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PLYWOOD AND THERMAL INSULATION BOARDS BASED ON THE MODIFIED PHENOL FORMALDEHYDE BINDER

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Abstract. The lower curing temperature of phenol formaldehyde binder allows reducing the costs of producing cellulose-containing materials, such as FSF plywood and thermal insulation composites made of plant wastes. However, the low-temperature production mode provides an insufficient degree of the phenol formaldehyde binder curing, leading to a decrease in water resistance of material based on the phenol formaldehyde binder. A modifier should be added to the phenol formaldehyde binder to reduce the amount of free hydroxymethylol groups in the cured binder and to form a stronger cross-linked structure in low-temperature curing conditions. Hydrogen peroxide was used as a phenol formaldehyde binder modifier in this research. The research results confirmed the hypothesis about the effect of hydrogen peroxide on the intensification of the structure formation of cellulose-containing materials based on the phenol formaldehyde binder. In conditions of low-temperature curing (120 °C for FSF plywood and 100 °C for thermal insulation composites made of plant wastes) modification of phenol formaldehyde binder with hydrogen peroxide reduced the binder curing time, the pressing time of FSF plywood, improved the physical and mechanical properties of FSF plywood and thermal insulation composites made of cellulose-containing filler (soft wood waste and irrecoverable flax spinning waste). When 1.0 % of hydrogen peroxide is added to the binder, the binder curing time reduces by 43.6 %. The addition of 1.0 % of hydrogen peroxide increased the shear strength of FSF plywood by 4.4 % and the static bending strength of plywood by 4.8 %. Modification of the binder with hydrogen peroxide increased water resistance of FSF plywood: plywood thickness swelling has been reduced by 2 % over 24 h in water. The strength of thermal insulation composites made of cellulose-containing wastes increased by 5.2 % with the addition of 1.0 % of hydrogen peroxide, thickness swelling decreased by 4.9 % over 24 h. The obtained research results allow recommending a modifying additive of hydrogen peroxide to phenol formaldehyde binder in an amount of 1.0 % of resin

mass to increase the strength properties of FSF plywood and thermal insulation composites made of plant wastes.
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Keywords: FSF plywood, phenol formaldehyde binder, hydrogen peroxide, physical and mechanical properties, thermal insulation boards, cellulose-containing waste, flax spinning waste.

ФАНЕРА И ТЕПЛОИЗОЛЯЦИОННЫЕ ПЛИТЫ НА МОДИФИЦИРОВАННОМ ФЕНОЛОФОРМАЛЬДЕГИДНОМ СВЯЗУЮЩЕМ

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Аннотация. Снижение температуры отверждения фенолоформальдегидного связующего позволяет уменьшить затраты на производство целлюлозосодержащих материалов: фанеры ФСФ, теплоизоляционных плиточных композитов из растительных отходов. Однако низкотемпературный режим не обеспечивает достаточной степени отверждения фенолоформальдегидного связующего, это приводит к снижению водостойкости материала на основе данного компонента. В условиях низкотемпературного отверждения для уменьшения количества свободных гидроксиметилольных групп в связующем и для формирования более прочно сшитой структуры необходимо введение в фенолоформальдегидное связующее модификатора. В качестве модификатора нами использован пероксид водорода. Результаты подтвердили гипотезу о его влиянии на процесс структурообразования целлюлозосодержащих материалов на фенолоформальдегидном связующем. Модификация фенолоформальдегидного связующего пероксидом водорода позволила в условиях низкотемпературного отверждения (120 °С для фанеры ФСФ, 100 °С для теплоизоляционных композитов из растительных отходов) уменьшить продолжительность отверждения фенольного связующего и прессования фанеры ФСФ, а также улучшить физико-механические показатели фанеры ФСФ и теплоизоляционных композитов из целлюлозосодержащего наполнителя – мягких

