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Effect of multicomponent mineral additives on the microstructure and strength of composite cement

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ABSTRACT
In the face of growing environmental and energy challenges, the cement industry is shifting towards the use of composite Portland cements containing hybrid mineral additives to reduce clinker consumption and CO_2 emissions. This study investigates the pozzolanic activity and hydration behavior of thermally activated aluminosilicate additives (TAFM), quartz-feldspar sand, apobasalt-orthoshale (APO), and limestone. The chemical composition and calcium oxide binding capacity of each component were examined using the lime saturation method. Results showed that TAFM exhibits the highest pozzolanic reactivity, significantly binding free lime (CaO), followed by APO and limestone. Composite cement mixtures were formulated according to GOST 31108–2020 standards, incorporating 20% hybrid additives. Mechanical tests revealed that such compositions improve long-term compressive and flexural strength, early setting times, and structural density. In particular, the combination of TAFM, APO, and limestone showed synergistic effects in enhancing hydration kinetics and final performance. The findings support the feasibility of using local mineral resources as effective components in sustainable cement production and highlight the benefits of hybrid additives in reducing clinker demand while improving mechanical and durability characteristics of cementitious composites.

Keywords: Thermally activated additives, clinker reduction, sustainable construction materials, cement hydration, hybrid mineral additives, composite Portland cement.

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Introduction

At present, the global construction materials industry faces the urgent challenge of developing and implementing new-generation materials. Key priorities in this domain include environmental protection, rational use of natural resources, reduction of production costs, and the adoption of energy-efficient technologies — all of which have become central components of modern scientific and technical policy. Among various industries, cement production is recognised as one of the leading contributors to carbon dioxide (CO_2) emissions, making the mitigation of its environmental impact a critical issue.

According to international research, the production of one ton of Portland cement releases on average 0.8 to 1 ton of CO_2 into the atmosphere, which accounts for approximately 7–8% of total global greenhouse gas emissions. As a result, the production and use of composite Portland cements — aimed at reducing CO_2 emissions, minimizing raw material and fuel consumption, and improving energy efficiency — has significantly increased worldwide [[1], [2], [3], [4]].

Composite Portland cements are a new generation of binders that offer notable environmental and economic advantages over conventional Portland cement. These binders incorporate two or more types of active mineral additives, which partially replace clinker in the composition. This not only reduces thermal energy consumption during production but also enhances resource efficiency and ecological safety.

Consequently, the use of composite cements contributes not only to environmental sustainability but also to improving the quality, durability, and long-term performance of construction materials. For this reason, the industrial-scale application of such cements, the careful selection of their mineral additives, and the detailed study of their hydration behavior have become key areas of research in modern cement chemistry.

Composite Portland cements are regulated under GOST 31108–2003, which defines the permissible types and proportions of mineral additives, as well as performance indicators and technical requirements [5]. Based on this regulatory framework, cement compositions may include a variety of active natural or technogenic mineral additives, such as volcanic rocks, pozzolans, industrial by-products, limestone, and sand-stone mixtures.

Numerous scientific sources have noted that such mineral additives actively influence the cement hydration process by accelerating the onset of hydration, modifying the reaction depth, and determining the types of hydration products that are formed [[6], [7], [8], [9], [10]]. As a result, denser phases develop within the microstructure of the cement paste, thereby enhancing its compressive strength, impermeability, frost resistance, and durability under aggressive environmental conditions. In addition, these additives reduce the tendency of concrete mixtures to segregate, lower heat generation, minimize shrinkage-induced deformations, and contribute to the overall structural stability and mechanical strength of the hardened material.

Nevertheless, in practice, many large-scale cement manufacturers remain cautious about implementing such composite products at the industrial level. The primary reason for this hesitance lies in the insufficient understanding of the combined mechanisms of action when multiple active mineral additives are used together. Most scientific studies focus on the individual chemical reactivity or hydraulic behavior of single additives. However, when used in combination, these additives may interact synergistically, antagonistically, or neutrally — and there remains a lack of experimental data supporting these complex interrelations [11].

This scientific uncertainty introduces increased risks related to quality assurance, standardization, and technological consistency. Therefore, before introducing composite cement products into industrial-scale production, it is essential to conduct detailed experimental investigations into the mutual interactions between additives, phase transformations, and the mineralogical structure of the resulting hydration products.

Among the most widely used additives in practice are aluminosilicate-based mineral components, such as natural pozzolans, metakaolin, fly ash from thermal power plants (TPPC), thermally activated clays, and other silicate–aluminate-rich materials. These additives significantly influence the hydration kinetics of Portland cement. The reactive silicon dioxide (SiO_2) and aluminum oxide (Al_2O_3) in their composition engage in secondary pozzolanic reactions with the free calcium hydroxide ($Ca(OH)_2$) released during cement hydration, resulting in the formation of calcium silicate hydrates (CSH) and calcium aluminate silicate hydrates (CASH) [[12], [13], [14], [15], [16], [17]]. These secondary reactions contribute to the development of a dense and durable microstructure in the hardened cement paste, which plays a critical role in long-term performance and durability [18].

