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Influence of Heavy Aggregates on the Qualities of Silicate Materials

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Abstract. The article is a part of the research devoted to modifications of autoclaved products, and it also refers to the theories widely researched by Harman Daly concerning balanced development and the problem of termination of aggregates and other components used in production of concrete and building materials as well as acoustics isolation and bulk density. The article discusses of the optimization of the composition of the mass and the effect of heavy aggregates on the microstructure of this elements. Heavy aggregates are important for designing of the bricks and for the right acoustics isolation of the material, and further for the construction wall. The studies conducted show that the utility parameters of the material reach the expected value of the additive in the amount of up to 25% of aggregate in relation to the mass of the product (basalt, barite aggregates) or up to 10% of diabase aggregate into the mass.

1. Introduction

The construction industry, along with automotive industry and IT are the branches which specifically influence human's life and environment, as well as they trigger changes in the look and functioning of our planet. It is estimated that:

- over 40% of international energy is used in construction sector,
- about 35% of greenhouse gases come from construction,
- about 50% recycled materials are used in building industry [1-7].

The results show that far reaching technical and technological advancement and rapid globalization require new development conditions and changes, also in the construction industry. The changes and new conditions involve developing new production methods and modifications in the process of production of building materials. The collected data show that every year gigantic amounts of matter is used in construction, as yearly worldwide production equals circa 20 billion tons of aggregate, 1,5 billion tons of cement, 0.8 tons of water. It does not come as a surprise that alternative methods of preventing excessive devastation of natural environment are still being searched for [4, 8-11,22]. Taking care of the environment and fighting for its wellbeing, and also acting according to balance development about which Herman Daly widely spoken in 80s, using recycled materials, such as aggregate from rebuilding or plastic elements (polystyrene HIPS, polymers) [3,8-11]. What becomes a problem is the rapid use of non-renewable resources, especially because of the common phenomenon known as overproduction. However, according to one of the commonly used norms is that the non-renewable sources cannot be used faster that their recycled substitute appear. Concrete, along with its modifications, is now the most popular building materials. However, from the ecological point of



view, sand-lime bricks, popular especially in Poland, Germany, Slovakia and France, are a beneficial material. Sand-lime bricks with the compressive strength of 15-25 MPa may be used in the construction of a single, as well as multifamily up to 5 storeys. Sand-lime brick production process is based on lime slaking, which is connected with exothermic reaction. As a result of this reaction, the temperature rises to 80-100°C. The heat ejected during the hydration process, absorbs the excess of water in the sand-lime mass. The main ingredient of a sand-lime brick is thus silica (Si), commonly occurring as a natural substrate. Content of Si in the external layers of the Earth equals circa 27% weight and is a second, just after oxygen, the most common element on our globe. Silica with chemical formula: SiO_2 in various polymorphic varieties (quartz, tridymite, cristobalite), silicates and aluminosilicate are the majority of rocks that create the Earth's crust. The name of the first, external layer of the globe—SiAl derives from silica [6, 12]. Silica is a safe material, which is even essential to the proper functioning of a human's organism. It may be thus implied that sand-lime bricks and their proper modifications are perfect material for construction of buildings, as well as they positively influence the quality of life and functioning in such places [1, 2]. Concrete and steel are nowadays good material for construction of industrial buildings, high-rises and engineering objects, however, when choosing life outside the working environment, one should mainly pay attention to the issue of health. The research performed by the scientists from the University of Manitoba in Canada, in cooperation with doctors and local hospitals, for instance, from the region of Manitoba, shows that there is crucial connection between diseases of the respiratory system with living in so-called "concrete capsules" [13], in which we are born, we live, work and stay in for the most of our lives. Possibility of using particular additions to the mass, were presented according to the multicriteria technical and economic analysis with the module of "Multidimensional exploration techniques" (STATISTICA 10.0). The analysis of the main compounds was chosen as the method for elaboration of the results. The conditioning factor for using a particular modifier were economy and ecology. Only modifiers with high density (higher than 1.7kg/dm^3) were taken into account. The subject material has the density of 1.7kg/dm^3 , absorbability of up to 16% related to its mass, its microstructure is porous, rough and mainly depends on the method and strength of compression and the method of its production. Heavy additives also positively influence the acoustic parameters of the building materials. Acoustic indicator of a material is also meaningful as it comes to the danger connected with noise causing stress (described as civilization disease of 21st century). Heavy additives with density exceeding 1.75kg/dm^3 were used). Sand-lime bricks show high resistance to biological corrosion, that is why they can be applied in interiors with strong alkaline levels. They provide friendly climate, preventing, at the same time, from development of fungi and bacterial flora. Because of its capacity of accumulation of heat and humidity, sand-lime bricks have proper qualities of regulating their conditions. The subject material is non-flammable (A1) and meet the requirements of the strictest regulations of fire resistance (fire resistance class 240 REI) [7]. Because of their durability, sand-lime products are admitted to be used in various environments. It is advised to always check the environmental conditions in order to make an adequate choice of sand-lime products suitable for particular environment. The use of particular sand-lime products depends on the class of exposition. Nowadays, there are various additives and admixtures, with which the building materials are modified. Among the most popular modifiers of the sand-lime mass are: microsilica, energetic waste (volatile ashes), additives thickening the sand-lime mass, surfactant additives which plastify the mass, additives that accelerate and delay lime slaking in the mass, combinations of additives, active mineral additives, post-metallurgical waste, and waste materials. The research performed with the use of sand-lime mass prove detrimental effects of using microsilica (in contrast with the research on concrete mass). When using X-ray generators the radiation energy is about 100-120 keV. Therefore, in areas where there are mentioned devices protective screens should be used. The effectiveness of various materials in weakening of gamma radiation depends on the number of Z elements forming the casing material, which is the number of electrons in the atoms of the elements and the radiation energy. According to [22] with photon energies less than 0.5 MeV, more effective in radiation weakening are these concretes which contain possibly large amount of elements of high Z number (the more effective it is,