древесных отходов и невозвратных отходов прядения льна. При введении в связующее добавки 1,0 % H_2O_2 продолжительность отверждения снижается на 43,6 %; прочность фанеры ФСФ при скалывании увеличивается на 4,4 %, при статическом изгибе – на 4,8 %, разбухание по толщине за 24 ч пребывания в воде снижается на 2 %; прочность теплоизоляционных композитов из целлюлозосодержащих отходов увеличивается на 5,2 %, разбухание по толщине за 24 ч снижается на 4,9 %. Результаты исследования позволяют рекомендовать для повышения прочностных показателей фанеры ФСФ и теплоизоляционных композитов из растительных отходов модифицирующую добавку H_2O_2 к фенолоформальдегидному связующему в количестве 1,0 % от массы смолы.
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Финансирование: Исследование выполнено при финансовой поддержке РФФИ и администрации Костромской области в рамках научного проекта № 19-43-440001.

Ключевые слова: фанера ФСФ, фенолоформальдегидное связующее, пероксид водорода, физико-механические показатели, теплоизоляционные плиты, целлюлозосодержащие отходы, отходы прядения льна.

Introduction

Phenol formaldehyde binder (PFB) is often used for the production of composites based on cellulose-containing materials. This is due to the favorable combination of cost and performance [6]. Producers use low-temperature production modes to reduce the expenses for manufacturing cellulose-containing materials based on PFB. This leads to a decline in the physical and mechanical properties of the materials, thus, restricting their use in construction.

Studies on improvement of operational parameters of building materials, such as FSF plywood and composites based on plant fillers and phenol formaldehyde binder are urgent both in Russia and all over the world. Mohd Asim and colleagues believe that in order to improve the use of phenol formaldehyde binder and composites on its basis in construction it is necessary to apply composite modification [1].

The most actively used methods of modifying plant components of composites studied so far, according to both Russian and foreign scientists, involve a chemical reaction of the reagent with the hydroxyl groups of the cell wall polymer [2, 14]. These hydroxyl groups are crucial in the interaction between wood or other plant filler and water. They represent the most reactive sites. In wet wood, as in other cellulose-containing fillers, water molecules interact with cellulose forming hydrogen bonds between hydroxyl groups and individual water molecules. Changing the amount of these water molecules causes shrinkage and swelling of the plant material [14].

The research is aimed at providing rational values of production factors of materials based on phenol formaldehyde binder – FSF plywood and thermal insulation composites made of cellulose-containing fillers (soft wood waste and flax spinning waste).

Modification of plant-polymer composites, such as FSF plywood and thermal insulation boards made of plant filler, affects the mechanism of interaction between the cell wall polymer and water.

Russian and foreign scientists have used hydrochloride, sodium thiosulfate and sodium hydroxide in the form of ionic liquid [8], iron(III) chloride, phthalic anhydride

$C_8H_4O_3$ [20], tannins from bark [9, 17], hydrolysis lignin [10], hydrogen peroxide [4, 15], hydrogen peroxide together with photocatalysts (at the resin synthesis stage) [16], and rice straw alkali lignin [12] as PFB modifiers. Phthalic anhydride shows the properties of aromatic compounds and condenses with phenols. Natural tannins are a mixture of gallic and digallic acids both in free form and in combination with monosaccharides; furthermore, different number of gallic acid molecules can be attached to a saccharide molecule. This property of natural polyphenols provides additional bonds with the polysaccharides of veneer or plant filler of composite boards when modified with phenol formaldehyde binder. However, no significant improvement in the water resistance of materials based on phenol formaldehyde resins (PF resins) has been achieved so far by using natural polyphenols and modified lignin.

One of the promising areas for improving the properties of FSF plywood and other building composites based on plant fillers is the use of hydrogen peroxide (H_2O_2) as a modifier. In addition to the modification of the binder itself, surface modification of veneer with a 5.0 % solution of H_2O_2 is also known [22]. However, this method requires the introduction of additional technological operations.

Peroxides are complex substances containing molecules with the functional group ROOR with the divalent O-O ion. Organic peroxides tend to easily decompose to free radicals in the form of RO. This very property ensures that H_2O_2 reacts with the hydrogen group of wood cellulose and plant fibers.