This process not only reduces the porosity of the cement stone but also improves its resistance to mechanical stress, water penetration, and freeze–thaw cycles. In addition, such mineral additives enhance the long-term durability of concrete by reducing microcracking and increasing resistance to sulfates and other chemically aggressive agents.

Moreover, recent studies have shown that carbonate-based additives — particularly finely ground and activated forms of limestone, dolomite, and other calcium carbonate materials — can significantly improve several key physical, mechanical, and technological properties of cement-based composites when incorporated into the cement matrix [[19], [20]]. These additives help mitigate segregation and bleeding in concrete mixtures, leading to greater homogeneity and stability within the fresh mix.

Carbonate additives also improve the water-retaining capacity of concrete, thereby enhancing its workability and finishing properties. Furthermore, their inclusion contributes to a reduction in the heat of hydration, which is particularly important for massive concrete structures, where high internal temperatures can lead to thermal cracking. The use of such additives reduces internal temperature differentials and mitigates the risk of thermal stress-induced cracking [[21], [22]].

Another significant benefit is that carbonate-based additives substantially enhance the resistance of concrete to aggressive environments, including water, freezing conditions, acids, and chloride ions commonly found in marine water [[23], [24]]. This makes them especially suitable for the production of concrete used in marine infrastructure, transportation facilities, and chemical processing plants, where long-term chemical durability is critical.

Experimental part

This study focused on evaluating the effectiveness of hybrid mineral additives based on local raw materials in composite Portland cement production. The following components were used in the preparation of the hybrid additives: thermally activated aluminosilicate material (TAFM), quartz-feldspar sand, apobasalt-orthoshale (APO), and limestone. TAFM was obtained by calcining a mixture of 70% tuff and 30% shale at 800–850 °C. APO is a volcanic-origin rock rich in Al_2O_3 and Fe_2O_3, while limestone is a carbonate-based material with a high CaO content (50.28%).

The chemical composition of all additives was determined using standard chemical analysis methods. Their pozzolanic activity was assessed through the lime saturation method developed by Y.M. Butt and V.V. Timashev, which evaluates the capacity of the materials to bind free calcium oxide (CaO) in saturated lime solutions, thus reflecting their reactivity.

Two composite cement formulations were developed in accordance with GOST 31108:2020. The first composition contained 75% Portland cement clinker, 12% limestone, 3% TAFM, 5% APO, and 5% gyPCum. The second composition included 75% clinker, 12% limestone, 3% TAFM, and 5% gyPCum. As a control, a standard cement (PÇ-D0) consisting of 95% clinker and 5% gyPCum was prepared. All cement mixtures were ground in a laboratory ball mill until achieving a fineness of 90–94% passing through a 008 sieve.

Testing involved determining the normal consistency (water demand) and setting times in accordance with GOST 310.3. The flexural and compressive strength of the cement mortars, prepared at a cement-to-sand ratio of 1:3, was measured using prismatic samples of 4×4×16 cm after 3 and 28 days of curing, following GOST 310.4 procedures. Additionally, to assess pozzolanic activity, the cement pastes were stored in saturated lime solutions for 30 days, after which the concentration of CaO in the liquid phase was measured using titration techniques.

Results and Discussion

In this study, hybrid additives based on locally available mineral raw materials were selected with the aim of enhancing the composition of composite Portland cement and improving its physicomechanical, hydration, and microstructural characteristics. As a primary hybrid additive, a thermally activated aluminosilicate component was used, obtained by calcining a mixture of 70% natural tuffaceous rock (tuffite) and 30% shale at 800 °C. The thermal activation process significantly enhanced the reactivity of the material, increasing its pozzolanic activity. This combination exhibits hydraulic properties, reacting with free calcium hydroxide during hydration and forming secondary calcium silicate hydrates (CSH) and calcium aluminate silicate hydrates (CASH).

In addition, quartz-feldspar sand, consisting primarily of SiO_2 and alkali feldspars (K/Na–feldspar), was introduced into the composition. This component contributes to physicochemical balance in the cement mixture, improves particle size distribution, and functions as a microfiller by reducing void phases and enhancing the density of the hardened cement matrix.

The third component utilized was apobasalt–orthoshale rock, characterized by a basaltic structure rich in iron- and magnesium-bearing silicates. These provide high mechanical strength, chemical stability, and thermal resistance. The wide range of mineral phases present in this rock type contributes to stable hydration and the formation of strong structural compounds within the cement matrix.

The synergistic effect of these three components was investigated in detail. Special emphasis was placed on their influence on Portland cement hydration behavior, including heat evolution, setting time, microstructural changes, and compressive strength. In addition, comparative measurements of compressive strength, water permeability, density, and porosity were performed on the hardened cement samples.

