Clean air technologies: modern approaches to reducing the toxicity of wood slab materials

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Abstract. The purpose of the work was to study and analyze existing methods for reducing the toxicity of wood slab materials. The requirements for the release of free formaldehyde from plywood of chipboard and other types of boards are constantly being tightened, since this toxicant is a strong carcinogen and can contribute to the development of a number of diseases in humans. Therefore, obtaining environmentally friendly wood slab materials in the process of their manufacture and operation, in order to prevent various human diseases, is an urgent problem. The paper presents the results of experimental studies on the development of formulations of adhesives based on urea-formaldehyde resins for gluing plywood and plywood products containing a filler that can simultaneously act as a formaldehyde acceptor and glue hardener. Waste from woodworking industries – oak bark flour, birch bast flour and wood flour are recommended as fillers. In order to increase their adsorption capacity, a rational mode of acid modification of these fillers has been established (activating agent – 12% solution of orthophosphoric acid, processing time 6-8 hours, processing temperature 80^0C). Acid activation of the selected fillers in the established mode allowed to increase their adsorption capacity for formaldehyde by about 80%. The introduction of acid-treated vegetable fillers into the CFS in the amount of 2-3 wt. h. per 100 wt. h. of resin allowed to reduce the content of free formaldehyde in liquid glue by 6-12 times. The use of these adhesive compositions allows you to completely eliminate the expensive hardener from the glue formulation when gluing plywood.

1 Introduction

Wood board materials such as plywood, particle board, MDF and others are made from wood raw materials and synthetic adhesives or binders. In Russia, fairly cheap and affordable urea-formaldehyde resins and adhesives based on them are most widely used. Wood glued materials have a number of advantages over natural wood, including the production of large-format plates, equal strength and the same behavior of plates (shrinkage, swelling, warping) both in length and width of the sheet, lower cost, etc [1]. But in addition to numerous advantages, one of the main disadvantages is the toxicity of wood board materials obtained on the basis of urea-formaldehyde resins. It is due to the presence of a free formaldehyde monomer, which has a very low MPC value, which also depends on the temperature, the duration of curing of the adhesive and the hardener used. The released formaldehyde is a colorless gas with a sharp unpleasant odor. It has an irritating and generally toxic effect, and is a strong allergen [2,3]. Even at a concentration of formaldehyde equal to 0.12 mg/m3, irritation of the mucous membranes of the eyes occurs, sore throat, cough [4]. According to the results of the conducted studies, scientists suggest that formaldehyde can cause or contribute to the occurrence of cancer. The chemical effects of formaldehyde are manifested in diseases of the nervous and immune systems. Therefore, certain hygienic requirements are imposed on wood glued materials in order to prevent various diseases of people in industrial and domestic conditions [2].

2 Reasons for the release of free formaldehyde

The amount of formaldehyde released from the finished product is due to various factors: the properties of CFS, their structural features, adhesive compositions, pressing modes of wood glued materials, the influence of external conditions during operation (air humidity, temperature and air exchange rate). During the curing process, formaldehyde is formed from methyl groups and ether bridges. In addition, the structured resin is subject to hydrolytic cleavage, the degree of which is determined by the spatial structure of the resin. Thus, part of the formaldehyde released from the adhesive or binder, which is not removed from the vapor-gas mixture during the pressing process, forms a source of its emission from the finished glued products into the environment.

Free formaldehyde is released into the environment from liquid resins and adhesives based on them, during the curing of adhesives, as well as from finished products during their operation. Scientists have proven that the higher the content of toxic substances in liquid resins and adhesives, the greater the toxicity of cured adhesives.

In accordance with the current "Requirements for polymer and polymer-containing building materials and furniture" (Uniform sanitary, epidemiological and hygienic requirements for products subject to sanitary and epidemiological supervision, chapter II, section 6) - the permissible level of migration of formaldehyde into the air should not exceed 0.01 mg/m3 [6].

