Influence of Modified Expanded Vermiculite on Adhesion to Cement Matrix in Thermal Insulation Composites

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**Abstract.** The article presents the results of a comprehensive experimental study aimed at increasing the adhesion strength between swollen vermiculite and cement matrix in lightweight thermal insulation composites. The influence of various methods of surface modification of vermiculite - alkaline activation, treatment with liquid glass, and silane coating - on the interface morphology and mechanical characteristics of the contact zone was studied. The adhesion strength was assessed by separating the grain from the cement matrix, and the microstructure was analyzed using scanning electron microscopy (SEM). The best results were obtained with a comprehensive modification combining all three approaches, which allowed for an increase in adhesion strength by 45% compared to the unmodified sample. The statistical significance of the differences was confirmed by analysis of variance (ANOVA, p < 0.05). The obtained data demonstrate the high effectiveness of the proposed approach and can be applied in the design of energy-efficient building materials based on cement composites.

**Keywords:** expanded vermiculite; cement matrix; adhesion; surface modification; alkaline activation; liquid glass; silane; SEM analysis; thermal insulation composites; contact zone; C-S-H hydrates; bond strength

# INTRODUCTION

Effective thermal insulation building materials play a crucial role in ensuring the energy efficiency of buildings, especially in climatic regions with pronounced seasonal temperature fluctuations. One of the promising lightweight aggregates is expanded vermiculite - an aluminosilicate mineral with a layered structure, characterized by low density   
(70-120 kg/m3), high heat resistance (up to 1200 °C), fire resistance, environmental safety, and resistance to biocorrosion [1, 3].

Due to its combination of physicochemical properties, vermiculite is actively used in lightweight concrete, plaster, and thermal insulation mixtures. However, its widespread application in cement composites is hindered by its low adhesion strength to the cement matrix, which is caused by several factors:

* low surface activity;
* layered structure with limited wettability;
* presence of closed and open pores on the grain surface;
* low reactivity in an alkaline environment [2, 10].

These characteristics lead to the formation of a defective contact zone, which is a porous layer between the vermiculite grain and the cement stone. Such a zone lacks full-fledged hydrate phases, possesses capillary permeability, and becomes the initiator of microcrack formation, ultimately reducing the durability and strength of the finished product [3].

To overcome these problems, methods for surface modification of vermiculite aimed at improving its adhesion properties have been actively researched in recent years. Among them, the following have been recognized as the most effective:

• alkaline activation using NaOH solutions, promoting the formation of reactive hydroxyl groups [2];

• treatment with liquid glass (Na2SiO3), forming silicate films that stabilize the contact zone [7];

• hydrophobization using silane compounds, which provides control over wettability and facilitates the formation of Si-O-Ca chemical bonds [1].

An additional relevant direction is comprehensive modification, which combines the mentioned methods in a single technological sequence.

Despite the familiarity of individual approaches, systematic comparison of various types of modification and their impact on microstructure and adhesion strength in the context of cement composites has been conducted to a limited extent. In particular, there is insufficient data on interface morphology, chemical structure of the contact zone, and statistical reliability of the observed effects.

The aim of this study is to experimentally evaluate the influence of various methods of modifying the surface of expanded vermiculite on its adhesion to the cement matrix. Special attention is paid to:

• determining the adhesion strength using the pull-off method;

• morphological analysis of the interface using SEM;

• studying the influence of the modified layer structure on the formation of hydrate phases;

• statistical confirmation of the effectiveness of each approach.

It is expected that the comprehensive modification will ensure the formation of a dense reactive zone and lead to a significant increase in adhesion strength, creating prerequisites for the development of new energy-efficient and durable construction materials.

# MATERIALS AND METHODS

**Source components**

Portland cement of grade PC 500-D0, complying with the requirements of GOST 31108-2020 [12], was used as the binding component. It is characterized by an increased content of clinker minerals and high hydration activity. The main aggregate is expanded vermiculite with a fraction size of 1-2 mm, obtained by thermal treatment of natural mineral at   
950-1100 °C. Structurally, vermiculite is a magnesium aluminosilicate with a layered crystal lattice capable of ion exchange and intercalation [9].

The initial density of the expanded vermiculite was 110-130 kg/m3, water absorption was approximately 300%, and the porosity modulus was > 70%.

**Modification methods**

The following methods of surface modification of vermiculite were employed in the study:

Alkaline activation (NaOH).

Treatment with a 1% sodium hydroxide solution at 60 °C for 60 minutes. The purpose of the treatment is to break down surface layers, remove impurities, and form reactive Si-OH groups [2].

Silicate coating (liquid glass).

Immersion in a 10% sodium silicate (Na2SiO3) solution for 30 minutes, followed by drying at 100 °C. The formed film reduces capillary water loss and increases structural stability [7].