To evaluate the effectiveness of the hybrid additives, their chemical composition and the relative proportions of oxide components were studied. These parameters are critical in determining the hydraulic or pozzolanic activity of the additives when blended with cement, as well as their reactivity during hydration and their influence on the microstructure and mechanical properties of the final product.

Particular attention was paid to the quantitative ratios of key oxides such as SiO_2, Al_2O_3, Fe_2O_3, CaO, and MgO. These indicators allowed for the classification of each additive's geochemical group, its natural or artificial origin, and its expected reactivity within the cementitious system. The chemical composition of the components used in this study — including thermally activated aluminosilicate (TAFM), apobasalt–orthoshale (APO), quartz–feldspar sand, and limestone — is presented below (Table 1).

According to the analysis results, the quartz–feldspar sand sample (abbreviated as QFS) primarily consists of silicon dioxide (SiO_2), which accounts for 88.72% of its composition. This high SiO_2 content classifies the material as a quartz–feldspathic rock and highlights its suitability as a microfiller that contributes to the densification of the cementitious matrix. The low content of Al_2O_3, Fe_2O_3, and CaO indicates that the material is chemically inert and does not significantly participate in hydration reactions, confirming its role as a non-reactive mineral filler.

The apobasalt–orthoshale (APO) sample, in contrast, contains substantial amounts of SiO_2 (46.61%), Al_2O_3 (14.91%), and Fe_2O_3 (8.20%), which are characteristic of dense volcanic extrusive rocks belonging to the andesite–basalt group. The high content of these oxides indicates potential pozzolanic or latent hydraulic activity. Furthermore, the presence of CaO and MgO suggests partial reactivity, enabling the formation of secondary compounds during the hydration process, which can enhance the mechanical strength and durability of cement composites.

The thermally activated mineral additive (TAFM) is composed of a mixture of tuffite and shale calcined at 800–850 °C, and it contains SiO_2 (51.45%) and Al_2O_3 (8.62%). These values confirm its classification as an aluminosilicate pozzolanic additive. Additionally, the content of CaO (12.00%) and MgO (2.20%) indicates that the material exhibits both pozzolanic and hydraulic activity, enabling it to react with free calcium hydroxide to form calcium silicate hydrates (CSH) and calcium aluminate silicate hydrates (CASH), which contribute to improved microstructure and mechanical performance.

The limestone sample is distinguished by its very high CaO content (50.28%), which makes it an effective carbonate-based reactive component for incorporation into cement. The loss on ignition (LOI) value of 39.61% reflects the release of CO_2 during decomposition of carbonates. During cement hydration, limestone promotes the formation of calcium carbonate hydrates and calcium aluminate carbonate (C–A–C) compounds. These products enhance the density, impermeability, and resistance to aggressive environments of the hardened cement paste.

Table 1 - Results of chemical analysis of the additives

Type of Additive	LOI (%)	SiO_2 (%)	Al_2O_3 (%)	Fe_2O_3 (%)	CaO (%)	MgO (%)	SO_3 (%)	Others (%)
Quartz–feldspar sand	1.51	88.72	1.60	1.99	0.83	0.90	0.44	4.01
Apobasalt–orthoshale (APO)	9.47	46.61	14.91	8.20	8.76	3.79	0.30	5.80
TAFM (Thermally activated)	16.14	51.45	8.62	2.39	12.00	2.20	0.58	6.62
Limestone ($CaCO_3$)	39.61	4.99	1.33	0.47	50.28	2.72	0.20	0.40

Note: LOI – Loss on Ignition.

To determine the pozzolanic or hydraulic activity of the additives, special tests were conducted to evaluate their capacity to bind free calcium oxide (CaO) during the hydration process. The pozzolanic and hydraulic reactivity of the selected additives — TAFM, quartz–feldspar sand (QFS), apobasalt–orthoshale (APO), and limestone — was assessed based on their ability to fix CaO released during cement hydration. The testing methodology was based on the determination of the lime-binding capacity of mineral additives within the cement system, which serves as a criterion for assessing their potential reactivity.

The experimental procedure was carried out in the following stages:

- Representative samples of each additive (TAFM, QFS, APO, and limestone) were ground to a particle size of ≤1 cm and blended with Portland clinker and gyPCum in a laboratory ball mill.
- The cement mixture for each sample contained: 700 g of clinker, 300 g of additive, and 30 g of gyPCum.
- The resulting cement compositions were ground to a fineness corresponding to 90–94% passing through a No. 008 sieve, achieving standard cement particle size.

To evaluate pozzolanic activity, the saturation level of free CaO in the liquid phase was determined after hydration. This analysis was conducted according to the methodology developed by Yu.M. Butt and V.V. Timashev, in which cement pastes were hydrated for 30 days, followed by titrimetric analysis of the alkalinity (meq/L) and free CaO concentration (mmol/L) in the liquid phase.

