



# Innovative Inorganic Insulation Materials (Mineral Slag Based Geopolymers)

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## Abstract

With the increase of energy price, the retrofitting of existing buildings with innovative insulation materials remains a key issue. Although a variety of insulation materials there is always a need for the development of new materials with improved thermophysical properties and low cost. Inorganic polymer materials are promising from this point of view. They are incombustible combining excellent physical and thermal properties with low production cost and significant environmental benefits. In this work, insulation materials with low density are produced with a low cost process and utilizing two different raw materials an industrial mineral and an industrial waste. In this paper, insulation materials with low density were produced with a low cost process utilizing two different raw materials: an industrial mineral and an industrial waste. The results indicated the potential of the application as insulation materials in the construction sector.

## Subject Areas

Material Experiment

## Keywords

Inorganic Polymers, Insulation Materials, Industrial Waste

## 1. Introduction

Although, the European citizens are becoming increasingly familiar with the concept of energy in use, that is the energy required by the occupants of an existing building, primarily for space heating, water heating and lighting, and of the need to reduce it, they have not realized yet that energy is needed not only to run a building but it also takes considerable amount of energy to create the building

products and the building itself. The embodied energy of a building, defined as the total energy required to produce the building components, transport them in place and construct the building, is a significant component of its lifecycle impact and can be the equivalent of many years of operational energy. An average household contains about 1000 GJ of energy embodied in the materials used in its construction, which is equivalent to about 15 years of operational energy consumption. Moreover, it is important to point out that when increasing the level of energy performance of buildings in operation, which is the current trend in the construction of new buildings dictated by many EC (EPBD 2010/31/EU) and member state directives, the embodied energy of the building materials represents a much higher percentage of the energy spent in the whole life cycle of the building. In modern times, one of the main goals in the construction industry is to increase energy efficiency by reducing energy losses, due to the high cost of cooling and heating a building. According to a study carried out in the 27 member states of the European Union, the high percentage of energy consumption in the last twenty years (250 - 300 Mtoe) is attributed to the requirements of the buildings for heating, cooling and ventilation due to their insufficient thermal insulation.

Conventional thermal insulation materials are divided into three subcategories: lightweight concrete (Aerated Concrete) based on cement, cellular concrete produced in autoclave (Autoclaved Aerated Concrete-AAC) also based on cement and organic thermal insulation materials (extruded polystyrene-XPS (DOW), expanded polystyrene-EPS and polyurethane foam). These materials are generally of high cost, including the cost of transportation to the construction site, as a result of which they are not preferred by manufacturers and consumers, and secondly, most of them are organic based, as a result of which they not only burn and decay in case of fire, but also propagate fire to the rest area of the building. A fire in a structure is an event which can have tragic consequences including loss of life, injuries, economic damage and environmental pollution. According to the statistics of the Greek Fire Service, the incidents of fires in building facilities for the year 2017 amounted to more than 3000. At this point, the case of the fire in Eastern Attica, which resulted in more than 100 victims, should be mentioned with 2500 destroyed houses. The severity of the effects of the fire is of the utmost importance also in terms of the structural body, mainly due to the risk of loss of stability and collapse of the bearing body but also due to the financial burden caused by the interventions, reinforcements after the fire.

The search for solutions to reduce and potentially restore the problems caused by a fire, leads to the development of new porous, fire-resistant, thermal insulation materials that reduce energy consumption in buildings and protect them from fire.

The geopolymerization technology [1] seems to be very attractive in developing new more effective insulation materials. Geopolymerization is a fast growing technology that involves a heterogeneous chemical reaction between several solid

aluminosilicate materials (naturally occurring minerals, industrial by-products or waste) and alkali metal silicate solutions at highly alkaline conditions and mild temperatures yielding amorphous to semi-crystalline polymeric structures. These structures constitute the matrix of a new family of innovative materials called geopolymers or inorganic polymers [1]-[7]. The inorganic polymeric materials possess excellent physical, chemical, mechanical and thermal properties such as high compressive and flexural strength, high surface hardness, micro- or nanoporosity, low water absorption and permeability, negligible shrinkage, thermal stability, fire and chemical resistance, which render them very promising materials for the construction sector.