High water resistance and mechanical strength are important for FSF plywood since it is used not only as an element of building structures, but also for shuttering, packaging production and car building. Hydrogen peroxide treatment improves the tribological properties of cellulose-containing materials [5].

It is known that the efficiency of H_2O_2 in oxidation reactions is determined by the concentration of its active decomposition products.

The mechanism of decomposition of H_2O_2, among other factors, is most influenced by the pH of the reaction medium. The first stage of the conversion of H_2O_2 in an alkaline medium (conditions of resole phenol formaldehyde resin) is the formation of a nucleophilic reagent – a hydroperoxide anion [21]:

$$H_2O_2 + HO^- \rightarrow HO_2^- + H_2O$$

The authors suggest that hydroxyperoxide anions during heating and hot pressing of plywood bind the hydroxyl groups of phenolic nuclei and accelerate curing of phenol formaldehyde binder.

Hydroxyperoxide anions are highly reactive compounds and can act as ligands in interactions with metal ions – complexing agents. There are always Fe^{3+} ions in phenol formaldehyde resin. Their trace values are formed when using an iron-molybdenum catalyst in formaldehyde synthesis.

The L.V. Pisarzhevskii Institute of Physical Chemistry of the National Academy of Sciences of Ukraine reported that chelated iron forms serve as catalysts for polymer formation from formaldehyde [13]. The interaction of chelate compounds “hydroxyperoxide anions–Fe^{3+}” with phenol formaldehyde oligomer initiates a polycondensation reaction with a higher rate at a lower temperature.

The electron configuration of Fe^{3+} ions corresponds to $1s^22s^22p^63s^23p^63d^54s^0$. The 3s, 3p and 3d subshells are occupied by electrons, the 4 subshells are free.

The common 4s, 4p and 4d hybrid orbitals are formed by interaction with the HO_2^- ligands. The Fe^{3+} ion, having donated these orbitals to the ligands, forms six donor-acceptor bonds that form an octahedron, which is characterized by the highest particle packing density and low energy reserve. Thus, six ligands (hydroxyperoxide anions) bind to the iron ion.

The theoretical assumptions put forward have been verified experimentally in this paper.

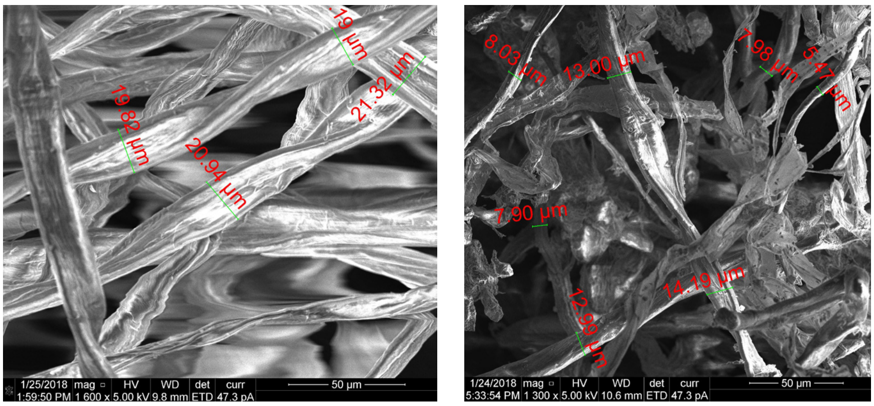
Objects and methods of research

The paper studies the influence of H_2O_2 on the curing time of the PFB based on the SFG-3014 resin, the shear strength and thickness swelling of FSF plywood, the static bending strength and thickness swelling of thermal insulation boards made of cellulose-containing wastes and the PFB. The addition of the modifier varied from 0.5 to 1.5 % of the binder weight (with a 0.5 % step).

The curing time of the PFB was detected at a temperature of 150 °C [19].

