand, the GPC mix has the highest average density and thermal conductivity coefficient as well. This relationship has also been confirmed in a work [34].

The mix GK, probably due to the very low strength of the structural framework, degrades upon intensive water removal from the sample under a high-temperature effect.

For all geopolymer mixes produced using “aged” alkaline solution (except for the GPC-24), high-temperature treatment provides strengthening degree from 0.65 for GMK-24 to more than 1.5 times (for mixes R-24 and GK-24).

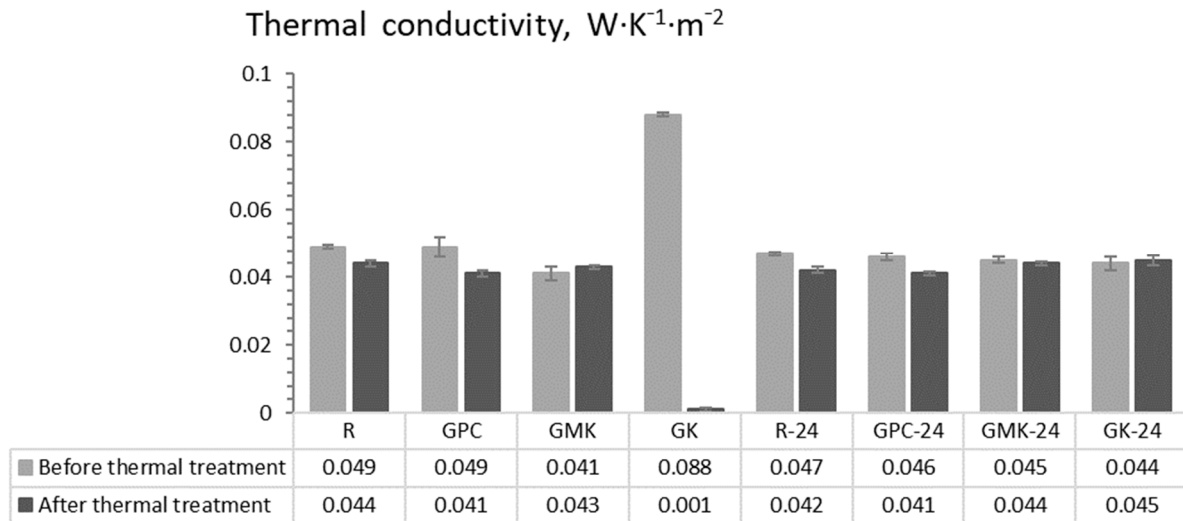
In the case of mixes GPC and GK, high-temperature treatment leads to a significant decrease in the compressive strength (up to 88% for GPC-24 vs. GPC), up to their complete destruction (for GK-24).

Although the previously obtained scientific results [35,36] indicate the effective use of MK in geopolymer systems, in this study, CGP specimens containing K and MK regardless of the introduction method of alkaline activator NaOH (mixes GMK, GMK-24, GK, GK-24) demonstrate low compressive strength vs. reference composition R. This can be explained by the fact that the reactivity of K and MK varies in a wide range of reactivity, depending on the ratio of basic oxides  $\text{SiO}_2/\text{Al}_2\text{O}_3$ . According to the classification presented in the study [37], increasing the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio from 2.25 to 3.25, the compressive strength significantly increases and reaches a maximum at a  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of about 3–3.5. A further increase in the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio again leads to a decrease in the compressive strength.

Additionally, the study [38] describes low reactivity of kaolin with the ratio  $\text{SiO}_2/\text{Al}_2\text{O}_3 = 1.7$ , which confirms the results of the current article. The introduction method of alkaline activator and high-temperature treatment showed reduction effects of the thermal conductivity coefficient (TCC) of the mix R-24 vs. R up to 4% and 10%, respectively. In case of the mix GMK-24 vs. GMK, the increase of TCC of  $\approx 10\%$  was observed; however, high-thermal treatment resulted in a 2% reduction for GMK-24 and a 5% increase of TTC (Figure 6). At the same time, in the case of the mixes GPC and GK, the introduction method of alkaline activator seems to be very influential parameters to control thermal



conductivity response of CGP. For the TCC of these mixes before high-thermal treatment, the values of TCC were 70% and 50% higher in comparison with the GPC-24 and GK-24, respectively. Regarding the effect of high-temperature treatment, the mixes GPC-24 and GK-24 demonstrated a 12% reduction and a 2% increase in the TCC values, respectively. A wide range of deviations of the TCC values among all experimental mixes indicates the relevance of the mineral modifying component used for CGP synthesis.



