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Green Building Materials for Circular Economy - Geopolymer Foams

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Abstract

This study aims to design and investigate foamed geopolymers as a green material dedicated to the circular economy. For synthesis as raw material, the main waste materials of two Polish coal mines, Wieczorek and Staszic, are applied. Additionally, various foaming methods are employed to utilize the by-product of energy production, especially the fly ash generated by the Skawina power plant. In this study, the main issues addressed are related to the selection of the most appropriate foaming agent and the optimization of the process parameters, including temperature, time, and mixture components. Hydrogen peroxide, aluminum powder, and a commercial foaming agent are selected as foaming agents in this research. During the process of sample preparation, stabilizers are applied in the form of polyglycol and cellulose. Through the conducted test, the results show that hydrogen peroxide and aluminum powder emerged as the two most optimal foaming agents.

Keywords: geopolymer, foamed building materials, lightweight materials, circular economy, foaming agents

1. Introduction

The current trends observed in the construction market emphasize the importance of developing multifunctional materials that serve an additional purpose alongside their structural properties. For instance, this material could provide thermal insulation [1-2]. The next anticipated achievement involves the development of intelligent materials, which will process the capability to control specific material parameters [2-3]. Moreover, these materials can exhibit additional functions or features, such as promoting a positive environmental impact or accumulating electricity [3-5].

The significance of additional materials features is particularly pronounced in the context of optimizing the management of raw materials and facilitating the transition from a global economy to a circular economy. It is noted that the construction industry is one of the sectors that have a significant impact on waste generation and pollution [2-6], therefore, the use of waste materials and low-carbon footprint materials is becoming increasingly important. The present technology used for Portland cement production, which has been inherited from the 20th century, suffers from various drawbacks [6]. These include remarkably high energy consumption, excessive utilization of natural resources, and significant environmental pollution, including large emissions of carbon dioxide and toxic nitrogen oxides. As a result, the most promising alternative solutions for Portland cement production currently are alkaline activation and geopolymerization-based technologies [6-8].

Foamed geopolymer materials are a solution that fits into the above trends due to their environmentally friendly nature. Geopolymer synthesis offers a significant advantage over the use of Portland cement, emitting approximately six times less carbon dioxide during the production process. Additionally, geopolymer production can rely on anthropogenic raw materials, such as fly ashes from coal combustion or mine tailings [7, 9]. Furthermore, these materials can fulfill multiple functions

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simultaneously, such as structural support and thermal insulation [10-11]. As a lightweight building material, geopolymers also exhibit additional characteristics, such as fire resistance, sound and vibration resistance, and the potential to stabilize pollutants or facilitate carbon capture and storage [2, 12-14]. However, it is essential to acknowledge that while geopolymers hold promise as construction materials, achieving a suitable porous structure remains a challenging task [15-16].

In the literature, various methods have been described to achieve a porous structure in geopolymers. Yu et al. [17] listed five basic methods for obtaining porous geopolymers:

(1) Self-forming method: This method relies on certain raw materials reacting during the geopolymerization process, leading to the development of an internal foamed structure. This reaction has been observed in cases involving raw materials containing aluminum [17-18].

(2) Direct foaming method: Considered the most popular technique for porous geopolymers. This approach involves directly adding foaming agents, surfactants, or both to the geopolymer paste during the production process, resulting in the formation of the foamed structure [17-18].

(3) Adding filler method: In this method, a foamed structure is achieved through the incorporation of porous filler or materials. Lightweight aggregates, such as microspheres, are often used for this purpose [11, 17].

(4) Particle stacking method: Traditionally, this method involves the formation of a porous structure within the geopolymer or between the aggregate and geopolymer paste. In modern technology, this structure can also be achieved between raw material particles, especially when using additive manufacturing techniques, such as binder jetting for production [17, 19-20].

(5) Other methods: This category encompasses various additional techniques, including modifications of the previously mentioned methods or the application of new approaches [17, 21].

