











## Opportunities for environmentally sustainable development of low-carbon technologies in cement production

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**Abstract.** In the context of the transition to a low-carbon economy, the development and implementation of environmentally sustainable technologies in cement production is becoming a key priority. Therefore, the development of new binding materials with reduced clinker content or no clinker at all is becoming a key area for reducing the carbon footprint in construction. Use of clinker-free binders, such as geopolymers and various equivalents based on mineral additives, can significantly reduce the carbon footprint of the construction sector in the environment. The most promising and appropriate benchmark is the disposal of industrial waste of aluminosilicate oxide composition with subsequent mechanical and alkaline activation. For the first time, the microstructure of geopolymers based on aspiration cement dust and tuff has been comprehensively studied. The theoretical prerequisite for the creation of a binder system of such a concept is the synthesis of sufficiently strong and resistant to external manifestations of alkali metals, including the structures of frame aluminosilicates with a hidden crystalline structure. The results of a comprehensive study (X-ray phase analysis, scanning electron microscopy, electron dispersion spectrometry, differential thermal analysis and IR spectroscopy) indicate the presence of characteristic hydration reaction products in the geopolymer paste. The following have been identified in the composition of the material: hydrated aluminosilicates; aluminates; silicate groups of sodium and calcium; mineral phases (quartz, calcite); feldspars of the albite-orthoclase series; micaceous components, etc. The data obtained confirm the typical composition characteristic of the processes of structure formation in geopolymer systems. The results obtained on the key results of the conducted studies confirm the high efficiency of the proposed technology and guarantee increased strength and durability of geopolymer concrete.

**Keywords:** industrial waste, sustainable development, low carbon footprint, man-made raw materials, clinker-free technology, activation, aspiration dust, structure

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## 1. INTRODUCTION

In the context of growing demand for cement, innovative approaches are needed to minimize the negative impact of its production on the environment [1]. One such solution is the development of new types of binders and concrete with their use with a reduced clinker content or complete rejection of it [2]. Current research confirms that the use of alternative binders, such as slag-alkali and geopolymer compounds based on various bases, contributes to a significant reduction in carbon dioxide (CO<sub>2</sub>) emissions compared with traditional cements. This makes them promising in terms of environmental sustainability in the construction industry. Such binders not only reduce the carbon footprint, but also demonstrate comparable, and in some cases improved, performance characteristics, which opens up new opportunities for creating environmentally friendly building materials. [3]. Such approaches are binary in nature in terms of the result obtained - they not only help to reduce the carbon footprint of the resulting composite, but also improve the physical-mechanical properties and performances of the final product [4].

In addition, investments in carbon dioxide capture and storage (CCS) technologies are a promising path to so-called "carbon neutrality" [5]. Such technologies are capable of capturing emitted CO<sub>2</sub> at the production stage and storing it in geologically stable formations [6]. Despite the high initial costs, successful implementation of CCS could be a game changer for the cement industry [7, 8].

Coordinated policies at the level of governments and international organizations are also essential [9]. Introducing strict emission standards and supporting sustainable practices in construction could significantly accelerate the transition to cleaner technologies [10]. By combining the efforts of the scientific community, industrial magnates, and the public sector, it is possible to achieve a more sustainable development of the cement industry and reduce its contribution to global warming [11].

The most promising and appropriate benchmark is considered to be the utilization of industrial waste of aluminosilicate oxide composition with subsequent mechanical and alkaline activation [12]. It is recommended to use alkali metal hydroxides of sodium and potassium, sodium (potassium) meta-, di- or orthosilicates, sodium aluminates, and soda ash as an alkaline sealant [13]. The theoretical basis for the creation of a bonding system in this concept is the formation of stable and stable structures resulting from the interaction of cryptocrystalline aluminosilicates of the  $R_2O \cdot Al_2O_3 \cdot nSiO_2$  type and finely dispersed solid phases that have aluminate and silicate ions in their structure under alkaline medium conditions [13]. The process of physicochemical reactions in the studied dispersed system "aluminosilicate solid phase – alkaline solution" can be described through a number of key transformations that occur sequentially and in parallel [14].

Firstly, dispersion and destruction of aluminosilicate bonds occurs under the influence of an alkaline environment, which leads to the formation of colloidal solutions [15].

Secondly, small particles of alkaline aluminosilicate and silica sols are converted into larger compounds as a result of their interaction [16].

Thirdly, the implementation of cation exchange ( $2Na^+ \leftrightarrow Ca^{2+}$ ) promotes the formation of new structures, which leads to the emergence of condensation-crystalline formations [17].

Thus, the described processes illustrate the complex mechanism of interactions in this dispersed system, where each of the transformations plays an important role in the formation of the result [18].

The discovered patterns in the formation of the structure and ensuring the properties of geopolymer paste using alkaline activation binders make it possible to obtain concrete composites for various purposes [19]. This includes quick-hardening and acid-resistant mixtures, cellular concretes with a density grade of D150–350, as well as heat-resistant, low-exothermic and high-strength materials of classes up to B140, shrinkage-free and many others [20]. Such possibilities significantly enrich the raw material base for the cement industry [21].