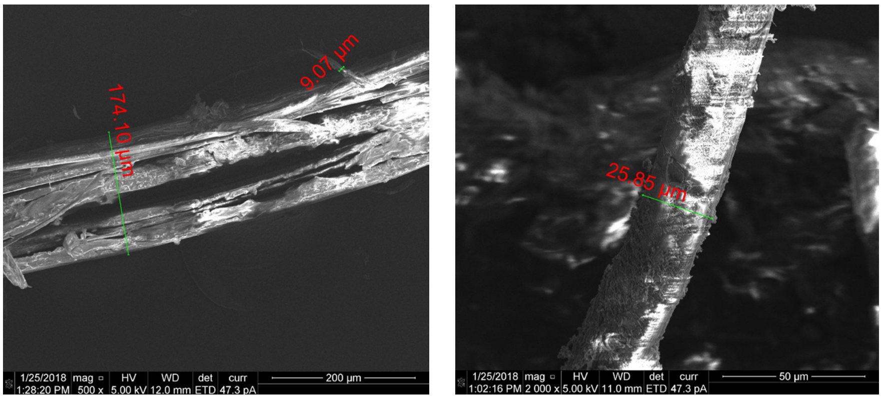
Five-layer plywood was manufactured on the basis of peeled birch veneer with a nominal thickness of 1.5 mm (State Standard GOST 99–2016). The veneer was pre-dried to a humidity of 7.0±1.0 %. After forming the package and applying the binder, hot pressing was carried out in a laboratory hydraulic press at a pressing temperature of 120 °C; pressing time – 5 min; specific pressing pressure – 1.6 MPa; binder consumption – 100 g/m^2. Manufactured plywood was cooled for 24 h and then cut into samples for testing.

Thermal insulation boards with an average density of 275 kg/m^3 were manufactured using the wet processing technology of fiberboard. The filler consists of 25 % of soft wood waste and 75 % of irrecoverable flax spinning waste. Cellulose-containing (irrecoverable) finely dispersed industrial wastes are disposed by incineration or landfilling. Cell damage is observed in processing of plant fibers of wood and annual plant (flax, cotton, etc.) wastes. Photos of plant wastes presented in the fig. 1 and 2 were obtained in the course of studies using an MS 20.1 Microscope [18].



a *b*

Fig. 1. Changes in the filler structure and dimensions during processing: *a* – cotton fibers; *b* – cotton processing waste fibers



a *b*

Fig. 2. Microstructure of flax processing waste: *a* – damaged fiber; *b* – fiber with the deposited binder

Samples of thermal insulation boards were dried at 100 °C to a humidity of 8±1.0 %.

The number of duplicated experiments in the tests *n* was equal to 10. A testing machine 2166 P-5 (scale value 0.1 N) was used to determine the static bending strength and the shear strength along the glue seam of the samples. The variation coefficient of the mechanical properties of materials was no more than 1.5 % when testing plywood samples and 2.1 % when testing samples of thermal insulation boards. The scatter of values in different duplicated experiments is due to the manual method of forming the material before pressing for plywood and drying for boards.

The maximum error (in accordance with the Russian State Standard GOST R 8.736–2011) for determining the shear strength of the samples along the glue seam is 0.02 MPa, and when static bending it is 0.03 MPa.

Research results and discussion

The results of determining the curing time of the binder are shown in table 1 and fig. 3, the parameters of plywood and composites based on the modified binder in are shown in table 2 and fig. 4–8.

Table 1

The results of determining the binder curing time

Content of H_2O_2 additive, %	0	0.5	1.0	1.5	4.0
Binder curing time, s	27.5	19.5	15.5	15.0	13.0

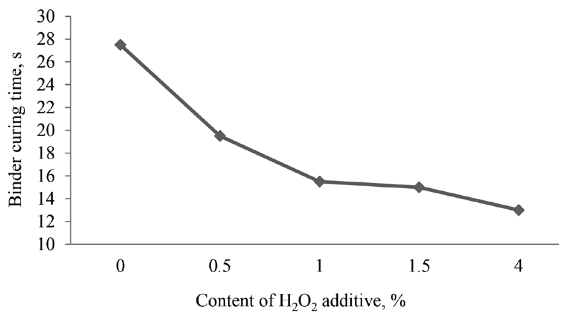


Fig. 3. Dependence of the binder curing time on the content of H_2O_2 additive

The addition of 1.0 % of H_2O_2 reduces the PFB curing time by 43.6 %. The results of determining the binder curing time confirmed the hypothesis that H_2O_2 is involved in cross-linking of the PFB. The acceleration of the binder curing time is the most intensive in the range of 0–1.0 % of the peroxide addition.