**Figure 6.** Effect of the introduction method of alkaline activator NaOH on thermal conductivity under different temperature conditions.

### 3.2. Calculated and Measured Porosity Characteristics

Using the values of the real density for the raw materials as well as their proportions in CGP compositions (Table 2), the values of the true densities for each experimental composition of CGP were calculated using formula (7).

Below, a demonstrative calculation of true density for the reference mix R is presented (Table 4).

**Table 4.** Component composition cellular geopolymer concrete, by wt.

Raw Materials	$\rho_{true}, g/cm^3$	Ratios, %
FA	1.87	53.5
NaOH	2.13	8.02
Water	1.00	38.3
Foaming agent	1.10	0.17

The true density of mix R was calculated using the formula (7):

$$\rho_{true, mixR} = 0.535 \cdot 1870 + 0.0802 \cdot 2130 + 0.383 \cdot 1000 + 0.0017 \cdot 1100 = 1550 \text{ kg/m}^3$$

The next step was to calculate the indicators of total porosity  $P_{total}$  according to the formula (3). Using the average density data (Table 4) for mix R ( $0.732 \text{ g/cm}^3$ ), the following equation was obtained:

$$P_{total} = \left(1 - \frac{0.732}{1.550}\right) \cdot 100\% = 52.8\%.$$

In addition, for experimental CGP specimens, the porosity was determined analytically (low-temperature adsorption-desorption of nitrogen N using the BET method). Since this method does not allow for directly determining the porosity values, it was calculated

from the following analytical data: the pore volume in the specimen at a relative pressure  $P/P_0 = 0.984699778$  (data obtained from the device) and the weight of the specimen. Analytical data obtained by the BET method are presented in Table 5.

**Table 5.** Initial data from Tristar II 3020 Micromeritics specific surface area and porosity analyzer for experimental geopolymers mixes.

Mix ID	Pore Volume in the Specimen at a Relative Pressure $P/P_0 = 0.984699778$ ( $V_{P/P_0}$ )		Specimen Weight ( $m_0$ ), g	
	Before TT	After TT	Before TT	After TT
R	0.002097	0.000052	1.1506	1.2142
GPC	0.011405	0.002097	1.0848	1.1620
GMK	0.005102	0.001563	1.0436	1.0876
GK	0.009027	0.004900	1.1244	1.1239
R-24	0.000790	-	0.9977	-
GPC-24	0.016408	0.001655	1.0054	0.9742
GMK-24	0.006274	0.001335	1.2268	0.9295
GK-24	0.004460	0.004193	1.0602	1.0391

To calculate porosity using data from the BET method, formula (8) was used:

$$P_{BET} = \frac{V_{pore}}{V_{general}} \cdot 100\% \quad (8)$$

where  $V_{pore}$  is the pore volume in a specimen with a mass of 1 g,  $\text{cm}^3$ ;  $V_{general}$  is volume of a specimen with a mass of 1 g,  $\text{cm}^3$ .

The pore volume  $V_{pore}$  in a specimen with a mass of 1 g was calculated with the formula (9) using the data from Table 5:

$$V_{pore} = m_0 \cdot V_{P/P_0} \quad (9)$$

where  $m_0$  is the weight of a specimen of CGP, g;  $V_{P/P_0}$  is pore volume in a specimen with a mass of 1 g,  $\text{cm}^3$ .

The volume of a specimen with a mass of 1 g  $V_{general}$  was determined by the formula (10):

$$V_{general} = \frac{1}{\rho_{average}} \quad (10)$$

A feature of the nitrogen adsorption–desorption method is the determination of porosity mainly in the nanoscale range (nanoporosity). Therefore, it is incorrect to carry out a comparative analysis of the calculated and analytical porosity values, because pores in the full-size range are taken into account in the calculated porosity values. At the same time, one of the tasks of this study was to determine the degree of correlation between the average density of the experimental CGP and their values of porosity, obtained by calculation and analytically using the BET method. Additionally, it was planned to study the effect of high-temperature treatment on the porosity and average density of the experimental mixes. The obtained data of the calculated true density, as well as the calculated and analytical values of porosity, are presented in Table 6.

