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Article

Effect of glass waste on ceramics and concrete production

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Abstract. The article is devoted to the study of using glass waste for ceramics and concretes production. The results of the spectral analysis of glass composition are presented, and phase changes and their impact on the microstructure and strength properties of ceramics and concrete are studied. Scanning electron microscopy and energy dispersive X-ray spectroscopy of the final material are discussed. As a result, it was revealed that after 28 days, concrete with added glass powder delayed the strength rise, but by day 112, the strength had considerably grown to 76.36 MPa. This results from pozzolanic reactions, where calcium hydroxide and glass combine to generate more hydration products that boost strength. Glass-based ceramic shows 13.20 MPa compressive strength, which satisfies construction material criteria, and was achieved by adding glass waste at a level of 10% of the clay mass. In addition to lowering the demand for natural mineral resources, the use of glass in ceramic blends promotes sustainable development and lessens the environmental load. Both the ceramic and concrete samples had a high content of SiO_2 and Al_2O_3 oxides. These are the main components that provide the material with high mechanical strength and chemical resistance. In the Spectral analysis of glass, in all the graphs, we see a high tendency for silicon dioxide (silica). This is explained by the fact that SiO_2 belongs to the group of glass-forming oxides, i.e., it is prone to the formation of supercooled melt-glass.

Keywords: production waste, concretes, ceramics, recycling, physical and mechanical properties.

1. Introduction

One of the top concerns for construction materials research is the development of efficient thermal and structural insulation materials, which is a difficult procedure. It is worth noting that the rational obtain of unprocessed materials and the full involvement of man-made waste in production are paramount in this area. Experience both domestically and internationally demonstrates that one of the most promising uses for industrial waste processing is the creation of construction materials, which makes it possible to meet the need of developed countries for raw materials up to 40% [1].

The use of industrial wastes in concrete products has certain benefits [2]. The combined use of fuel-containing waste and chemical compounds that prevent water evaporation can compensate for the shrinkage deformation of monolithic concrete and increase its crack resistance [3].

[4] used waste containing saponite in the production of fine-grained concrete with high levels of strength, frost resistance, and workability. In experimental models of vibrating reinforced concrete columns, the type of technology used in [4] had a major impact on the physical, mechanical, and structural features of concrete. According to [5], [6], the slag and fly ash are useful supplementary materials for enhancing the qualities of cement-concrete. These works show that an increase in cement consumption and the amount of filler in self-compacting concrete causes the concrete mixture's viscosity to drop. They found that if the amount of filler exceeds 15% by weight of cement, the viscosity is practically independent of cement consumption. Additional cementitious materials (ACMs) such as fly ash, ground blast furnace slag, fumed silica, and metakaolin are being utilized more frequently as binders for high-strength and high-performance concretes [7]. [8] created research

and technical bases to produce and use the concrete based on ash-slag mixtures of thermal power plants. They divided the ash and slag combination into a fine-grained fraction with slag passing through a 5 mm sieve. Next, the fine-grained fraction, slag, cement, and mixing water are dosed in the mixer individually based on the concrete's composition. It was discovered that this technological advancement enables concrete to be strengthened by 20–30% while consuming 15.20% less cement in the concrete production with the same strength. [9] attempted to fully substitute the coarse aggregate with a broken concrete and a portion of a fine aggregate with crushed glass, obtaining compositions replacing these aggregates by 20%, 25%, and 30%. The primary findings of this study demonstrated that the maximum compressive strength, which considerably surpasses the strength of concrete grade B35 and approaches the strength of concrete grade B40 and B45, is achieved when glass is used as a fine aggregate replacing sand by 30%. This suggests that glass is suitable for use in a variety of structures. In addition, they showed that strength growth does not occur linearly as the amount of glass in concrete samples increases, but instead, the rate of strength growth slows down with time. [10] developed a technology for producing high-strength concrete based on industrial and household glass waste. Glass as a filler for concrete compositions usually reduces the strength of the material due to the chemical interaction of glass and cement. Therefore, the researchers presented a new approach to create composite glass concrete, crushing broken glass. Glass particles of about 50-60 micrometers in size, obtained in a ball mill, were used as a filler. Glass was also used as a binder, but crushed finer up to one micrometer. As a result of a series of experiments, they managed to increase the strength of the glass concrete composition by 2.5 times.

