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Influence of glass components on the properties and structure of sand-lime materials

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Abstract: The use of recycled glass sand in building materials requires the assessment of its strength and thermodynamic properties. The paper presents the results of research on the influence of glass sand on the structure and properties of silicate brick and the composition of the material modified by introducing a glass composite. The structure of the sand-lime material was assessed on the basis of scanning electron microscopy analyzes using the SEM QUANTA FEG 250 microscope. From the obtained results, it was found that glass sand had a positive effect on the compressive strength, keeping the material at a level not exceeding 20 MPa. Research based on SEM shows that the modification with glass sand leads to the crystallization of the C-S-H phase towards gyrolite and natrolite in the modified brick and tobermorite in the reference brick. Silicates, as natural materials, fit into the criteria of ecological construction and due to this fact, the work has relevance on the subject of sustainable construction.

Keywords: glass sand, C-S-H, tobermorite, amorphous material, sustainable construction

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Introduction

The construction industry, as a major global industry, has a significant impact on the planet and the environment. Currently, the discussion has moved on from construction itself to construction as a sustainable industry, which is closely related to environmental protection, recycling (e.g. mine aggregate) protecting natural deposits against over-exploitation and reducing the carbon footprint associated,

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among other things, with the production of building materials (Recykling, 2022). One of the materials that is subject to the recycling process is glass (Krawczyk, 2018; Kuśnierz et al., 2019). By using this product, sustainable construction is supported. Recycling is an important aspect of ecological construction, because industrial waste covers huge areas of the Earth. In 2020, industrial waste amounted to almost 110,000 tonnes, of which 48% was recycled (GUS, 2021). According to the research carried out (Kuśnierz et al., 2019), in 2013, only in Poland, about 200,000 tons of glass was collected, mainly from the demolition of houses. The construction materials industry, although providing immeasurable benefits, largely contributes to the burden placed on the natural environment.

The use of autoclaved materials is beneficial from an ecological point of view (due to the raw materials used: sand, lime, water), and after demolishing the silicate building, the material can be used in the form of silicate aggregate. These materials are also durable and resistant to changing environmental conditions (e.g. temperature) due to the method of their production (high temperature and pressure) (Zapotoczna--Sytek, 2020). Solutions are being sought that will enable the relief natural deposits of natural aggregates (sands). One such solution is the possibility of using sand with recycled glass in the production of silicate. Analyzing the scientific literature on the results of already conducted research into the impact of glass components in autoclaved materials, it can be concluded that the topic deserves further exploration (Stępień et al., 2019a; Stępień et al., 2019b; Stępień et al., 2020). Glass recycling produces many composites, the most popular being: glass sand, glass powder or glass fibers. Glass sand is made of 95% quartz grains SiO_2 with a diameter of 0.1 mm to approx. 2.0 mm and admixtures of iron oxide, cobalt or chromium and it is characterized by an amorphous structure (similar to C-S-H). This structure means that the atoms cannot move freely, and the melting of the body takes place in a wide range of temperatures (crystallization process). Such behavior of the body is called thermodynamic metastability – the body is stable up to a certain temperature, and as the temperature changes, the body crystallizes, changing the proportions in the amorphous structure in favor of a crystalline body (Stępień et al., 2019a). Glass sand, which is used for the production of silicates, is rich in sodium. The sodium present in the glass components may cause the formation of crystalline phases in the direction of natrolite (due to the high temperature – about 100°C) or gyrolite, as well as xonotlite (Stępień et al., 2019a; Stępień et al., 2020). Building materials are composites composed of at least two different phases, amorphous and crystalline. The division of materials depends on the type of chemical bonds (Czarnecki et al., 1994). In autoclaved materials, the main phase is CSH (Calcium Silicate Hydrates) and crystalline tobermorite. The CS-H phase undergoes crystallization under the influence of temperature (Stępień et al., 2020; Taylor, 1997). In concrete materials, CSH occurs in gel and in fibrous form and it is related to the C/S molar ratio. At the temperature of 347°C there is a critical point of water, at higher temperatures the water-vapor pressure reaches 40 MPa, and above that anhydrous compounds are formed (Zapotoczna-Sytek, 2013). As the molar ratio increases, the compressive strength increases and the shrinkage value, when the concrete dries out, decreases. The share of a given phase and the degree of crystallization determines the performance properties of both concrete and autoclaved materials (Taylor, 1997). Tobermorite, which is the main phase in autoclaved articles, is formed at a C/S molar ratio of about 0.83 and is the crystalline phase. At a higher molar ratio, it is unstable and recrystallizes into xonotlite. Together with the C-S-H phase, it is responsible for the strength of concrete and the materials in which it occurs (Stępień et al., 2019a; Taylor, 1997). Due to the increasing development of technological possibilities, research related to the modification of traditional building materials are carried out. The aim of the article is to analyze the properties and structure of silicate modified with recycled material, which was carried out on the basis of the obtained SEM tests, EDS spectra and strength results.