In accordance with GOST 10632-2014, the content of harmful chemicals released by stoves in industrial premises should not exceed the maximum permissible concentration for air in the working area of industrial premises equal to 0.3 mg/m3 [7]. The content of free formaldehyde in finished products is determined in production conditions according to GOST 27678-2014 by the perforating method and the method of gas analysis EN 717 (European standard).

The following formaldehyde emission classes are currently provided in production, as shown in Table 1.

Table 1. Impregnation composition content and contact angle wetting for birch wood

Formaldehyde emission class	**The maximum permissible norms of formaldehyde content in the plate, established by the perforating method, mg/100g abs.dry.plates**	**Maximum permissible standards for the release of formaldehyde from the plate into the air, established by the test method in the climatic chamber, mg/m3 of air Wood with out impregnation**
E 05	up to 4.0 incl.	up to 0.08 incl.
E 1	up 4.0 to 8.0 incl.	up 0.08 to 0.125 incl.
E 2	up 8.0 to 20.0 incl.	up 0.124 to 0.5 incl.

3 Main methods of reducing the toxicity of wood board materials

In the course of our work, we conducted a study and analysis of existing methods for reducing the toxicity of wood slab materials. If you look into the issue of reducing formaldehyde in manufactured products, then there are many ways to solve this problem, but each method has its pros and cons.

3.1 The use of low-toxic resins

Until recently, the most common KFS for plywood production were resins of the following brands: KF-Zh(F), KF-MT, KF-O, which did not allow the production of products with low release of free formaldehyde [8]. Currently, a very large number of low-tar low-toxic carbamide-formaldehyde resins of various brands have been developed in our country: KF-NP, KF-A, KF-60P KF-MT-15, KF-MT-10, KF-MT-15KP, KF-MT-PP, KF-MTU, KFS-1, KF-NFP, KF-015, KF-015 M, KF-02, KF-02E, KF-MNP, KF-53D and others [9], but the molar ratio of carbamide to formaldehyde (K:F) in the listed resins ranges from 1:1.3 to 1:1.12. Their use makes it possible to obtain products (plywood, chipboard, etc.) mainly of the E2 class and for individual plywood brands − E1. Among the listed resins there are evacuated KF-NFP, KF-MT and others (dry residue 63% or more), semi-evacuated and non-evacuated resins KF-60P, KF-02E, KF-53D, KF-NV, SKF-NM and others (dry residue content from 50 to 60%). The content of free formaldehyde in these resins is 0.3% (KF-MT, KF-A), depending on the brand, and 0.1% (KF-NP) in some brands [9].

The experience of using domestic low-grinding urea-formaldehyde resins with a molar ratio of urea and formaldehyde 1:1.2...1.25 has shown that they are suitable for use as a binder in the production of chipboard and plywood [10]. Thus, evacuated CF-NFP resin and semi-evacuated KF-60P, KF-N-54, KF-NV, KF-NP resins allow the manufacture of slab materials, providing toxicity classes E1 and E2 [5,11]. However, when using the CF-NFP resin developed by TSNIIF LLC, a number of problems are noted: the complexity of resin synthesis, the impossibility of transporting it due to limited viability (shelf life), practical unsuitability for plywood enterprises using cold pressing of veneer packages, a decrease in the production capacity of workshops, and most importantly, a decrease in bonding strength [5]. KF-MT, KF-MT-U, and KF-MT-15 resins have the same disadvantages in plywood production, although they provide a reduction in toxicity to 10 mg per 100 g of absolutely dry plywood. New types of domestic carbamide-formaldehyde resins KF-A, KF-115-53, KF-115-55 have also been developed, which are synthesized by deep condensation in an acidic environment without the formation of wastewater. But in the production of such grades of resins, there is a high gas content of the air with methanol (up to 100 mg/m3 at PDKworking area = 0.5 mg/m3) [5,11].