In addition, alkaline binders allow the use of substandard fine sands and fillers with a high content of clay and dust particles, including waste from paste crushing industries [22]. As a result of the research, frost-resistant concretes of grade F1000 and waterproof up to W30 were developed [23]. Their shear strength characteristics, as well as resistance to corrosion and biological influences, are comparable to the same indicators of products made of ordinary Portland cement [23, 24]. The conducted studies confirm that concretes based on alkaline binders have exceptional physical and mechanical properties and high performance characteristics. This makes them a promising material for use in various fields of construction where traditional cement mixtures are unable to meet the specified requirements [25]. Such an innovative approach creates significant prospects for the introduction of modern composite materials in the construction industry, guaranteeing increased reliability and operational durability of the structures under construction [26]. The new generation of concrete composites demonstrates: high functional efficiency due to an optimized material structure; an expanded scope of application – from responsible load-bearing structures to specialized structures; competitive market advantages, including improved technical and economic indicators. These characteristics allow us to consider these materials as a promising alternative to traditional construction solutions that meet modern requirements for sustainable development and resource conservation [27].

Moreover, at the end of the 20th century, regulatory and technical documentation and recommendations regarding raw materials, recipes, indicators and the procedure for preparing slag-alkaline binders and concretes based on them were approved [28, 29]. The combination of these factors, supported by the key advantages of the technology, indicates the need for intensive development of clinker-free binders and their widespread introduction into construction practice. Precast reinforced concrete structures and products made from them are particularly promising areas of application [30], road [31] and monolithic construction [32], especially in an aggressive sulfate environment [33], hydromelioration [34, 35], subway tunnel tubing [36, 37].

Thus, in light of the increasing emphasis on environmental sustainability and responsible treatment of nature, progress in the study and use of technologies that do not require clinker is becoming not only preferable but critically important for the future of the construction industry. This is particularly relevant in the context of the depletion of the planet's natural resources [38, 39]. A research gap lies in the comprehensive study of the microstructure of geopolymers based on aspiration cement dust and tuff [40, 41].

The goal of this work was to develop a low carbon, non-clinker binder using a technological raw material. The following tasks were achieved:

- study of the characteristics of raw materials as potential components of the binding bond of alkaline clogging;
- selection of the optimal composition of the geopolymer composition using clinker production waste;
- comprehensive examination of the obtained alkaline activate paste microstructure.

## 2. METHODS AND MATERIALS

### *Materials*

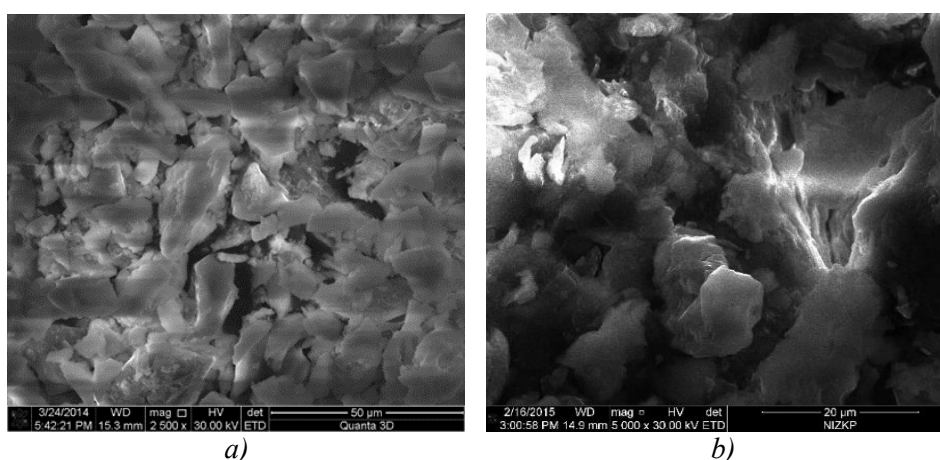
The authors of the work offer their vision in solving the identified issues, and the proposed development results aimed at creating a clinker-free binder system will reveal the positive possibilities of alkaline mixing of finely dispersed additives of various origins. Aspiration dust from a cement plant was used as a dispersed powder of the binder composition. This product is not secondary in cement production and is not returned back to the technological cycle of the wet method of preparing the raw mix, since attempts led to thickening of the sludge, which negatively affects the production technology and properties of clinker. The origin algorithm of aspiration dust is accumulation in a dust precipitation system, extraction and storage on the territory of the plant as it accumulates, followed by removal to nearby agricultural lands. Being in a finely dispersed state, aspiration dust is weathered in the environment, harming all living beings. But a weighty reason to find an application for this waste is that aspiration dust is obtained from a carefully selected and adjusted raw mix of clinker production, in which clay minerals and calcium carbonate (calcite) are burned at  $t = 400\text{--}500^\circ\text{C}$  and at the same

time retain an amorphous state. It is also important that it can be used without mechanical activation in its natural form (specific surface area  $S_{sp} = 220 \text{ m}^2/\text{kg}$ ).

An aqueous solution of low-modulus sodium liquid glass with a density of  $1.42 \text{ g/cm}^3$  and a silicate modulus of 2.8 (in terms of dry matter  $\text{Na}_2\text{O} - 6.2\%$ ) was used as an alkaline grout.