The results showed that the TAFM-containing cement sample absorbed 287.14 mg of CaO from the saturated lime solution after 15 titration cycles, indicating a high level of pozzolanic activity. At the end of the test period, the TAFM cement sample exhibited the following values in the liquid phase:

- Free CaO concentration: 3.86 mmol/L
- Total alkalinity: 58.00 meq/L

These results confirm the high reactivity of TAFM, attributable to its rich content of reactive aluminosilicate components, and its active participation in pozzolanic reactions during hydration.

In contrast, the cement sample containing limestone exhibited a notable increase in the alkalinity of the liquid phase. While limestone is only weakly reactive, it affects the ionic balance of the solution, thereby exerting an indirect influence on the formation of hydration products (Table 2).

As shown in Table 2, the thermally activated mineral additive (TAFM) demonstrated the highest lime-binding capacity among the tested additives. The concentration of CaO in the liquid phase was measured at 3.86 mmol/L, while the total alkalinity reached 58.00 meq/L. These values confirm the high pozzolanic reactivity of TAFM, indicating its strong ability to actively react with free calcium hydroxide. This behavior reflects the material's high hydration reactivity and justifies its classification as a highly active pozzolanic additive.

On the other hand, the quartz–feldspar sand (QFS) sample exhibited the highest residual CaO concentration in the liquid phase (8.39 mmol/L) and the lowest amount of bound CaO (94.2 mg). These results indicate that QFS has very low pozzolanic or hydraulic reactivity and primarily behaves as an inert microfiller within the cement system. According to the standard GOST 24640–91 "Additives for Cement. Classification", such low-reactivity additives are typically used to economize clinker consumption without significantly contributing to hydration reactions.

Table 2 - Results of chemical analysis of the additives

Active Mineral Additive	Origin of Additive	CaO Content (mmol/L)	Total Alkalinity (meq/L)
TAFM (thermally activated)	Technogenic (artificial)	3.86	58.00
QFS (quartz–feldspar sand)	Natural (sedimentary)	8.39	56.80
APO (apobasalt–orthoshale)	Natural (volcanic)	6.80	57.20
Limestone	Natural (carbonate-based)	3.00	68.00

Note: The table presents the concentration of free calcium oxide (CaO) and total alkalinity (in meq/L) in the liquid phase in contact with cement pastes containing different additives.

The apobasalt–orthoshale (APO) additive showed intermediate reactivity, with a CaO concentration of 6.80 mmol/L and alkalinity of 57.20 meq/L, suggesting limited pozzolanic activity. Due to its content of reactive oxides (SiO_2, Al_2O_3, Fe_2O_3), APO can still positively influence the density and mechanical strength of cement composites and may be classified as a partially reactive or borderline pozzolanic additive.

Although the limestone sample showed the lowest CaO concentration in the liquid phase (3.00 mmol/L), this should not be interpreted as an indication of high pozzolanic activity. Instead, this result reflects the carbonate nature of limestone, which indirectly influences the alkalinity balance within the hydration environment. The high alkalinity value of 68.00 meq/L suggests that limestone contributes to ionic equilibrium, supporting the stable formation of hydration products. Rather than acting as a reactive catalyst, limestone typically functions as a stabilizing additive, improving final properties such as density and durability of the hardened cement paste.

Based on the results in Table 2, the concentration of CaO and total alkalinity for each cement sample containing different additives were analyzed. These data were used to construct comparative graphical plots, visually depicting the relative reactivity of each additive within the hydration environment. The plot allows for the assessment of the additives' alignment with pozzolanic or hydraulic activity criteria (Figure 1). The positions of points 1 (QFS), 2 (APO), 3 (TAFM), and 4 (Limestone) on the graph with respect to the lime solubility isotherm (curve A) were used to evaluate the qualitative performance of each additive. The concentration of free calcium oxide (CaO) in the liquid phase in direct contact with the cement paste serves as a key indicator of the additive's reactivity—i.e., its pozzolanic or hydraulic activity.

A point located below the isotherm indicates that the additive has a strong capacity to bind CaO from the liquid phase, which implies high reactivity. Conversely, a point on or above the isotherm reflects limited or negligible chemical interaction with CaO, indicating low reactivity.

The analysis of the plotted data reveals that all tested additives demonstrated some degree of CaO absorption during the hydration and hardening processes, but their levels of reactivity varied significantly:

- Point 1 (TAFM) is positioned well below the isotherm, indicating the highest pozzolanic and hydraulic activity among all additives. As a thermally activated artificial material, TAFM actively reacts with free lime in the hydration environment and forms strong secondary hydrates.