the lower the radiation energy). With concretes it causes reducing their dimensions and the weight of casing material relative to the casing of lighter concretes. However, at 0.5-5.0 MeV energies the differences in weakening efficiency are proportional to the differences in bulk density of materials. Assumed is thesis that, in the case of silicate products, also at the X-ray energy less than 0,5 MeV, the weakening efficiency depends on the bulk density of the product [2, 3, 18].

2. Methodology, the laboratory tests and materials

Laboratory experiments carried in Sand-lime production plant in Ludynia belonging to the SILIKATY Group. Samples of traditional and modified elements were taken to the forms with the size of 4x4x16 cm. Basalt, barite and diabase aggregates were used. During the experiment, respectively 10, 20, 30, 40, 50% of modifier was added to the sand-lime mass. The best results were observed when the participation of the modifier did not exceed 25-30% in relation to the product's mass. The experiment was carried out according to polish standards.

2.1. The sand-lime bricks and hydrothermal conditions

Sand-lime bricks commonly called "Silicates" are construction materials providing a solid structure and a comfortable interior microclimate (table 1). The initial phase of their production involves mixing the: sand, lime and water. The mix is then placed in steel silos like a reactors, where it is left for minimum four hours or more (it depends on the slaking lime process). Here the process of slaking takes place, accompanied by an increase in temperature to around 60-80°C. It is recognized that 56 g of lime falls 18 g of water. At this stage silica loses its crystalline structure, which in turn facilitates the subsequent formation of products. Next, the silicate mix is directed to the press, in which it is compressed at a pressure of 15-20 MPa, and formed into bricks and blocks of suitable size and shape. Two types of presses are applied: pneumatic and hydraulic. Hydraulic press is better for silicate production. In the final phase the compressed blocks are placed in autoclaves and subjected to a hardening process in the temperature around 200°C at the pressure of 1.6 MPa (232 PSI). During the next 6-12 hours of autoclaving the lime reacts chemically with sand and the mixture undergoes the process of recrystallization.

Table 1. General characteristics of sand-lime products

Compressive strength [MPa]	Bulk density [kg/dm ³]	Absorbability [%]	Microstructure [phase structure]	Color
15-20	1,73	16	C-S-H phase, tobermorite	Milky white

Table 2. Bulk density in particular attempts

L.p.	Bulk density for 6 samples [kg/dm ³]
1	1,71
2	1,69
3	1,81
4	1,73
5	1,72
6	1,75

Volume density of sand-lime products was determined according to the PN-EN 772-13:2001 norm "Methods of examining masonry units – Part 13: Determining net and gross density of masonry units in dry state (except for natural stone)". Volume density of traditional sand-lime products equals about 1,73 kg/dm³ [15-19]. Table 2 shows bulk density in particular attempts.

