

# THE STUDY OF THE INTERACTION OF ADHESIVE WITH THE SUBSTRATE SURFACE IN A NEW COMPOSITE MATERIAL BASED ON MODIFIED GYPSUM AND TREATED RICE STRAW

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**Abstract:** *The article considers the issues of thermochemical treatment of the surface of plant aggregates to ensure strong adhesive interaction with a gypsum matrix in a composite building material. A selection of additives contributing to the improvement of adhesive strength and physical and mechanical properties of gypsum boards was conducted. A comparative analysis of the effect of moisture sorption on the durability of the composite material at the processing of fiber aggregates was also performed.*

**Keywords:** *adhesion, fiber-matrix, gypsum, straw, delignification, moisture deformation*

**Introduction.** Scientific and technological progress in the construction industry is based on the development of new and effective building materials that possess a set of required properties for various purposes. For a long time, wood, ceramics, steel, concrete and reinforced concrete were primarily used as building materials. In Uzbekistan, with the development of the construction industry in the last decade, there is now a wide adoption of composite building materials at the construction of most facilities.

Composite building materials are the multiphase systems comprising two or more monomaterials with various properties. Through rational combination of starting components, and a synergetic effect that follows, new materials are formed, which still retain the individual characteristics of each of the starting heterogeneous component [1,10-12].

The purpose of creating composite building materials is to improve certain properties of the starting components, for instance, their mechanical, thermophysical properties, chemical resistance, durability, as well as to decrease the cost of materials, including through the use of various wastes. Composite building materials include mortars, concrete, ceramics, mastics, adhesives, putties, paints and varnishes, fiberglass and other artificial multicomponent materials.

The idea of creating composite materials is not a novelty. Since ancient times people have been traditionally using adobe, consisting of clay as a bonding substance (matrix) and straw as a stiffening reinforcement. Furthermore, the asbestos cement, consisting of cement as a matrix and natural fibrous asbestos material serving as reinforcement has been used in construction for a long time. The properties of such composites are determined by the high strength of the reinforcing fibers, the rigidity of the matrix, and the bond strength at the matrix-fiber interface. The ratio of these parameters characterizes a whole set of mechanical properties of the material and its destruction mechanism as well.

In general, an adhesive interaction of fiber and matrix determines the properties of composites and their behavior during operation. The local stresses in the component reach maximum values at or near to the interface, wherein the destruction of the material usually occurs. The interface should ensure an efficient transfer of load from the matrix to the fibers.

The adhesive interaction at the interface should not break up under the action of thermal and shrinkage stresses induced either by the differences in temperature coefficient of linear expansion of the fiber and matrix or by the chemical shrinkage of the binder during its solidification [2,13-15].

The adhesion strength depends on the binding energy, completeness of contact determined by the relief of the surface, the interphase surface energy, wetting, and other surface phenomena, as well as the conditions for forming the contact (e.g. pressure, temperature, etc.) [3-4].

Today in Uzbekistan, some enterprises have already adopted the production of gypsum fiber boards, which is a rather new composite material for the local market. Such a composite contains a recycled cellulose fiber, which is evenly distributed in the gypsum mass and serves as reinforcement. Yet, the deficiency of recycled cellulose forces local entrepreneurs to apply various manufacturing technologies and use plant-based raw materials, which greatly affects the production process, primarily the operations to ensure high adhesion of the fiber to the matrix e.g. processing of fibrous raw materials or modification of the binder. In many cases, the straw of cereal crops is used as alternative raw material due to its affordable price and availability in large quantities as required. However, similar to many organic cellulose aggregates, straw has both the inherent valuable properties and negative ones, which make it difficult to produce composites of high strength [4-5,14-15]. The specific properties of such an organic cellulose aggregate include an increased chemical aggressiveness, significant amounts of moisture deformation and developing swelling pressure, a high degree of anisotropy, high permeability, low adhesion to the matrix, larger elasticity at mixture compaction. All of these properties adversely affect the matrix hardening, the structure formation, the strength and durability of the composite material to moisture-induced influences.

The article contains the experimental data on adhesive interaction with the substrate surface in the new composite material. Exploring the nature of the interaction is of fundamental importance for the understanding of the adhesion mechanism of fibers, in particular, straw fibers to gypsum crystals, and also for the development of solutions to enhance the adhesion between them.

First of all, there is a need to more extensively explore the catalytic effects at the adhesive - substrate interface as well as the molecular and chemical forces in the contact zone. Since the degree of negative effect of moisture deformations of the fibrous aggregate on the strength of the gypsum fiber board is largely determined by the adhesion rates of different materials (straw and gypsum), it is thus advisable to analyze the interconnection of these factors [6].