In Veliky Novgorod, Acron synthesized carbamide-formaldehyde resins of the new generation KF-EU(D), KF-EU(F), with a free formaldehyde content of 0.05%, the use of which allows to obtain plywood of class E1 while reducing binder consumption as a result of improving the properties of the film structure of the cured polymer, and chipboard on These resins are produced of class E1 and E2 [1, 2]. When using KF-EC(F) resin for plywood production, the bonding strength is at least 1.5 MPa. In order to reduce toxicity in liquid urea-formaldehyde resins at chemical enterprises producing them, the content of free formaldehyde is reduced by introducing compounds binding formaldehyde and urea into the resin at the last stage of condensation. A formulation and technology for the synthesis of an innovative urea-formaldehyde resin with a molar ratio of F : K=0.9 : 1.0 have been developed. Chipboards on this resin with 1% formaldehyde in the hardener have a formaldehyde content of 3.7 mg/100 g. This meets the requirements of emission class E0.5.

3.2 Changing the gluing and hot pressing modes

The temperature and duration of pressing have a great influence on the release of formaldehyde from wood slab materials. An increase in the temperature and duration of pressing contributes to an increase in the release of formaldehyde from the plates during their pressing, and therefore reduces its amount in finished plates [1].

3.3 Using methods of processing finished slabs

It is possible to reduce the content of free formaldehyde in finished products not only during their manufacture. It is possible to use methods to reduce toxicity by post-pressure treatment. For example, the FD–EX method of the Vercors company (Belgium) is known based on three-stage processing of finished slabs in chambers sequentially arranged one after another. First, the plate is treated with gaseous ammonia, and then the ammonia is removed from the surface layers of the plates by ventilation. At the last stage, the stove is treated with air saturated with formic acid vapor to avoid the release of ammonia from the stove. As a result of the reaction, formic acid ammonium is formed, which then reacts with free formaldehyde located in the inner layers of the plate. Thus, the emission of formaldehyde into the environment is reduced. According to the Verkor company, the physical and mechanical properties of the plates do not change after such processing of the plates in the chamber. It is possible to place the plates in the chamber immediately after their manufacture, as well as from the finished product storage warehouse [13].

Another method of processing plates in an ammonia atmosphere is also known. The treatment of slabs in an ammonia atmosphere is based on the exposure of slabs indoors in an ammonia atmosphere. The amount of ammonia is about 0.8...1.0% of the mass of the plates. This processing method reduces the amount of formaldehyde released from finished plates by 3.3 times. The author suggests [5] that ammonia binds latent acids that are found in wood particles and thereby helps to reduce the release of formaldehyde from slabs.

It is possible to reduce the content of free formaldehyde in finished particle boards by applying the method of processing plates with moisture and sulfur dioxide. To do this, the plates must be placed in a chamber into which steam is supplied. Mixing with air, steam is distributed by fans over the volume of the chamber (about 13 kg of water per 1 kg of dry air). Then sulfur dioxide is fed into the chamber (the ratio of sulfur dioxide to air is 50:106). When using this method, the plates are kept in the chamber for a fairly long time - about 80 hours. But it should be noted that after such treatment, the release of free formaldehyde from chipboard decreases by 50...75% [1].

Other methods are also used – treatment with carbamide dissolved in polyvinyl alcohol, casein or hydrazine sulfate, dispersions, polyvinyl alcohols, treatment with hot and cold air. Some of these methods are based on the evaporation of moisture from the plates, along with which free formaldehyde is removed when blown with air. Others are based on the binding of formaldehyde released from the plates with a chemical substance. But it is worth noting that such methods have not been widely used, since they reduce the release of formaldehyde to a small extent.