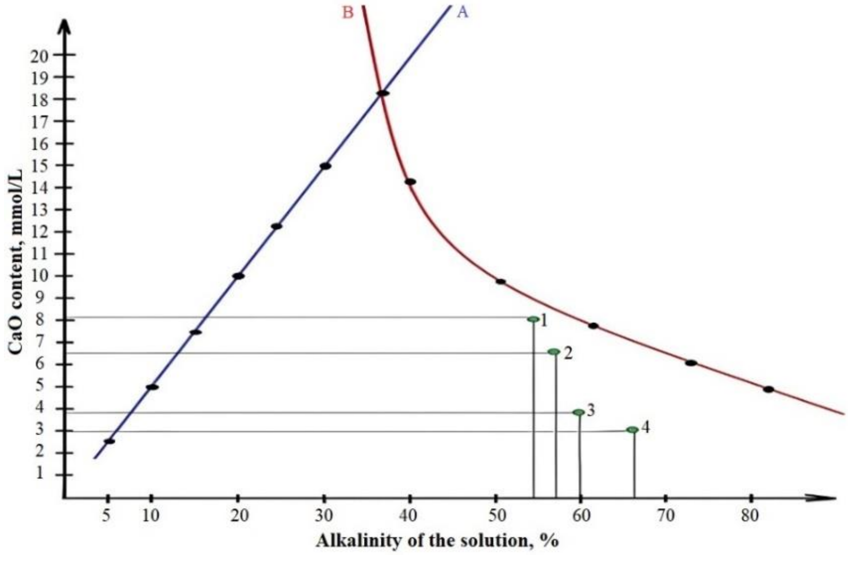

Figure 1 - Pozzolanic activity of mineral additives

A – Solubility isotherm of lime at 40 °C
B – Total alkalinity of the solution (excluding the contribution of CaO)
1 – Cement sample with QFS (quartz–feldspar sand)
2 – Cement sample with APO (apobasalt–orthoshale)
3 – Cement sample with TAFM (thermally activated mineral additive)
4 – Cement sample with limestone

- Point 2 (QFS) lies very close to the isotherm, suggesting that this additive exhibits very low pozzolanic reactivity. Quartz–feldspar sand behaves primarily as an inert filler, contributing little to CaO consumption.
- Point 3 (APO) is located between TAFM and QFS, signifying that it possesses moderate pozzolanic activity. APO may be classified as a semi-reactive or borderline pozzolanic material, due to its intermediate CaO-binding ability.
- Point 4 (Limestone) appears at a low CaO concentration but should not be interpreted as a sign of high pozzolanic activity. Instead, this is attributed to the carbonate nature of limestone, which influences the chemical balance of the hydration medium indirectly, rather than through direct lime binding.

Thus, the positioning of the data points relative to the solubility isotherm in Figure 1 provides a clear visual assessment of the pozzolanic or hydraulic activity of the studied additives. TAFM clearly stands out as the most effective and reactive component, whereas QFS (quartz–feldspar sand) demonstrates the lowest reactivity and can be classified as an inert mineral filler.

Based on the conducted research, the following conclusions were drawn:

- According to the Student’s criterion for evaluating hydraulic activity, the tested raw materials do not meet the requirements specified in UzDSt 901:1998. Therefore, they cannot be used independently as active mineral additives for cement production.
- However, all studied mineral additives — TAFM, APO, limestone, and QFS — demonstrated a certain level of pozzolanic activity, which supports their potential application as reactive-filler components in the production of various types of Portland cement. These additives can bind free calcium oxide (CaO) released during cement hydration and setting.
- The highest lime-binding capacity was shown by TAFM, a thermally activated technogenic material. In contrast, QFS exhibited very low hydraulic activity, while the pozzolanic reactivity of APO fell between TAFM and QFS, indicating intermediate behavior.
- Based on the amount of CaO absorbed from the solution in which the cement samples were immersed, the additives may be ranked in the following descending order of pozzolanic activity:

TAFM → APO → QFS → Limestone

- To enhance the pozzolanic and hydraulic performance of these additives, it is recommended to formulate composite additives by combining them with various modifiers. This approach aims to improve their reactivity potential and contribute more significantly to the strength development of the cement matrix.

To produce composite Portland cements, raw material mixtures (clinker blends) were formulated in accordance with the requirements of GOST 31108:2020. The prepared mixtures were jointly ground in a laboratory ball mill to obtain cement samples for further testing.

Table 3 - Cement composition and its effect on setting time

No.	Type of Cement	Grinding Time, min	Residue on Sieve No. 008, %	Standard Water Demand (W/N), %	Initial Setting Time (h: min)	Final Setting Time (h: min)
1	PC-D0	30	10.0	25.7	3 h 20 min	5 h 00 min
2	PC-KD20	20	6.0	27.0	2 h 40 min	5 h 00 min

Notes:

- PC-D0 – pure Portland cement without additives (control sample).
- PC-KD20 – Portland cement with 20% composite additive (limestone + TAFM + APO or QFS).
- The reduction in grinding time for PC-KD20 is attributed to the softer structure and higher grindability of the ceramic components.
- Standard Water Demand (W/N) indicates the percentage of water required for normal consistency according to GOST methods.