Nanazashvili I. Kh. et al. studied the adhesive properties of composite materials from waste wood and plant materials based on polymer and mineral binders [2]. According to authors the adhesive properties of wood with mineral binders depend on the tree species, the chemical composition, additives, treatment conditions, specific surface area, etc. It was found that with a decrease in the specific surface area of straw aggregate to a certain limit, the strength of the composite increased. The decrease in strength when using aggregates with coarse aggregates can be partially explained by the influence of large moisture deformations, which cause stresses in the contact zones during hardening and drying. When using a fine fraction, such a decrease can be explained by a significant decrease in the thickness of

the gypsum crystalline layers in the structure due to the large specific surface of the aggregate [7,16-17].

With an increase in surface roughness, the adhesion of straw with a gypsum matrix increases as well. At the same time, the increase in adhesive strength is proved to be associated with the appearance of a large number of active centers, an increase in the true contact area, and mechanical adhesion of the fiber and cavities serving as dowels and rivets of a kind [8].

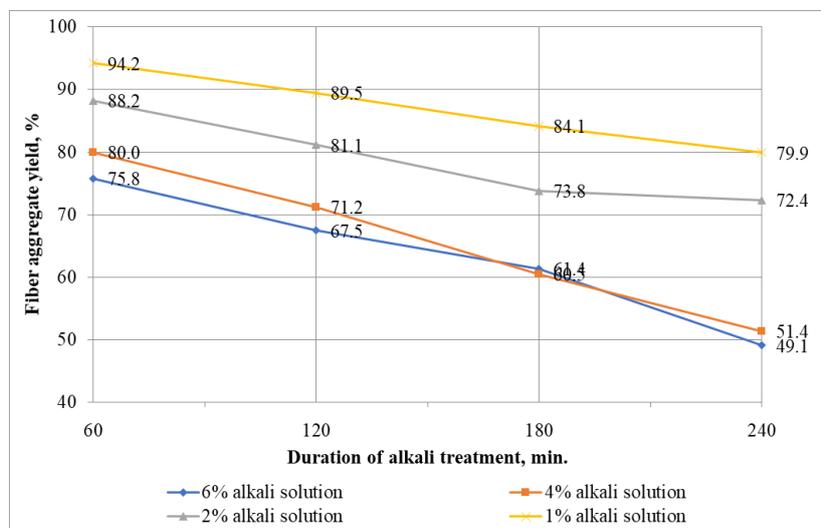
The true contact area can be enlarged by removing the fat and wax layer from the straw surface and thus allowing the appearance of additional fibers and depressions. It is known, that rice straw contains from 10 to 30% of mineral components, which can be removed through the alkaline treatment. The alkali solution affects the ligno - carbohydrate complex resulting not only in the removal of the mineral component and part of the lignin, but also in the destruction of polysaccharides [9].

With regards to the abovementioned, an attempt was made to explore the regularities between improvement of raw material (straw treatment) and adhesion of the fibers to the matrix (gypsum modification). The study therefore had to achieve the following tasks:

- Ensure the maximum extraction of mineral components and lignin from the ligno - carbohydrate complex of rice straw with minimal destruction of polysaccharides;
- Ensure the formation of denser gypsum crystal lattice, which increases the bonding strength at the fiber-gypsum interface.

The experimental tests were carried out using modifiers, plasticizers, desugared straw broths, mineral modifiers, which influence the hardening of the adhesive bonding of straw with gypsum to a different degree. Furthermore, the rice straw of the last year harvest containing 58 % cellulose, 14 % lignin, 5,4 % resin, 3,2 % soluble substances, 19,4 % mineral substances and construction gypsum were used.

Dry and chopped into 12–20 mm rice straw was cooked with an aqueous solution of sodium hydroxide under the following conditions: straw – solution ratio - 1/8; NaOH concentration - 1-6%; processing temperature - 90 ° C; the duration of the temperature rise - 15 minutes; the duration of alkaline treatment - 60-240 minutes. The resulting material was washed with distilled water to a neutral medium, sieved, ground, dried and weighed. Preliminary studies on alkaline treatment of rice straw and dependence of the product yield from the alkali concentration and duration of treatment are presented in Fig. 1.



**Fig. 1. The dependence of the yield of fibrous aggregate on the alkali concentration and duration of treatment**

The results obtained indicate a significant decrease in the product yield at the alkali concentration of 6% and treatment duration of 240 minutes (Fig. 1), therefore the further increase of the alkali concentration and treatment duration was seen as unnecessary.