Hydrophilization (silane GKZh-11).

Treatment with a 5% aqueous emulsion of a silane agent at room temperature (contact time - 15 minutes), followed by drying at 80 °C. This modification reduces surface energy and improves interfacial compatibility [1, 2].

**Complex modification**

Sequential application of the three aforementioned methods: NaOH → liquid glass → GKZh-11. This scheme allows for obtaining a structure on the grain surface that promotes the growth of cement hydrates and the formation of a strong contact zone.

**Sample preparation**

Cement paste (W/C = 0.35) was mixed in laboratory conditions and poured into molds with a diameter of 25 mm and a height of 40 mm. At the fresh stage, one modified vermiculite grain was immersed vertically in the paste to a depth of 5 mm. The samples were kept in a humid chamber for 28 days at a temperature of (20 ± 2) °C and a humidity of at least 95%.

**Methodology for determining clutch strength**

Adhesion strength was evaluated using the grain separation method from the cement matrix, similar to GOST 5781.2-2022 [13] and ISO 4624. The tests were conducted on a universal tensile testing machine with a loading rate of 0.5 mm/min. The maximum separation load was recorded and converted to stress using the formula:

(1)

where: F - force of detachment (N); A - grain contact area (mm2).

Each type of modification was tested in three replications, after which the mean values and standard deviations were calculated.

**Microstructural analysis**

The morphology of the contact zone between vermiculite and the cement matrix was investigated using scanning electron microscopy (SEM) with a TESCAN VEGA 3 microscope. Prior to analysis, samples underwent vacuum metallization. Concurrently, local elemental analysis (EDS) and Vickers microhardness measurements were conducted near the interface (load - 0.1 N, dwell time - 10 seconds).

**Statistical processing**

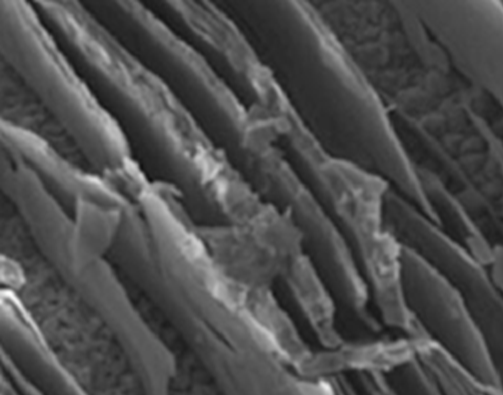
To assess the significance of differences between the groups, a dispersion analysis (ANOVA) was used with the post-hoc Tyuki criterion at a significance level of p < 0.05. The results were processed in the OriginPro 2023 and Python (SciPy, stats models) software environment.

# RESULTS AND DISCUSSION

**Unmodified vermiculites**

The test results showed that unmodified expanded vermiculite exhibits low adhesion strength to the cement matrix - an average of 0.62 MPa. The separation occurred strictly along the grain-matrix interface, indicating the absence of effective interaction. SEM analysis revealed the presence of a defective layer up to 40 μm thick, practically devoid of hydrate compounds (Fig. 1). The contact zone was loose, with pronounced porosity and weak penetration of hydrates into the grain structure.

This behavior corresponds to literature data [4], where the absence of functional groups on the vermiculite surface and its layered structure prevents the formation of C-S-H or other hydrate bonds.

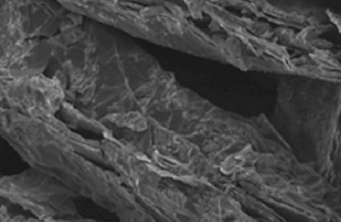


**FIGURE 1.** Without treatment

**Alkali treatment (NaOH)**

After alkaline activation with a 1% NaOH solution, a significant improvement in adhesion was observed: the strength increased to 0.74 MPa. The nature of the failure became partially cohesive: traces of cement paste remained in the contact zone. SEM images (Fig. 2) revealed the formation of a rough surface structure with localized activation of Si-OH groups [2]. The presence of such groups promotes the nucleation of C-S-H hydrates and partial infiltration of the gel into the grain micropores.

Similar behavior is confirmed by studies [5], where alkaline treatment enhances the reactivity of vermiculite without radically altering its structure.

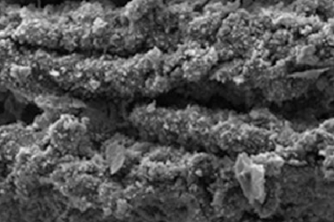


**FIGURE 2.** Alkaline activation (NaOH)

**Silicate coating (liquid glass)**

Treatment with a 10% solution of liquid glass (Na2SiO3) resulted in the formation of a smooth, thin silicate film on the grain surface. The adhesion strength increased to 0.70 MPa, but the fracture still occurred along the boundary. SEM analysis (Fig. 3) revealed a smoothed surface with sparse zones of cement gel penetration. This indicates partial inertness of the formed film, despite its moisture-protective properties [7].