3.4 Finishing by applying films, paints, veneers Using methods of processing finished slabs

It is known that finishing with veneer, paper, polymer films, varnishes, and paints leads to a decrease in the release of free formaldehyde from wood slab materials. With such finishing methods, it is possible to reduce formaldehyde emissions up to 6 times compared to control samples. is possible to reduce the content of free formaldehyde in finished products not only during their manufacture. It is possible to use methods to reduce toxicity by post-pressure

3.5 Modification of resin or binder with various additives

Currently, a large number of different formaldehyde acceptors have been developed and widely used in the domestic industry as additives to resin, hardener. According to many scientists, this method of reducing the toxicity of wood slab materials is the most effective. The most common acceptors of formaldehyde are carbamide, ammonia water and other amino compounds [1,9,14]. Under certain conditions, these acceptors make it possible to obtain wood glued materials and plates of class E1, but in most cases their use leads to a deterioration in physical and mechanical parameters [4]. If acceptors are used in small quantities, then a sufficient reduction in toxicity is not provided [14].

In order to solve these problems, at one time VNIIDrev developed and tested in production conditions a formaldehyde acceptor of the AF brand (TU OP 13-0273643-94-93), which provides the possibility of obtaining low-toxic particle board. The acceptor is a white or slightly colored crystalline powder. In its composition, in addition to traditional carbamide, there are other components that provide, along with the binding of free formaldehyde, a more in-depth structuring of the resin during its curing. The advantages of this acceptor also include non-toxicity, non-deficiency and the comparative cheapness of its components [14]. The use of this acceptor does not lead to a significant deterioration in the physical and mechanical properties of the plates. The AF acceptor can be used both in a solid state and in the form of an aqueous solution (depending on the specifics of production). When using a powdered acceptor, processing lines must be equipped with additional dispensers of bulk materials. When using an aqueous solution, it is necessary to combine the operation of its application with the introduction of a combined hardener and, as a result, it is necessary to maintain certain conditions in the binder supply system necessary for the use of highly concentrated solutions.

To solve the problem of the safety of wood slabs, it is possible to use melamine as additives of adhesives and impregnating resins. For a long time, Russia simply did not have its own production of this substance. The first melamine production started working only in 2012 – its capacity was 50 thousand tons per year.

In [11, 15], in order to reduce the toxicity of particle board based on KFS, aluminosilicates with a rigid frame structure of natural origin - ASP were used as a modifying additive. The ASP additive was introduced into the resin of the KF-MT-15 brand in an amount of 5-15% by weight of the resin with a formaldehyde content of 0.15-0.25% directly during the preparation of the binder before pressing. It has been experimentally established that when aluminosilicates are introduced into the binder in an amount of 10-15%, the degree of purification from formaldehyde is 25-30% with a single use of the additive. The effect of the sequence and number of resin modifier treatments, as well as the duration of their contact, on the toxicity of the resin was studied. The authors [11] found that the best results were obtained when the resin was treated with an ASP modifier three times for 3 hours, in this case, the toxicity of the initial resin decreases from 0.15-0.25% to 0.05-0.06%. At the same time, the degree of purification of KF-MT-15 resin from formaldehyde is 60-80%. The advantages of introducing this modifier into the resin include the fact that it does not worsen the physical and mechanical properties of particle board (strength indicators even increase), as well as the fire hazard of the latter decreases. Thus, when TSA is introduced in an amount of 5-15% into a urea-formaldehyde binder for particle board, the formaldehyde emission from them is no more than 8 mg/100 g of the plate, which corresponds to class E1. The disadvantages include the fact that when introducing aluminosilicates of different deposits, as well as different degrees of dispersion, it affects the activity of reducing the toxicity of CFS in different ways.

There are known works to reduce the toxicity of CFS-based adhesive compounds using lignosulfonates as a modifying additive [16]. The use of technical lignosulfonates (OST 13-183-83) or ammonium-based alkalis, which are by-products of pulp and paper production by the acid sulfite method, is promising. Lignosulfonates are physiologically acceptable, completely non-toxic, have a pleasant smell of coffee, and are colored dark brown. They combine well with CFS and are well diluted with water, have a thickening ability, which allows you to use kaolin in smaller quantities, or completely abandon it. Lignosulfonates are reactive, characterized by low viscosity with high dry matter content.