Among the various methods mentioned, the most popular technique is the direct foaming method with the application of different foaming agents. This method enables the production of materials with meso- and macro porosity, making them suited for thermal insulation applications [22]. In the direct foam method, two of the most popular used are hydrogen peroxide and aluminum powder. Both of these agents are favored for their widespread accessibility and effectiveness in the foaming process [22-25]. Both agents are added during the materials manufacturing process, but their mechanisms for material creation vary slightly [18]. Hydrogen peroxide is usually applied in conjunction with the alkali solution, resulting in the generation of bubbles within the materials [18, 22-23]. It is a common practice to use aluminum powder with solid ingredients. This reaction is generally faster compared to the hydrogen peroxide process, and it demands meticulous attention, especially in determining the appropriate amount of added frontiers [18, 22-23].

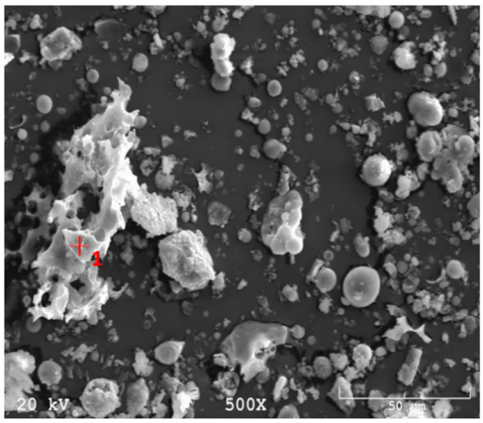
Other frontiers, such as elemental silicon-containing materials or sodium hypochlorite, also hold potential as foaming agents. However, the research in this area is relatively limited, indicating the need for further investigation [18, 24]. The foaming process itself is another challenge that requires some improvements and further investigation, especially regarding the stability and repeatability of the obtained results [18, 26].

This study aims to produce lightweight materials employing various foaming agents. Hydrogen peroxide, aluminum powder, and a commercial foaming agent were used as the foaming agents under investigation. During the sample preparation process, stabilizers in the form of polyglycol and cellulose were added. Subsequently, the physical and mechanical properties of foamed materials were determined. The expectation density and mechanical properties were found to be lower compared to those of solid geopolymers [27-28]. The obtained results support the correlation between these values. Moreover, the obtained results have been discussed with selected literature. The tests carried out led to the determination of hydrogen peroxide and aluminum powder as the two most optimal foaming agents, considering the physical and thermal properties of the obtained materials. Efforts utilizing a commercial foamier for foam concrete did not yield satisfactory results.