The visual inspection of the material boiled in 1-2% alkaline solution indicated that treatment even with a duration of 240 min. did not result in the required yield and quality values. The analysis of the first treatment allowed to identify a range for further studies, meaning alkali concentration - from 3 to 5%, processing time - from 60 to 240 minutes.

After a series of experiments, the optimal conditions for the alkaline treatment of rice straw were identified. Based on the optimized conditions (treatment duration - 150 min; alkali concentration – 3,5%), the material visually resembling cotton fibers with a product yield of 57.1% was obtained.

In order to study the degree of adhesion of the obtained material with a gypsum binder, the strength characteristics of specimen mixtures, which also contained various types of additives to improve the bonding strength of fiber-gypsum, since the strength of the gypsum fiber composite, in our opinion, depends on the bonding strength between adhesive and substrate.

*Table 1: The effect of additives on the characteristics of the gypsum-fiber mixture*

No	Straw,%	Additive,%		W/G	Spreading, mm	Bending strength, MPa	Compressive strength, MPa
		C -3	Frem nanogip s				
1.	0.5	-	-	0.66	180	3.84	9.80
2.	0.5	-	0.2	0.66	250	4.69	15.4
3.	0.5	-	0.4	0.43	182	4.12	15.6
4.	0.5	-	0.7	0.33	180	4.34	16.1
5.	0.5	-	1	0.43	250	4.81	17.9
6.	0.5	-	2	0.36	260	6.84	18.6
7.	0.5	0.7	-	0.66	220	4.02	9.82
8.	0.5	1	-	0.66	236	4.02	10.6
9.	0.5	1,5	-	0.66	244	4.02	11.4
10.	0.5	2	-	0.66	258	6.03	12.6
11.	1	-	-	0.72	171	3.35	8.24
12.	1	-	0.2	0.72	245	3.68	10.8
13.	1	-	0.4	0.48	172	4.98	12.6
14.	1	-	0.7	0.36	171	4.98	16.1
15.	1	-	1	0.48	244	4.01	11.8
16.	1	-	2	0.40	249	4.82	13.5
17.	1	0.7	-	0.72	212	3.54	9.60
18.	1	1	-	0.72	218	3.98	9.80
19.	1	1,5	-	0.72	232	4.64	10.4
20.	1	2	-	0.72	238	5.40	12.6

The data analysis leads to several conclusions:

- adhesive strength depends on the gypsum paste density and its chemical activity, while the higher is the viscosity of the solution, the lesser it can penetrate into the straw pores.
- additives reduce the water demand of the mixture, which leads to the increase of the matrix density, and as a consequence, to the increased strength of dry material.

When mixing with the additives that increase the density of the gypsum matrix and the strength of gypsum-fiber interaction the rate of compressive strength of gypsum fiber composites significantly increased, and this indirectly indicates that without additives

bulk and moisture deformation may lead to destructive processes occurring in the board during its hardening.

The experimental studies show the "FremNanogips" additive for gypsum mixtures (manufactured by JSC "Plant of additives and lubricants "FRAME") to be the most effective for the formation of specimens, which have the high degree of adhesion throughout storage. Increased adhesive strength of specimens is the result of the increased density of the gypsum crystal lattice, which ensures a more solid bonding with a fiber surface.

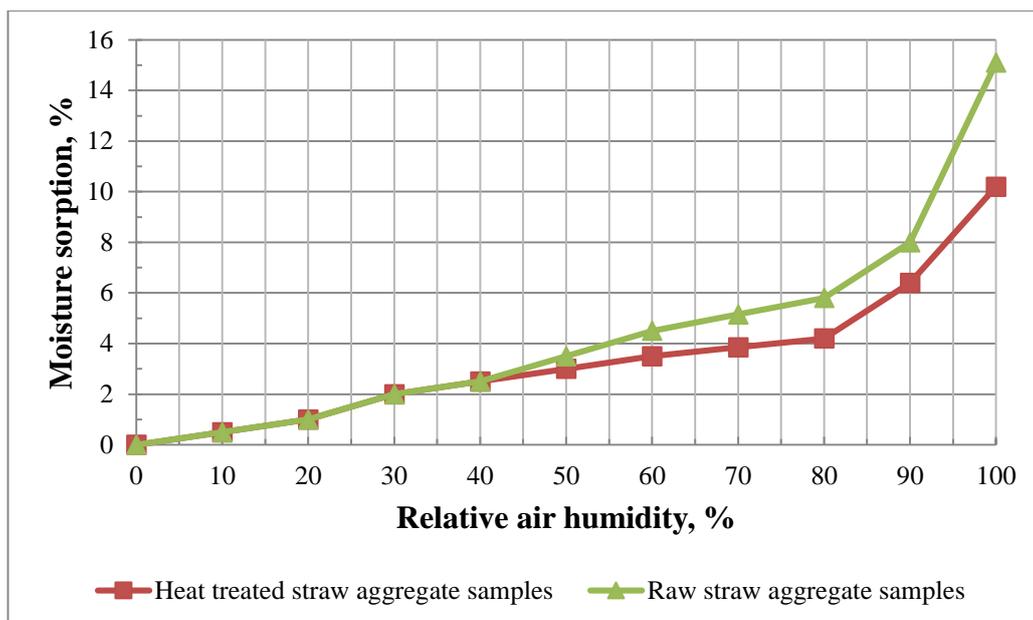
It is assumed that the adhesive interaction between straw and gypsum results from the interaction of ion calcium formed during hardening of the gypsum paste in the contact zone with the polar functional groups of straw components such as cellulose, lignin, and hemicellulose.