Ceramic manufacturing is one of the industries that utilize the materials the most. The loss of natural resources used as raw materials for the ceramic industry is a serious problem for this sector of the economy and affects the price and quality of ceramic products. In this regard, [11] took into consideration the possibilities of employing mine waste in the form of ultramafic rocks (wehrlites and dunite) as an additive in the manufacturing of building ceramics. [12] investigated the prospect of creating facing ceramics from the waste of the metallurgical and glass sectors. According to the findings of the study, a batch of 40% clay and 60% cullet must be fired at 1050°C to produce laboratory samples of facing ceramics having a density of 2064 kg/m^3, a compressive strength of 42.24 MPa, and closed-pore formation. The impact of partially replacing the basic mix of ceramic floor tiles with waste urban glass is covered in [13]. Because of the high percentage of fluxing oxides in the waste, which promotes the formation of the glassy phase, increasing the waste addition to 20% results in an increase in linear shrinkage during firing, which subsequently gradually decreases to less than 3%. Bulk density rises as temperature and waste addition percentage increase. Water absorption diminishes as temperature and waste addition rise, with temperature having a greater impact than waste addition. [14] considered the prospect of producing facing ceramics and clinker based on brick clays utilizing large-tonnage waste glass products. The study selected a composite additive based on glass waste as a salt mineralizer required for sintering the clay-glass batch. The crystalline and amorphous phase is derived from sodium aluminosilicates. The X-ray phase analysis of the sintered ceramic composition revealed the presence of approximately 40% glass phases, which are uniformly distributed over the crystalline substance based on the composite's microphotography. [15] proposed to utilize waste in the form of finely ground slag, which is added to the raw material mixture to create wall ceramics that are stronger and have better thermal insulation properties. For wall-building materials, including ceramics, thermal conductivity remains a pressing issue. Therefore, as one of the particular solutions to this problem, the study suggested the use of porous ceramics in enclosing structures. Thus, while burnable additives added to the clay mass of ceramic wall materials effectively increase their porosity, they also control the firing temperature and encourage consistent sintering [16]. To enhance the thermal characteristics of ceramic products, the initial batch is commonly modified by incorporating a porous additive. For example, the use of a cullet clay - sodium hydroxide system (in a clay-glass cullet component ratio of 50:50, sodium hydroxide 4-6%) and a 2% additive of anthracite, limestone or a gas-forming agent allows expanding the scope of application of glass-ceramics as a structural heat-insulating material [17], [18], [19]. [20] studied the chemical composition of various wastes applicable to partially replace natural components in ceramics. The

waste from galvanic manufacture is a paste-like material with a moisture content of 65–85% and a density of 1160–1240 kg/m^3. The sludge's composition and moisture content determine whether or not it can be used in a ceramic charge. The characteristics of ceramic products are mostly influenced by the bulk content of calcium carbonate in the sludge. In this regard, [21], [22] studied the effect of galvanic slurries on ceramic materials. They revealed on test samples that as the sludge content increases above 9%, substances such as calcium carbonate and iron oxides increase, which leads to an increase in the melt and intensive sintering of the shard. Thus, the introduction of up to 9% galvanic sludge into the batch helps preserve the porous structure and can be used as an additive for ceramic materials used in construction. To create ceramics, [23] proposed compositions based on red clay, including cullet and waste from the pulp and paper industries. The study showed that when burnt at 1050 °C and 1100 °C, ceramic materials based on cullet and additives from the pulp and paper sector showed superior mechanical and physical qualities than materials made entirely of clay. [24] supports the possibility of using large-scale, widely distributed scrap brick waste produced when replacing outdated brickwork or crushing waste to increase the raw material base for the production of ceramic bricks with good mechanical and physical qualities as well as a low thermal conductivity coefficient. They investigated the effect of waste on the technological characteristics of clayey raw materials. [25] proposed the addition of red or bauxite mud to clay. They showed that bricks made only of clay have a lower compressive strength than bricks made with 50% red mud added to clay and sintering at 950°C for an hour. Eventually, they obtained a clayey material with compressive strength of 52.54 MPa, water absorption of 21%, and linear shrinkage of 0.46%. [26] made in-depth research of the sludge from titanium and ilmenite pigment manufacturing. They concluded that leftover brick wastes can be used as a technogenic raw material to manufacture ceramic bricks. Ceramic stone, made with the addition of crushed brick powder, is characterized by fairly high mechanical strength and is graded M100 and M150, which meets the regulatory requirements of [27]. The optimal content of crushed broken ceramic bricks is 10-30 wt. %. When it increases to more than 30 wt. %, the compressive strength is significantly reduced, and the samples' water absorption rate increases, and when its content decreases to less than 10 wt. %, properties remain virtually unchanged [28]. [29] performed X-ray diffraction (XRD) of glass, and differential thermal analysis (DTA) and scanning electron microscopy (SEM) of samples containing 10-15% glass, obtaining an improvement in mechanical and thermal properties but a deterioration in the appearance of samples containing 25% or more glass.

Previous studies showed numerous possibilities of using waste for various purposes. However, the constant increase of industrial waste in the world suggests the need for further improvements in this direction. Besides, the large amounts of silicon oxide found in glass can react with calcium hydroxide in concrete to produce an alkali-silicate reaction that reduces the material's durability. If the proportions of adding glass powder are not correctly selected, a decrease in the strength and frost resistance of ceramic and concrete products can be observed. Due to the addition of glass to the composition of building materials, new technological processes are added, including grinding, heat treatment, etc., and accordingly, they require significant costs. The lack of knowledge in the aforementioned aspects suggests that it is necessary to continue research, improving the composition and properties of the final material, and develop more effective methods incorporating glass waste for concrete and ceramic production. To overcome these issues and to make further attempts to utilize the waste and supplement the existing knowledge, this study aims to study the potential of glass wastes for concretes and ceramics.

2. Methods

Concrete and ceramic samples were prepared in laboratory conditions incorporating glass as an industrial waste.