1. Materials and methods – the scope of laboratory tests

The conducted research concerns the determination of the influence of glass components on selected properties of autoclaved building materials (silicate bricks). Two types of bricks were tested: traditional silicate bricks produced on the basis of quartz sand and 90% of bricks modified with recycled glass sand from bottle glass granulation of 0.08-0.16 mm. The following tests were made:

– Scanning electron microscopy analysis (QUANTA FEG 250) – observed the topography of the tested material – shape, presence and location of characteristic points, on a nanometric to micrometric scale. The microscope has a high imaging resolution and is also used to analyze the composition of the tested material using energy dispersion X-ray microanalysis – EDS spectra (Rafalska-Łasocha et al., 2011).
– Compressive strength tests [MPa] – the tests were performed on samples with dimensions of 50x50x50 mm, after 28 days from their production.

2. Research results

Figure 1 shows the microstructure of autoclaved materials obtained by SEM analysis showing a traditional silicate brick.

Figure 1a shows a grain of sand (SiO_2) marked with the number "1". The grain is surrounded partly by the C-S-H phase and partly by crystalline tobermorite, which is in the form of plaque, denoted by the figure "2" in Figure 1b shows approximately 2500x tobermorite, also known as C-S-H (I). Tobermorite is the basic phase that creates the structure of autoclaved materials. During the production of sand-lime materials, other phases may appear at high temperatures, e.g. xonotlite, natrolite or gyrolite (Stępień et al., 2019a). Figures 2a and 2b show a silicate brick modified with glass sand at 250x and 2500x magnification. The microstructure of the material appears to be compact and very tightly knit. When compared to traditional brick, the structure is lumpier.

a) b)

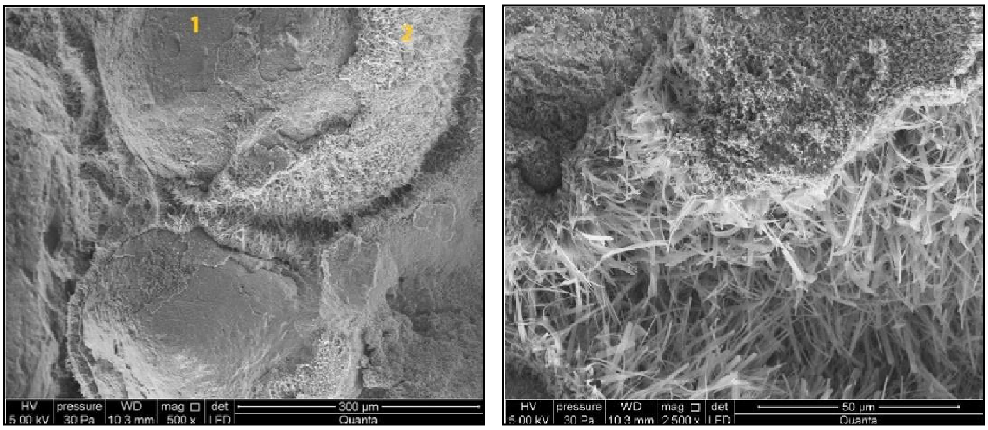


Fig. 1. Scanning electron microscope (SEM) image of the microstructure of traditional silicate bricks: a) high pressure, 500x magnification, b) high pressure, 2500x magnification (*own research*)

a) b)