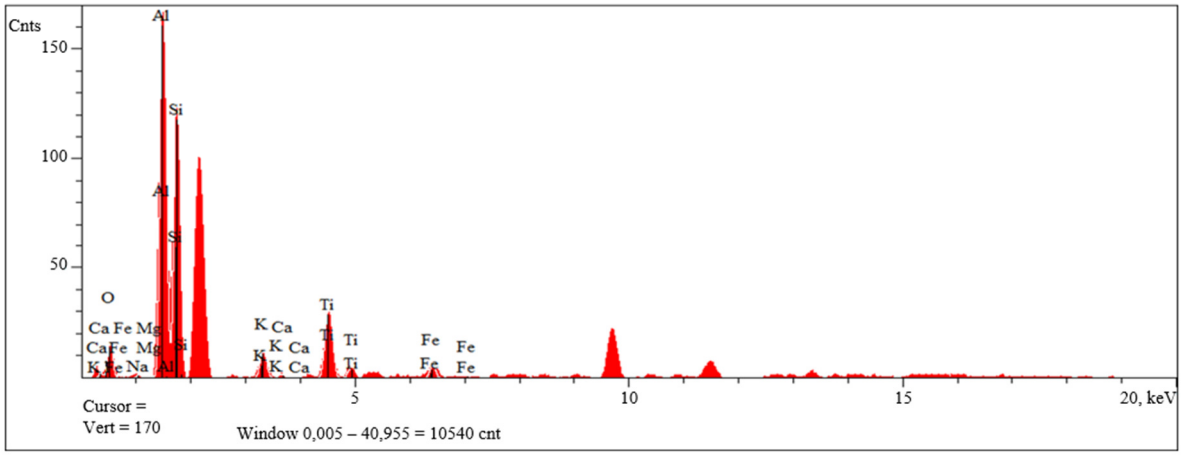
2. Materials and Methods

2.1. Materials

The study employs three distinct raw materials, including two variants of mining tailings from coal production, known as coal shales, and a by-product generated during energy production. The first raw material employed in this study is fly ash obtained from the "Skawina" CHP power plant located in Lesser Poland, Poland. This power plant generates electricity through cogeneration with heat and provides technological steam, drinking water, and industrial water to Skawina companies. With a total installed electric power of 330 MW, the power plant exhibits significant technical and technological capabilities. It includes two hydrogen generators with a combined power of 2.46 MW and heating nodes that facilitate the production of heat energy. Moreover, the power plant has a flue gas desulfurization installation, resulting in a significant reduction of sulfur dioxide emissions by 92%, and it also helps in curbing emissions of other pollutants like chlorine, fluorine, and mercury. Fly ash, which is a by-product of heat production, is the key component in this study. The morphology of the raw material is shown in Fig. 1(a), and the results of the EDS analysis for point 1 are presented in Fig. 1(b) and Table 1.



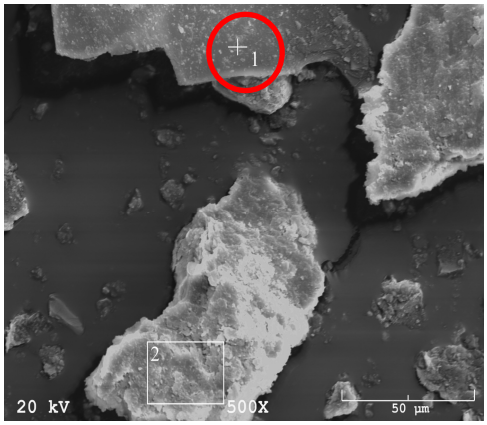
(a) SEM image for fly ash from "Skawina" power plant



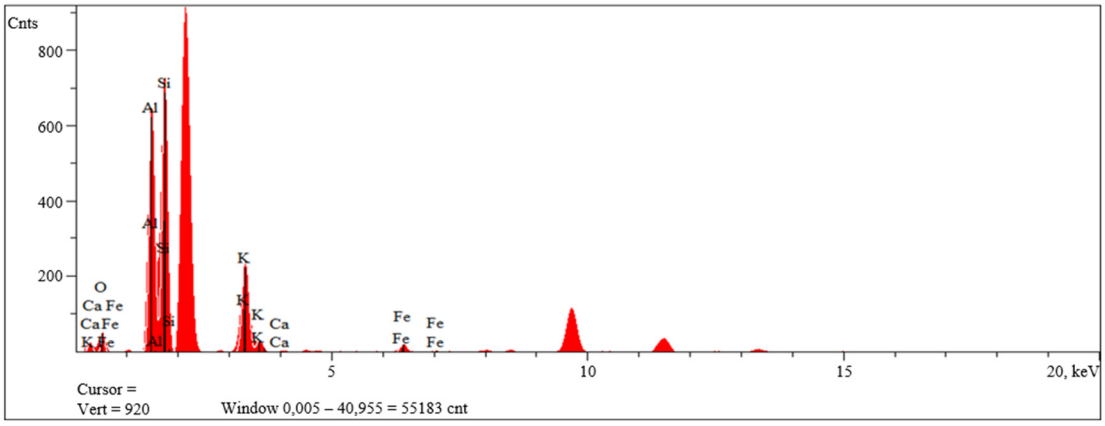
(b) EDS for fly ash from the "Skawina" power plant (elemental analysis)

Fig. 1 Microstructure analysis for fly ash from the "Skawina" power plant

The second raw material utilized in this study is coal shale obtained from the "Murcki-Staszic" hard coal mine located in Katowice. This coal mine is a product resulting from the merger of the Murcki and Staszic mines. The estimated lifespan of the coal resources in this mine is approximately 50 years, with a daily extraction rate of around 23,000 tons of coal. The mined coal is of low quality because of its elevated sulfur content and low chlorine levels. However, after undergoing the geopolymerization process, which involves grinding and milling the materials, the particle size was reduced to a range between 2 and 100 μm. The morphology of the raw material is shown in Fig. 2(a), and the results of the EDS analysis for point 1 are presented in Fig. 2(b) and Table 2.