This work aims at demonstrating the insulation properties of inorganic polymeric materials for their application as insulation materials on buildings which achieve low density and thermal conductivity (0.06 - 0.07 W/mK) with satisfactory mechanical properties.

## 2. Experimental

### 2.1. Materials

The slag used in the present study was provided by the metallurgical plant of the Greek company LARCO G.M.M.S.A. that treats laterites to produce ferronickel. The slag is generated during the reductive smelting of laterites in electric arc furnaces and is granulated using a flash water cooling process. For the synthesis of inorganic polymers, an adequate quantity of granulated slag was grinded to  $-100\ \mu\text{m}$  and the resulted powder had a mean particle size ( $d_{50}$ ) of  $15.05\ \mu\text{m}$  measured on a MALVERN Laser Particle Size Analyzer. The chemical analysis of the slag (Table 1) was performed by fusion with a mixture of  $\text{Li}_2\text{B}_4\text{O}_7/\text{KNO}_3$  followed by nitric acid digestion; the concentration of metals in the produced solution was determined with Atomic Absorption Spectroscopy (Perkin Elmer 2100 Spectrophotometer). According to Table 1, the slag is a siliceous material very rich in iron oxides and rich in alumina. It also contains substantial amounts of trivalent chromium, magnesium and calcium oxides, as well as traces of nickel. The mineralogical analysis of the slag determined with the X-ray diffraction

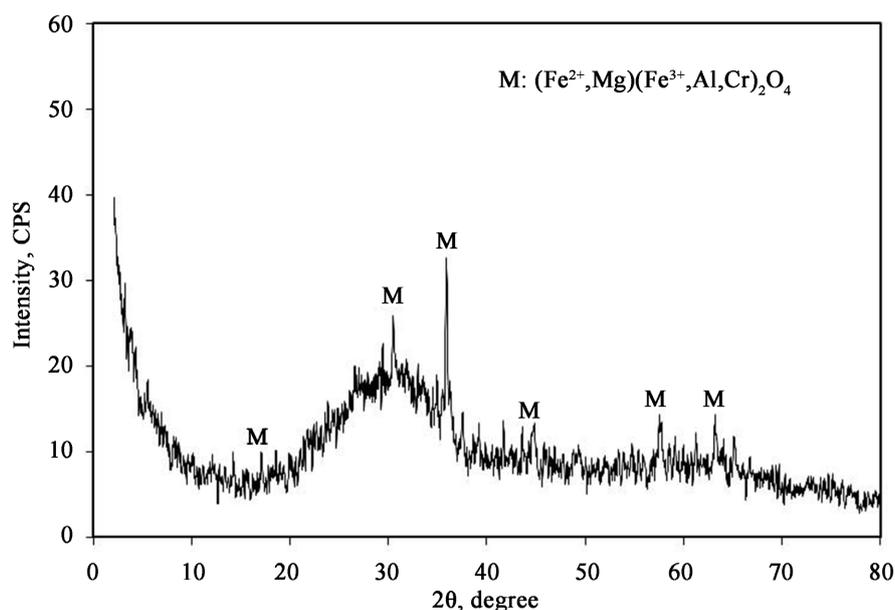
**Table 1.** Chemical analysis of slag (atomic absorption spectroscopy).

Species	% w/w
$\text{SiO}_2$	41.14
$\text{Al}_2\text{O}_3$	13.79
$\text{FeO}$	34.74
$\text{Cr}_2\text{O}_3$	5.41
$\text{MgO}$	3.59
$\text{CaO}$	0.71
$\text{Ni}$	0.14

method on a SIEMENS D 5000 diffractometer is shown in **Figure 1**. The slag is mainly constituted of an X-ray amorphous silicate or aluminosilicate phase indicated by the broad band registered between  $2\theta = 20^\circ$  and  $2\theta = 40^\circ$ . Ferrite-spinels  $[(\text{Fe}^{2+},\text{Mg})(\text{Fe}^{3+},\text{Al},\text{Cr})_2\text{O}_4]$  were detected as the only crystalline mineralogical phases in the slag (**Figure 1**). Magnetite ( $\text{Fe}_3\text{O}_4$ ) was the principal constituent of spinels accompanied by magnesioferrite ( $\text{MgFe}_2\text{O}_4$ ) and magnesiochromite ( $\text{MgCr}_2\text{O}_4$ ).