To improve the processes of forming the geopolymer paste structure and reducing pore space in the binder, mineral powder from volcanic tuff was added. The choice of the additive of volcanic origin is justified, since, firstly, it can be classified as an aluminosilicate raw material with the presence of a glass phase; secondly, this raw material has a sufficient degree of “ordering” of the structure; thirdly, when interacting with alkali metal cations, the formation of the necessary structures occurs. The matrix’s structural elements are connected to them. The specific surface area of the volcanic tuff powder studied varied in the range  $480\text{--}500 \text{ m}^2/\text{kg}$ . The reaction activity was observed even when the samples were compressed with water. The strength limit of the compression test was  $3.2 \text{ MPa}$ .

A “rough” relief of the surface of the grains characterizes the microstructure of the particles of the volcanic tuff additive (Fig. 1) with closed porosity of the internal structure. This confirms the high solubility and activity of ash particles.



**Fig. 1.** SEM images of volcanic tuffa particles: a)  $\times 2500$  b)  $\times 5000$ .

Therefore, the addition of volcanic tuff (10% of the binder) will make up the percentage of aluminosilicates in the system due to the reduction of the concentration of the clinker component. The setting time of the alkaline binder has increased by 40 minutes, which facilitates high-quality preparation and molding of products.

The addition of sodium  $\text{Na}_2\text{SiF}_6$  silicate (in the investigations was determined a dosage of 6% by mass of  $\text{Na}_2\text{SiO}_3$ ) assists in the secretion of gel of polymerized silicon silica  $\text{Si}(\text{ON})_4$ , a Parallel formed  $\text{NaF}$  increases the strength of the artificial paste.

#### Mix design

Preliminary studies [2,6] have established effective proportions for the combination of the following components: “Aspiration cement dust – volcanic tuff –  $\text{Na}_2\text{SiO}_3$  – sodium silicate”. The binding agent formulation is shown in Table 1.

**Table 1.** Mix proportions, wt. %.

Dust	Volcanic tuff	$\text{Na}_2\text{SiO}_3$	$\text{Na}_2\text{SiF}_6$
56	10	28	6

#### Methods

From the binding composition “aspiration dust – volcanic tuff –  $\text{Na}_2\text{SiO}_3$  – sodium silicofluoride” (Table 1), sample series of  $40 \times 40 \times 160 \text{ mm}$  were prepared, using standard Volsky sand as the fine aggregate. The specimens were demolded the next day and then placed in a drying oven for heat

treatment at a temperature of 80–110 °C for several hours (2–2.5 hours), with the process duration being one week.

The specific surface area of the volcanic tuff powders was determined using a PSKh-12 instrument (Sudakov–Khodakov device, Russia).

The alkaline activator – as the main carrier of binding properties – was examined using an electronic pH meter with a measurement accuracy of 0.01 pH units; as a result, the hydrogen index of  $\text{Na}_2\text{SiO}_3$  was  $\text{pH} = 12.6$ .

The microstructure of the prepared samples was studied using modern equipment; electron probe analysis was conducted using a Vega II LMU scanning electron microscope (Czech Republic), and X-ray phase and structural analyses were carried out using an ARLX'TRA diffractometer (Switzerland). Infrared spectroscopy was also performed using an IR Fourier spectrometer (Japan). The physical and mechanical properties of the binder material samples were determined in accordance with GOST 30744-2001.

### 3. RESULTS AND DISCUSSION

Analyzing the obtained data, it can be noted that the studied compositions are characterized by a fine-crystalline and heterogeneous crystalline structural organization (see Fig. 2c), with rounded closed pores with a radius of 0.25 mm (see Fig. 2a, b). The close interaction of the cement matrix and fine aggregate particles is clearly visible.

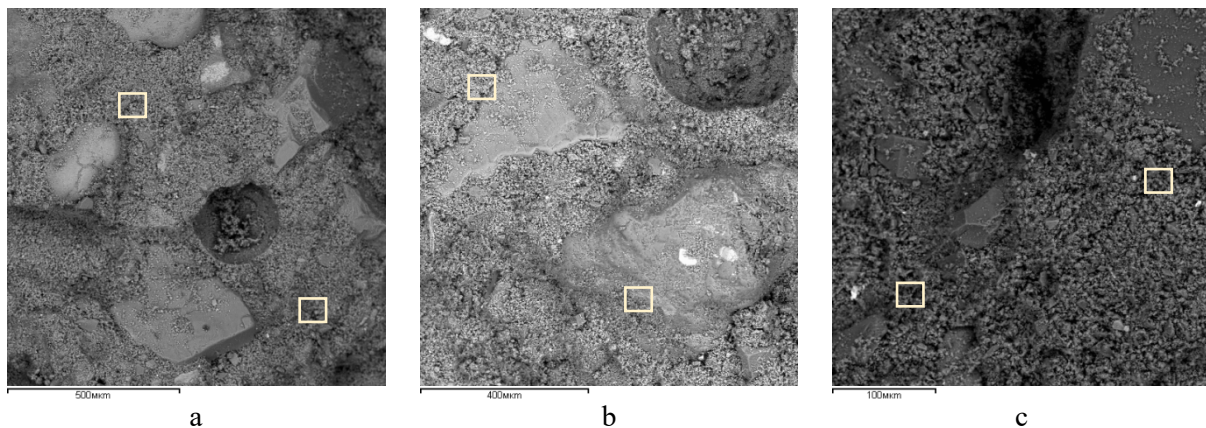
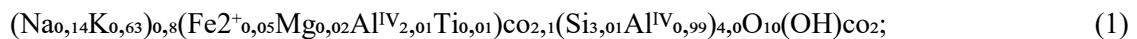


Fig. 2. SEM images of the studied binder in geopolymer concrete.