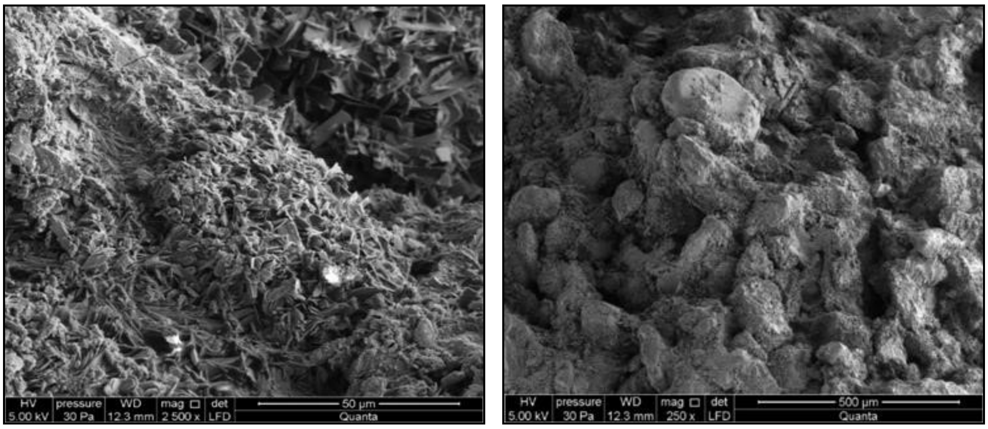
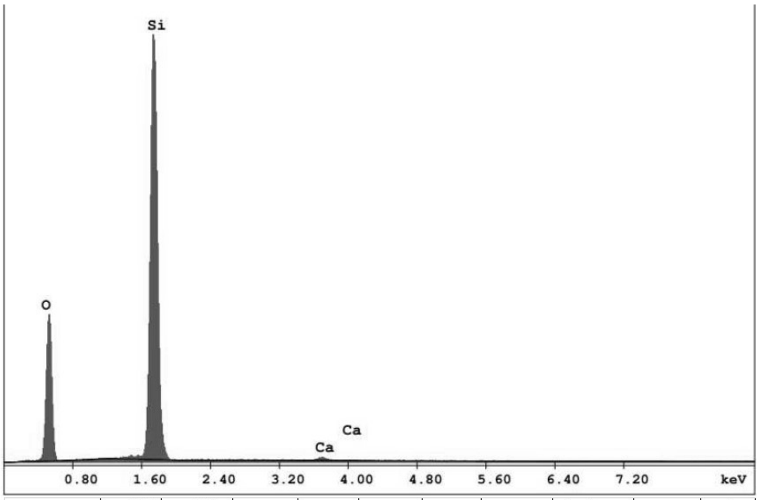


Fig. 2. Scanning electron microscope (SEM) image of the glass sand-modified silicate brick microstructure: a) high pressure, 250x magnification, b) high pressure, 2500x magnification (*own research*)

Figures 3 and 4 show the EDS spectra at a selected point for the reference material (traditional autoclaved brick) and for the material modified with glass sand, respectively. Tests were carried out on porous sputtering samples. The x axis is energy in keV and the y axis is intensity. EDS analysis can be qualitative and quantitative. The results of the quantitative analysis contain the following information: Element – identified lines that are characteristic of X-ray radiation, Wt – weight% of an element in the material, At – atomic% of the element in the sample, K-Ratios – is the ratio of the radiation intensity of the characteristic line element in the material to the intensity from a pure element under the same conditions, Z, A, F – correlation coefficients for atomic number, absorption, fluorescence, Net Intensities – intensity of characteristic lines after background subtraction, Background Intensity – continuous radiation intensity, which is considered the background subtracted in the determination of the net intensity, Intensity Error – the relative error of the determination of the intensity of the peaks, expressed as standard deviation, Peak to Background Ratio (p/b) – the ratio of the number of counts in the peak to the number of counts in the subtracted background. In the first case (Fig. 3), i.e. in traditional brick, silicon and oxygen are the most abundant, and calcium was also present. On the other hand, in the brick with the glass sand (Fig. 4), more additional elements appeared (the main ones are silicon and oxygen), e.g. sodium, magnesium, aluminum. It depends on the composition of the glass sand, and, in principle, on the impurities it has in its structure.

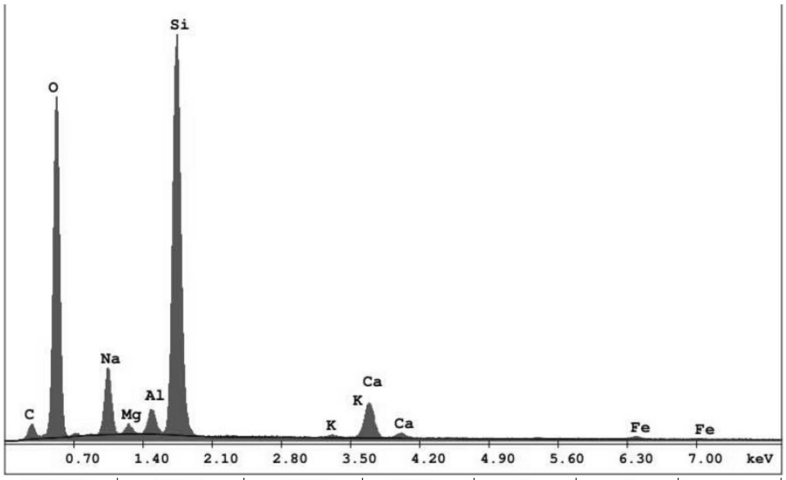


ELEMENT	Wt [%]	At [%]	K-Ratio	Z	A	F	Net Inte.	Bkqd Inte.	Inte.Error	P/B
O K	40,51	54,57	0,1326	1,0254	0,3189	1,0007	230,07	1,94	0,61	118,49
Si K	58,55	44,92	0,5097	0,9827	0,8858	1,0001	944,09	6,90	0,30	136,82
Ca K	0,94	0,50	0,0081	0,9542	0,9063	1,0000	8,82	5,86	4,69	1,51
Total	100,00	100,00								