The metakaolin that was used in this study was provided from AGS Mineraux, a member of Imerys Minerals. Metakaolin had a mean particle size ( $d_{50}$ ) of  $13.68\ \mu\text{m}$  measured on a MALVERN Laser Particle Size Analyzer. As is shown in **Table 2**, it is a material rich in silica and alumina oxide. Its mineralogical analysis showed that it is mainly composed of an X-ray amorphous silicate or aluminosilicate phase which is accompanied by crystalline mineralogical phases that belong to Illite  $[(\text{K}, \text{H}_3\text{O})(\text{Al}, \text{Mg}, \text{Fe})_2(\text{Si}, \text{Al})_4\text{O}_{10}[(\text{OH})_2, (\text{H}_2\text{O})]$ , Quartz ( $\text{SiO}_2$ ), Anatase ( $\text{TiO}_2$ ) and Tridymite ( $\text{SiO}_2$ ).

Also, pure silicon dioxide (particle size  $< 64\ \mu\text{m}$ ) provided by Sigma Aldrich



**Figure 1.** X-ray diffractogram of the slag.

**Table 2.** Chemical analysis of metakaolin.

Species	% w/w
$\text{SiO}_2$	52.66
$\text{Al}_2\text{O}_3$	40.97
$\text{FeO}$	1.83
$\text{K}_2\text{O}$	1.18
$\text{TiO}_2$	1.43
$\text{Na}_2\text{O}$	0.71

was used for the synthesis of metakaolin geopolymer. (Figure 2) Finally, an alkaline sodium hydroxide solution was also used for the synthesis of inorganic polymers. The solution was prepared by dissolving pellets (Merck, 99.5% purity) of anhydrous sodium hydroxide in deionized water. Hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) is a widely known inorganic foaming agent which is thermodynamically unstable and therefore can be easily decomposed to water and oxygen gas [Equation (1)], with the latter playing the role of the blowing agent [1].



## 2.2. Experimental Procedure

Inorganic polymers were prepared according to the following procedure (Figure 3).

A homogeneous viscous paste was initially prepared by mixing mechanically the solid materials with sodium hydroxide solution at a solid to liquid ratio equal to 4 g/mL. Then the foaming agent was added in the paste, under high speed mixing and then was casted in appropriate open plastic (ERTACETAL<sup>®</sup>) moulds

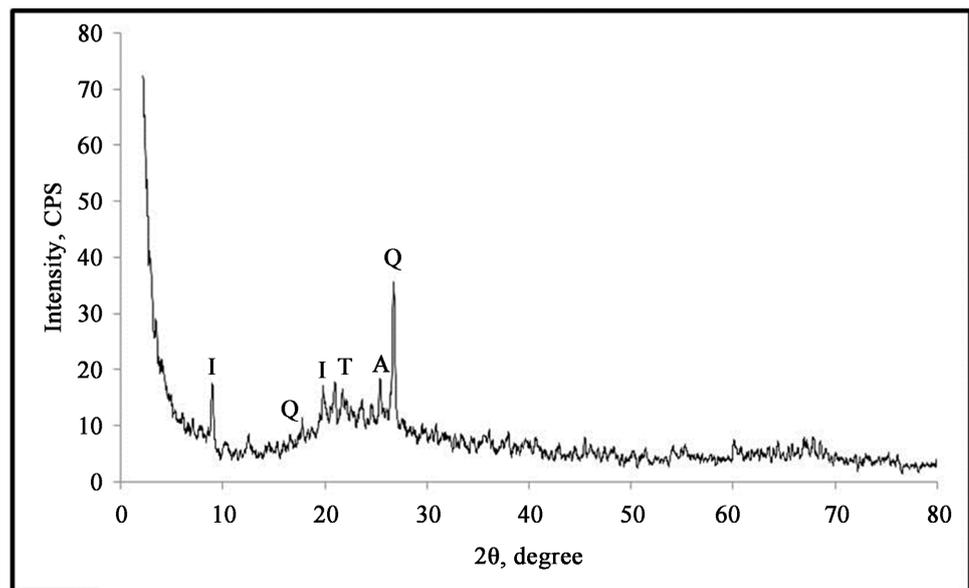


Figure 2. X-ray diffractogram of metakaolin.



