

# **Development of polystyrene-geopolymer composite for thermal insulating material and its properties with special regards to flame resistance**

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**Abstract.** As a first part of the research, systematic experimental series were conducted in order to develop an appropriate fly ash-based geopolymers binder focusing on the workability of the paste. In these series, the NaOH molar ratio and water glass/NaOH ratio were investigated and the fineness of the fly ash was optimized presented in a recent paper. Based on these results the effect of metakaolin on the mechanical properties was studied. After developing the appropriate binder, experimental series were carried out using ground polystyrene as light aggregate in various concentration (from 30 V/V% up to 98 V/V%) and geopolymers as a binder in order to develop a heat insulating material. Compressive and flexural strength, specimen density, flammability, freeze-thaw resistance were determined in order to characterize the composite product. As a result of the experimental investigation, it was found that the flexural strength of the composite was found to be ~400 kPa which is as high as the original polystyrene heat insulating panel. Additionally, the flammability was much better than the original pure PS product, the sample was not ignited even at higher PS content (90%). Furthermore, the freeze-thaw resistance of the composite improved compared with the neat geopolymers.

## **1. Introduction**

Geopolymers are inorganic polymers prepared by the reaction of alumino-silicate bulk material and alkali activator solution at high pH. The products have high early compressive strength, low drying shrinkage, good fire resistance and superior durability in the aggressive environment compared to Portland cement concrete [1,2]. Geopolymers can be used as a binder of lightweight aggregate for producing heat insulating composite [3], especially for organic compounds to improve its flame resistance.

It was proved in recent papers that geopolymer properties are controllable by mechanical activation (grinding) of the secondary raw materials [4–6]. Furthermore, chemical activation parameters (molar ratio of NaOH, composition of alkaline activator) affected the characteristics of the resulted geopolymers, i.e. compressive strength, specimen density, structure, ... On the other hand, these secondary material based geopolymers with engineered properties might be appropriate to use as a binder for lightweight aggregate.

The main objective of the project reported in this paper is to develop a new environmentally friendly heat insulating material, polystyrene-geopolymer composite, using secondary raw materials. The starting material of the polystyrene (PS) was waste from a construction site and package material.

Regarding the geopolymers binder, fly ash was used as the main component activated mechanically as well as chemically using various alkaline liquids.

## 2. Materials and methods

### 2.1. Materials

Landfilled brown coal fly ash (BCFA from Tiszaújváros, Hungary), fresh lignite type fly ash (LFA) (from Mátra power plant, Visonta, Hungary) and metakaolin (MK) were used as the main component for geopolymers binder development. To increase material reactivity and fineness, the raw fly ash was mechanically activated by using continuous operating ball mill. Characteristic particle size and the geometric “outer” specific surface area (SSA) values of fly ashes and metakaolin can be found in Table 1. The chemical composition of raw materials is given in Table 2.

**Table 1.** Characteristic particle size values and specific surface area (SSA) of raw materials.

	Visonta raw fly ash (LFA)	Tiszaújváros raw fly ash (BCFA)	Visonta ground fly ash	Tiszaújváros ground fly ash	Metakaolin (MK)
$x_{10}$ ( $\mu\text{m}$ )	13.2	22.1	8.4	5.5	3.2
$x_{50}$ ( $\mu\text{m}$ )	46.7	78	26.3	17.3	6.1
$x_{80}$ ( $\mu\text{m}$ )	85.4	145.6	58.6	49.4	8.7
SSA ( $\text{cm}^2/\text{cm}^3$ )	2018	1182	3330.1	4977.4	22119

It can be seen from Table 1 that LFA represents a much finer particle size distribution than BCFA. However, due to the same grinding residence time, BCFA became finer than LFA. On the other hand, MK has the finest size distribution.

**Table 2.** The XRF chemical composition of the main raw materials (w/w%).

Chemical component	BCFA	LFA	MK
$\text{SiO}_2$	61.32	45.5	51.4
$\text{Al}_2\text{O}_3$	26.71	14.2	45.9
$\text{MgO}$	0.89	3.58	0.26
$\text{CaO}$	1.5	12.8	0.40
$\text{Na}_2\text{O}$	1.06	0.4	0.05
$\text{K}_2\text{O}$	1.72	1.59	1.34
$\text{Fe}_2\text{O}_3$	4.27	11.1	1.12
$\text{MnO}$	-	0.174	0.033
$\text{TiO}_2$	-	0.48	0.050
$\text{P}_2\text{O}_5$	-	0.297	0.064
$\text{SO}_3$	0.25	1.7	<0.013
LOI, 1050°C	1.92	2.33	-

Before geopolymers-polystyrene composite production, the PS was comminuted in two stages by using rotary shear shredder (first stage) and hammer mill (second stage). As a result of crushing unique polystyrene beads were obtained. For geopolymers binder production waterglass and NaOH solution were used as an alkaline activator.