The mixture for the concrete samples was prepared according to [30] using M400-graded cement from Heidelberg [31], sand and gravel from a quarry deposit 15 km from Astana, tap water, and glass. The sand was cleaned of foreign matter and sieved through a 0.63 mm sieve. It was

dehydrated in a drying chamber at a temperature of 100 °C. The gravel was washed thoroughly to remove various impurities and dried in similar conditions. The glass was taken from the remains of bottles crushed to a size of 1-2 mm and turned into powder (Figure 1a). The chemical composition of the powder glass was preliminarily revealed by XRD spectral analysis using a 5EDX9000B spectrometer [32]. Chemical element concentrations were assessed in relation to reference samples. Table 1 below shows the composition of the prepared concrete mixtures.

Table 1 – Composition of concrete samples, %

No.	Cement	Sand	Gravel	Water	Glass waste
1	10.20	23.49	50.02	9.16	7.12
2	10.20	20.43	50.02	9.16	10.18
3	10.2	17.42	50.02	9.16	13.2
4	10.2	14.51	50.02	9.16	16.11
5	10.2	10.73	50.02	9.16	19.89

The concrete samples were prepared in 10×10×10 cm cubic forms (Figure 2b) and were kept for 28 and 112 days, after which tests were carried out for compressive strength according to [33].

a) Crushed glass

Figure 2 – Concrete samples

Figure 1 – Preparation of concrete samples

The mixture for the ceramic samples was prepared using the crushed bottle glass as above and clay from a quarry deposit owned by the SG Brick factory [34] in Astana. The selected clay belongs to the group of medium-dispersed clays and is technologically characterized as moderately plastic and insensitive to drying and firing processes. The clay was preliminarily crushed using a jaw crusher before being mixed with glass powder. Several compositions of clay-glass mixtures were prepared with a glass fraction of 5%, 10%, 15%, and 25%. The ceramic samples were molded in a cylindrical shape with a diameter of 5 cm and a height of 10 cm. The samples were placed in a drying chamber and fired in a kiln at 1000-1200 °C, according to [35]. After that, the samples were tested for compressive strength according to [36] (Figure 2).

Figure 2 – Ceramic sample tested for compressive strength

Next, we selected the crushed particles of both concrete and ceramic samples and subjected them to SEM likewise [29] using a Hitachi TM4000Plus microscope in magnifications ranging from 25 to 2000 times to examine the microstructural characteristics of materials. The research on phase distribution and the characteristics of the concrete surface received the most attention. SEM analysis assisted in determining the surface's morphology and structure; the existence of cracks and other flaws, which can impact the strength and resistance to additional impacts; and the size and distribution of the materials particles. Additionally, we conducted an energy-dispersive spectroscopy (EDS) to reveal the composition of the concrete and ceramic materials measured in mass and atomic fractions of each element detected. This analysis helped identify the main chemical elements and see how they are distributed across the material at different points, and how the material looks from the inside at different magnifications.

3. Results and Discussion

The test results of the glass spectroscopy showed a high tendency of silicon dioxide (silica). Most of the types of glass mentioned in the literature contain significant amounts of SiO_2 (>70%). This is explained by the fact that SiO_2 belongs to the group of glass-forming oxides, and it is prone to the formation of supercooled melt-glass. The glass with such a percentage of SiO_2 has high strength, hardness, heat resistance, and chemical inertness. Thus, the chemical composition of glass relative to cement is presented in Table 2.

Table 2 – Chemical composition of glass relative to cement, %

Na_2O	MgO	Al_2O_3	SiO_2	SO_3	Cl	K_2O	CaO	TiO_2	Fe_2O_3
0.136	0.61	2.4981	90.124	0.0401	0.0028	0.1674	12.7689	0.1418	1.0772

Table 2 above shows that the glass contains a significant amount of silica. High SiO_2 content usually ensures high chemical inertness and thermal stability of the material. As noted in [37], Al_2O_3 and CaO play a key role in the closure of hydrate phases in the cement paste. The presence of Al_2O_3 (about 4.3%) can ensure the formation of additional aluminate phases that increase early strength. A small amount of alkali oxides (Na_2O, K_2O) can, on the one hand, hinder hydration and, on the other hand, increase the risk of alkali-acid filling reaction. However, in this case, their content is relatively small.

Figure 3 shows spectra of glass compared to cement.