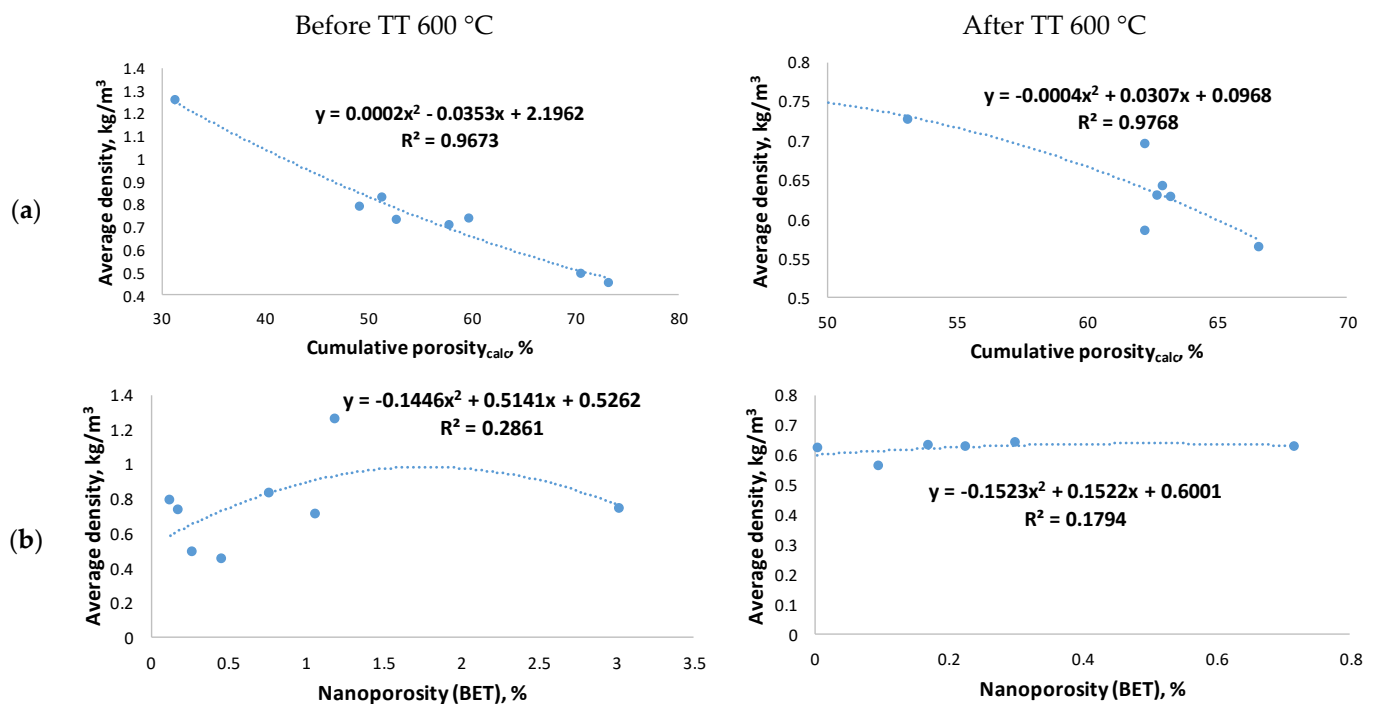
Data in Table 6 shows the differences in porosity before and after high-temperature treatment at 600 °C in both cases:

- Porosity determined by calculation method;
- Porosity determined using analytical equipment according to the BET method.

Figure 7 shows the correlation between the average density (Table 3) from the calculated and analytical values of porosity (Table 6) for the experimental CGP composites.