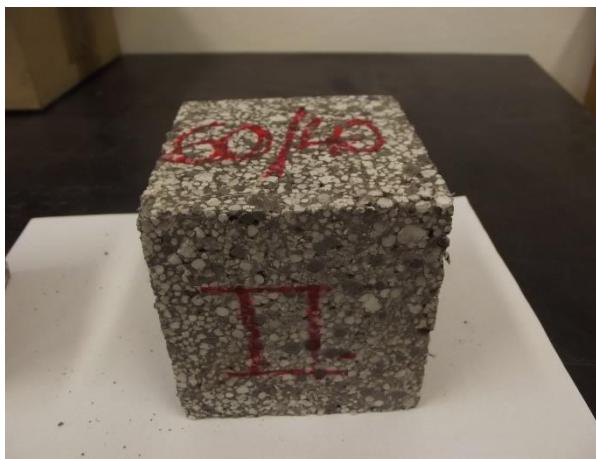
## 2.2. Methods

**2.2.1. Geopolymer and composite technology.** Systematic experimental series were conducted in order to develop an appropriate binder focusing on the workability of the geopolymer paste. As a first step NaOH molar ratio (6; 8; 10; 12; 14 M) and water glass/NaOH ratio (0/100; 25/75; 50/50; 75/25; 100/0 %) was investigated and presented in a recent paper [4]. In the case of optimal NaOH molar ratio and waterglass/NaOH ratio, the effect of metakaolin dosage on the mechanical properties of geopolymer binder was studied in the present work. The ratio of metakaolin to fly ash was 5; 10; 15 and 20 m/m%. The fly ash part consisted of ground BCFA and LFA in 50-50%. In each case, heat curing of geopolymer was carried out at 30°C for 6 hours.

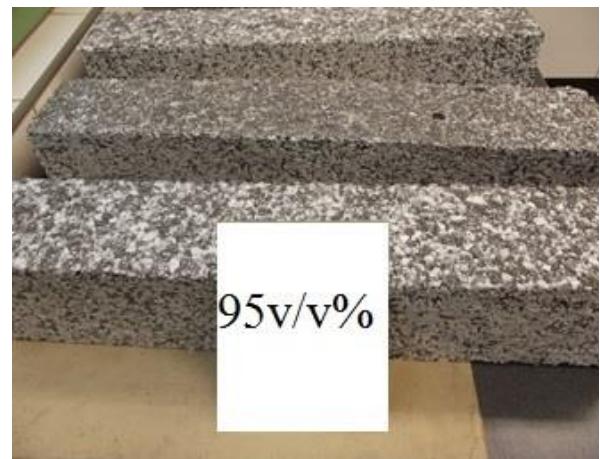
After developing the optimal geopolymer binder, experimental series were carried out using comminuted waste polystyrene as lightweight aggregate in various concentration (from 60% up to 98v/v%) and geopolymer as a binder. The geopolymer paste-polystyrene mixture was prepared in a forced action mixer. In the case of high polystyrene content (98 v/v %) the mixture was compacted by using cyclic pressure compaction. In other cases, the vibrating table was used for compaction. After compaction, the composite products were conditioned at 30°C for 1 hour. Then the finished specimens were demoulded and stored.

Compressive- and flexural strength, specimen density, flammability, freeze-thaw resistance were determined in order to characterize the composite product.

**2.2.2. Strength test.** The first stage of strength test of the polystyrene-geopolymer composite was the analysis of the behaviour of the binder. The influence of the following factors on the uniaxial compressive strength of the binder was analysed separately: 1) ratio of the added metakaolin and 2) age of the specimen.