The addition of 10-20% lignosulfonates (LST) to the KFS reduces the release of a vapor-gas mixture during polycondensation, including formaldehyde, by 2-2.5 times. Moreover, the toxicity of the adhesive compound and the gas contamination of the air in the workshop with free formaldehyde are reduced. The introduction of lignosulfonates into a carbamide-formaldehyde binder during the selection of simultaneous selection of hardeners (ammonium persulfate, aluminum sulfate, sodium hydrosulfate) and technological modes of plate manufacturing allows to obtain chipboard of the E1 emission class.

In the studies conducted by the St. Petersburg Forestry Academy, modified lignosulfonates (MLS) on a mixed calcium-sodium base were used, in which calcium ions were replaced by aluminum ions. The best results were obtained when using MLS in combination with ammonium orthophosphates and carbamide. At the same time, the emission of formaldehyde decreased from 4.5 to 2.7 mg/g of resin. The use of these compositions for particle board showed a decrease in the release of formaldehyde by 1.5-2.0 times while maintaining the physical and mechanical properties of the plates [10]. These works [10, 17] also provide information that LST and MLS have sufficient stickiness and can partially replace resin from 20 to 40%.

Doronin Yu.G., Kondratiev V.P. [11,18] resorcinoformaldehyde resin RM-1, with a minimum content of free phenol and formaldehyde, developed at NPO Nauchfanprom, is considered as a modifier of urea-formaldehyde resins in the production of plywood. RM-1 resin was introduced into the KFS in order to increase the water resistance of plywood, as well as to reduce its toxicity. When RM-1 was introduced into low-tar resins such as KF-MT, KF-NFP, etc., low-toxicity plywood "Carbophane C" was obtained, corresponding to classes E1 and E2, depending on the brand of resin. But the introduction of RM-1 has its drawbacks, because the additive is scarce and expensive, and this, accordingly, leads to a significant increase in the cost of plywood products. The introduction of 15 wt. h. RM-1 per 100 wt.h. of carbamide resin into the resin is optimal for reducing the emission of formaldehyde from cured adhesives, and with an increase in the amount of RM-1, the release of formaldehyde from samples increases sharply, since the curing time of the binder increases and at the same time the activation energy of the glue curing reaction increases.

In the works of Paznikova S.N. and Leonovich A.A., it was proposed to reduce the toxicity of CFS by modifying it, during synthesis, with inorganic electrolytes [19,20] (LiCl, NaCI, KCI, NaBr, NaJ, KBr, KJ, CaCI, MgCI2) and the OHN reagent. It has been established that low-toxic particle board with improved performance properties are obtained using carbamide-formaldehyde binders synthesized in the presence of sodium chloride or potassium chloride. At the same time, the mass fraction of free formaldehyde in modified resins is 0.07-0.08%, but the technological characteristics deteriorate slightly.

Abroad, in order to reduce toxicity, modifying additives such as urea, resorcinol, a mixture of urea with resorcinol, polyvinyl alcohol, borohydrites of alkaline and alkaline earth metals, as well as salts of amines and imines formed by sulfuric acid and phenolsulfonic acids with urea, dicyandiamine, melamine and other amines are also introduced into the resin

[21,22]. But data on the use of the above-mentioned KFS modifiers were found only for the manufacture of environmentally friendly particle board.

In Japan (Japanese patent No. 5217048), a method has been proposed for the introduction of carbamide and carboxymethylcellulose (CMC) into the binder in the amount of 100 m.h. urea and 4 m.h. CMC per 1000 m.h. binder. An additional 50 m.h. of wheat flour (smet) is added to the binder (hardener is a 20% solution of ammonium persulfate). The release of formaldehyde from the binder 2 days after its curing is only 0.00019% [1].