Hydrated sodium and calcium aluminosilicates play a key role in the structure formation. The observed crystalline aggregates are characterized by high density and reach sizes of 10-20 microns (Fig. 3, 4). A semi-quantitative analysis revealed their complex component composition (Tables 1, 2; analyses 1-2), including interacting phases:

1. Basic Interactions:
  - Reaction zones with calcite • Potential contacts with thin layers of  $\text{Ca}(\text{OH})_2$  (Table 2, analyses 3-5);
  - Interfaces with muscovite mica scales (~20 microns)
2. Crystal chemical features of muscovite: • Set the formula:



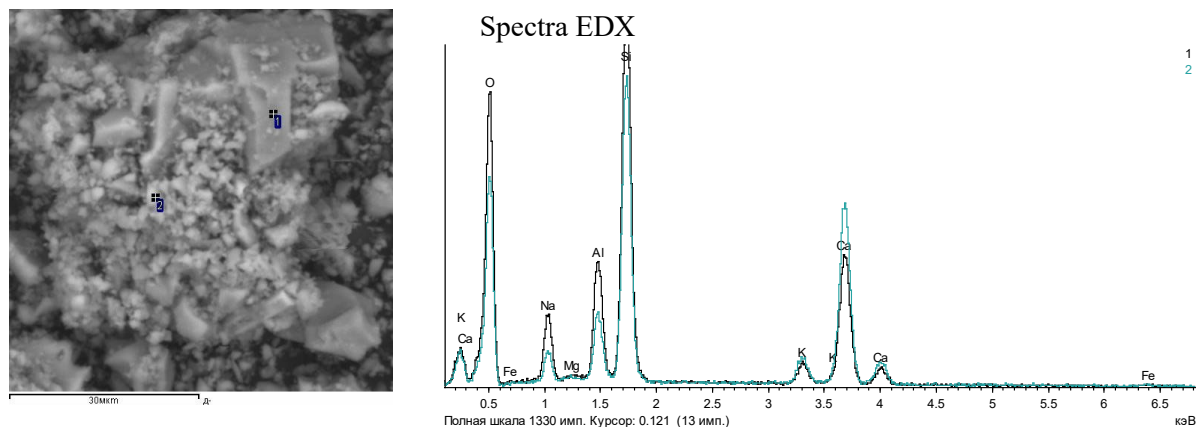
- Key Features: o Predominance of the potassium component ( $\text{K}^+/\text{Na}^+ \approx 4.5:1$ );
- High aluminum content in octahedral (2.01 at units) and tetrahedral (0.99 at units) positions;
- Ideal hydroxyl group  $(\text{OH})_2$ .

3. Analytical confirmations:

- X-ray phase analysis: identification of muscovite;

- Electron probe microanalysis (EZA): quantitative compliance with the formula;
- Preservation of the stoichiometry of the silicon-oxygen tetrahedral layer ( $\text{Si}+\text{Al} = 4.0$ ).

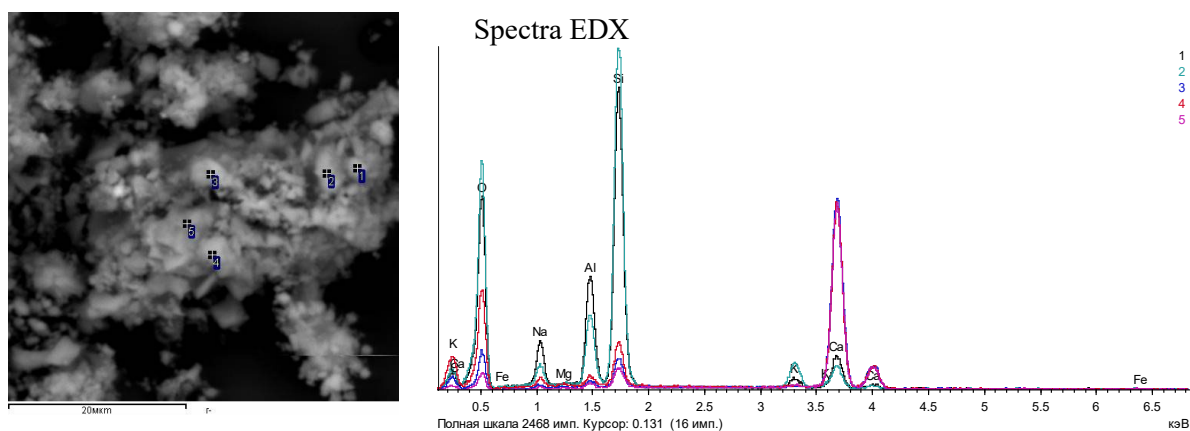
The data obtained strongly indicate the formation of stable aluminosilicate structures with a well-defined crystallochemical organization, which significantly affects the mechanical properties of the composite.