The addition of heavy aggregates usually results in an increase in density and thus sound insulation of the bulkhead. The autoclave process did not change the structural properties of the introduced modifiers (barite, basalt and diabase). Because of the similar mineral composition of the diabase and basalt aggregate, as well as the ambiguous chemical composition of this type of aggregates, radiological resistance measurements were made only for barite aggregates i.e. barium sulphate (BaSO_4). The size of the atomic number should be taken into account when selecting heavy aggregates. Barium sulphate and basalt and diabase aggregates are used in the silica mass, which are characterized by the presence of the following calcium and magnesium compounds (usually magnesium oxide is maintained at 5-12% and calcium is about 10%) [6].

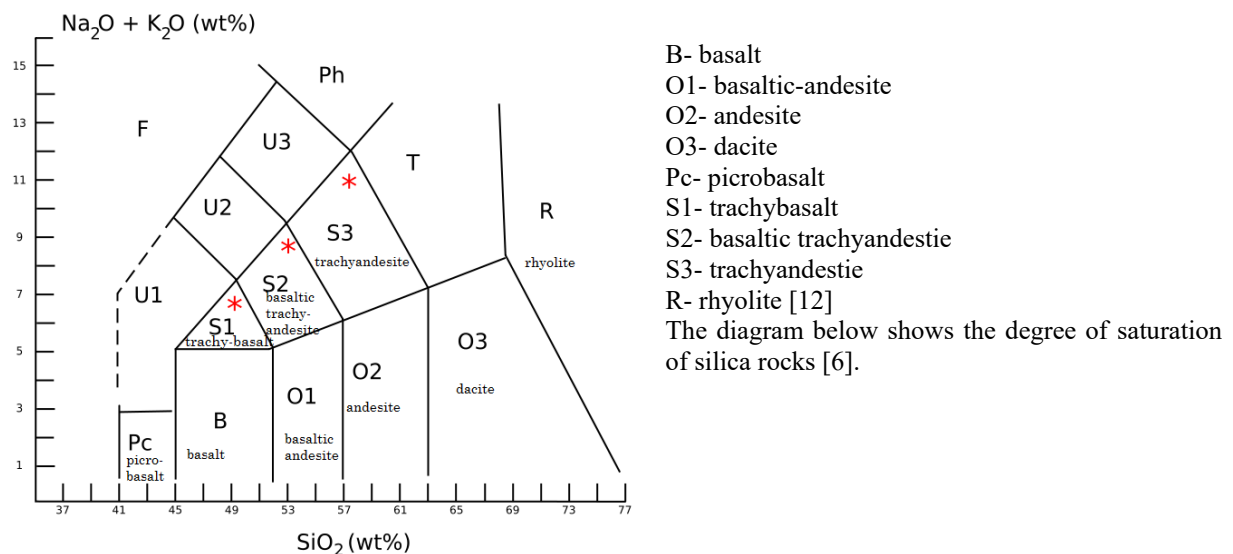


Figure 1. TAS classification with field symbols showing degrees of silica saturation (Total Alkali Silica).

2.1.1. Microstructure of the traditional sand-lime brick

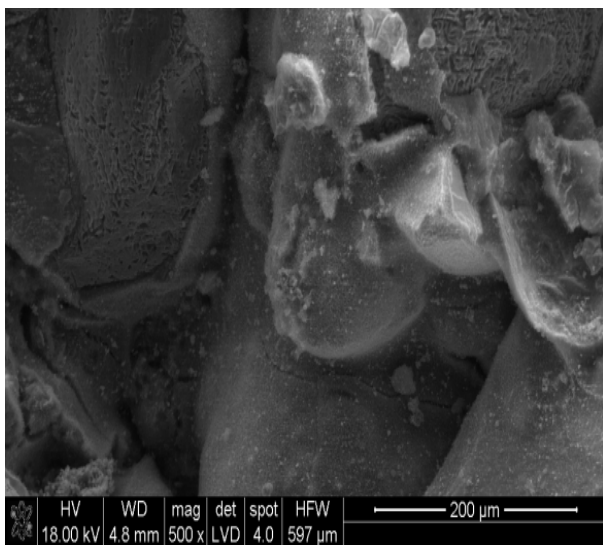


Figure 2. Microstructure of the sand-lime product.

