Study of the forming processes of the arbolite structure during the chemical activation of flax shove

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Abstract. As one of the ways of directed formation of structures in arbolite (wood concrete), the use of properties of organic additive might serve. A capillary-porous, chemically active material is used for this purpose. The structure of capillaries and pores determines the perspective of directed mass transfer within the “binding substance-additive” system, and the chemical activity of flax shove fibers can replace the physical bonds of the components with stronger ones, the chemical bonds. To determine the ways of modification of the additive, an aqueous solution of calcium sulfate hemihydrate (CaSO₄·0.5H₂O) was applied along with liquid glass. The substantiation of the chemistry of the processes occurring in the wood-cement compositions during their strengthening was given. Analysis of the dynamics of strengthening of the material has shown that as a result of the chemical activation of flax fibers, the internal structure of capillaries and pores experience colmatage, which inhibits water absorption inside them, and also stronger bonds of flax fibers with cement stone are formed. With the application of the proposed technology, the compressive strength of arbolite increases almost 1.4 times, and the strength under transverse bending increases 1.6 times.

1. Introduction
The modern level of technology in civil engineering places high demands on building materials in terms of improving their thermal protection, durability, and efficiency. The development of composite materials with good thermal insulation properties and their use in construction technology in conditions of economy of heat and power resources are of great technical and economic importance. Cement-based wood composite materials largely meet these requirements. The main direction of development of the production of building materials and products is the widespread use of cellulosic waste of organic nature, which appears after the harvesting of agricultural cultures. These requirements have appeared as the outcome of limited resources, the need for long-distance transportation, high level of material consumption, and energy intensity of several technological processes of extraction and processing of raw materials. The above factors significantly hinder the development of the industry of building materials, which are made of natural raw materials.

2. Literature sources review
In recent years, the production and processing of flax with enhanced environmental and operational qualities has remarkably increased, both in Ukraine and in other countries of the world [1, 2]. The...
article [3] presents an explorative study analyzing the market potential for timber-concrete composites (TCC) in the German construction industry. A co-operation between concrete and timber constructors becomes necessary, which experts call a “clash of cultures.” But, TCC involve sticking together timber beams and concrete slabs to add stiffness and to resolve the acoustic and vibration issues that can afflict pure timber constructions [4]. That is why, researchers [4] have devised a “recipe” for a lightweight, pourable concrete that uses wood chips as aggregate. Ordinary concrete is one of the most common materials in buildings. However, due to its low ductility and flexural strength, its seismic behavior can be improved upon by different additives (sawdust and wood shavings). In this regard, wood-concrete composites exhibit desirable structural properties not achievable by either wood or concrete alone, making it an interesting material from a seismic point of view. The results [5] show how blocks with wood aggregates comply with current regulations for structural materials in the seismic countries, while also considerably outperforming traditional concrete blocks in the event of an earthquake.

Wood fiber had a positive impact on the mechanical strengths of autoclaved aerated concrete [6]. Using fiber produced with wood waste could reduce the negative impacts on environment and improve the mechanical property of autoclaved aerated concrete. In the most case, the effect of adding wood fiber was better than that of polyester fiber. In the experimental investigation [7] an attempt was made to recycle wood waste. Concrete-containing wood aggregate in percentages of 0, 15, 20 and 25 in place of crushed stone was developed with a characteristic compressive strength of 25 MPa, with a mix proportion of 1:1.26:2.76 and with water/cement ratio of 0.45.

Lignocellulosic lightweight concretes are a potential contributor to sustainable development. Due to their low cost, lightweight and thermal insulation properties, lignocellulosic by-products received a particular attention, in the recent years, for manufacturing lightweight concretes. Proper mixture of flax stalk fiber with mortar can improve and enhance thermal insulation performance of buildings and the crack resistance capability of plaster layers. The result [8] shows that this method can lower the weight of plastering by 42.46 per cent and reduce the cost of plastering by 33.36 per cent. This new material has shown its superior energy saving and environmentally friendly features. Therefore, the material has a broad practical potential in constructions. However, lignocellulosic aggregates are not always fully compatible with cement matrices leading to setting delays, significant dimensional variations and low mechanical strengths. To solve this problem, it is necessary to carry out treatment of the fiber to prevent the fiber/matrix debonding. In [9] flax fibres surface were treated by different treatment in order to enhance the interfacial bonding force between flax natural fibres and vinyl resin matrix. The experiment of contact angle and mechanical property were done to characterize the surface topography and bonding force and choose the best modification. Because of their hydrophilic nature, flax fibers absorb water during the mixing stage and release it gradually during the curing step. Thus, the rheological properties and the setting of the cement paste are deeply disturbed. In addition, the diametrical shrinkage of the fiber at the desorption stage can lead to a weakening of the fiber–matrix bond. It is therefore necessary to treat of the fiber mixture to prevent the debonding. The results [10] obtained from these formulations show an improvement in the rheology and the mechanical performances of the composites.