Figure 3. Foamed geopolymer production steps.

and cured at ambient temperature for 48 hours. After curing, the specimens were de-moulded and the mechanical and thermophysical properties of materials were measured through a set of tests.

### 2.3. Analysis and Tests

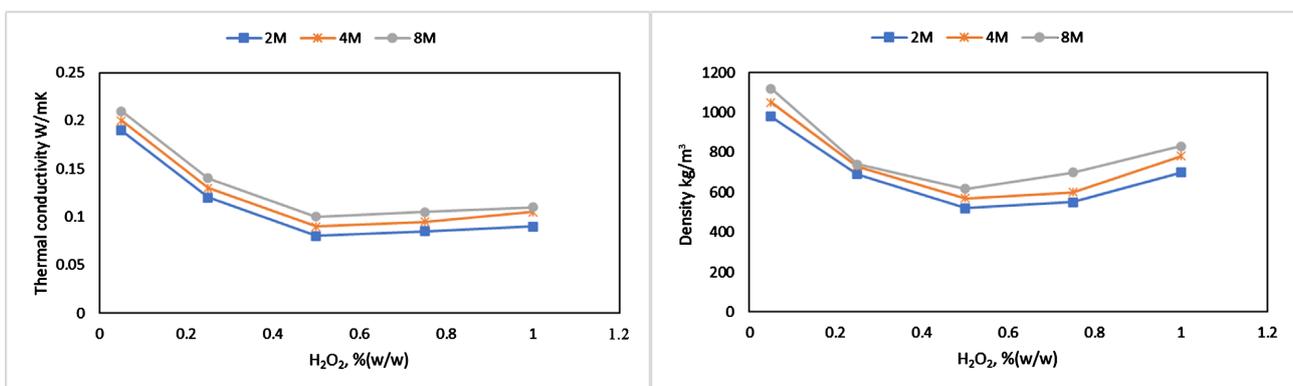
The geopolymer properties studied in this work include density measurement, uniaxial compressive strength and thermal conductivity. Compressive strength was measured using cubic specimens with dimensions  $50 \times 50 \times 50$  mm. The thermal properties of the geopolymer were assessed through its thermal conductivity, which was measured according to EN 12667:2001 on a  $15 \times 15 \times 2$  cm specimen.

## 3. Results and Discussion

Initially the foaming process was evaluated utilizing metakaolin as the main raw material. On this purpose the alkalinity of the solution was investigated in order to evaluate the effect on the foaming process. Three different alkalinities of sodium hydroxide were investigated (2 M, 4 M and 8 M) with a range of hydrogen peroxide 0.05% - 1% w/w to the solid mass. The solid to liquid ratio used was 4.0 g/ml which was the optimum for the foaming of the materials.

The results shown in **Figure 4** where a) the alkalinity seems to have a light effect on the thermal conductivity and final density of the material b) the 0.5%w/w quantity of hydrogen peroxide seems to be optimum value since a plateau is developed while further increase does not affect the density and thermal conductivity of the material. The optimum thermal conductivity achieved was 0.08 W/mK (with a relevant density of  $520 \text{ kg/m}^3$ ), which however is higher than our target of 0.06 - 0.07 W/mK.

The same procedure was followed also for the case of the slag as raw material. The results (**Figure 5**) were similar observing however an increase in the density and the thermal conductivity. This is attributed to the higher apparent density of the raw material compared to the metakaolin case ( $3.19 \text{ g/cm}^3$  to  $2.33 \text{ g/cm}^3$ ) which renders the foaming process more difficult comparing to the metakaolin case.