The incorporation of ceramic-based additives into the cement composition has a significant impact on its physical-mechanical and technological properties. In particular, the PC-KD20 cement composition, which includes 20% composite additives (a mixture of limestone + TAFM + apobasalt–orthoshale or quartz–feldspar sand), exhibits a shorter grinding duration—only 20 minutes—compared to the reference pure Portland cement (PC-D0).

This behavior is attributed to the relatively softer texture and higher grindability of the ceramic additives, which facilitate faster milling to achieve the target fineness (Table 3).

In the PC-KD20 cement sample, the fineness is significantly higher, as evidenced by the fact that only 6.0% residue remains on sieve No. 008. This indicates a greater degree of particle dispersion and, consequently, higher reactivity of the cement. However, such fineness also leads to an increased water demand during the hydration process. As a result, the required standard water-to-cement ratio (W/N) for PC-KD20 is 27.0%, which is slightly higher than that of pure Portland cement (PC-D0), which stands at 25.7%.

Analysis of the setting times reveals that PC-KD20 cement initiates setting earlier (2 h 40 min) compared to the control PC-D0 (3 h 20 min). This is attributed to the increased surface area and higher number of reactive sites, which accelerate the hydration kinetics. However, the final setting time for both cement types remains the same at 5 hours (Figure 2). This suggests that the composite additives included in the cement matrix primarily enhance early-stage activity, without significantly altering the overall setting duration.

Thus, the incorporation of ceramic waste into the cement composition alters its physicochemical and rheological properties, which play an important role in optimizing construction material formulations.

The normal consistency (W/N) of the composite Portland cement (CPC) was slightly higher than that of the control (reference) cement. This increase is attributed to the enhanced water demand caused by the plasticizing effect of limestone and TAFM (thermally activated mineral additive). These components increase the mix's need for water due to their surface activity and specific particle structure.

The initial setting time of the experimental cement occurred significantly earlier compared to the control cement. This phenomenon is likely due to the rapid binding of free calcium hydroxide ($Ca(OH)_2$) released during hydration by the composite additives, which accelerates the formation of hydration products.

As a reference sample, plain Portland cement with 95% clinker and 5% gypsum was used, containing no mineral additives. The compressive strength of the composite Portland cement (CPC) samples was evaluated and compared with the reference sample (PC-D0) following the methodology outlined in GOST 310.4. Test specimens were prepared from a 1:3 cement-to-sand mixture, molded into 4×4×16 cm prisms, and cast using Volsk construction sand as the standard fine aggregate (Table 4).

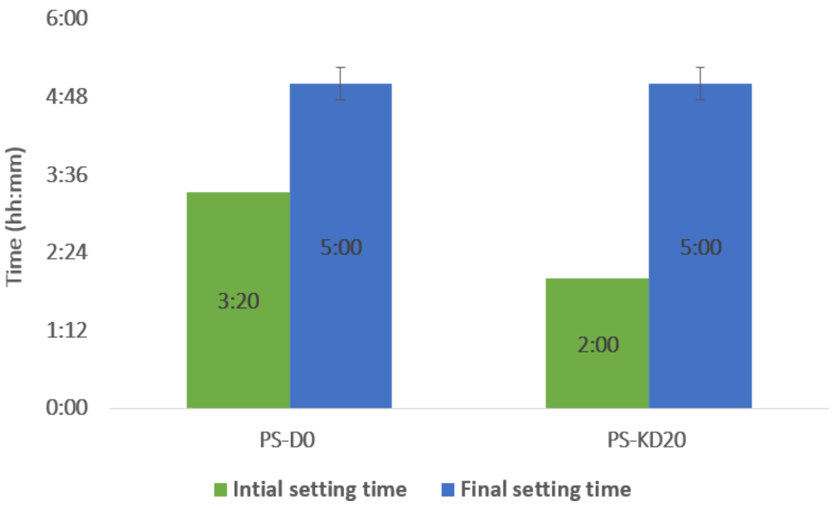

Figure 2 - Comparison of cement setting times

Table 4 - Effect of composite additives on flexural and compressive strength of portland cement (MPa)

№	Type of Cement	Flexural / Compressive Strength after 3 Days (MPa)	Flexural / Compressive Strength after 28 Days (MPa)
1	PC-D0	5.60 / 21.40	6.20 / 40.20
2	PC-KD20	4.65 / 21.40	7.25 / 41.00