**Table 6.** Physical characteristics of CGP composites.

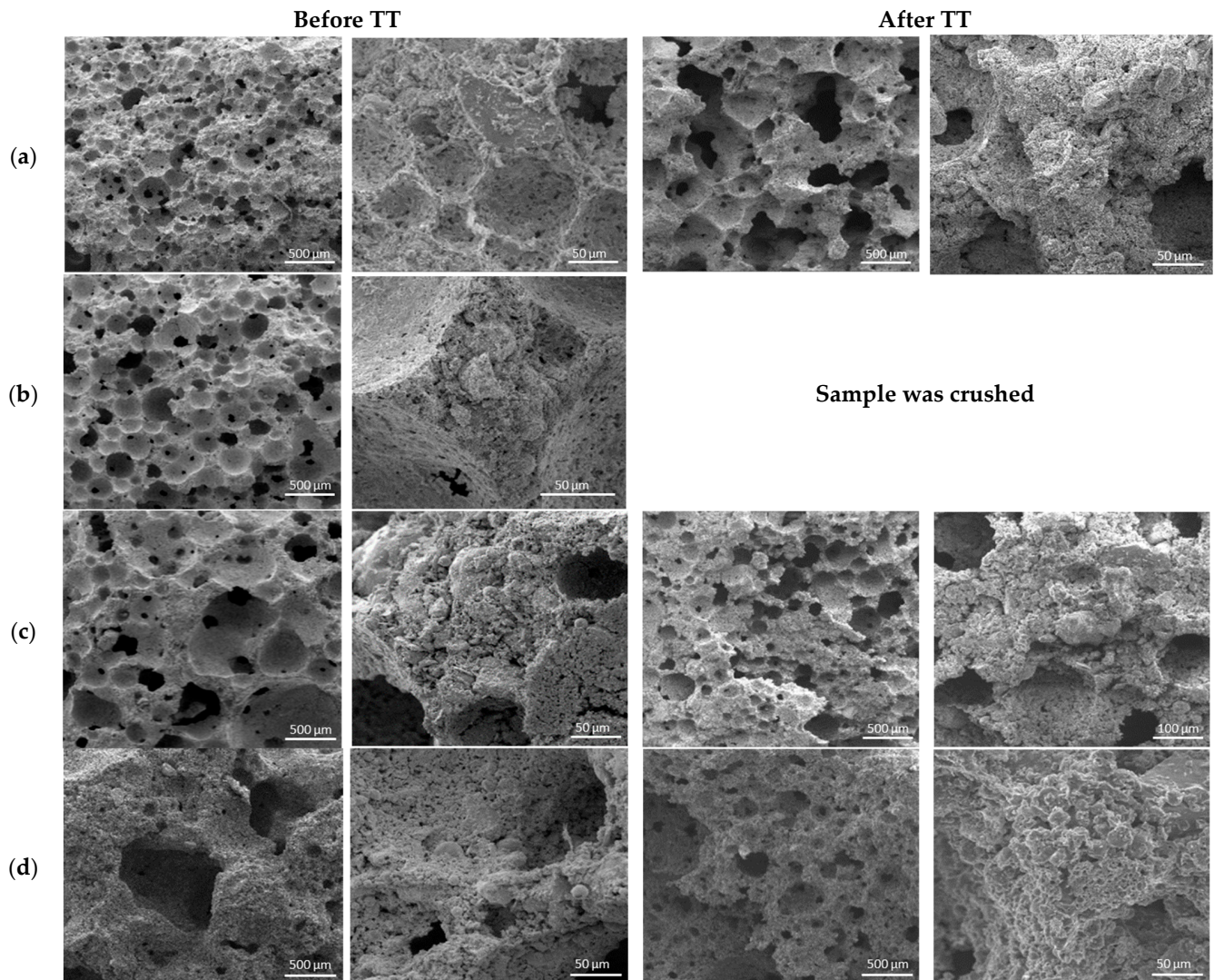
Mix ID	True Density $\rho_{true.composites}$ g/cm <sup>3</sup>	Calculated Porosity, %		BET Nanoporosity	
		Before TT	After TT	Before TT	After TT
R	1.55	52.8	62.2	0.177	0.0037
GPC	1.84	31.3	62.2	1.19	0.169
GMK	1.69	70.6	66.6	0.263	0.096
GK	1.71	73.3	-	0.456	-
R-24	1.55	49.2	53.1	0.122	-
GPC-24	1.84	59.8	62.9	3.02	0.3
GMK-24	1.69	57.9	62.7	1.06	0.225
GK-24	1.71	51.3	63.2	0.76	0.716

**Figure 7.** Correlation between average density and total porosity for CGP: (a)—for calculated total porosity; (b)—for nanoporosity measured by the BET method.

