Original papers
Advanced structural materials, materials for extreme conditions

УДК 691.175.743 DOI: 10.17277/jamt-2025-10-01-049-056

Wood-polymer composites based on polyvinyl chloride reinforced with chrysotile asbestos

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Abstract: The problem of reinforcing wood-polymer composites (WPC) is currently being solved by the introduction of bonding agents that improve adhesion between wood filler and polymer. However, the introduction of such additives does not strengthen these composites to the extent that they can be used in new areas of construction. Filling of composites with fiber modifiers shows its high efficiency. In this paper, the effect of chrysotile-asbestos fibers of different lengths on physical and mechanical characteristics of polyvinyl chloride (PVC)-based wood-polymer composites was studied. The study showed that the most effective fibers for reinforcing WPC were 2 mm long, with a 25 % increase in tensile strength and a 38 % increase in flexural strength. The optimum asbestos content was 7.5 pbw for all fiber lengths. The proposed compositions of PVC-based WPCs will make it possible to expand the range of products based on them, including in the field of structural plastics.

Keywords: polyvinyl chloride; wood-polymer composite; chrysotile asbestos; reinforcement; modification; filling.

For citation: Khantimirov AG, Abdrakhmanova LA, Islamov AM, Nizamov RK, Suchkova EA. Wood-polymer composites based on polyvinyl chloride reinforced with chrysotile asbestos. *Journal of Advanced Materials and Technologies*. 2025;10(1):049-056. DOI: 10.17277/jamt-2025-10-01-049-056

Древесно-полимерные композиты на основе поливинилхлорида, армированные хризотиловым асбестом

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Аннотация: Проблема усиления древесно-полимерных композитов (ДПК) в настоящее время решается путем введения в их состав связующих агентов, улучшающих адгезию между древесным наполнителем и полимером. Однако введение подобных добавок неспособно усилить данные композиты в той степени, чтобы применять их в новых областях строительства. Наполнение композиционных материалов волокнистыми модификаторами показывает свою высокую эффективность. В данной работе изучалось влияние хризотил-асбестовых волокон различной длины на физико-механические характеристики древесно-полимерных композитов на основе поливинилхлорида (ПВХ). Исследования показали, что наиболее эффективными для усиления ДПК являются волокна длиной 2 мм, при этом прочность при растяжении увеличилась на 25 %, при изгибе – на 38 %, а модуль упругости при растяжении увеличивается в два раза. Кроме того, выявлено снижение показателей водопоглощения и истираемости, причем данный эффект в большей степени выражен для составов с более длинными волокнами. Оптимальное содержание асбеста составило 7,5 м.ч. для всех длин волокон. Предлагаемые составы ДПК на основе ПВХ позволяют расширить ассортимент изделий на их основе, в том числе и в области конструкционных пластмасс.

Ключевые слова: поливинилхлорид; древесно-полимерный композит; хризотиловый асбест; армирование; модификация; наполнение.

Для цитирования: Khantimirov AG, Abdrakhmanova LA, Islamov AM, Nizamov RK, Suchkova EA. Wood-polymer composites based on polyvinyl chloride reinforced with chrysotile asbestos. *Journal of Advanced Materials and Technologies*. 2025;10(1):049-056. DOI: 10.17277/jamt-2025-10-01-049-056

1. Introduction

The dramatic growth in demand for wood-polymer composites (WPC) in the construction materials industry causes an increase in production volumes of these materials, an expansion of the range of products manufactured on their basis, and a comprehensive study of the modification by the scientific community and manufacturers.

One of the main goals of researchers and practitioners is to improve the physical and mechanical properties of WPC, which can be achieved by introducing various modifiers, mainly binding agents of various natures, improving the compatibility of the wood filler with the polymer matrix. However, today, fibrous modifiers are increasingly used to strengthen WPC, playing the role of micro-reinforcing fillers. This trend is not accidental, since, in general, fiber-filled polymer composites have been known for a long time and have a number of advantages over dispersed-filled ones [1–4]. The effect of strengthening fibrous composites is based on the high strength of the fibers, which are often tens and hundreds of times higher than the strength of the polymer.

There are studies on the properties of reinforced WPCs. For wood-filled composites based on polyethylene (PE) and polypropylene (PP), compositions with basalt [5–8], glass [9, 10], carbon [11, 12], organic [1, 13, 14] and asbestos fibers [1, 15, 16] are known.