The bonding forces between the gypsum paste and the straw cell walls can be explained by the adsorption theory of adhesion. It is known that the constituent parts of the straw - the cellulose in the first place, are characterized by the structural polarization (i.e. the surface of the molecular chains of the cellulose, hemicellulose and lignin carry a negative charge) and therefore should be well connected with polar substances. Furthermore, the treated straw fibers contain a small amount of water-soluble extractive substances, which reduces the humidity of deformation in the contact zones [8, 11-15].

Does the strength of the obtained gypsum fiber specimens increase due to a decrease in the moisture deformations of the fiber aggregate? Assumingly it may be achieved owing to the reduced negative charge of straw fibers induced by the hot alkali treatment and blocking of polar groups, the hydroxyl ones in the first place, which are located on the surface of the cellulose molecular chains, hemicellulose and lignin of the fiber aggregate. With the aim to confirm these considerations, the effect of moisture on gypsum fiber specimens was analyzed.

The following method was applied to measure the moisture sorption. Pre-dried to constant weight the gypsum fiber specimens with aggregates from the conventional rice straw and heat-treated in alkali solution rice straw (in the manner described above) were placed in desiccators. The vapor-air medium in the desiccators was created artificially using a chemical solution of sulfuric acid of various concentrations, providing a relative humidity of 40 - 96 %. The humidity level in specimens was measured by weighing every two weeks during the first 2 months, and then every week until a constant specimen weight on the scale was achieved.

The experimental results are shown in the diagram (Fig. 2). The moisture sorption of all specimens reaches 2,5 % in a desiccator at a relative air humidity of 40%. The moisture sorption rate of untreated straw specimens is 22 % higher compared with the treated straw specimens and makes up 3,5 % at a relative air humidity of 60%. The moisture sorption of gypsum fiber material from the treated straw makes up 6,4 % at a humidity of 90%, while the moisture content of untreated straw specimens increases by 8,0 %. The relative air humidity of 96 % causes an increase in the moisture sorption of conventional straw specimens to 15,1 %, which is 33 % higher than that of the treated straw specimens (10, 2 %).



**Fig.2. Water vapor sorption isotherms of gypsum fiber composites**

Data analysis shows that heat treatment of rice straw with the alkaline solution reduces the water absorption of gypsum fiber composites. The rationale for such operation is determined by the polar nature of this particular high molecular compound. The increase in the hydrophobicity of heat-treated straw fibers is caused by the blocking of adsorption-active hydroxides of cellulose macromolecules and other straw components due to the hydrogen bonds established between metal groups and straw hydroxides.

The experimental studies lead to the following conclusions:

- the adhesive strength between the straw and gypsum depends on the specific surface of the aggregate, aggregate shape coefficient, the roughness of the fibrous aggregate, the chemical activity of the composite components, water/gypsum ratio, amounts of chemical additives, level of transformation processes of the structural and chemical characteristics in the straw in the course of its thermochemical processing;

- at high level of ambient humidity the untreated straw-based composite materials may reach the moisture sorption index equal to 15,1 %, that may lead to lower operational characteristics in the future and may cause destruction of wall panels and structures in general.

Our studies suggest an effective method for the complex preparation of the fiber aggregate by the preliminary heat treatment in alkaline solution followed by mixing with gypsum and modifier, which in turn reduces the paste viscosity and contributes to the clogging of open pores and depressions in the aggregate and thus ensures the high degree of adhesion of the matrix and aggregate.

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