**Figure 1.** Probe to determine the uniaxial compressive strength of the composite.



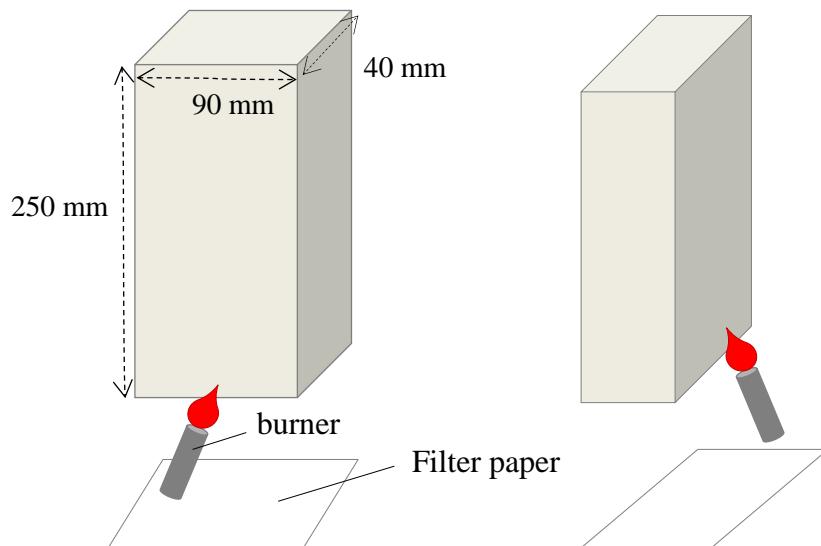
**Figure 2.** Probes to determine the flexural tensile strength of the composite.

The design of the binder composition was based on previous results [4,6]. Uniaxial compressive- and flexural strength tests of the specimens made of various composition of geopolymer binder and polystyrene were carried out. The method of the measurements was as usual in the construction materials industry [7]. Cylindrical probes of the binder and cubic ones (Figure 1) of the composite were used for the measurements of uniaxial compressive strength. Three-point bend testing was used to determine the flexural tensile strength of the composite prisms in the size of 100x100x500 mm (Figure 2).

**2.2.3. Flammability test.** The method of the test was described by the EN ISO 11925-1:2010 standard (Part 2. Single-flame source test) [8].

This fire test method has been developed to define the reaction of products to fire. The method specifies a test for determining the ignitability of products by direct small-flame impingement under zero impressed irradiance using vertically oriented test specimens.

Although the method is designed to assess flammability, this is addressed by measuring the spread of a small flame up the vertical surface of a specimen following application of a small (match-sized) flame to either the surface or edge of a specimen for either 15 s or 30 s. The determination of the flaming formation droplets depends on whether or not the filter paper placed beneath the specimen ignites (Figure 3).



**Figure 3.** Schematic of the Flammability test.

The following parameters were examined and documented during the test:

1. Heating mode: the heating time (15 s or 30 s) and the heating position (side surface or edge of the sample).
  2. If the sample ignites: height the peak of the flame, and the time from the start of heating to the ignition.
  3. Is there observed any burning drops, which could ignite a filter paper, placed under the sample?
- Usually, 6-12 flammability tests are carried out which are classified into categories according to the EN 13501-1 standard [9].
- The flame was prepared by the controlled gas burner, the test specimen was positioned into the flame with a suitable stand (under fume cabinet) so that the edges of the specimen were horizontal and vertical positions. The flame reached the bottom edge of the specimen. At the end of the test, the specimens were visually inspected, the weight of the dripping product was measured. The absolute weights are given since the density of the test specimens is significantly different.

**2.2.4. Freeze-thaw resistance.** The freeze-thaw resistance of both geopolymers and geopolymers-polystyrene composite were measured. Three series of the investigation were carried out. During "A" series, standard size ( $\varnothing 35 \times 65$  mm) geopolymers cylinders were produced. The mortar itself was made of 55 m/m% ground fly ash and 45 m/m% activation liquor. The liquor was made of 75 m/m% Betol 39T water glass and 25 m/m% 12 M NaOH. Half of the test cylinders were stored under water for 28 days and strength of the test cylinders was measured. The other half of the test cylinders were stored under water for 24 hours at age of 18 days. Water saturated test cylinders were exposed to freeze cycles