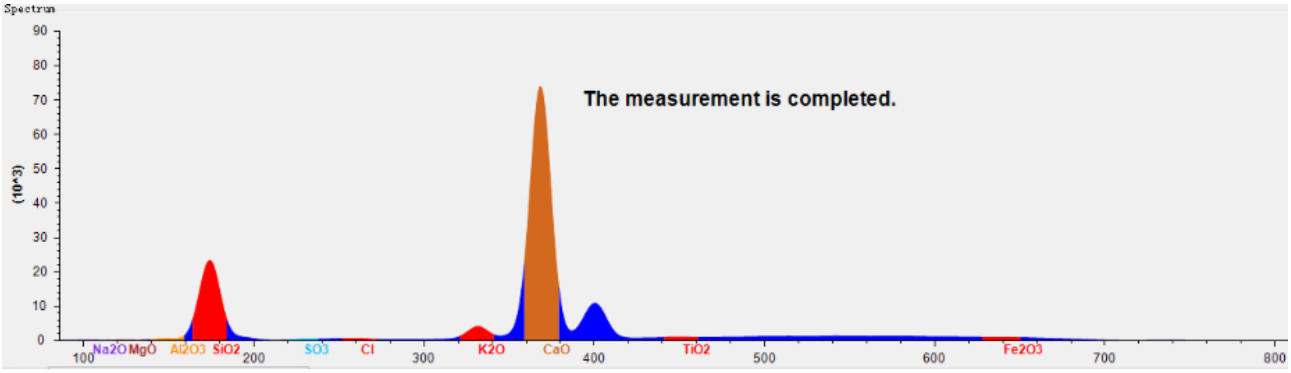


Figure 3 – Spectra of glass compared to cement

The high SiO_2 content in glass (around 70%) promotes a pozzolanic reaction when interacting with the cement matrix, which can lead to improved mechanical properties of concrete due to the additional formation of cement compounds [38]. High CaO content in cement promotes intensive hydration, which ensures the formation of a strong cement matrix. A slight decrease in CaO (our 62% compared to 64–66% in the literature) can slow down hydration reactions and affect early strength gain [39]. The lowest Cl content may indicate high chemical purity. Chlorine is usually added to glass

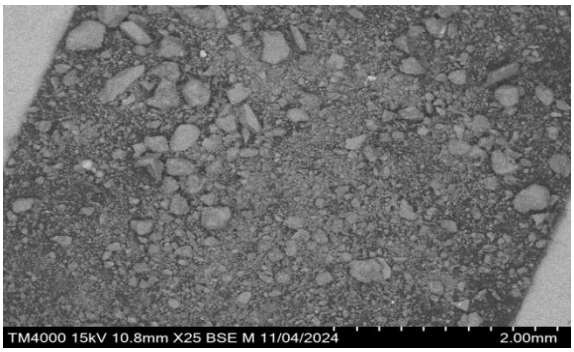
as a flux to improve fusibility and remove gas bubbles during melting. The presence of 2.5% Al_2O_3 in the glass composition shows improved mechanical and thermal properties of the glass.

The compressive strength of concrete was 20.50 MPa after 28 days, but after 112 days, it was 76.36 MPa. The maximum compressive strength in this concrete is achieved by using glass as a fine aggregate instead of sand at 20% replacement, while in [9], it reached 30%. Several studies suggest that a longer curing time is beneficial for strength growth and that a higher specific surface area of recycled and broken glass may lead to a bigger increase in compressive strength [37]. Because the "alkali-silicate reaction" is negligible and may even result in a decrease in strength because of the initial weak bond between the glass and the cement matrix, adding glass powder to concrete can slow down the strength gain process in the early stages while simultaneously contributing to an increase in strength later on. Due to the hydration of cement, concrete becomes stronger over time, and the effect is enhanced in later stages when glass particles containing amorphous silica are present. This helps to generate other hydration products, including hydrated calcium silicates (C-S-H), which boost strength. Pozzolanic reaction: Glass acts as a pozzolanic additive since it is an amorphous substance with a high silica content. This indicates that it can generate more C-S-H gels by interacting with calcium hydroxide that is released during cement hydration.

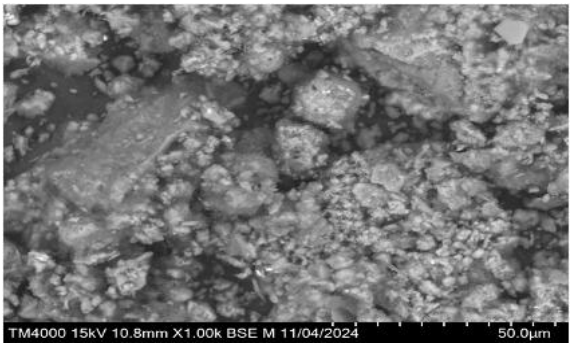
The compressive strength of the ceramic material was 13.14 MPa. This was achieved with the addition of glass in the amount of 15% of the total mass of clay. Since the typical strength values for wall materials are between 10 and 15 MPa, a result of 13.14 MPa might suggest reduced porosity and efficient compaction. Ceramic materials with a strength of 13.14 MPa are used as heat-insulating blocks, decorative wall panels, cladding for buildings, and load-bearing walls.

To make the material more appealing for a range of climates, thorough research must be carried out to ascertain its resilience to cold, durability, and aggressive environment behavior.

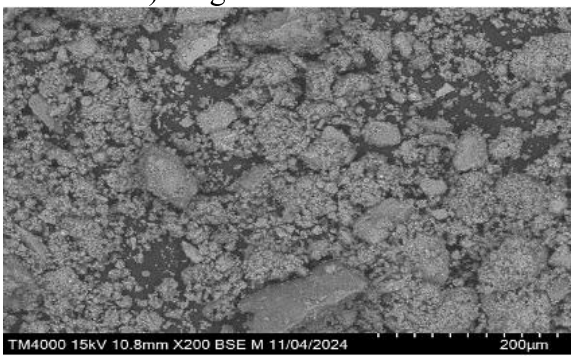
Figure 4 shows concrete's microstructure analysis report by Scanning electron microscope.