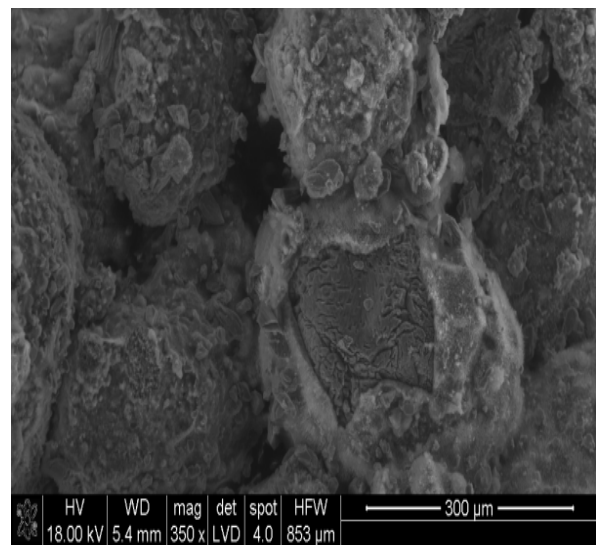


Figure 3. Microstructure of the sand-lime product

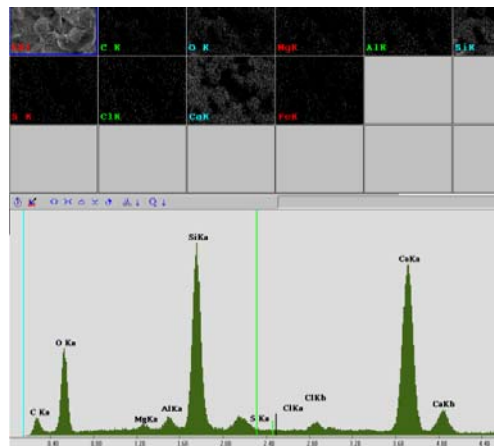


Figure 4. EDS spectrum of a selected microbe of a traditional silicate product.

It is a result of notable speed increase in reaction of silica and calcium hydroxide or calcium silica with creation of hydrous phases. So far, thorough analysis has been performed on materials with higher CaO rate than sand-lime products, in which the amount of CaO compared to SiO_2 is minimal and essential for calcium hydration reaction (2-10% depending on the quality of sand) [3, 8, 9, 21]. Characteristic for the C-S-H phase is its high amorphity. Level of structure arrangement, depending on fusion conditions and rise of temperature, changes from amorphous into crystalline state (tobermorite, xonolite). In the figure 2 SiO_2 sand grains surrounded by particular phases are visible. Sand-lime products are porous materials. Presence of phases in their microstructure does not only enable their more thorough analysis, including frequency of their occurrence, but also their influence on the porosity of the subject product. Sand grains are surrounded by amorphous C-S-H phase. Under the hydrothermal treatment and favourable technological and humidity conditions (saturation of vapor in autoclave), hydrated calcium silica with disordered structure transform into crystalline ordered form. Depending on the modifier, it is possible not only to interfere into physio-economic features, but also into microstructural (phase) ones. Figure 2 and figure 3 present the built of traditional sand-lime product. The picture above presents EDS spectrum (figure 4), thus elemental composition of the subject material. According to the analysis, in the examined micro area of the sample, the largest portions are: silicon (Si), calcium (Ca) and oxygen (O). Trace quantities of magnesium (Mg) and aluminium (Al) were observed. The presence of magnesium and aluminium is identified with sand deposits for industrial production of autoclaved bricks.

2.2. Essential and microstructural characteristics of sand-lime brick modified with heavy additives

The compressive strength test was based on Polish standards (figure 5,6) [15-19].



Figure 5. Acoustic brick modified by basalt aggregate (grain size up to 2 mm).



Figure 6. Barite aggregate used for modification of sand-lime mass (particle size up to 2 mm).



Figure 7. Diabase aggregate used for modification of sand-lime bricks.