Elemental analysis detected the predominance of Na and Si in the boundary zone, which is also observed in publications on the stabilization of lightweight aggregates with silicates [7].

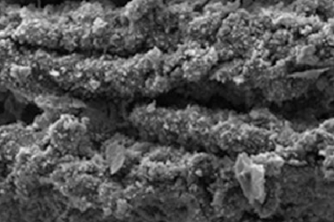


**FIGURE 3.** Liquid glass treatment

**Silane treatment (GKZh-11)**

The application of silane (GKZh-11) in the form of an aqueous emulsion ensured surface hydrophilization, reduced wettability, and the formation of strong Si-O-Ca interfacial bonds. The adhesion strength was 0.76 MPa, and the tear-off exhibited a mixed character (Fig. 4).

SEM images revealed zones of fine reaction where C-S-H hydrates transition to the modified grain surface. These data are in good agreement with the results of [1, 2] regarding the ability of silane groups to chemically bind with cement gel components.



**FIGURE 4.** Hydrophilization (silane GKZh-11)

**Complex modification**

The most significant effect was achieved through complex treatment: NaOH → liquid glass → silane. The adhesion strength increased to 0.91-0.92 MPa, which is approximately 45% higher than that of unmodified grain. The nature of failure was cohesive: destruction occurred within the cement paste (Fig. 5).

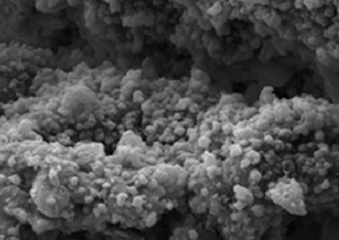
SEM analysis revealed:

• absence of voids and microcracks in the contact zone;

• a pronounced structure of C-S-H and C-A-S-H hydrates;

• formation of a transitional chemically bonded zone [11].

Such morphology indicates a reaction-integrated interface stabilized by multiple Si-O-Si and Si-O-Ca bridges, as well as the possible involvement of NH2 groups from the silane [5, 11].



**FIGURE 5.** Complex modification

**Statistical assessment**

The results of statistical analysis using the ANOVA method showed that the differences between groups are significant (p ≈ 3.37 × 10−15). Tukey's post hoc test confirmed that the complex processing is statistically superior to other methods (table-I). This indicates high reliability of the experimental observations and confirms the effectiveness of the proposed approach.

**TABLE 1.** Adhesion under Various Modification Techniques

|  |  |  |  |
| --- | --- | --- | --- |
| **Processing method** | **Proposed System** | **Nature of discontinuity** | **Comment** |
| Without processing | 0,62 | Along the border | Porous layer, weak interaction |
| NaOH | 0,74 | Partially by matrix | Active groups, local reactivity |
| Liquid glass | 0,70 | Along the border | Stabilization, but the film is inert |
| Silan (GKZh-11) | 0,76 | Mixed | Stable wetting, chemical bonding |
| Complex modification | 0,91–0,92 | Cohesive | Close contact zone, phase integration |

## CONCLUSIONS AND PRACTICAL SIGNIFICANCE

1. The conducted research demonstrated that surface modification of expanded vermiculite significantly increases its adhesion strength to the cement matrix and improves the morphological characteristics of the contact zone.

2. Unmodified vermiculite exhibits low adhesion (0.62 MPa) and forms a porous, defective interface, leading to a decrease in the strength and durability of composites.

3. Various types of treatment:

Alkaline activation (NaOH) increases adhesion to 0.74 MPa due to the formation of reactive groups on the surface;

Treatment with liquid glass reduces capillary activity but creates an inert film (0.70 MPa);

Silane modification provides hydrophobicity and partial chemical bonding with cement (0.76 MPa).

4. The greatest effect is achieved through complex modification, which includes stepwise activation, silicate strengthening, and silane treatment. The adhesion strength increases to 0.91-0.92 MPa, and the failure mode becomes cohesive, indicating a strong chemically bonded zone.

5. Scanning electron microscopy (SEM) confirmed the formation of a dense, integrated interfacial zone saturated with C-S-H and C-A-S-H crystals, without voids or cracks. Such structures ensure stable material behavior under operational loads.

6. Statistical analysis (ANOVA) demonstrated highly significant differences between processing methods   
(p ≈ 3.37×10−15), with the complex method proving to be statistically significantly more effective compared to the others.

7. The practical significance of the results lies in the fact that the proposed approach can be recommended for industrial application in the production of lightweight and heat-insulating cement composites with enhanced strength, water resistance, and frost resistance. The modification methods do not require complex equipment, are cost-effective, and can be easily scaled up in typical production environments.

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