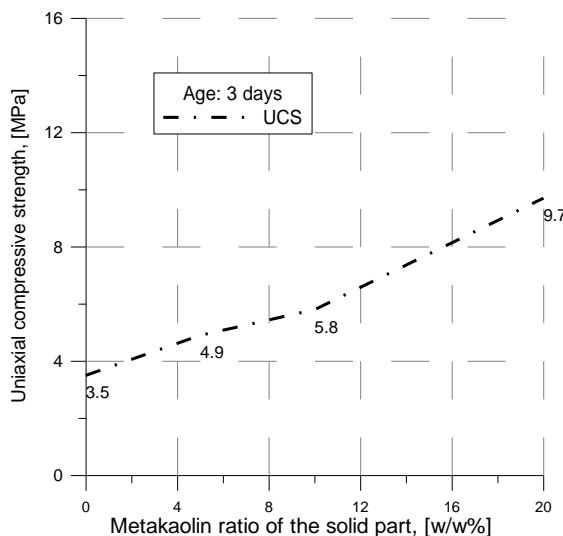
10 times in series. One freeze cycle was determined as 10 hours at -12°C temperature followed by 4 hour linear warm-up to 20°C and then maintain at the same temperature.

The same procedure has been carried out during measurement series "B", where geopolymers-polystyrene composite was produced. The geopolymer mortar was prepared according to the same recipe as it was in series "A", however, comminuted polystyrene was added and mixed. The rate of the polystyrene to the geopolymer binder was 90 V/V%. Measurement protocol was also the same, and the strength of original and frozen test specimens was measured and determined. A third measurement series were also carried out. In this series "C", the same mixture of mortar and polystyrene mixture was used creating cuboid specimens. Also, instead of full saturation, the test specimens between freeze cycles were only stored at 22°C temperature and under 90% relative humidity in a climate chamber modeling realistic scenario an insulation material normally exposed. In this case, strength for both original and frozen test cuboids was measured.

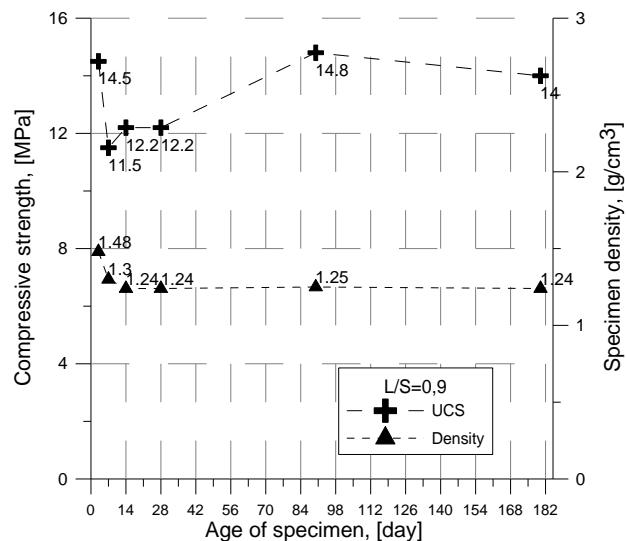
### 3. Results and discussion

#### 3.1. Strength of geopolymer and composite

Based on our present and previous results [4, 5, 6], it was found that the following factors influenced significantly strength of the geopolymer binder: origin of the fly ash, its activation type, composition and quantity of the activating solution, origin quantity of the added metakaolin, and age). Although, the results describe the tendency of the influence of the different factors on the strength of the geopolymer. Effects of the ratio of metakaolin and age of geopolymer are demonstrated in Figure 4 and Figure 5. It can be seen clearly that the addition of metakaolin has a positive effect on the fly ash based geopolymer compressive strength due to its high reactive components. More or less a linear relationship was found between metakaolin ratio and compressive strength based on the measured points.



**Figure 4.** Effect of metakaolin on the strength of the binder.

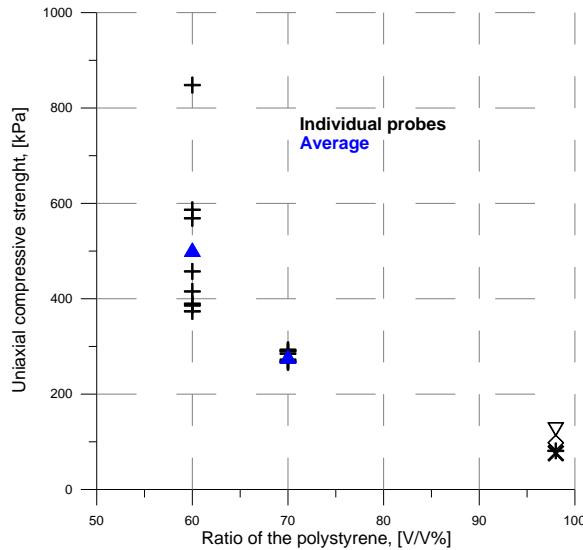


**Figure 5.** Effect of age on the strength of the binder (L/S: ratio of liquid to solid, alkaline activator to solid material).