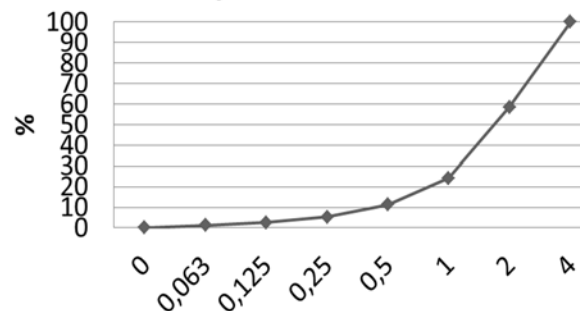


Figure 8. Diabase aggregate from the Niedźwiedzia Góra with a fine-afanite structure.

The most optimal granulation of aggregates in the sand and lime technology is assumed to be the share of grain fractions: $2.5 \div 0.5$ mm - up to 30% and $0.5 \div 0.05$ mm - at least 65% [9]. The used diabase aggregate (figure 8) is derived from the diabase rock exploited in the Małopolska mine - Niedźwiedzia Góra. Diabase in terms of mineral composition and origin belong to the group of gabras and basalt [23].

Diabase rock, derivation of which is aggregate, is exploited in Niedźwiedzia Góra Mine – that is where the subject material comes from. In terms of mineral composition and genesis, diabase belongs to the group of gabbro and basalt. Over the years, the qualities and structure of the rock were analyzed precisely, on the basis of microscope descriptions and the results of the chemical analysis performed by R. Zuber and S. Zaręczny. In 1956 rock coming from Niedźwiedzia Góra was classified as quartz diabase by geologist Z. Rozen [23]. The content of SiO_2 oscillates between 49,69% and 53,20%. The amount of Al_2O_3 equals between 15,1 – 17,5%, FeO rate is between 4 – 6,6%, Fe_2O_3 is more varied and equals 4,2 – 9,1%, similar to the amount of CaO 5,7 – 8%. Another research [23] prove that diabase rocks, according to their chemical and geochemical terms, are not alkaline, but their form is in the middle, closer to acid rocks. This feature may be the cause of change in the production of sand-lime bricks because of the fact that sand-lime products are alkaline. Figure 7 diabase from Niedźwiedzia Góra has fine-grain aphanitic structure. It disintegrates into sharp-edged pieces with grains thicker than basalt. On the fresh break its hue is grey steel.

With bulk density is related to the protection from the harmful effects of radiation [1, 2]. Due to the high cost of research, only blocks modified with barite aggregates were analyzed. The result of measurements of the ionizing radiation of the element is the rate of weakening the X radiation beam. It was denoted by K and this is a quotient of power of the photon dose equivalent $H_{x,0}$.

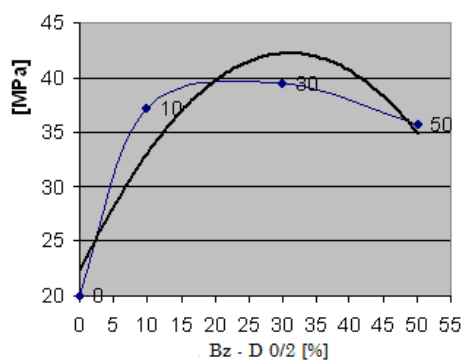


Figure 9. Relationship between basalt aggregate and compressive strength.

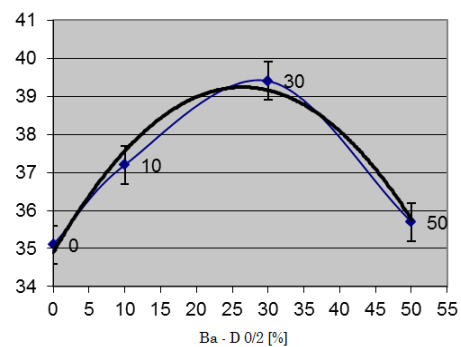


Figure 10. Relationship between barite aggregate and compressive strength.

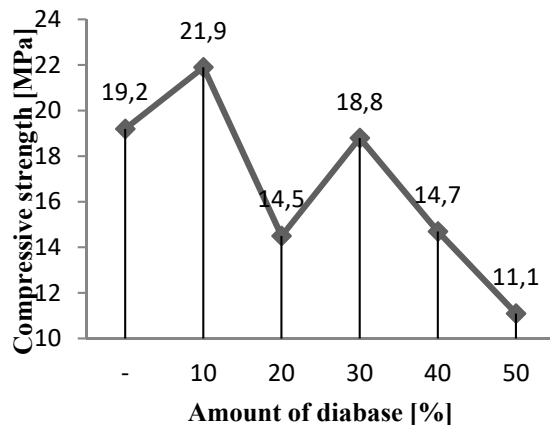


Figure 11. Relationship between diabase aggregate and compressive strength.