(a) SEM image for the mine tailing from the "Murcki-Staszic" hard coal mine



(b) EDS for fly ash from the "Skawina" power plant (elemental analysis)

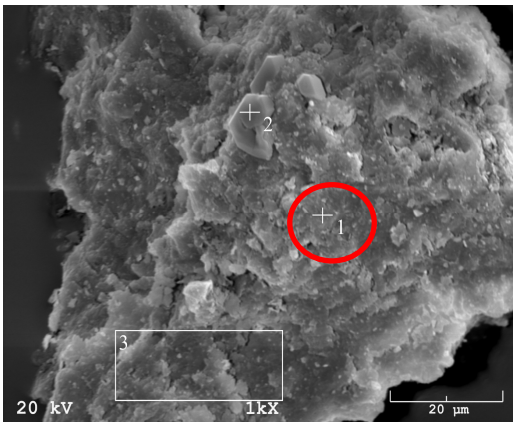
Fig. 2 Microstructure analysis for the mine tailings of the "Murcki-Staszic" hard coal mine

Table 1 Summary of EDS analysis for fly ash from the "Skawina" power plant

No	Element	% by mass
1	O	22,190
2	Na	0,637
3	Mg	0,308
4	Al	31,941
5	Si	30,916
6	K	2,850
7	Ca	0,401
8	Ti	8,576
9	Fe	2,181

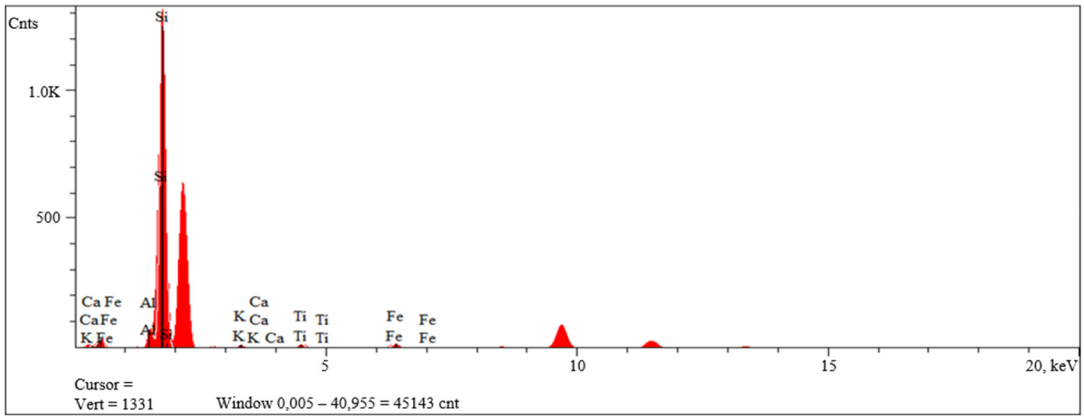
Table 2 Summary of EDS analysis for the tailing from "Murcki-Staszic" hard coal mine

No	Element	% by mass
1	O	17.103
2	Al	26.695
3	Si	39.330
4	K	14.315
5	Ca	0.236
6	Fe	2.320



(a) SEM image for the mine tailing from the "Wieczorek II" hard coal mine

Fig. 3 Microstructure analysis for the mine tailing from the "Wieczorek II" hard coal mine



(b) EDS for the tailing from the "Wieczorek II" hard coal mine (elemental analysis)

Fig. 3 Microstructure analysis for the mine tailing from the "Wieczorek II" hard coal mine

Table 3 Summary of EDS analysis for mine tailing from the "Wieczorek II" hard coal mine

No	Element	% by mass
1	O	14.181
2	Al	4.357
3	Si	75.900
4	K	1.220
5	Ca	0.226
6	Fe	1.476

The third raw material employed in this study is coal shale sourced from the "Wieczorek II" hard coal mine located in Katowice. The mine was founded on 9th October 1882, and it is connected to 11 smaller mines. However, it has currently undergoing liquidation since 2018. After the preparation of the material for the geopolymerization process, which involves grinding and milling, the particle size was reduced to a range between 30 and 200 μm. The morphology of the raw material is illustrated in Fig. 3(a), and the results of the EDS analysis for point 1 are presented in Fig. 3(b) and Table 3.