**Fig. 3.** Ground mass hydrates.

**Table 2.** Composition of the bulk hydrates, in wt.% (locations of analysis are shown in Fig. 3).

	Na <sub>2</sub> O	MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	K <sub>2</sub> O	CaO	FeO	Total
1	8.21	0.19	12.62	46.89	2.02	17.69	0.49	88.11
2	4.09	0.22	6.69	35.91	2.49	24.61	0.52	74.53



**Fig. 4.** Phases of the main mass.

**Table 3.** Composition of the main mass crystals, in wt.% (locations of analysis are indicated in Fig. 4).

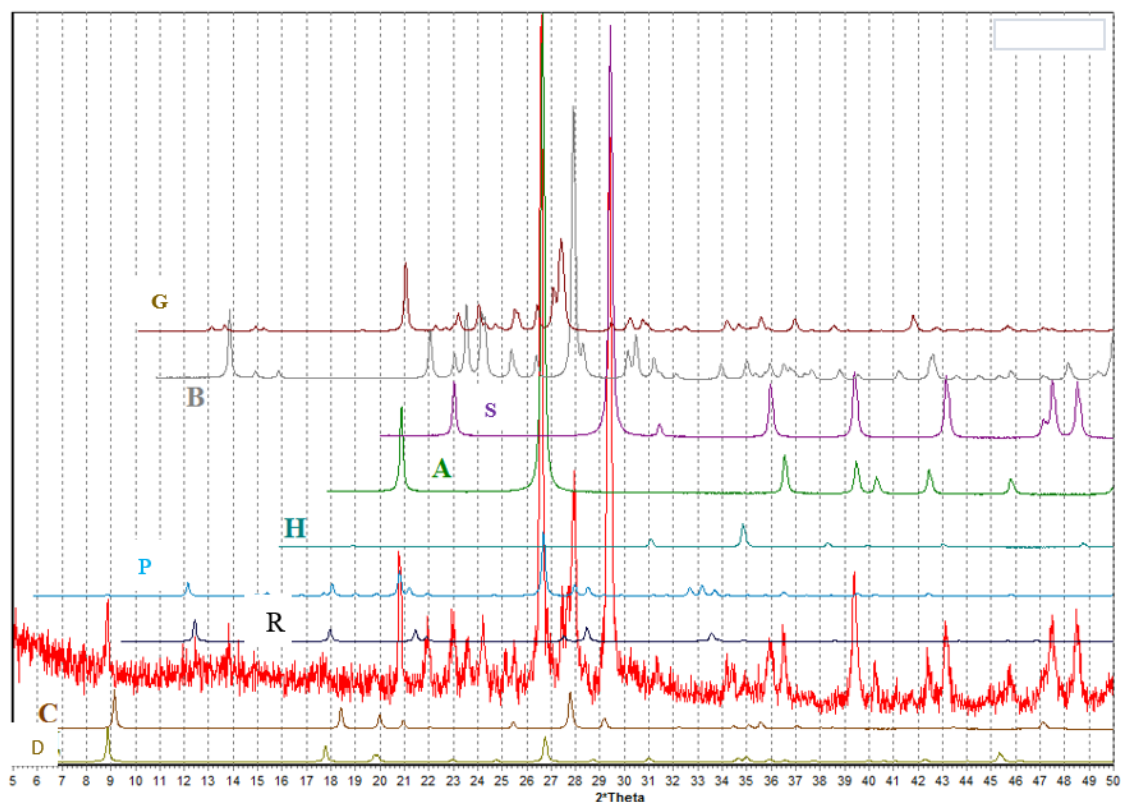
	Na <sub>2</sub> O	MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	K <sub>2</sub> O	CaO	FeO	Total
1	8.59	-	18.68	61.71	1.55	7.58	0.52	98.63
2	4.25	0.27	12.09	71.51	5.11	5.49	0.15	98.87
3	0.93	0.19	1.2	5.75	0.25	44.67	-	52.99
4	2.31	0.52	1.27	4.29	0.22	43.24	-	51.85
5	0.69	-	1.12	4.11	0.29	42.48	-	48.69



The results of X-ray phase analysis (XFA) confirmed the presence of the following crystalline phases in the sample (Fig. 5):

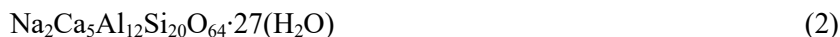
1. Main mineral components:
  - Quartz ( $\text{SiO}_2$ ) is the dominant phase
  - Feldspars:
    - Albite series (Na-plagioclase)
    - Orthoclase ( $\text{KAlSi}_3\text{O}_8$ )
  - Mica group (mainly muscovite)
  - Calcite ( $\text{CaCO}_3$ )
2. Specific components:
  - Zeolites (hydroaluminosilicates of the framework structure)
  - Possible secondary phases:
    - Amorphous aluminosilicate formations
    - Calcium hydroxide compounds
3. Features of the phase composition:
  - Feldspars exhibit isomorphic substitutions in the Na-K series
  - Zeolites indicate the processes of hydration and ion exchange
  - The combination of calcite with silicate phases indicates complex carbonation reactions
4. Technological consequences:
  - The presence of zeolites confirms the occurrence of low-temperature hydrothermal processes
  - The quartz/feldspar ratio affects the reactivity of the system
  - Calcite acts as a mineral filler and a reaction component

This mineralogical composition is characteristic of geopolymer systems that have passed the stage of alkaline activation, and explains the observed physico-mechanical properties of the material.