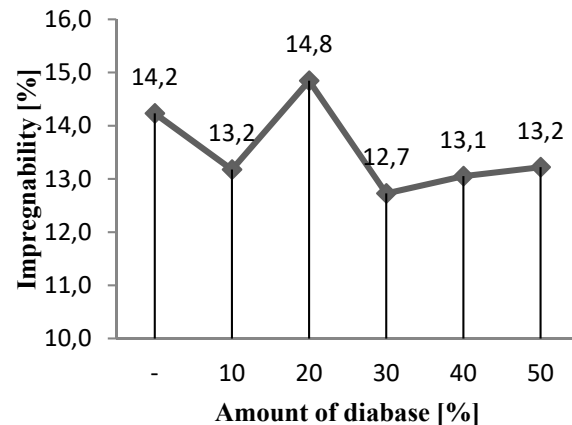


Figure 12. Relationship between diabase aggregate and impregnability.

Table 3. The results of weakening the X radiation beam test performed on the silicate products.

Silicate brick	X radiation energy[keV]	Value of $H_{x,0}$ [Sv/h]	Value of H_x [Sv/h]	Rate of weakening, K
Traditional brick	100	70,11	2754	0,0255
	118	598,51	16090	0,0371
20% Ba	100	3,49	2753	0,0012
	118	35,00	16097	0,0021

The calculations of the thickness of radiation shield have been done (table 3). The required multiplication factor of the weakening off this kind of radiation beam. Data was used for the tests:

- $L = 1.5$ m - for the distance from the X-ray source to ionization chamber
- 100 keV - X-ray energy
- $K = 1740$ - the required rate of weakening, which corresponds to the thickness of ionizing radiation shield equal to 1,5 mm for a plate of lead [3].

Table 4. The results of the calculations of required thickness of radiation shield.

Silicate brick	X radiation energy [keV]	Weakening multiplication	Weakening multipl. ratio $[\sigma]$	Required thickness of rad. shield, d [mm]
Trad.brick	100	39	44,60	154
20% Ba	100	807	2,16	7,5

3. Results and discussions

The studies showed an increase in density to 2.35-2.65 kg /dm³ in the case of basalt and barite aggregate and increase the strength to 42 MPa in the case of modification of silicate mass with basalt aggregate and 37 MPa in the case of modification with barite aggregate. Taking into account the compressive strength of the silicate sandwiched silica, the sample shows the highest value with a 10% share of diabase grains. The value of the strength of the modified item reaches a value of 21.9 MPa. Increasing the additive content (20-50%) causes a gradual decrease in strength, which in turn lowers the test property by almost 50%. Causes should be found in the shape of grains and their diameter (a large proportion of aggregates with a fraction greater than 1.5-2mm (figure 8)). The proper size of the aggregate used for the production of silicates is considered to be characterized Continuous grading curve, without explicit dominance of any of the grain fractions.

3.1. Microstructure of bricks modified by different kind of aggregate

SEM observation (figure13-17), along with EDS analysis of breaks in the modified sand-lime products show similar microstructure to the one of sample with modification of 10% diabase grit. The phases visible in the photograph which were created as a result of reactions between the elements of a sand-lime sample may be compared to moss covering a stone, with disorderly placed grass blades in various sizes and angles. Particular phases in those specific examples were given numbers attributing the EDS. Large quantities of Si and Ca were observed, little amounts of Al and Mg along with trace amounts of Fe, which are a part of quartz sand. It is natural, because sand is the basic element of brick. Increase of the addition of diabase grit to 30% compared to the rest of the elements of the mass influences the internal structure of the final product by its unification. Figure13 presents creation of aggregate of C-S-H phase close to the surface with thickness of 14-20 μm .