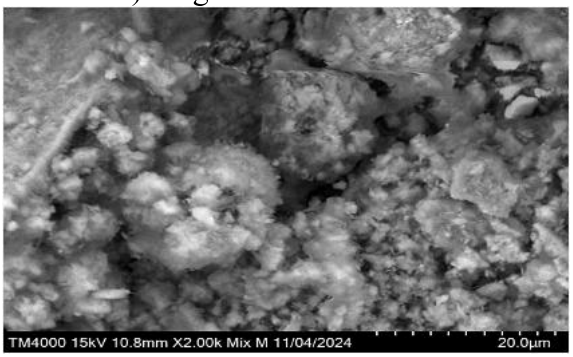
a) Magnification of 25x



c) Magnification of 1000x



b) Magnification of 200x



d) Magnification of 2000x

Figure 4 – Micrograph of concrete material surface

The above microphotograph of the concrete surface at different magnifications allows us to see the structure of the material. Individual particle detection, measurement, and analysis were made feasible by the capacity to extend to various sizes: a) The material's heterogeneity and porosity are demonstrated by 1 mm-sized particles. b) A more intricate structure is seen here, with individual filler

and cement matrix grains ranging in size from 50 to 200 microns. c) Particles range in size from 5 to 50 microns, and hydration products are apparent. d) The hydrated calcium silicate phases that give C-S-H gel its mechanical strength are apparent as crystalline structures with particle sizes ranging from 1 to 5 microns.

Compared to classic concrete, capillary pores can reach several micrometers, while in concrete with glass their number and size are reduced. This is confirmed by studies where glass acts as a microfiller, reducing porosity [40]. The area between the filler and the cement paste is usually more porous and vulnerable to moisture and ion penetration, and the addition of glass can result in a more even distribution of fine particles in the cement matrix [41]. The use of glass as an additive to the concrete mix has a positive effect on its microstructure. Compared to classic concrete, concrete with glass has a denser structure of cement stone, reduced capillary porosity, and potentially higher strength and durability. However, it is necessary to take into account the possible alkali-silicate reaction with an excessive amount of alkaline components, therefore, it is necessary to observe the optimal proportions of the glass additive to eliminate negative effects.

Figure 5 illustrates various elements found in the concrete with the peaks in the spectrum.

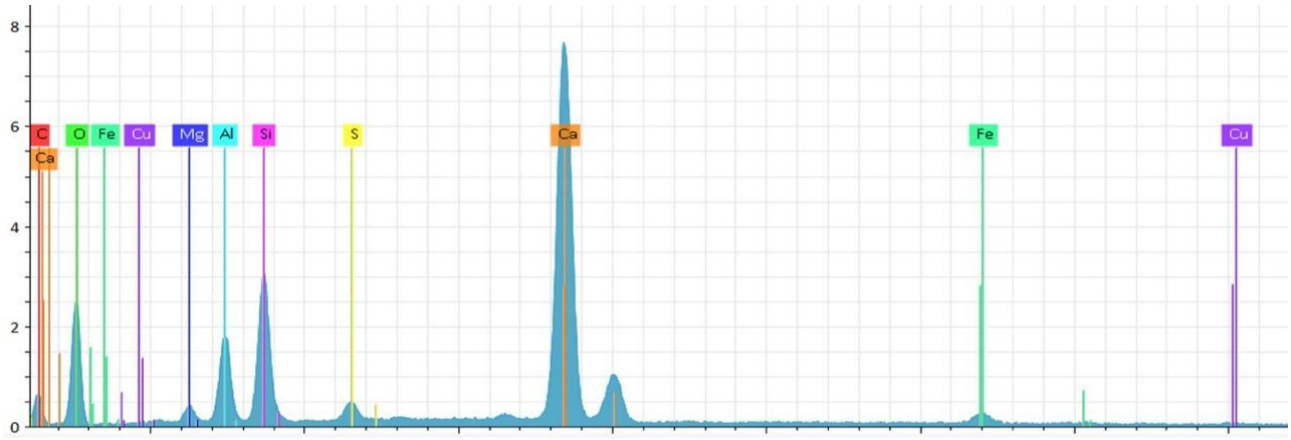
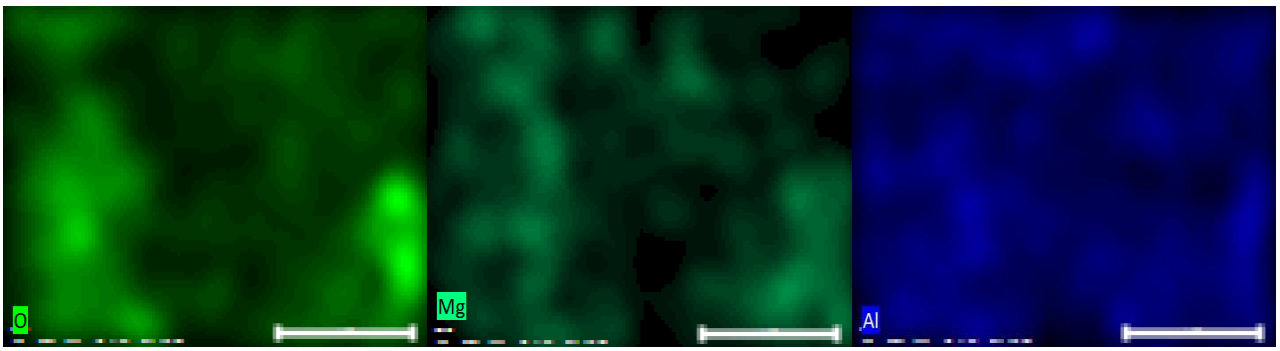