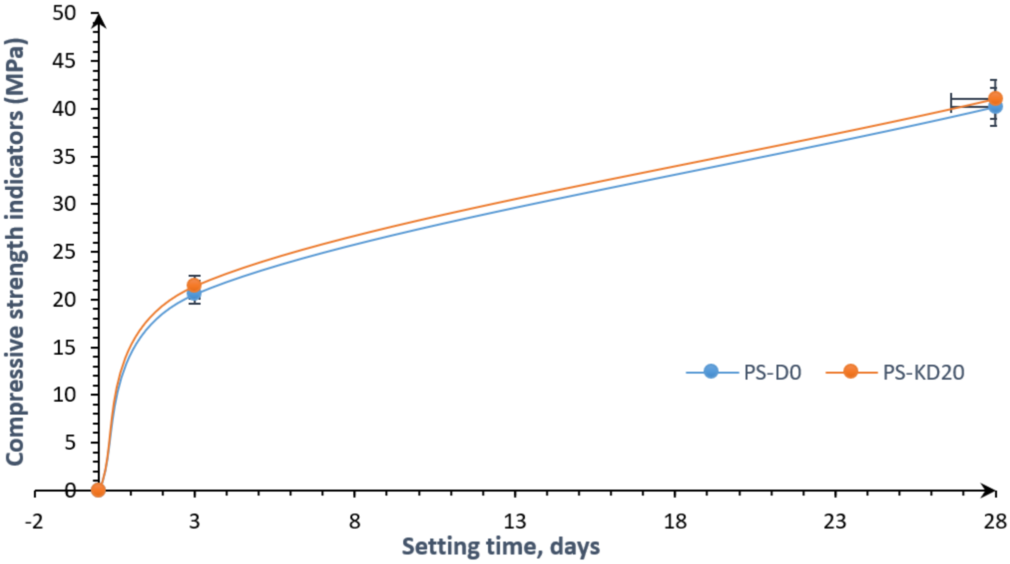

Figure 3 - Effect of KGCh-Based Composite Additive on Compressive Strength of Portland Cement (MPa)

The mechanical performance of Portland cement is significantly influenced by the type, quantity, and reactivity of mineral additives incorporated into its formulation. In the present study, a composite additive system comprising limestone, thermally activated mineral additive (TAFM), and either apobasalt-orthoshale (APO) or quartz–feldspathic sand (QFS) was introduced to partially replace clinker in Portland cement, aiming to improve sustainability without compromising performance.

The early-age flexural strength (3 days) of the composite cement (PC-KD20) was measured at 4.65 MPa, which is lower than that of the control cement (PC-D0) — 5.60 MPa. This reduction is attributable to the delayed reactivity of certain mineral phases within the composite additives. Specifically, materials like feldspathic sand and orthoshale may require a longer induction period to develop their pozzolanic or hydraulic potential. Moreover, the lower early-age strength may result from dilution of clinker content, which directly contributes to early hydration and strength gain.

In contrast, compressive strength at 3 days remained unchanged (21.4 MPa) across both cement types. This parity suggests that the filler effect and microstructural densification provided by the composite additives may partially compensate for the reduction in reactive clinker. These additives may act as nucleation sites, accelerating the formation of hydration products such as calcium silicate hydrate (C–S–H), thereby maintaining compressive performance.

By 28 days, the composite cement not only recovered but surpassed the control sample in flexural strength (7.25 MPa vs. 6.20 MPa) and slightly exceeded it in compressive strength (41.0 MPa vs. 40.2 MPa). This confirms the long-term pozzolanic contribution of TAFM and APO, which, over time, react with free calcium hydroxide to form additional C–S–H and calcium aluminosilicate hydrates (C–A–S–H). The synergy between limestone and active silicate/aluminate phases may also contribute to the refinement of pore structure and enhancement of interfacial transition zones (ITZs) within the cement matrix.

Furthermore, the observed strength development indicates that the composite additives do not negatively impact the long-term performance of cement but instead facilitate gradual densification and strength improvement, making them viable for sustainable cement design. These results align with previous studies demonstrating that carefully optimized hybrid mineral additives can simultaneously support the mechanical integrity and ecological performance of composite Portland cements.

The 28-day strength results clearly illustrate the long-term effectiveness of the composite additives. Specifically, the flexural strength of PC-KD20 cement reached 7.25 MPa, significantly higher than the 6.2 MPa observed for the control sample (PC-D0). This improvement is attributed to the progressive hydration activity of the mineral additives, which enhance the formation of strength-contributing hydrates, densify the cement matrix, and strengthen internal microstructural bonds over time.

In terms of compressive strength, PC-KD20 cement achieved 41.0 MPa, slightly exceeding the control sample's 40.2 MPa. This marginal gain further indicates that the composite additives contribute to refined microstructure, reduced microcracking, and improved compactness of the hardened cement paste.

Overall, the combination of limestone, TAFM, and either apobasalt-orthoshale or quartz–feldspathic sand demonstrates a substantial pozzolanic contribution, reacting with $Ca(OH)_2$ to form additional C–S–H and C–A–S–H gel phases that improve durability and mechanical performance. Particularly, TAFM and limestone play crucial roles due to their plasticizing effect and contribution to structural stability, respectively.

Therefore, these composite additives not only enhance cement quality but also contribute to environmental sustainability by incorporating industrial by-products. Their application in Portland cement production represents a technologically sound, mechanically efficient, and ecologically viable solution for the future of sustainable construction materials.