The curves of the dependence of the average density and total porosity in Figure 7 show that the values of the calculated porosity have a high degree of correlation with the average density of the experimental compositions of CGP without high-temperature treatment ( $R^2 = 0.967$ ) and after high-temperature treatment at 600 °C ( $R^2 = 0.969$ ) as well. In both cases, the correlation in the combination “average density—calculated porosity” is best described by a polynomial function. Analyzing the relationship between the average density and nanoporosity measured using the BET method (Figure 7b), it should be noted that the correlation dependence is very low in both cases: without high-temperature treatment and after treatment at 600 °C ( $R^2 = 0.286$  and  $R^2 = 0.179$ , respectively). Probably, such low correlation is not only due to the limited size range of measurements by the Tristar II 3020 analyzer, but also due to the prevalence of closed pores in the studied cellular geopolymer structures, which are not taken into account when using the BET method. Thus, a comparative analysis of the correlation between average density and porosity demonstrated an obvious advantage of using the computational method vs. the analytical BET method in terms of the degree of accuracy of the results obtained.

### 3.3. Microstructure of Cellular Geopolymers

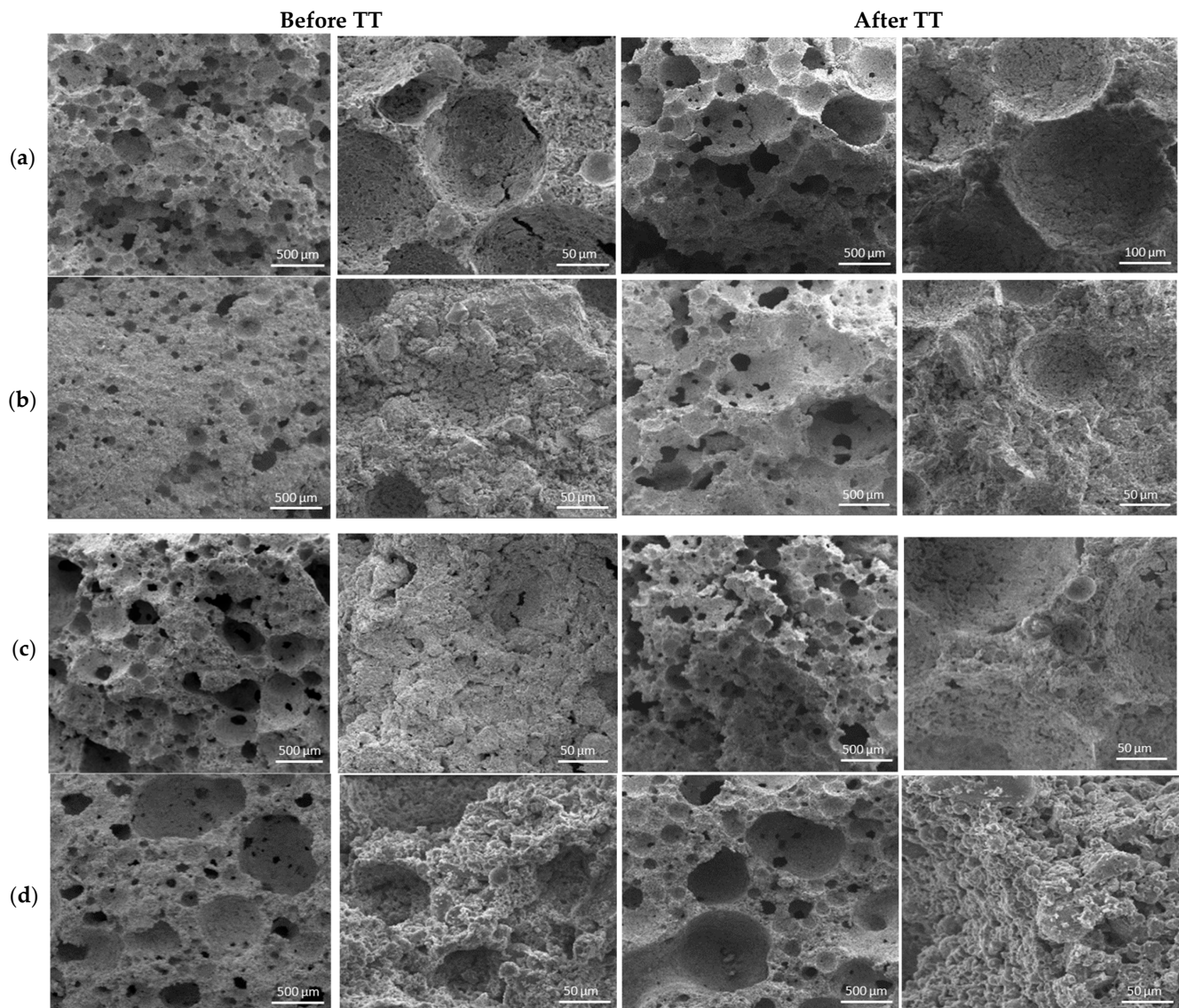
The microstructure of CGP with different compositions was studied (Figures 6 and 8), as well as using two introduction methods of alkaline activator, which are freshly prepared aqueous NaOH solution (Figure 8) and 24 h “aged” aqueous NaOH solution. The microstructure of cellular geopolymers before and after high-temperature treatment at 600 °C is presented in Figure 9.