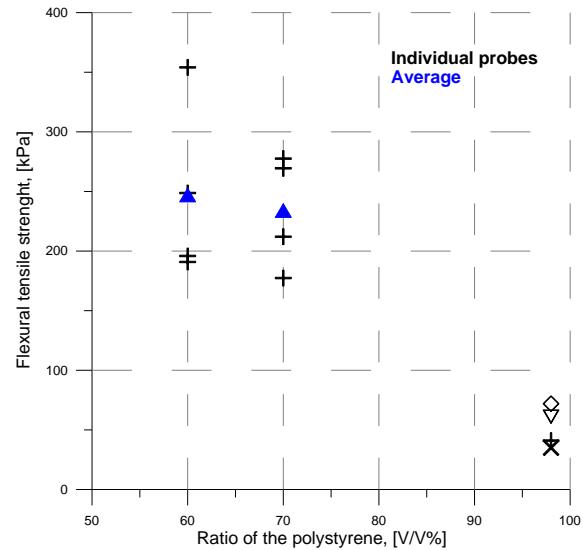
It can be established that the compressive strength of geopolymer binder decreased after 7 days but it starts to increase as a function of time and reaching 14.8 MPa after 90 days of aging. Possible explanation might be that the unreacted NaOH decreases the strength of the cross linked 3D network of the resulted product by dissolution of the freshly generated geopolymer. The reason of density decrease is due to the elimination of water from the specimen due to drying. This combined effect of remained water content and unreacted NaOH causes the decrease of strength.

The strength of the polystyrene-geopolymer composite depends basically on the strength of the binder and the ratio of the components. Strength properties of the composite can exceed the ones of the

polystyrene itself used as an additive (Figures 6 and Figure 7). The ideal ratio of the components can be determined considering other factors as well as strength is just one of the quality requirements.



**Figure 6.** Uniaxial compressive strength of the composite.

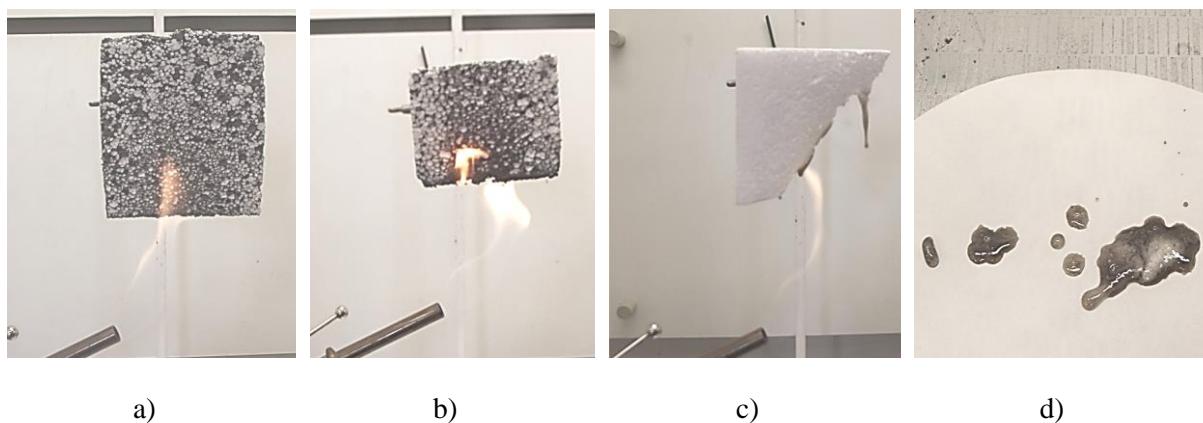


**Figure 7.** Flexural tensile strength of the composite.

The above compressive- and flexural strength results are comparable with those of other mineral based heat insulating panel products (perlite, mineral wool) [10].

### 3.2. Flammability

Figure 8 and Table 3 show the flammability test results of specimens made of polystyrene-geopolymer composite with various concentrations.