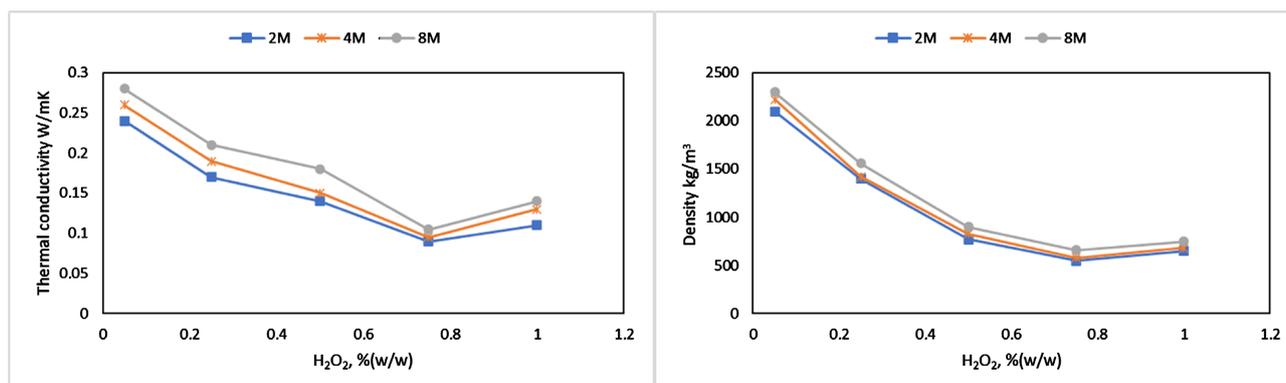


**Figure 4.** Effect of the alkalinity on the foaming process.

Based on this data there was no sense to investigate further materials utilizing exclusively slag since the density would be high and too far from our target for insulation materials (around 350 - 450 Kg/m<sup>3</sup>).

For both cases of raw materials, increase in the percentage of hydrogen peroxide did not provide materials with lower density, since the higher amount created more and larger bubbles and voids which were not stable and collapsed, thus the developed materials had finally higher density from the ones presented above.

In order to achieve lower density and thus lower thermal conductivity, a surfactant was added, assisting in maintain the voids and bubbles in the polymeric gel, preventing them from collapsing and thus creating more porous and homogenous materials. For this purpose, Hostapur OSB surfactant was used, which is a high foaming primary surfactant (anionic wetting and air entraining agent based on Alpha-olefin sulfonate, sodium salt). As it can be shown from **Figure 6** the addition of the surfactant retains the porosity of the geopolymer, since there is no collapse of the bubbles and voids and the cured material reaches lower density. In **Figure 6** both materials are synthesized with 2 M NaOH and 0.2%w/w to solid phase hydrogen peroxide as foaming agent, while in the b case 0.2%w/w to the solid phase, of surfactant has been added which seems to boost the foaming of the final material. The final density of the left picture cured



**Figure 5.** Effect of the alkalinity on the foaming process (slag case).



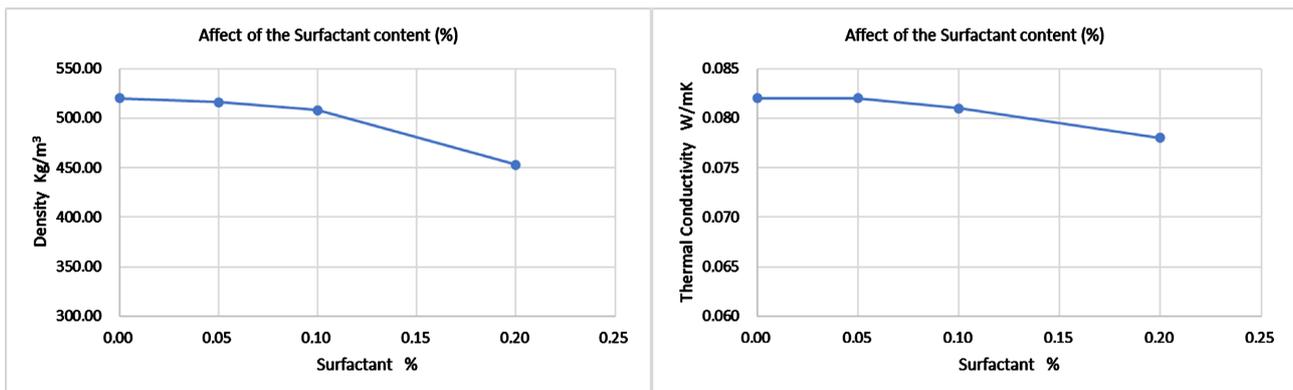
**Figure 6.** Effect of surfactant addition. Right picture geopolymer with 0.2%w/w surfactant, left no surfactant.