**Figure 8.** Microstructure of CGP composites freshly prepared alkaline NaOH solution before and after high-temperature treatment (TT): (a)—GPC; (b)—GK; (c)—GMK; (d)—R.

Analysis of the microstructure of the mix GPC (Figure 8a) demonstrates that high-temperature treatment leads to an increase in the number of communicating and open pores due to the destruction of weak inter-pore partitions. As a result, there is an integration of the pores, a defect of their regular spherical shape. The structure becomes much more friable. When using an aged NaOH solution (mix GPC-24, Figure 9a), high-temperature treatment has a much less destructive effect on the cellular structure of the specimens. The microstructure of GPC and GPC-24 mixes is in agreement with their compressive strength values (Table 3). For mixes GPC and GPC-24, high-temperature treatment at 600 °C leads to a decrease in compressive strength by 89% and 39%, respectively.





**Figure 9.** Microstructure of CGP with different compositions using an aged alkaline NaOH-solution, before and after high-temperature treatment (TT): (a)—GPC-24; (b)—GK-24; (c)—GMK-24; (d)—R-24.

Similar changes in microstructure are observed in the mixes GK and GK-24 using kaolinite (Figures 8b and 9b). When using a freshly prepared NaOH solution (mix GK, Figure 8b), a structure is formed with a uniform pore volume distribution and a high concentration of closed pores with a regular spherical shape and approximately the same size. However, when analyzing the compressive strengths for mixes GK and GK-24 (Table 3), the skeleton of the specimens is observed as very weak (0.059 MPa), which is completely destroyed under the high-temperature treatment. Most likely, the destruction is caused by the intense removal of free water from the friable cellular skeleton. However, the use of aged NaOH solution for the mix GK-24 (Figure 9b) leads to the formation of a significantly denser structure and wider distribution of pores in size. The thickness of the interpore partitions has greatly increased, and the number of communicating and open pores have also increased. In this case, high-temperature treatment reduces the concentration of unreacted raw components and thereby leads to the compaction of the structure. Additionally, for mix GK-24, the formation of a stronger matrix structure is observed, which hardens as much as 1.5 times at temperatures of 600 °C.

In mixes GMK and GMK-24 using metakaolin (Figures 8c and 9c), a uniform porous structure is formed, with a high concentration of closed pores; pore sizes vary over a

wide size range. For these mixes, the method of using an alkaline activator and high-temperature treatment does not affect the pore structure. The SEM photomicrographs in Figures 8 and 9 for the mixes GMK and GMK-24 are identical. However, for mix GMK after high-temperature treatment, and for composition GMK-24 before and after high-temperature treatment, a more monolithic structure of the interpore partition and matrix is observed. A denser and more-monolithic structure is observed for mix GMK-24 after high-temperature treatment. An analysis of the compressive strength of these mixes (Table 3) showed that there is a correlation with the features of the microstructure, where there is a strengthening of the structural framework during high-temperature treatment in the range from 0.65 to 2 times. Additionally, the use of an aged alkaline NaOH solution provides the structure hardening by more than 2.5 times. The microstructure of the reference mixes R and R-24 before high-temperature treatment is identical. There is a low degree of porosity, an uneven distribution of pores in volume and size. The pores are predominantly large, with an irregular shape and friable intrapore walls. However, for both mixes, high-temperature exposure contributes to the formation of a more developed pore structure with distinct interphase boundaries “pore–interpore partition”. Additionally, for compositions after high-temperature treatment, a more formed, dense geopolymer structure is observed vs. the structure of mix R without high-temperature treatment. These features of the microstructure confirm the effect of increasing the strength of mixes R and R-24 after high-temperature treatment up to 4 times. The influence of the method of using an alkaline activator for an unmodified geopolymer (mix R) on the morphology of the pore structure is insignificant, but it clearly affects the strength characteristics. An increase in compressive strength is observed when using an aged NaOH solution.