Conclusions

Based on the results of chemical analysis and pozzolanic activity evaluation, it was found that all tested mineral additives (TAFM, APO, and limestone) possess varying degrees of pozzolanic and hydraulic activity. Among them, the thermally activated aluminosilicate material (TAFM) demonstrated the highest reactivity by effectively binding free CaO in the hydrated cement matrix.

The experimental cement compositions prepared according to GOST 31108:2020, which included hybrid additives, showed improved physical and mechanical performance compared to the control cement. Notably, cement mixtures containing TAFM, APO, and limestone exhibited accelerated setting times, increased long-term strength, and better structural compactness.

The use of limestone as a partial clinker substitute is viable from an economic and ecological perspective, although its pozzolanic effect is limited. However, when combined with more active additives like TAFM, synergistic behavior was observed, enhancing the performance of the overall cementitious system.

The research confirms the potential of utilizing locally available mineral materials in hybrid form to develop environmentally sustainable and technically effective composite portland cements.

Conflicts of interest. On behalf of all authors, the corresponding author states that there is no conflict of interest.

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Көп компонентті минералды қоспалардың композициялық цементтің микроқұрылымы мен беріктігіне әсері

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ТҮЙІНДЕМЕ

Қоршаған орта мен энергияға қатысты артып келе жатқан қиындықтар жағдайында цемент өнеркәсібі клинкерді тұтынуды және CO_2 шығарындыларын азайту мақсатында гибридті минералдық қоспалар қосылған композициялық портланд цементтерін пайдалануға бет бұруда. Бұл зерттеуде термиялық белсендірілген алюмосиликатты қоспалар (ТБАҚ), кварц-далалық шпаттық құм, апобазальт-ортоалевролит (АПО) және әктас сияқты компоненттердің пуццоландық белсенділігі мен гидратациялық әрекеті зерттелді. Әр компоненттің химиялық құрамы мен кальций оксидімен байланыстыру қабілеті әкпен қанықтыру әдісімен анықталды. Нәтижелер ТБАҚ ең жоғары пуццоландық реакциялық белсенділікке ие және бос әктасты (CaO) айтарлықтай байланыстыратынын көрсетті, одан кейін APO және әктас. Композициялық цемент қоспалары GOST 31108–2020 стандартына сәйкес, 20% гибридтік қоспаларды қосу арқылы дайындалды. Механикалық сынақтар мұндай қоспалар қысымға және иілуге беріктілікті арттыратынын, ерте қату уақытын қысқартатынын және құрылымдық тығыздықты жоғарылататынын көрсетті. Әсіресе ТБАҚ, APO және әктас комбинациясы гидратация кинетикасын жеделдетуде және соңғы беріктік сипаттамаларын жақсартуда синергиялық әсер етеді. Бұл нәтижелер жергілікті минералдық ресурстарды орнықты цемент өндірісінде тиімді компонент ретінде қолданудың мүмкіндігін растайды және гибридтік қоспалардың клинкер тұтынуын азайта отырып, цементті композициялардың механикалық және ұзақ мерзімді қасиеттерін жақсартудағы артықшылықтарын айқындайды.

Түйін сөздер: Термиялық белсендірілген қоспалар, клинкер мөлшерін азайту, орнықты құрылыс материалдары, цементтің гидратациясы, гибридті минералдық қоспалар, композициялық портландцемент.

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Влияние многокомпонентных минеральных добавок на микроструктуру и прочность композиционного цемента

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АННОТАЦИЯ

В условиях растущих экологических и энергетических вызовов цементная промышленность переходит к использованию композиционных портланд цементов, содержащих гибридные минеральные добавки, с целью сокращения потребления клинкера и выбросов CO_2. В данном исследовании изучена пуццолановая активность и гидратационное поведение термически активированных алюмосиликатных добавок (ТААД), кварц-полевошпатового песка, апобазальт-ортоалевролита (АПО) и известняка. Химический состав и способность связывать оксид кальция каждого компонента были определены методом известковой насыщенности. Результаты показали, что ТААД обладает наивысшей пуццолановой реакционной способностью и значительно связывает свободную известь (CaO), за ним следуют АПО и известняк. Композиционные цементные смеси были сформулированы в соответствии с требованиями стандарта ГОСТ 31108–2020 с добавлением 20% гибридных минеральных добавок. Механические испытания показали, что такие составы улучшают прочность на сжатие и изгиб в долгосрочной перспективе, сокращают ранние сроки схватывания и повышают структурную плотность. Особенно комбинация ТААД, АПО и известняка продемонстрировала синергетический эффект, способствуя интенсификации гидратации и улучшению конечных характеристик. Полученные данные подтверждают целесообразность использования местных минеральных ресурсов в качестве эффективных компонентов для устойчивого производства цемента и подчёркивают преимущества гибридных добавок в снижении потребления клинкера при одновременном улучшении механических и долговечных свойств цементных композитов.

Ключевые слова: Термически активированные добавки, снижение содержания клинкера, экологически устойчивые строительные материалы, гидратация цемента, гибридные минеральные добавки, композиционный портландцемент.

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