Flax shelve is effective for processing into materials for various purposes, due to the peculiarities of its physical and chemical structure and low cost. Flax stalks are destroyed during fiber extraction in the process of munching and scutching, and their woody parts, which are falling out, and form a flax shelve. The dimensions of these particles vary from 1 to 15 mm in length; most often there are particles with a length of about 5 mm. The thickness of them ranges from 0.3 to 1.5 mm. The average density of flax shelve fibers is 120-140 kg/m³. The chemical composition is similar to wood. Flax shelve contains up to 45-58 % cellulose, 21-29 % lignin, 23-26 % pentosanes. The use of flax shelve in the production of walling items with mineral binding substances, for example, with cement, is fully justified only if the impact of the so-called “cement poisons” on the process of structure formation of the material is reduced. Therefore, when designing the composition of arbolite for walling items, one should carefully approach the selection of various chemical additives used as mineralizers. The aim of the research
project [11] was the development of a wood-cement concrete used as local filler and stiffening element in wooden ceiling elements. Three different ways to accelerate the hydration of the cement and therefore to counteract the effect of the so-called cement poisons were examined. The filling material consists of water-glass and/or Portland cement mineralized wood particles. Through the use of only 15 mass % wood particles as filler, the gross density of the concrete can be significantly reduced by 36 to 39 %. The fresh concrete mixed with water, at a water/cement ratio of 1.0 to 1.1, has a crumbling, easily malleable consistency. The achieved compressive strength of the wood concrete of 4 to 5 N/mm² is higher than the required compressive strength transverse to the fibre of softwood (3.2 N/mm² for C50 strength class).

The increase in production and the expansion of the range of walling materials and structures based on organic waste is an important task, part of which can be resolved by organizing the production of wall wood concrete structures.

3. Study aim
The work aims to increase the physical and mechanical characteristics of arbolite products based on flax shive, to obtain an aggregate that is inert in physical and chemical terms to prevent the eventual impact by extractive substances appearing due to colmatage of the filler surface by a mineralizer. The saturated aqueous solution of calcium sulfate CaSO₄·0.5H₂O and water glass are applied as the latter.

4. Materials and methods
One of the ways of directional structure formation of arbolite can be the use of properties of organic additive, such as a capillary-porous chemically active material. The structure of capillaries and pores determines the prospect of purposeful transfer in the “binder-aggregate” system, and the chemical activity of wood can create the necessary conditions for this purpose. Besides, the physical bonds of the components can be replaced with more durable ones – chemical ones. The main task, in this case, is the determination of activators of properties of wood and the study of the processes taking place during the formation of the structure of the material under study. The task is achieved by the fact that the raw mix for the manufacture of arbolite, which includes portland cement, wood filler, and water glass, additionally contains a saturated aqueous solution of hemihydrate calcium sulfate (CaSO₄·0.5H₂O).

The proposed raw material mixture is prepared as follows. 8 g of hemihydrate calcium sulfate has been added to 1 liter of water, and the mixture has been left for 1-2 hours until the pH of the solution which is equal to 6.8-7 does not change to pH 5-6 of the solution. It is known that the solubility of hemihydrate calcium sulfate in water is 8 g per liter of water. To the necessary amount of the saturated solution of calcium sulphate (for 1m³ of arbolite) water glass has been added in the amount of 4 % of the mass of cement (the density of water glass is 1.3-1.4 kg/l), and the solution has been stirred for 1-2 minutes until a whitish, viscous gel is formed. Wood aggregate has been processed with the gel obtained, leaving the former for 10-15 minutes for clogging of the pores of the wood filler due to the impact of the gel, containing silicic acid and calcium sulfate in aqueous solution. As a result, the release of extractive substances from wood filler is reduced. Then binding substance was injected, namely Portland cement. The composition is thoroughly mixed until indiscrete mass appears. The resulting arbolite mixture is characterized by hardness of 5-15 sec according to the technical viscometer. Samples of 10×10×40 cm dimensions were formed from the resulting mixture. Compaction of the arbolite mixture was carried out by the tamping method. After strengthening under normal conditions, the samples were tested both in compression and bending. The compositions of the samples are presented in table.1.
Table 1. The compositions of arbolite samples.