Figure 5 – Energy dispersion spectrum of concrete by elements

Based on the EDS results above, the percentage of elements found in concrete are as follows: O – 38.04%, Mg – 1.51%, Al – 6.95%, Si – 11.35%, S – 1.6%, Ca – 37.04%, Fe – 2.59%, and Cu – 0.92%. The oxygen is the primary ingredient of concrete's oxides and silicates. It contributes to the creation of oxide compounds (silicates and aluminosilicates), which make up the majority of concrete's structure and provide it with stability, mechanical strength, and water resistance. The primary ingredient in cement is calcium, which also helps to generate calcium hydrates (C-S-H) and portlandite ($Ca(OH)_2$), which give concrete its resilience. Because magnesia compounds are formed, magnesium has an impact on frost resistance. Iron, copper, sulfur, and aluminum all speed up the cement's hardening process and offer resistance against chemical impacts.

The element distribution map in Figure 6 shows how the elements are distributed in the concrete, with each color representing a different element: O, Mg, Si, Fe, Al, and C.



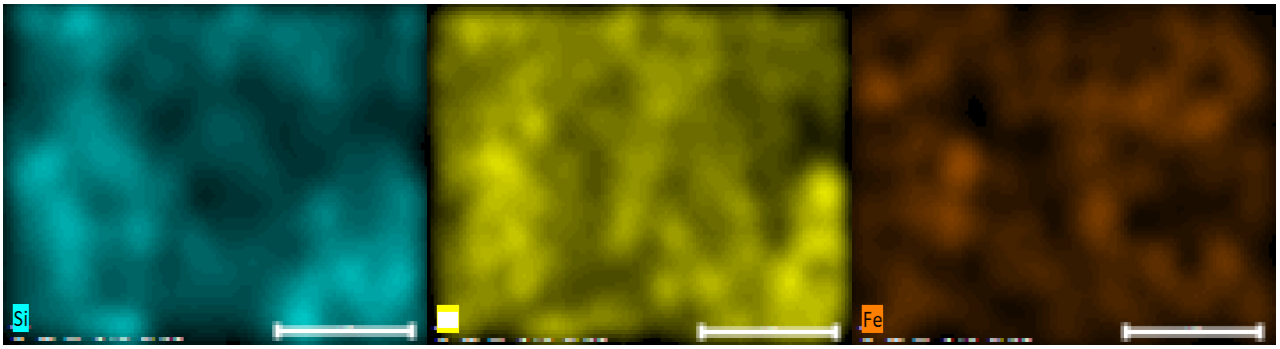


Figure 6 – Energy dispersive spectroscopy elements indicated in colors

Table 3 provides information about the chemical composition of concrete by oxides.

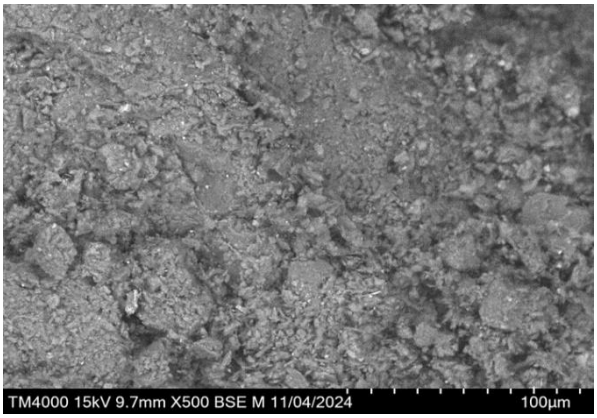
Table 3 – Oxide chemical composition in concrete, %

MgO	Al_2O_3	SiO_2	SO_3	CaO	FeO
10.17	13.66	33.07	1.96	29.71	4.40

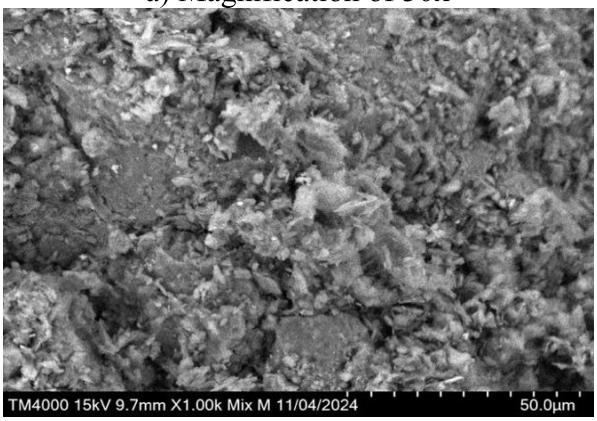
The decrease in CaO (29.71%) compared to conventional concrete (60-67%) confirms the presence of pozzolanic materials such as glass. The increased content of Al_2O_3 (13.66%) improves the thermal resistance of concrete, it exceeds the values of the classic composition of concrete on Portland cement (4-8%). MgO (10.17%) in this composition is also higher (1-5%), which affects swelling and shrinkage. SiO_2 is the main component of the pozzolanic reaction, which increases the density of the cement stone structure [40].