**Figure 8.** Pictures of the flammability tests, a) polystyrene content is 90 v/v%, b) polystyrene content is 98 v/v%, c) polystyrene content is 100 v/v%, d) dropping product of 100% polystyrene.

Figure 8 shows the images of the geopolymer – polystyrene composite and only PS during the flammability test series. These images indicate the good flame resistance of geopolymer: PS aggregate is protected against flame in the composite specimen (Figure 8 a). At the same time neat polystyrene is totally melted (Figure 8c-d).

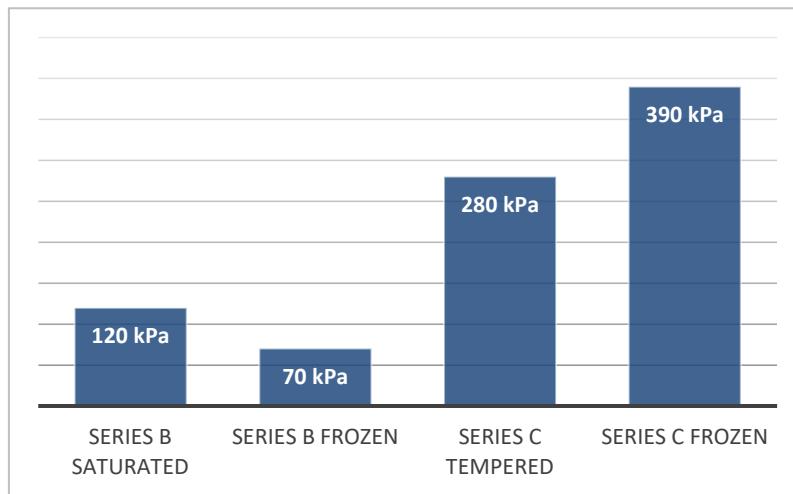
**Table 3.** Description of the specimen behaviour during flammability test.

Polystyrene content, [v/v%]	Average weight of dropping material [g]	Comment
100	0.41	The specimen was not burnt. It was melted and dripped.
98	1.79	The specimen was ignited until the flame reached it.
97	0.59	The specimen was ignited until the flame reached it.
94	0.35	The specimen wasn't ignited, only the edge of the bottom was smoldered.
90	0.24	The specimen wasn't ignited, only the edge of the bottom was smoldered slightly.
70	0.13	The specimen wasn't ignited, only the edge of the bottom was smoldered slightly.
60	0.08	The specimen wasn't ignited, only the edge of the bottom was smoldered slightly.

Results show the composite specimens with 60-94 v/v% polystyrene content were not ignited, were not fueled the flame, only the lower part of the samples was smoldered. In the case of 97-98 v/v % polystyrene content, specimens were ignited until the flame reached them. The weight of dropping product of polystyrene - geopolymers composites was increasing with polystyrene content, but the filter paper was not ignited by the dropping product. The sample of 100% PS was practically melted in the flame, the filter paper was not ignited by the dropping product. The filter paper was not ignited by the dropping product in any cases. In contrast to the 100% polystyrene, it is obvious that the geopolymers content of the samples inhibits the destruction caused by direct flame.

### 3.3. Freeze-thaw resistance

According to our measurement results, significant strength decrease was recorded in most cases during freezing. In series "A", the compressive strength of geopolymers mortar was around 7 MPa. Mortar test cylinders completely deteriorated by the freezing process after 3 cycles.



**Figure 9.** Compressive strength of composite after freeze-thaw test.

In series "B", the original composite compressive strength was 200 kPa, while water saturated cylinder had an average strength of 120 kPa. Freezing process did not deteriorate these specimens, however, final strength becomes lower 70 kPa as an average. Most surprising results has been achieved in series "C", where original composite strength was the same as in series "B" which average value elevated to 280 kPa after 10 days of climatic conditioning and become even higher, 390 kPa after 10 cycle freezing.

## 4. Conclusions

Based on this experimental investigation the following conclusions can be drawn. It was found that appropriate binding forces occurred between the geopolymer matrix and the polystyrene aggregate, hence composite material could be developed. The uniaxial strength of the geopolymer and the composite material can be improved by the addition of metakaolin due to its high reactivity. Geopolymer has a positive effect on the flammability of the polystyrene-based composite by improving flame resistance. Due to the curing of the geopolymer-polystyrene composite in a climate chamber under high humidity (simulating the real conditions of a heat insulating panel) its freeze-thaw resistance improved significantly.

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