specimen was  $650 \text{ kg/m}^3$  while in the right picture (specimen with the surfactant), the density of the cured material reached  $450 \text{ kg/m}^3$ . The latter specimen reached a thermal conductivity value of  $0.078 \text{ W/mK}$ .

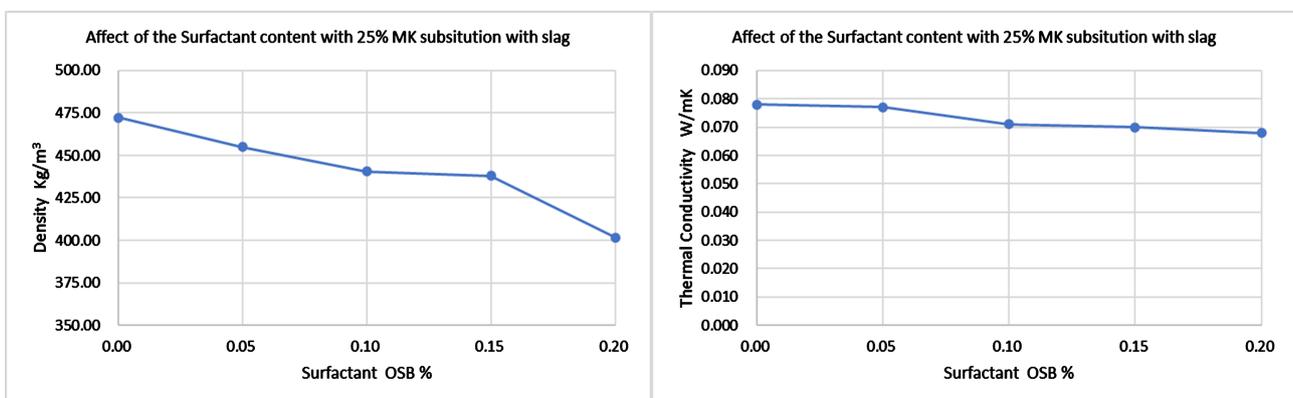
The summary of the analysis of the effect of the surfactant addition, is presented in **Figure 7** where it is clear that the 0.2% w/w was the optimum value while higher amounts resulted to unstable materials.

The last step of the analysis was to investigate the addition of slag in the synthesis which for sure would reduce the final cost of the material due to the fact that the slag is an industrial waste facing deposit problems. On this purpose the substitution of metakaolin with slag was investigated for two different percentages, 25% (**Figure 8**) and 50% substitution (**Figure 9**).