**Fig. 5.** Diffraction pattern of the sample “aspiration dust – volcanic tuff –  $\text{Na}_2\text{SiO}_3$  – sodium fluorosilicate” compared with the PDF-2 database data. The comparison phases are given: A – quartz, S – calcite, B – albite, G – potassium feldspar, D – muscovite, H – analcime, P – gismondite, R – garronite, C – paragonite.

X-ray structural analysis shows that the zeolite under study has a structure similar to garronite, whose theoretical formula is:



although the actual composition may vary significantly. The key reflection of this phase is clearly defined at  $2\Theta = 12.4$  ( $7.10 \text{ \AA}$ ). Electron probe analysis reveals the presence of hydrated amorphous Na-Ca-Si compounds rich in water (see Figure 6 and Table 3). Additional studies revealed the presence of aluminum-containing phases structurally and chemically similar to zeolites (Fig. 7, 8; Table 4). Characteristics of these phases:

1. Morphological features:

- Submicron particle sizes (less than 1-2 microns)
- Pronounced surface roughness
- Aggregated status

2. The elemental composition:

- Typical zeolite-like  $\text{SiO}_2/\text{Al}_2\text{O}_3 \approx 2.0$
- The presence of calcium as a framework – forming cation
- Possible analogues:

Ca-phillipsite:



Garroni:



3. Analytical limitations:

- Quantitative analysis errors (EDS):
- Small particle size effect
- The influence of surface topography
- Possible overlap of the spectra of neighboring phases

4. Technological implications:

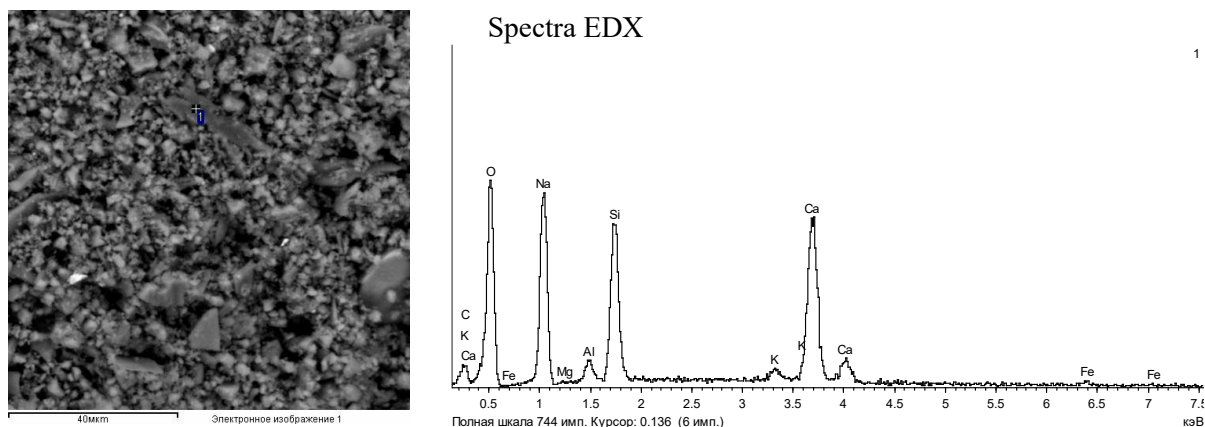
- Formation of nanoscale zeolite - like phases:
- Increases the ion exchange capacity of the system
- Helps to strengthen the matrix
- Affects the long-term stability of the material

5. Genetic interpretation:

- Formation of secondary hydroaluminosilicates
- The result of the interaction of an alkaline solution with aluminosilicate components
- A sign of low - temperature synthesis.

These phases, despite the difficulties of quantitative analysis, play a significant role in shaping the performance characteristics of the material, especially in terms of chemical resistance and durability.