In [7], a study was conducted to examine the dependence of the WPC properties on the amount of basalt fiber with fiber lengths of 3 and 12 mm. It was found that with a fiber content of 15 to 25 %, the tensile strength reaches its maximum value. At the same time, the bending strength also increases with a fiber content of 20 to 30 %, regardless of the fiber length. It was also found that with a fiber content of more than 30 %, its distribution becomes uneven. The best comprehensive improvement in the WPC properties is achieved with a basalt fiber content of 12 mm in the range between 15 and 30 %.

Studies were conducted [8] with a mixture of basalt fiber and wood fiber in a 1 : 1 ratio being introduced into WPC based on PP. The addition of 10 % of the mixture of these fibers led to a significant improvement in the characteristics of the composite. The elastic modulus doubled, and the tensile strength increased by 30 %. For composites with a total fiber content of 20 %, the linear coefficient of thermal expansion decreased almost three times.

In the literature, there is data on the reinforcement of WPC – polyvinyl chloride (PVC) with glass [17, 18], basalt [19–21], aluminosilicate, zirconium [22] and wood fibers [23, 24]. For example, the introduction of glass fiber with a diameter of 13 μm [12] increased the flexural modulus and strength by 52–129 and 21–93 %, respectively.

In [14], PVC composites filled with rice husk and reinforced with basalt fiber were studied. The fibers were introduced into the composites instead of part of the rice husk, with their maximum content being 12 % of the total mass of the material. With an increase in the content of basalt fibers from 0 to 8 %, the hardness increased from 45.9 to 51.9 HRC, and the impact strength from 3.9 to 5.5 $kJ{\cdot}m^{-2}$. However, with a further increase in the fiber content to 12 %, these indicators decrease sharply. The authors explain this phenomenon by the agglomeration of basalt fibers, which occurs with an excess of fiber content in the composite.

In our studies [15, 16] the properties of PVC-based WPCs with 50 pbw of wood flour reinforced with basalt fibers were studied. The optimal fiber content was found to be 10 pbw. The highest physical and mechanical properties were found for fibers 6.4 and 12.7 mm long. It was shown that an important condition for the effectiveness of basalt fibers is the presence of a lubricant on their surface.

Chrysotile asbestos fibers are often used as a reinforcing component in plastics [25]. It is known that chrysotile fibers improve the elasticity, strength, and thermal and chemical resistance of plastics [26].

Chrysotile is a mineral with a parallel-fibrous structure consisting of layers with the formula $Mg_6[Si_2O_5](OH)_8$. Depending on the deposit, location of asbestos zones and type of ore, the outer diameter of the fibrils can vary within 100–650 angstroms, the inner diameter is 20–300 angstroms, and the wall thickness is 100–220 angstroms [27]. Thus, chrysotile is a tubular nanostructure consisting of magnesium hydrosilicates.

Any asbestos fiber is an aggregate of many smaller fibers, which in turn consist of fibrils [28].

The aggregate of fibers has a lower modulus of elasticity and tensile strength than an individual fiber. This is due to the fact that the load is transferred from one fiber to another only due to friction, resulting in a lower specific load on the bundle than on an individual fiber [29]. However, it is difficult to separate the aggregates into individual fibers, so in composite materials containing chrysotile asbestos, the main load is taken by bundles of asbestos fibers, which does not allow the full potential of this type of fibrous filler to be revealed.

Traditionally, asbestos fibers are actively used to reinforce thermosetting plastics and are less commonly used in composites based on thermoplastics: PE, PP, ABS and PVC. Thermoplastic WPCs based on PE and PP reinforced with asbestos fibers are known [16, 30].

The study [16] was aimed at reinforcing WPC based on PP with asbestos fibers with an average length of 2.5 mm. Combination of chrysotile with wood flour was carried out through an aqueous dispersion in a vibratory mill, which after drying to a constant mass was combined with PP and samples were formed by hot pressing. Dispersion of asbestos in an aqueous medium contributed to the splitting of aggregates into elementary fibers, but complete splitting could not be achieved. The optimal content of reinforcing fibers was 10 %, while a decrease in the flammability of composites and an increase in bending strength by 60 % were noted.

There are no published data on the use of asbestos in the composition of WPC based on PVC, although it is WPC-PVC that are characterized by the greatest physical and mechanical properties [31, 32]. Thus, the conducted analysis of the literature allows us to conclude that the use of asbestos fibrous fillers in the composition of PVC-based WPC is promising.