Figure 7 shows micrographs of deep (BSE) and mixed (Mix) surfaces of ceramic material burned at 1000 ℃ with 15% waste addition, in 4 different magnifications.

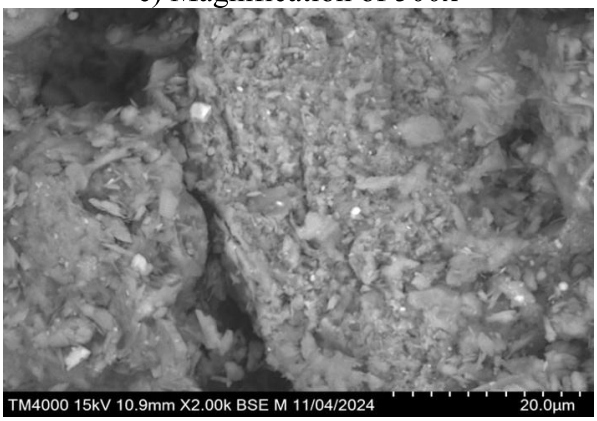
a) Magnification of 30x



c) Magnification of 500x



b) Magnification of 1000x



d) Magnification of 2000x

Figure 7 – Micrograph of ceramic material surface

In Figure 7a, particles are large, angular, and ranging in size from 500 microns to 1 mm. In Figure 7b, particles range in size from 1 micron to 50 microns with fine grains and thin lamellar formations. In Figure 7c, particle size varies from 50 microns to 100 microns. We see a porous structure and heterogeneity in the distribution of the material. In Figure 7d, particles range in size from 0.5 microns to 20 microns. Here, we can see fine structures, microcracks, and areas of dense particle accumulation. Sizes from 0.5 microns to 1 mm affect its mechanical properties, such as strength and resistance to external influences.

It can be assumed that the appearance of microcracks and pores is associated with the addition of glass components, which leads to a change in the structure of the material during sintering. Probably, glass helps to reduce porosity and increase the strength of the material.

Comparing the obtained results with other studies, it can be noted that the addition of glass components similarly affected the microstructure of ceramic materials in the works of other authors [14]. With an increase in the cullet content from 0% to 10%, water absorption decreased from 15.67% to 15.10%, porosity decreased from 28.77% to 28.29%, and the maximum strength of 31.68 MPa was achieved with a cullet content of 7.5%, which is associated with the formation of a glassy phase filling the pores and microcracks in the ceramic structure. Studies conducted using spectral analysis also showed a decrease in porosity and an improvement in mechanical properties with the addition of a glass phase [42].

The sintered body displays the liquid phase as a result of the glass waste melting, which closes holes and reduces porosity, reducing water absorption and enhancing the mechanical properties of fired samples [43].

Mapping the surface of the ceramic sample is in view at Figure 8.