Table 2

Properties of plywood and composites on the modified binder

Content of H_2O_2 additive, %	FSF plywood			Thermal insulation composites	
	Thickness swelling, %	Static bending strength, MPa	Shear strength along the glue seam, MPa	Thickness swelling, %	Static bending strength, MPa
0	10.93	117.83	1.81	10.3	0.58
0.5	10.66	121.38	1.82	10.1	0.59
1.0	10.71	123.44	1.89	9.8	0.61
1.5	9.84	117.25	1.69	9.6	0.64
4.0	9.78	112.15	1.52	9.5	0.62

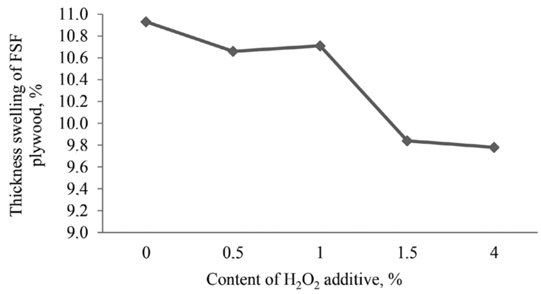


Fig. 4. Dependence of thickness swelling of FSF plywood on the content of H_2O_2 additive

The addition of 1 % of H_2O_2 reduces the thickness swelling of plywood by 2 % over 24 hours in water. The maximum increase in water resistance is realized by the addition of H_2O_2 in an amount of 1.5 %. At the same time, thickness swelling of plywood reduces by 9.97 %. A further increase in the content of the peroxide additive has a negligible effect on the reduction of thickness swelling of plywood after exposure to water.

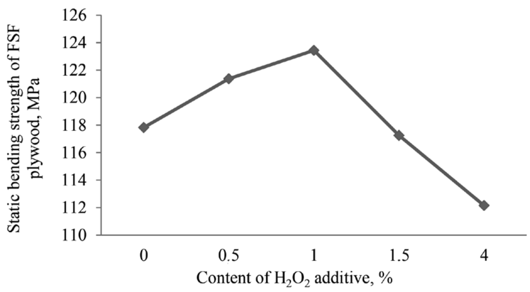


Fig. 5. Dependence of the static bending strength of FSF plywood on the content of H_2O_2 additive

The addition of 1.0 % of H_2O_2 to the adhesive composition increases the static bending strength of plywood by 4.76 %, then the static bending strength of plywood reduces. The introduction of 4 % of peroxide is excessive and reduces the static bending strength by 4.8 %.

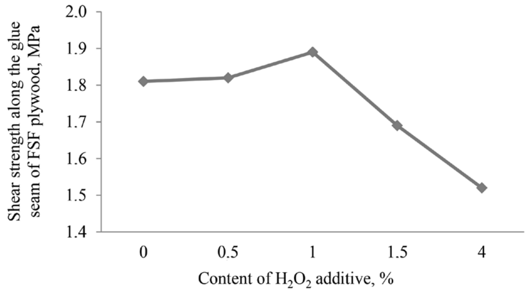


Fig. 6. Dependence of the shear strength along the glue seam of FSF plywood on the content of H_2O_2 additive

The shear strength along the glue seam of FSF plywood increases by 4.4 %, with the addition of 1.0 % of H_2O_2, then the shear strength along the glue seam of plywood decreases. Since this parameter characterizes the adhesive ability of the binder, the conclusion is that the addition of H_2O_2 over 1.0 % is excessive.