2. Materials and Methods

2.1. Methods for preparing compositions

The initial formulation of the polymer compositions includes suspension PVC grade S-7059-M (100 parts by weight), acrylic impact strength modifier (7 parts by weight), calcium stearate stabilizer-lubricant (3 parts by weight), dibasic lead stearate heat stabilizer (5 parts by weight), and M180 coniferous wood flour (50 parts by weight).

The following fibrous fillers were used:

– chrysotile asbestos grade 6K-30 with an average length of 1 mm (Russian Standard 12871–2013) produced by JSC Orenburg Minerals, the average diameter of fiber aggregates is 20–100 μm;

– chrysotile asbestos with an average fiber length of 2, 6 and 12 mm, obtained from asbestos cord (Russian Standard 1779–83), the average diameter of aggregates is 10 μm.

Each type of asbestos was introduced in quantities of 0.5; 2.5; 5; 7.5; 10 and 12.5 pbw.

Chrysotile was introduced by adding and mixing it in the required quantity into a small portion of wood flour (30 % of the required quantity), then the fibre content was gradually reduced by introducing the remaining portion of the wood filler.

To study the WPC, extruded samples were obtained on a laboratory twin-screw extruder LabTechScientific LTE 16–40 with a temperature profile from 183 to 200 °C and a screw speed of 20 rpm for 10–15 min.

2.2. Methods for determining properties

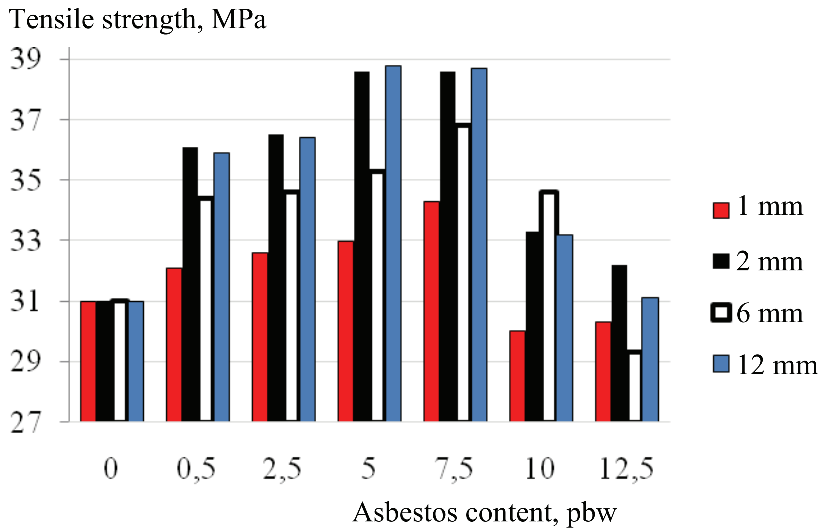
In this study, the following parameters were tested on WPC samples: tensile strength according to Russian Standard 11262–2017 (ISO 527-2:2012), bending strength according to Russian Standard 4648–2014 (ISO 178:2010), average density (Russian Standard 15139–69), water absorption and abrasion (Russian Standard 11529–2016). To determine the tensile strength, the clamp movement speed was 100 mm·min^{-1}, for static bending strength – 50 mm·min^{-1}. The tests were carried out on five samples at room temperature. The error in load readings on scales within the working measurement range is up to 2 %.

The study of the microstructure of WPC composites and the interaction of fibers with the polymer matrix was carried out using scanning electron microscopy on an AURIGA microscope combined with an IncaEnergy 350 X-Max energy dispersive spectrometer (EDS).