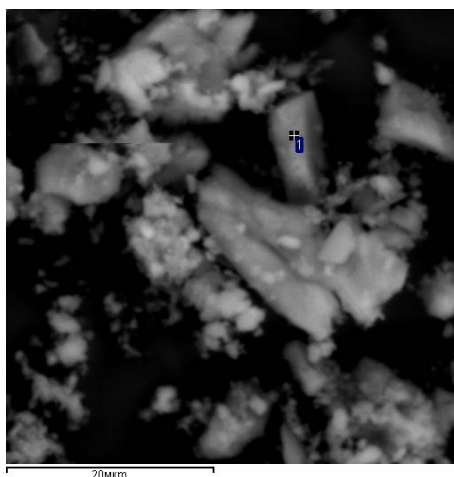




**Fig. 6.** Hydrated Na–Ca–Si compounds in the structure of the material.

**Table 4.** Composition of hydrated Na–Ca–Si compounds (shown in Fig. 6), in wt.%.

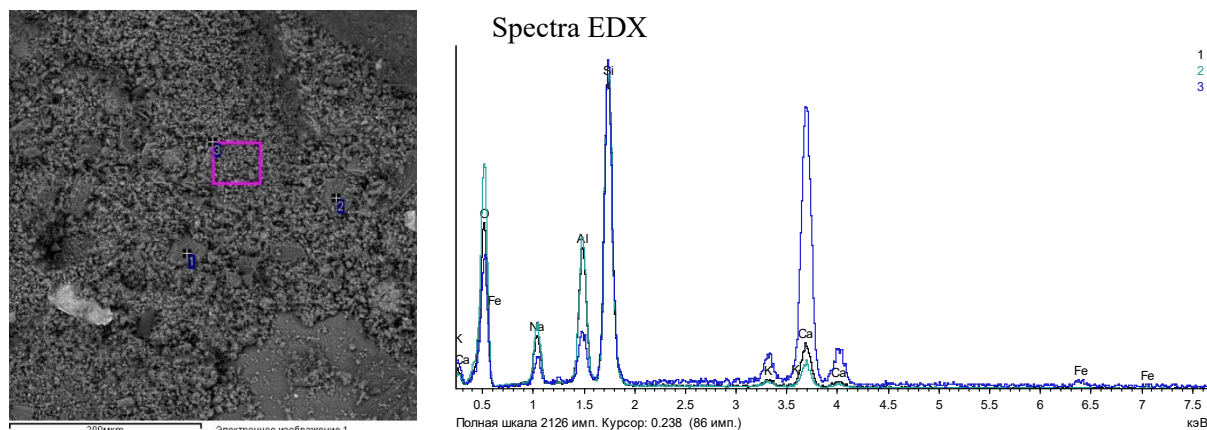
	Na <sub>2</sub> O	MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	K <sub>2</sub> O	CaO	FeO	Total
1	13.49	0.04	1.51	10.98	0.47	13.26	0.64	40.39



**Fig. 7.** Hydrated Na–Ca–Si compounds in the structure of the material.

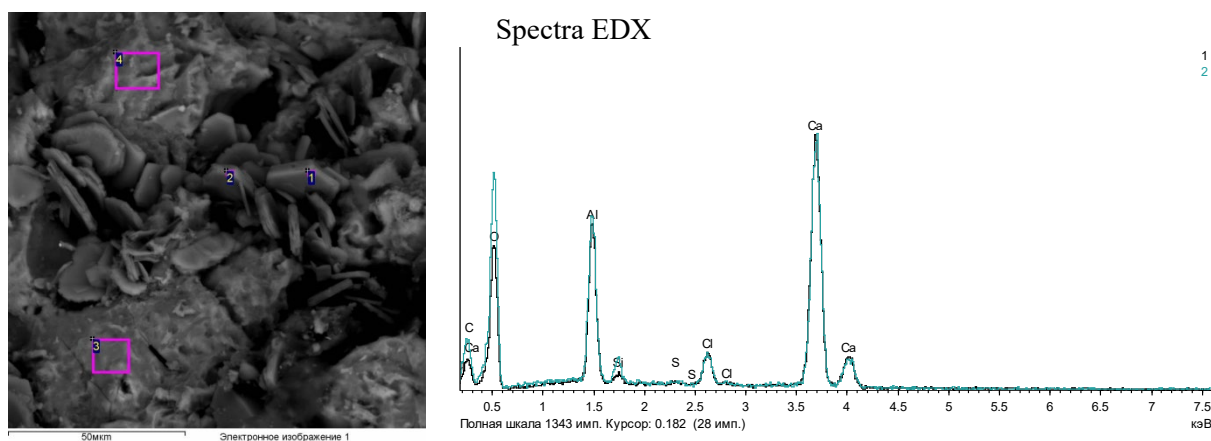
**Table 5.** Example of the composition of the zeolite phase (shown in Figure 7), wt.%.

	Na <sub>2</sub> O	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	K <sub>2</sub> O	CaO	FeO	Summary
1	5.13	17.06	40.92	0.38	6.48	0.11	71.09



**Fig. 8.** EDX spectra of crystals close to zeolites.

Crystals of calcium hydroaluminates are developed in the microcavities (Fig. 9).



**Fig. 9.** Crystals of calcium hydroaluminates (1 and 2).

**Table 6.** The results of the complex analysis of crystals (1 and 2) of calcium hydroaluminate and the host matrix 3 and 4 (Fig. 9), wt.%.

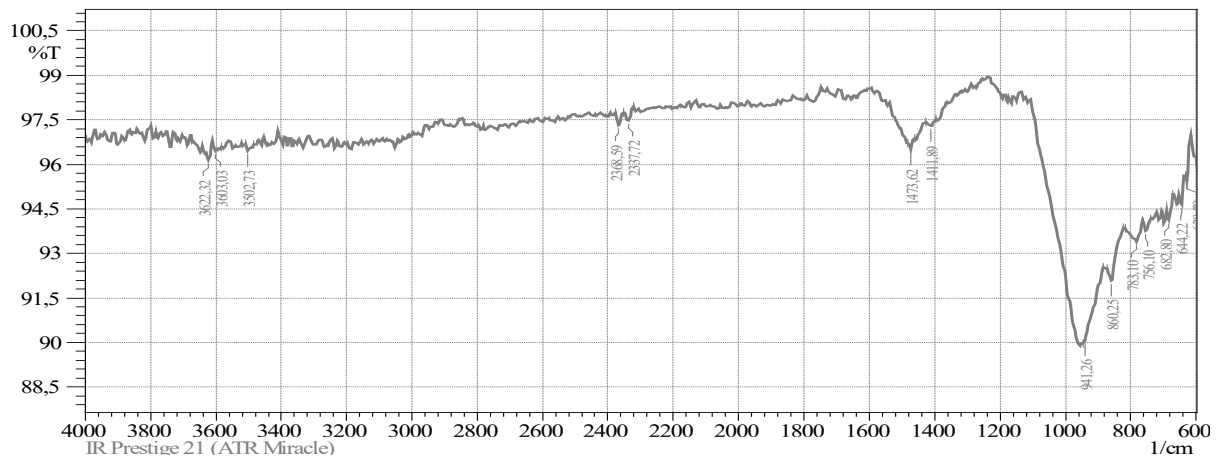
	Na <sub>2</sub> O	MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	SO <sub>3</sub>	K <sub>2</sub> O	CaO	TiO <sub>2</sub>	MnO	FeO	Total
1	-	-	17.82	1.27	0.40	-	34.45	-	-	-	53.94
2	-	-	18.01	3.31	0.48	-	33.76	-	-	-	55.56
3	0.44	1.27	5.23	38.09	0.25	0.12	37.31	0.16	0.06	1.24	84.17
4	0.36	1.31	4.25	33.53	-	0.16	36.85	0.03	0.07	0.54	77.09