Fig. 3. EDS spectrometry spectrum in point 1 for traditional silicate brick and analysis of the elemental composition for the studied area (*own research*)

In addition to the SEM analysis, the compressive strength was tested. The silicate brick has been modified with glass sand, used as a replacement for quartz sand in the amount of 30, 50, 70 and 90%. Table 1 presents the results of the obtained tests. The sample E is characterized by the highest compressive strength, reaching a value of over 20 MPa. This sample was modified with glass sand in the amount of 90%. Sample A, which had no modifier in its composition, achieved a strength slightly above 5 MPa.

Glass sand turned out to have a very good effect on the silicate product. Due to the glass compounds in its composition, as well as lime and sodium, it improves strength properties and durability. Used in the production of silicate bricks, it can reduce the amount of lime and quartz sand used. By analyzing the data, it is concluded that with the increasing amount of glass sand – the compressive strength increases.



ELEMENT	Wt [%]	At [%]	K-Ratio	Z	A	F
C K	5,64	9,24	0,0090	1,0405	0,1531	1,0006
O K	46,98	57,83	0,1550	1,0231	0,3224	1,0005
Na K	6,69	5,73	0,0295	0,9576	0,4583	1,0039
Mg K	0,81	0,66	0,0045	0,9816	0,5573	1,0074
Al K	1,97	1,44	0,0131	0,9528	0,6913	1,0134
Si K	31,44	22,05	0,2424	0,9806	0,7855	1,0008
K K	0,34	0,17	0,0030	0,9295	0,9169	1,0088
Ca K	5,17	2,54	0,0465	0,9516	0,9449	1,0005
Fe K	0,95	0,34	0,0082	0,8642	0,9987	1,0000
Total	100	100				

Fig. 4. EDS spectrometry spectrum in point 1 for silicate brick modified with glass sand and analysis of the elemental composition for the studied area (*own research*)

Table 1. Comparison of the results of compressive strength of the modified silicate brick depending on the content of glass sand in the raw material mass (*own research*)

Samples	QUARTZ SAND – OS [%]	GLASS SAND – GS [%]	Compressive strength [MPa]	VARIANT
A	90	0	5.25	OS 90% + GS 0%
B	60	30	15.03	OS 60% + GS 30%
C	40	50	18.31	OS 40% + GS 50%
D	20	70	19.50	OS 20% + GS 70%
E	0	90	20.23	OS 0% + GS 90%

The autoclaving time in laboratory conditions, due to the autoclave capabilities, was – 4.5 hours compared to the traditional production of silicate bricks – 8 hours. For this reason, some properties of silicate bricks may differ slightly from those produced in bulk. In order for the silicate material to achieve the appropriate parameters, including strength, the final process, i.e. the aforementioned autoclaving, is very important. At this stage, the material is subjected to steam under very high pressure (up to 1.6 MPa) in hermetic tanks (autoclaves) at a temperature of 200°C. Due to such hardening and as a result of chemical reactions, the material achieves a high level of compressive strength and hardness (Produkcja silikatów, 2022).

Conclusions

Sustainable construction aims to reduce energy consumption and bases its idea on natural resources. Silicates are 100% natural materials with many advantages. They have high compressive strength compared to ceramic red bricks, very good insulating and acoustic properties, and high fire resistance (Stępień et al., 2019a). The tests carried out on traditional silicate brick and silicate brick modified with glass sand showed that the substitute for quartz sand, in this case – recycled glass sand, has a positive effect on the modified material. The difference between sample A, which only contained quartz sand, and sample E, which was 90% modified with glass sand, is 74%. According to the research, this means that the more glass sand, used as a replacement for quartz sand in the composition, the better the compressive strength. The sodium present in the glass sand may cause the material to swell, but this is dependent on the component composition and moisture and pressure variations within the autoclave. The SEM analysis showed that the C-SH phase mainly appears, which is an amorphous phase that transforms to a varying degree into tobermorite. In addition to the above-mentioned hydrated calcium silicates, there may also appear: gyrolite, natrolite, yennite or xonotlite. These phases arise depending on the molar C/S ratio and temperature.

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