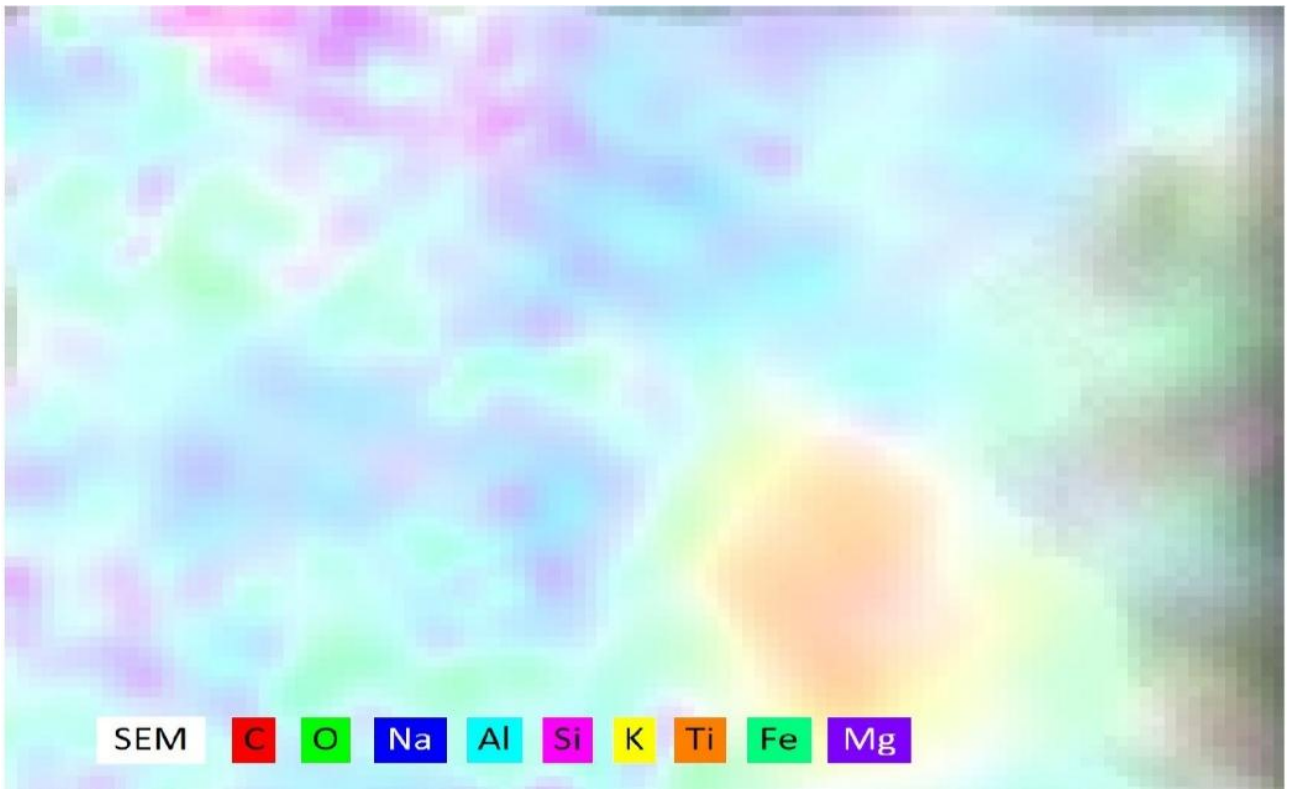


Figure 8 – Energy dispersive spectroscopy elements are indicated in colors

The elements of the ceramic sample presented on the map with magnification 4000x are highlighted in different colors, and we also see the designation of each element in a specific color. The predominance of green, blue and pink colors is evident, which corresponds to the high O, Al, Si content. We see this in the table below.

Table 4 shows the chemical composition of ceramic material by elements.

Table 4 – Element-by-element composition of ceramic material, %

O	Na	Mg	Al	Si	K	Ti	Fe
46.03	1.01	0.51	12.83	21.54	2.85	11.62	3.61

In Table 4 above, the high Si (21.54) and Al (12.83) content confirms the presence of the main ceramic phases, such as quartz and alumina, which is consistent with the data presented in many studies. The presence of K and Na in small quantities (2.85% and 1.01%, respectively) promotes sintering and the formation of a glassy phase. This improves the structural integrity of the material and facilitates the production process. Similar observations are given in works on the optimization of the composition of ceramic mixtures [44]. This ratio is important for ensuring the thermal stability and strength of ceramic products. The Ti percentage of 11.62% may suggest the presence of phases that boost the material's strength. Research suggests that some titanium-containing phases might aid in enhancing mechanical properties. More study is necessary to determine whether much titanium might lead to the production of undesirable pollutants. Although the Fe level is quite modest (3.61%), its presence can influence the final product's color and, in some situations, aid in the production of particular crystalline phases. According to the research, even trace levels of iron can alter ceramics' mechanical and optical properties [42].

Table 5 shows the chemical composition of ceramic material by oxides.

Table 5 – Oxide chemical composition of ceramic material, %

MgO	Al_2O_3	SiO_2	K2O	TiO_2	FeO	Na_2O
0.77	22.14	42.09	3.14	17.70	4.24	1.24

As can be seen from Table 5 above, the next oxides are responsible for the strength characteristics: Al_2O_3, SiO_2, TiO_2, SiO_2, Al_2O_3, and Na_2O. Aluminum provides mechanical strength and chemical resistance, especially at high temperatures, and silicon indicates resistance to acids. Titanium and iron oxide improve the corrosion properties of the material, and potassium oxide chemical resistance. The presence of alkaline oxides (K_2O, Na_2O) and MgO in small concentrations allows the reduction of the baking temperature, improves the process of glassy phase formation, and increases the illumination of the finished product [45].

4. Conclusions

The study's findings support the potential applications of cullet in the manufacturing of concrete and ceramic construction materials. The implementation of these technologies not only lessens the environmental impact of waste disposal but also enhances the end goods' performance attributes. However, the following crucial factors must be considered for successful industrial implementation:

1. Taking into consideration the existence of cullet, technological parameters for hardening concrete and firing ceramic goods are adjusted.
2. Production cost estimation that accounts for the substitution of glass waste for conventional raw materials.
3. Carrying out experimental studies at already-existing businesses to evaluate the true properties of materials.
4. Develop a scheme for the delivery of glass waste to building materials manufacturers, calculate all costs associated with transportation, etc.
4. It is necessary to develop appropriate technological schemes for the production of ceramic and concrete products using waste glass for various purposes.
5. Conduct experiments to define mechanical load resistance, moisture resistance, and frost resistance.
6. Try mixing glass waste with other industrial waste in certain proportions and see their effect on the properties of ceramic and concrete materials.

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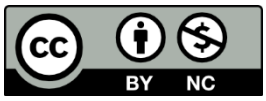

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