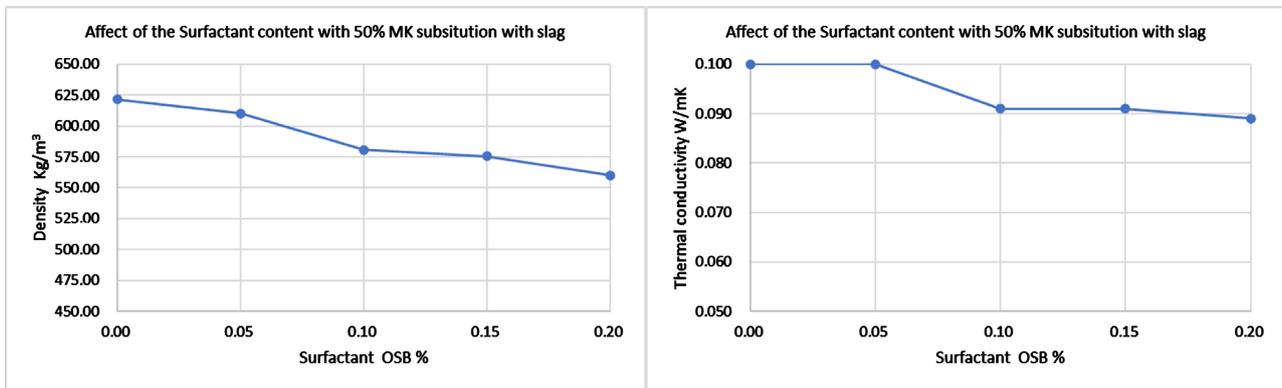
As it can be shown from **Figure 8**, the addition of 25 % of slag improved the thermal conductivity and the density of the material which is attributed to the improvement of the rheology during foaming. More precisely the addition of a raw material with a higher density but in low percentage enhanced the stability of the voids and bubbles during foaming acting in compensation with the surfactant. Thus, the cured material reached a density of  $400 \text{ kg/m}^3$  with a thermal conductivity of  $0.068 \text{ W/mK}$ . This material seems to be a very competitive product for the market and thus the mechanical properties were further investigated. As it can be shown from the below **Figure 10** the increase of the surfactant



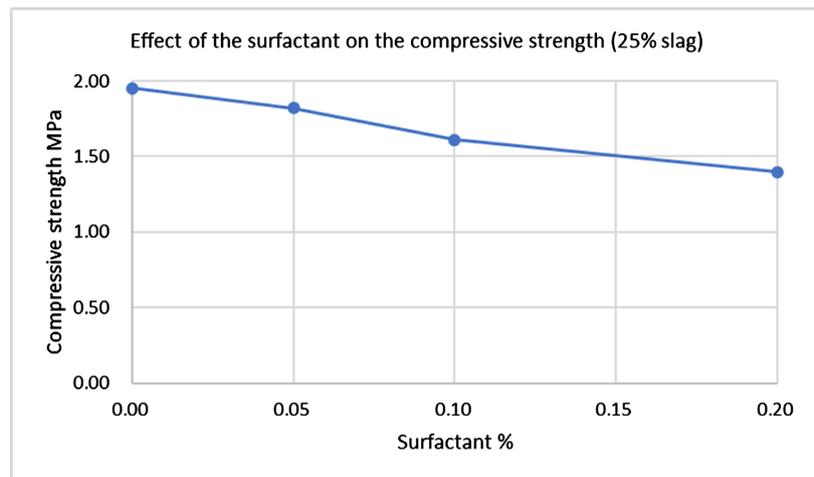
**Figure 7.** Effect of the surfactant on the density and thermal conductivity of metakaolin foamed geopolymer.



**Figure 8.** Effect of the 25% substitution of MK with slag on density and thermal conductivity.



**Figure 9.** Effect of the 50% substitution of MK with slag on density and thermal conductivity.



**Figure 10.** Effect of the surfactant on the compressive strength of the developed materials.

results to a decrease of the compressive strength. The optimum synthesis with the lower density reaches a compressive strength of 1.5 MPa which on the first side seems low but it should be taken into consideration that the specific material will not load bearing capacity and will only be applied as superficial material or facade/insulating material.

#### 4. Conclusions

During the specific research inorganic polymers for applications as insulation materials were developed utilizing both an industrial mineral (metakaolin) and an industrial waste (metallurgical slag from the treatment of laterite ores). The research showed the following results:

- Both raw materials can be foamed utilizing hydrogen peroxide as a foaming agent.
- The alkalinity of the aqueous phase has a light effect on the foaming process of the geopolymers while at the same time the lower alkalinity results to lower density materials.
- The materials utilized slag as raw material reach higher density comparing the ones with metakaolin which is attributed to the higher bulk density of the

raw materials.

- The lower achieved density with hydrogen peroxide and metakaolin as raw material was  $450 \text{ kg/m}^3$  which was improved by adding a surfactant which maintained the bubbles stable without collapsing phenomena.
- The substitution of 25% of metakaolin with slag on the upper synthesis improved further the density of the final material to  $400 \text{ Kg/m}^3$ .

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## Conflicts of Interest

The authors declare no conflicts of interest.

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