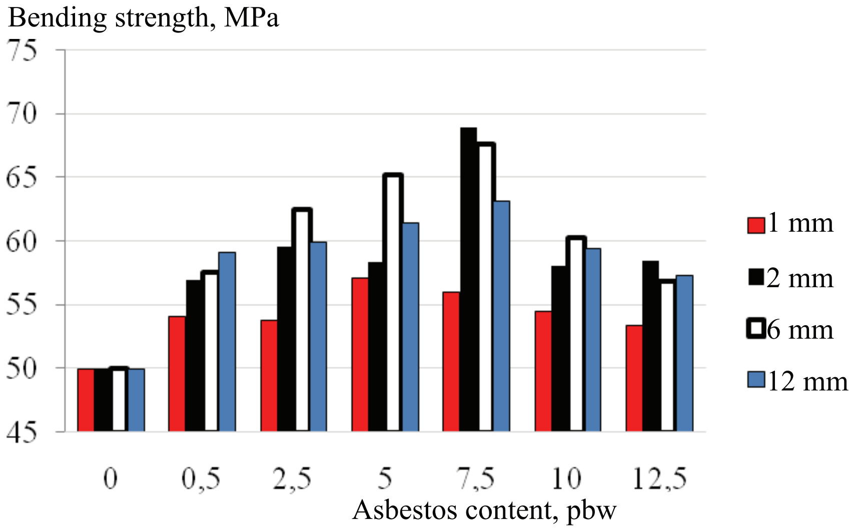
3. Results and Discussion

At the first stage, the mechanical properties of WPC-PVC were analyzed using asbestos fibers of different lengths (Fig. 1).

For all samples, an increase in tensile and bending strength is observed, with maximum values being achieved at 7.5 pbw. However, the bending strength remains higher than that of the original WPC in the entire range of concentrations. From all the data presented, it is clear that the efficiency depends on the fiber length. Composites containing very short fibers (1 mm) and long ones (12 μm) are less effective. Obviously, due to the small aspect ratio (length 1 mm), this type of asbestos fiber practically



(a)



(b)

Fig. 1. Concentration dependence of tensile strength (*a*) and bending strength (*b*) of WPC composites filled with asbestos fibers of different lengths

does not exhibit anisotropy of properties and behaves like a typical dispersed filler. It is worth noting that the introduction of chrysotile fibers through mechanical dry mixing (p. 2.1) cannot solve the problem of splitting fiber aggregates into fibrils (elementary fibers), which leads not only to the need to use a larger number of fibers to achieve high physical and mechanical properties, but also to additional interweaving of aggregates and fibrils with each other. This effect is more pronounced, the longer the average length of chrysotile and the higher the concentration, which leads to the formation of a non-uniform heterogeneous structure of the composite. For example, even for WPC with the inclusion of asbestos fibers 2 mm long, inclusions in the form of bundles with a diameter of up to 150 μm can be seen (Fig. 2). The most effective distribution

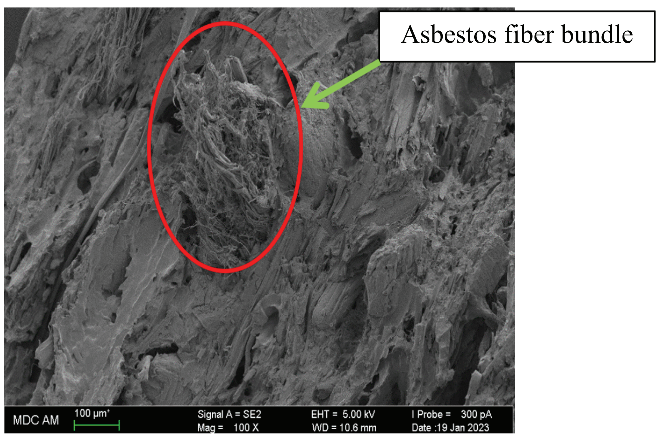


Fig. 2. Micrograph of a brittle chip of a WPC extrudate with 10 pbw asbestos fiber with a length of 2 mm (×100)

of chrysotile fibers can be achieved in aqueous dispersions using dispersants and ultrasonic treatment [33, 34], but this method is economically justified for introduction into materials in the production of which water and its various dispersions are used. The introduction of chrysotile into wood flour through aqueous dispersions is quite energy-intensive, since the filler after this treatment must be dried to residual moisture of 1–2 %.

The main product made of WPC is decking, which is used as a covering for paths, promenades, summer verandas, gazebos and other similar structures. It is exposed to atmospheric precipitation and intensive abrasive wear in places with high foot traffic [3, 4].

The results of tests of WPC for abrasion and water absorption depending on the length of asbestos fibers at a content of 7.5 parts by weight (from mechanical test data) are shown in Fig. 3.

Abrasion decreases with the increasing length of asbestos fiber. Obviously, longer fiber lengths are more difficult to “pull out” due to the larger area of contact with the polymer matrix.