Thus, when comparing the microstructure and compressive strength, the following conclusions can be drawn about the influence of the introduction method of alkaline activator and high-temperature treatment on the structure of experimental cellular geopolymers:

- The effect of high-temperature treatment on the PC-containing CGP shows a negative effect on the formation of the pore structure, regardless of the way the alkaline activator is used. This is because the hydrated PC is not a thermal resistant material. Therefore, the destruction of the cement structure at 600 °C inevitably initiates the degradation of the integrity of the geopolymer cellular structure as a whole. The 24 h aged e NaOH solution leads to the formation of a less-strong structure before high-temperature treatment, but provides higher thermal resistance;
- In the mix containing kaolinite, the 24 h aged NaOH solution has a positive effect on the implementation of structure-forming processes, but negatively affects the formation of a correct uniform pore structure. In this case, high-temperature treatment helps to strengthen the structure and increase its thermal resistance;
- For the mixes using metakaolin, the introduction method of alkaline activator and the high-temperature treatment have no significant effect on the pore morphology. However, these two factors significantly intensify chemical reactions, which leads to the formation of a denser and more-monolithic structure of the inter-pore space and, as a consequence, to the strengthening of the framework CGP;
- Such as in the case of metakaolin, in the mix without a MMC, the introduction method of alkaline activator and high-temperature treatment does not have a noticeable effect on the morphology of the pore space but contributes to the formation of a more-monolithic structure of the inter-pore space and the strengthening of the structure. However, 24 h aged NaOH solution promotes the formation of a more developed pore morphology during high-temperature treatment.

#### 4. Conclusions

In this research study, the influence of several modifying methods, such as the introduction method of alkaline activator, the introduction of a mineral modifying component and its nature, as well as high-temperature treatment on the physical, mechanical and thermal insulation properties of geopolymer foams was established. The effect of the



modifying methods on formation of the pore structure and pore morphology was also studied. The obtained experimental results and relations allow the formulation of the following conclusions:

- The nature of mineral modifying agent dramatically affects pore structure formation, compressive strength, average density and porosity of cellular framework, as well as thermal conductivity. Plus, the hardening effect of cellular geopolymers is provided only by PC as a MMC in case of using a freshly prepared NaOH aqueous solution. However, PC-containing CGP are not thermal-resistant composites;
- The way the alkaline activator is used for CGP synthesis plays a significant role in compressive strength development for all experimental mixes. The 24 h aged NaOH solution contributes to the formation of a more durable matrix structure of CGP; the formation of a more regular pore structure, which leads to reduction of average density, yet enables to increase compressive strength of CGP and also contributes to an increase thermal-resistant properties of the studied CGP specimens;
- The 24 h "ageing" of NaOH aqueous solution contributes to a more-complete realization of chemical processes. This statement is supported by significant areas of a monolithic structure formed within the interpore space. In addition, a more-regular pore volume and size distribution, as well as a higher degree of pore integrity, is a good indicator of lower thermal conductivity for mixes produced with 24 h aged alkaline NaOH solution vs. specimens produced with freshly prepared alkaline solution;
- Lower values of the thermal conductivity coefficient are typical for unmodified CGP and MK-containing CGP composites, as well. This is because FA and MK are characterized by a predominantly vitreous structure, the concentration of which increases during geopolymerization;
- The introduction method of alkaline activator did not have a noticeable effect on thermal conductivity for all studied CGP. However, the high-temperature treatment of CGP at 600 °C led to decrease in thermal conductivity;
- A high degree of correlation was revealed between the average density and the calculated total porosity for the studied CGP before ( $R^2 = 0.967$ ) and after ( $R^2 = 0.969$ ) high-temperature treatment at 600 °C. At the same time, the correlations between the average density and the nanoporosity measured by the BET method were  $R^2 = 0.286$  and  $R^2 = 0.179$ , for the mixes before and after high-temperature treatment at 600 °C, respectively. Thus, the proposed computational method for determining total porosity is more efficient and practical.

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