On the DTA curve (Fig. 10), the formation of the specified compounds is confirmed by the presence of endoeffects: 80–2000 °C – intense mass loss with heat absorption,  $\Delta m \approx 30\text{--}35\%$ , max – 1300 °C. At temperatures of 220 – 4700 °C, there is a uniform mass loss, at 330 °C, an endoeffect is observed. Starting from 470 °C, there is a more intense mass loss, at 580 – 590 °C there is a small endoeffect. Starting from 740–860 °C – there is a very intense mass loss  $\Delta m \approx 30\%$  with heat absorption, max – 8400 °C.



**Fig. 10.** DTA of hydration products “aspiration cement dust – volcanic tuff –  $\text{Na}_2\text{SiO}_3$  – sodium fluorosilicate.

The readings of the infrared spectrometry (IR) peaks of the cement paste hydration products on the binder “aspiration dust – volcanic tuff –  $\text{Na}_2\text{SiO}_3$ – sodium fluorosilicate” are given in Table 7. In the IR analyses, 870-500  $\text{cm}^{-1}$  are attributed to (Si-O-Al) (Fig. 11).



**Fig. 11.** IR spectra of geopolymer paste hydration products.

**Table 7.** Peaks of IR spectra of cement paste hydration products.

	Peak	Intensity	Corr. intensity	Base(H)	Base(L)	Area	Corr. area
1	628.7	95.426	0.537	632.6	617.2	0.272	0.019
2	644.2	94.656	0.568	651.9	632.6	0.421	0.017
3	682.8	94.136	0.509	690.5	671.2	0.475	0.024
4	756.1	93.768	0.295	759.9	740.6	0.525	0.016
5	783.1	93.387	0.631	817.8	763.8	1.537	0.083
6	860.2	92.093	0.709	871.8	821.6	1.583	0.056
7	941.2	89.981	0.159	945.1	883.4	2.438	0.010
8	1411.8	97.296	0.170	1427.3	1396.4	0.356	0.012

Continuation of Table 7

9	1473.6	96.515	0.269	1481.3	1462.0	0.281	0.009
10	2337.7	97.486	0.353	2353.1	2322.2	0.317	0.024
11	2368.5	97.330	0.405	2376.3	2353.1	0.251	0.021
12	3502.7	96.469	0.222	3510.4	3483.4	0.410	0.015
13	3603.0	96.459	0.272	3610.7	3583.7	0.414	0.017
14	3622.3	96.143	0.461	3637.7	3614.6	0.374	0.025

Comprehensive studies (electron microscopy, X-ray diffraction, microprobe analysis, DTA and IR spectroscopy) revealed the following phases in cement stone [42, 43]:

- Hydrated aluminosilicates of Na and Ca;
- Na/Ca silicates;
- Calcium aluminates;
- Calcite;
- Feldspar (quartz, albite, orthoclase);
- Muscovite.

Submicron crystals are formed by the interaction of calcite with an alkaline hydroaluminosilicate gel. Volcanic additive [44, 45]:

- improves the composition of hydration products in contact areas and pores;
- due to the amorphous  $\text{SiO}_2$ , it slightly reduces the pH, contributing to the formation of quartz and polymerized silicic acid.

Thus, the process combines crystallization and gelation with a noticeable effect of mineral additives.

#### 4. CONCLUSIONS

The results of the studies confirm the effectiveness of the development data, which shows:

1. The components of the binder system “aspiration dust – volcanic tuff” exhibit a certain degree of amorphousness, and when mixed with an alkaline activator, the processes of hydration product interaction and geopolymer transformations are intensified.
2. The most effective formulation of the binding bond «Aspirational cement dust - volcanic tuff -  $\text{Na}_2\text{SiO}_3$  - siliceous sodium» was established, the presence of aluminosilicate with the presence of glass phase promotes the development of own structural formations, matrix binding to structural elements.
3. Comprehensive study of microstructure confirm the presence in of difficult to dissolve and strong joints in geopolymer paste guaranteeing its durability.

The growing environmental and energy challenges of recent years have stimulated the active development of energy-efficient alkali-activated binders in science and industry. These new materials represent an alternative to traditional cements, which require significant energy costs during production. The research shows that the use of alkali-activated binders, such as geopolymers and various equivalents based on mineral additives, can significantly reduce the carbon footprint of the construction sector in the environment

#### 5. ACKNOWLEDGEMENTS

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