Minimum water absorption values are also typical for compositions with 12 mm long fibers, the decrease was 44 %. The average density of the samples had an extreme at 7.5 pbw, the values of this indicator are at the same level for all lengths (an increase of 12 %).

Thus, water absorption and abrasion resistance of WPC naturally decrease with increasing fiber length in the composition. However, the change in strength properties is extreme and, as has been shown, the greatest strengthening effect is characteristic in the presence of fibers 2 and 6 mm long. And since in all cases water absorption and abrasion resistance remain significantly higher than the WPC indicators without the introduction of asbestos fibers, for practical implementation it is possible to recommend the use of fibers 2 to 6 mm long.

For the composite with the maximum increase in mechanical properties (with asbestos fibers 2 mm long), additional operational and technical indicators were established (Table 2).

In general, asbestos-filled polymer compositions are usually characterized by a decrease in impact strength, but in the developed composition a fairly significant effect of increasing this indicator was found. A similar effect is observed for the modulus of elasticity in bending.

Comparing the obtained results on the introduction of chrysotile asbestos with the data that the authors of the article published using basalt fibers [21], it can be concluded that the maximum increase in strength indicators for asbestos is achieved with a lower content (7.5 pbw versus 10 pbw) and a shorter fiber length (2 mm versus 6.4 mm). The absolute indicators of the strength properties of WPC are higher when using asbestos fibers. Obviously, this is due to the higher modulus of elasticity and fine organization of the chrysotile structure compared to basalt fiber, which allows for effective transfer of loads from the polymer matrix to the fibers.

Table 2. Operational and technical characteristics of the composite with the highest strength

Indicator	Basic WPC	Asbestos 2 mm 7.5 pbw
Tensile modulus, MPa	2709	3435
Bendingmodulus, MPa	1426	1796
Impact strength, kJ·m^{-2}	13.7	16.9
Hardness, kg·cm^{-2}	104	111

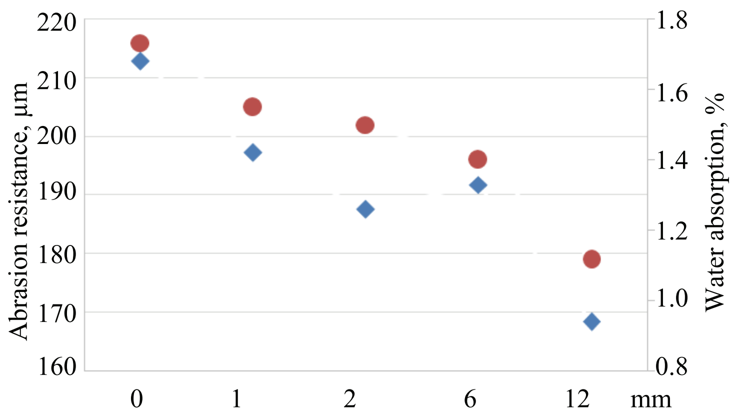


Fig. 3. Dependence of the abrasion resistance and water absorption indices of WPC composites on the length of asbestos fibers

4. Conclusion

The use of asbestos fibers in wood-filled PVC-based composites has proven to be effective in strengthening them.The best results were achieved with the introduction of 7.5 pbw of chrysotile fiber 2 mm long. Composites with such a filler demonstrated an increase in tensile strength by 25 %, in bending strength by 38 %, and the tensile modulus of elasticity increases twofold. With an increasing fiber length, a decrease in water absorption and abrasion resistance was observed. Compositions with chrysotile fibers of maximum length (12 mm) reduced these indicators by 44 and 17 %, respectively, which helps to increase the service life of products. The introduction of asbestos fibers in quantities greater than 7.5 pbw was ineffective and can lead to deterioration of operational properties due to entanglement of fibers and possible release of crystallization water from the structure of chrysotile asbestos during high-temperature treatment. With dry mechanical mixing of chrysotile, it is impossible to achieve complete splitting of fiber aggregates into fibrils, which does not allow the reinforcing properties of this modifier to be fully revealed. Despite this, chrysotile asbestos is a more effective modifier compared to the fairly popular basalt fiber, showing a greater increase in strength (by 2–3 MPa) with a shorter fiber length (2 mm versus 6.4 mm) and a lower concentration (7.5 pbw versus 10 pbw).

5. Funding

This study received no external funding.

6. Conflict of interest

The authors declare no conflicts of interest.

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Received 03 December 2024; Revised 28 December 2024; Accepted 04 February 2025