The analysis of raw materials confirms the essential chemical composition for the production of geopolymers, including a large amount of silica and alumina [11], and the mineralogy composition of the raw material was also provided by XRD. Table 4 provides a summary of the results obtained from the analysis [29]. The analysis results demonstrate the potential of these raw materials for the geopolimerization process, as they contain minerals rich in silica and alumina, which are essential for creating a geopolymer network [27].

Table 4 Summary of XRD analyses for raw materials

No.	Mineralogical composition	Fly ash "Skawina" [%]	Mine tailing "Staszic" [%]	Mine tailing "Wieczoerk" [%]
1	Quartz SiO_2	42.3	34.4	49.9
2	Mulite $Al_6Si_2O_{13}$	54.8	---	---
3	Hematite Fe_2O_3	0.6	---	---
4	Magnetite Fe_3O_4	0.5	---	---
5	Anhydrite $CaSO_4$	1.4	---	---
6	Rutyl TiO_2	0.4	---	---
7	Muskowit-2M1 $KAl_2(Si_3Al)O_{10}(OH,F)_2$	---	13.6	7.3
8	Kaolinit-1Ad $Al_2Si_2O_5(OH)_4$	---	37.4	28.3
9	Illit-2M1 $(K,H_3O)Al_2Si_3AlO_{10}(OH)_2$	---	14.6	14.5

2.2. Samples preparation

Foamed geopolymer composites were produced from the three selected raw materials. As foaming materials, the selected component included hydrogen peroxide, aluminum powder with a maximum size of 60-micron particles and a purity of 99.9%, along with a commercial foam concrete foamier. During the sample preparation process, stabilizers in the form of polyglycolic acid and cellulose were used to stabilize the foam. All samples were prepared using a 10-molar solution of sodium hydroxide mixed with sodium silicate in a 1:2 ratio and tap water. The trials with a commercial foam concrete foamier did not allow us to obtain satisfactory results, and the samples were not porous because the testers resigned. Table 5 displays the list of foamed material samples that have been prepared for analysis.

Table 5 Samples designation

No.	Sample designation	Raw material	Foaming agent	Stabilizer
1	FA1	Fly ash “Skawina”	Aluminum powder 1.5 g	---
2	FA2	Fly ash “Skawina”	Perhydrol 36 %	polyglycolic acid
3	FA3	Fly ash “Skawina”	Perhydrol 36 %	cellulose
4	MTS1	Mine tailing “Staszic”	Aluminum powder 4.5 g	---
5	MTS2	Mine tailing “Staszic”	Perhydrol 36 %	polyglycolic acid
6	MTS3	Mine tailing “Staszic”	Perhydrol 36 %	cellulose
7	MTW1	Mine tailing “Wieczoerk”	Aluminum powder 1.5 g	---
8	MTW2	Mine tailing “Wieczoerk”	Perhydrol 36 %	polyglycolic acid
9	MTW3	Mine tailing “Wieczoerk”	Perhydrol 36 %	cellulose

The samples were prepared according to the following scheme:

(1) The alkali solution was prepared and stabilized, ensuring the proper temperature and equilibrium concentrations were achieved.

(2) The alkali solution was mixed with the raw materials for approximately 5 minutes.

(3) After adding the foaming agents and stabilizer, the mixing process was continued for approximately 30 seconds, and the paste was left to foam.

(4) The Geopolymers were subjected to heat treatment in a laboratory dryer, covered with foil to prevent evaporation, for 24 hours at 75 °C.

(5) After 24 hours of heat treatment, the samples were removed from the molds and stored under laboratory conditions.

After 28 days, the obtained samples were subjected to an evaluation of their parameters. This evaluation included visual inspection and structural tests, microstructural investigation, porosity assessments, and an analysis of their strength properties.