| Component                           | Mixture 1 | Mixture 2 | Mixture 3 |
|-------------------------------------|-----------|-----------|-----------|
| Portland cement, kg                 | 350       | 350       | 350       |
| Wood aggregate, kg                  | 250       | 250       | 250       |
| Water glass, % (kg)                 | 4 (14)    | 4 (14)    | 4 (14)    |
| Calcium sulphate hemihydrate (CaSO₄·0.5H₂O), % (kg) | -         | 5 (17.5)  | -         |
| Water, kg                           | 360       | 360       | -         |
| Water saturated with hemihydrate calcium sulfate (CaSO₄·0.5H₂O) 8 g per 1 l of water | -         | -         | 360       |

5. Results and discussion

The reaction originating from Na-water glass by modifying it with semi-aqueous gypsum proceeds according to the following reaction:

\[ \text{Na}_2\text{O} \cdot n\text{SiO}_2 + \text{CaSO}_4 \cdot 0.5\text{H}_2\text{O} \rightarrow \text{CaO} \cdot n\text{SiO}_2 \downarrow + \text{Na}_2\text{SO}_4 + 0.5\text{H}_2\text{O} \]

As the gel coagulates, its composition approaches the composition of a difficultly soluble compound - wollastonite CaSiO₃, and the process is facilitated by chemical interaction with polysaccharides of flax shove; it leads to their encapsulation in internal spaces of flax shove. At the same time, wollastonite is capable of activating the processes of early strength development in arbolite due to the formation (in the presence of Ca(OH)₂) of calcium hydrosilicates of a colloidal degree of dispersion and the phases, which are similar by their crystallochemical structure:

1. tobermorite 5CaO·6SiO₂·nH₂O;
2. hillebrandite 2CaO·3SiO₂·2H₂O;
3. nekoite 3CaO·6SiO₂·8H₂O;
4. foshagite 4CaO·3SiO₂·2H₂O;
5. gyrolitic phases (for example, 8CaO·12SiO₂·10H₂O);
6. compounds of mixed type: calcium hydrocarbon silicates (for example, scawtite 6CaO·6SiO₂·CaCO₃·H₂O), potassium - calcium hydrosilicates (for example, K₂O·28CaO·48SiO₂·14H₂O), and other CSH-phases (figure 1) [12].

Figure 1. Conditional scheme of encapsulation of polysaccharides in flax shove.

Figure 1 shows the involvement of portlandite in the process of early strength development with the formation of calcium hydrosilicates (the dotted line indicates the separation of the chemically active colloid system by its composition depending on contact with the polysaccharides inside the capillaries and contact with the products of clinker hydration at the outlet from the capillary mouth). Sulfate-sodium component of the sol-gel binder also is not inert, or ballast. Hydroxyl groups of
individual elements of the lignin structure in the surface dense layers of flax campfires can be replaced by sulfate groups, which later act as active crystallization centers for low and highly basic forms of calcium sulfohydroaluminate in the form of ettringite of needle-like morphology. Thus, the smooth surface of flax shove gets activated in local places and enjoys stronger adhesion contacts due to chemical interaction with the hydroxyl groups of lignin: guaiacyl propane-, syringyl propane-, and 3-n-hydroxyphenyl propane hydroxyl (figure 2).

![Figure 2. Structural fragments of lignin (I, II, III).](image)

The test results of the samples are presented in Table 2.

**Table 2.** The mechanical characteristics of arbolite samples.

| Characteristic                  | Mixture 1 | Mixture 2 | Mixture 3 |
|--------------------------------|-----------|-----------|-----------|
| Transverse bending strength, MPa | 1.2       | 1.1       | 1.7       |
| Compressive strength, MPa      | 1.2       | 1.8       | 2.95      |

As it can be seen from the table 2, the introduction of a saturated solution hemihydrate calcium sulfate makes it possible to increase the strength characteristics of arbolite 1.4 times for bending and more than 1.6 times for compression.

6. Conclusions

Thus, on the basis of the studies performed, the use of silica sol-gel and calcium sulfate in an aqueous solution as coagulation approaches the composition to a difficultly soluble compound – wollastonite CaSiO₃, which is enhanced by chemical interaction with flax shove polysaccharides and leads to their encapsulation in internal spaces of flax fibers that is able to regulate arbolite structure formation processes.

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