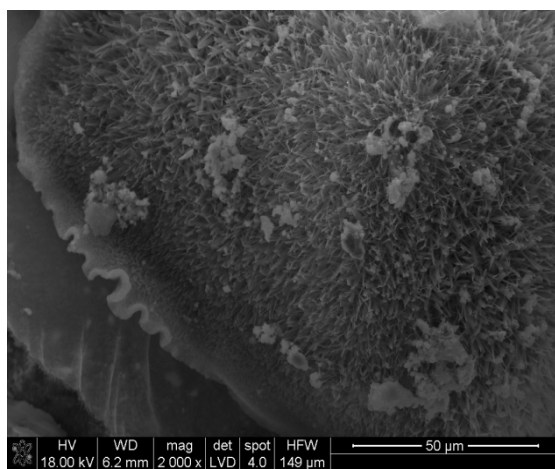


Figure 13. Modification of brick by basalt aggregate.

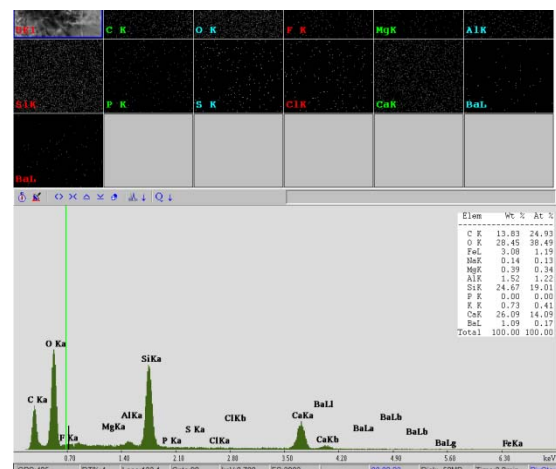


Figure 14. EDS spectrum of the brick with basalt aggregate.

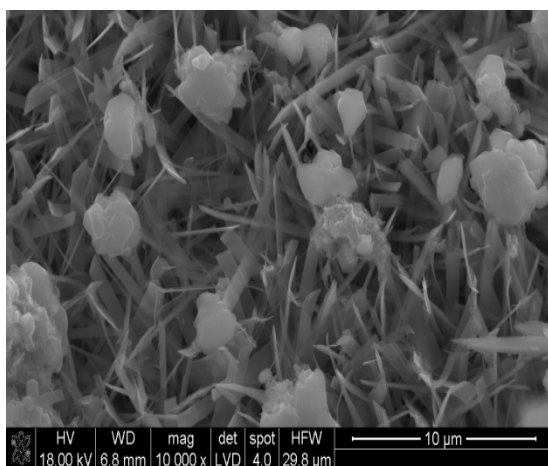


Figure 15. Modification of brick by barite aggregate.

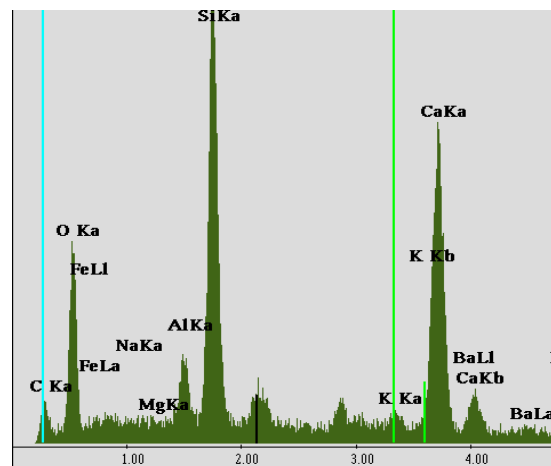


Figure 16. EDS spectrum of the brick with barite aggregate.

4. Conclusions

The share of basalt and barite aggregate should not exceed 25% of the aggregate in relation to the weight of the product. Increasing the aggregate contributes to a decrease in strength. The best values of silicate brick reach when the silicate mass is modified with aggregate in the amount of 23%. The strength value is then about 40 MPa. Adequate modification of the silicate mass with barite aggregate

is adequate. Bulk dens. of products reached values of 1.73kg/dm^3 to an average of 2.5kg/dm^3 . Modification of silicate mass with diabase aggregate suggests the use of aggregates up to 10%, otherwise there is a decrease in strength. Diabase aggregate are not chemically and geochemical in nature, only indirectly closer to acidic rocks. This feature and the presence of magnesium can cause changes during the modification of silicate bricks (silicates are alkalis) and further decrease in strength. Due to the above, further investigations of modified articles, including mercury intrusion porosimetry, precise chemical analysis and determination of hydrated calcium silicate and C/S ratio should be carried out.

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