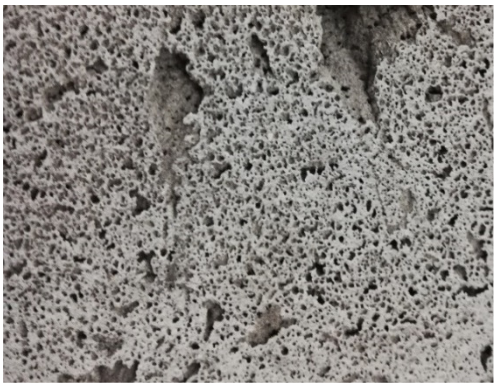
2.3. Methods

The microstructure investigation of the raw materials was performed using a JEOL JSM-820 scanning electron microscope. Before the investigation, the powdered samples were covered with a gold layer to enhance their conductivity, and the gold-coated samples were positioned on the carbon pod to facilitate the analysis. The application of the gold layer was performed using a JEOL JEE-4X sputtering machine. The information on the mineralogical composition was obtained using the X-ray method, specifically the Debye–Scherrer technique for phase analysis, applied in the PANalytical AERIS device. The compressive strength test was conducted on the MATEST 3000 kN test machine, following the standard PN-EN 12390-3:2019-07, which pertains to Testing Hardened Concrete - Part 3: Compressive Strength of Test Specimens.

3. Results and Discussion

The exemplary structure of the pores is presented in Fig. 4. There is a slight difference in the visible structure of both materials. When using aluminum powder, a structure with tiny pores was achieved. However, there are also visible irregular large voids, as shown in Fig. 4(a). On the other hand, when using hydrogen peroxide, the voids are larger but exhibit a more regular pattern, as

shown in Fig. 4(b). It is noted that the presented structure can not only depend on the raw materials and foaming agents but also on various other factors that influence the material preparation process, including atmospheric pressure and temperature [17-18]. Table 6 demonstrates that the samples foamed with hydrogen peroxide resulted in the lowest densities. On the other hand, the samples foamed with aluminum powder exhibited the highest compressive strength among all the tested materials.



(a) Fly ash-based geopolymer using aluminum powder

(b) Fly-ash-based geopolymer fed using hydrogen peroxide

Fig. 4 Examples of the foam structure obtained with the application of different foaming agents

Table 6 Results obtained from the feeding process.

No.	Sample designation	Density [kg/m^3]	Compressive strength [MPa]
1	FA1	612	2.28
2	FA2	267	0.82
3	FA3	298	0.85
4	MTS1	861	4.30
5	MTS2	533	0.76
6	MTS3	501	0.81
7	MTW1	532	0.81
8	MTW2	529	0.62
9	MTW3	535	0.69

The correlation between density and compressive strength is evident, as observed in the results. As the density decreases, the strength properties also show a decrease. This relationship is well-established and documented in the literature [22-25, 30]. Consequently, designing foamed materials becomes a challenging task, as it involves striking a compromise between achieving desired insulation properties and maintaining adequate mechanical strength [17-18].

4. Conclusions

In this study, various foaming methods were employed to investigate the use of lightweight materials. The results indicate that foamed geopolymers can be effectively synthesized using the most appropriate foaming agents, such as hydrogen peroxide, aluminum powder, and a commercial foamier for foam concrete. To ensure appropriate material structure and achieve low density, stabilizers were applied in the form of polyglycol and cellulose during the sample preparation process. Based on the test conducted on the prepared samples, the following conclusions can be drawn:

(1) Based on the physical properties of the materials obtained, the two most optimal foaming agents are hydrogen peroxide and aluminum powder.

(2) The study confirmed that the materials achieved a low density, with values ranging between 267 and 861 kg/m^3.

(3) The finding of the study confirmed that the foamed materials exhibited reasonable mechanical properties, specifically, the compressive strength of the materials ranged between 0.62 and 4.30 MPa.

(4) The results of the study indicate a significant correlation between the density of the materials and their strength.

(5) The study reveals that the attempts made with a commercial foamier for foam concrete did not yield satisfactory results.

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Conflicts of Interest

The authors declare no conflict of interest.

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