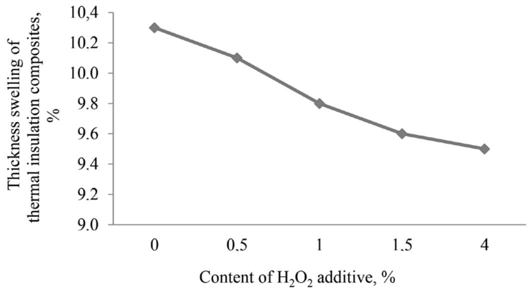


Fig. 7. Dependence of thickness swelling of thermal insulation composites on the content of H_2O_2 additive

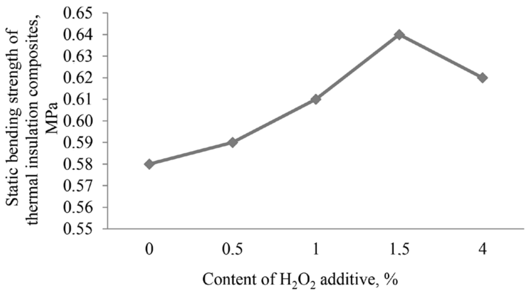


Fig. 8. Dependence of the static bending strength of thermal insulation composites on the content of H_2O_2 additive

Strength of thermal insulation composites made of cellulose-containing waste increases by 5.2 % with the addition of 1.0 % of H_2O_2 and thickness swelling decreases by 4.9 % over 24 h.

The static bending strength of FSF plywood increases with the addition of 0.5–1.0 % of H_2O_2 and then begins to decrease. The reason for this, in the authors' opinion, is the effect of H_2O_2 on both the adhesive composition and the wood components.

There is data [11] that hydrogen peroxide exposure to wood veneer initiates oxidative delignification of wood, while microcrystalline cellulose elements are formed for filler made of waste of plant annuals, such as flax and cotton [11].

At the initial stage oxidative delignification veneer has a positive effect on the elasticity of plywood, increasing the relative mobility of the lingo-carbohydrate complex elements. This process increases the static bending strength of plywood. The addition of 1 % of H_2O_2 to the PFB increases the degree of cross-linking of phenol formaldehyde resin. This increases the shear strength of plywood at the glue seam. A further increase in hydrogen peroxide addition leads to a reduction in strength.

The effect on the adhesive composition is due to the fact that an excess amount of H_2O_2 increases the amount of hydroxyperoxide anions, and the complex becomes unstable with an excessive amount of ligands. The presence of unstable complexes reduces the static bending strength of plywood by 4.8 %.

The microcrystalline cellulose presence in the polymer composition increases the composite strength [7]. Due to this, as well as due to the lower production temperature of thermal insulation composites based on the PFB modified with H_2O_2, the strength of thermal insulation boards made of cellulose-containing wastes increases in the range up to 1.5 % of H_2O_2 additive.

According to Rafáł Czarnecki and Janina Łęcka [3] modification of PFB with H_2O_2 increases the strength of particle boards; however, it has little effect on water resistance. Differences with the results of determining the water resistance of plywood and composites based on the modified binder, obtained in this work, are explained by the different type of cellulose-containing filler, as well as by the differences in technological factors of composite production processes.

Conclusion

The use of H_2O_2 as a modifying additive to phenol formaldehyde resin reduces the binder curing time and therefore makes it possible to reduce the pressing time of FSF plywood, and thus reduce the costs of plywood production.

The addition of H_2O_2 affects not only the binder cross-linking, but also veneer and cellulose-containing filler. Addition of 0.5–1.5 % of H_2O_2 improves the strength properties of FSF plywood and thermal insulation composites made of plant wastes; the addition of 4.0 % of H_2O_2 weakens the components of wood veneer and reduces the static bending strength of plywood.

The positive effect of H_2O_2 addition on water resistance of plywood and thermal insulation boards made of plant wastes was revealed. This is due to an increase in the cross-linking degree of the binder, as well as, apparently, the interaction of the hydroperoxide anion with the components of the cell wall.

The recommended hydrogen peroxide content is 1.0 % for FSF plywood and 1.5 % for thermal insulation boards. This will reduce the pressing (drying) time of the material and thus the production costs.

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