In the works of Perminova D.A. [23], to reduce the release of formaldehyde from chipboard, it was proposed to modify carbamide-formaldehyde resins with safe and affordable additives such as glyoxal and glycouryl. Moreover, with the help of glycouryl, free formaldehyde is bound, and glyoxal is necessary for the formation of a spatial grid at the stage of hot pressing. The proposed method for modifying the adhesive composition with glyoxal, in an amount from 0.5 to 1% by weight. It allows for a 20% reduction in thickness swelling and a 40% increase in the tensile strength of the chipboard. Resins modified with glycoluryl, produced according to the method developed by scientists, make it possible to obtain chipboard with a formaldehyde content 33% lower than the base one at the enterprise without changing the technological cycle of pressing and preserving the physico-mechanical characteristics of particle board [23].

At VGLTU, in the works of Razinkov E.M. and other [24,25], when developing the technology for the production of non-toxic biosecurity particle board, modifying chemical additives were studied, representing a chromomednobor preparation (HMBB-3324) and sodium pentachlorophenolate (PCPHN) belonging to the group of chlorophenolic compounds. In addition to these drugs, the effect of ammonium silicofluoride (CFA), which is a formaldehyde acceptor, antiseptic and hardener for CFS, is considered. It is advisable to introduce CFA and PCFN into particle board together with resin, while these preparations do not worsen the physical and mechanical properties of the plates.

3.6 Introduction of formaldehyde adsorbent fillers

Traditionally, in the production of plywood and plywood products, fillers are added to the glue that change their properties, such as viscosity, shrinkage of the adhesive seam, strength properties, etc. It turned out that when various types of fillers are introduced, they may also have an adsorption activity to formaldehyde (equal to 15-17%), but this indicator is not high enough for the prospective use of adsorbents in industrial conditions. It is necessary to enhance the adsorption capacity of adsorbent fillers due to thermal and acid treatments [9].

Our research consisted in evaluating the adsorption capacity of fillers that can simultaneously perform the function of an active filler of adhesives and a formaldehyde acceptor, as well as the choice of modes of their modification. As fillers of urea–formaldehyde adhesives for gluing plywood, we recommend waste from woodworking industries - oak bark flour, birch bark flour and wood flour (it is possible to use grinding wood flour) [26,27,28]. In order to increase their adsorption capacity, we have established a rational mode of acid modification of these fillers (activating agent – 12% solution of orthophosphoric acid, processing time 6-8 hours, processing temperature 800C). Acid activation of oak bark flour, birch bark flour and wood flour in the established mode allowed to increase their adsorption capacity for formaldehyde by about 80% [29,30].

In order to reduce the toxicity of urea-formaldehyde glue, adhesive compositions have been developed and recommended for practice, which differ from those traditionally used by introducing acid-treated oak bark flour (2.5-3 wt. h. per 100 wt. h. resin) or acid-treated wood flour (2-2.5 wt. h. per 100 wt. h. resin) or acid-treated birch bast (2.5-3 wt. h. per 100 wt. h. resin), which reduced the content of free formaldehyde in liquid glue by 6-12 times (compared with the glue recipe without adding filler). The use of these adhesive compositions makes it possible to completely eliminate the expensive hardener from the glue formulation when gluing plywood [31]. To regulate the viability of adhesives with acid-treated fillers (if they are introduced in an amount of more than 2 wt.h. per 100 wt.h. resin), it is advisable to additionally introduce 1 wt.h. ammonia water of 20% concentration into the glue. Ammonia water (NH_4OH) also acts as an acceptor of formaldehyde, further reducing its amount in liquid glue. We controlled the content of free formaldehyde in liquid adhesives using the sulfite method.

We also conducted studies to determine the release of free formaldehyde from finished plywood by the laboratory chamber method [9], according to the results of which it can be concluded that the introduced acid-treated vegetable fillers reduce the emission of formaldehyde from finished products by about 1.5 times and ensure the production of E1 emission class products. It should be noted that the advantages of introducing fillers treated with orthophosphoric acid into the resin include the fact that they do not worsen the physical and mechanical properties of plywood (strength indicators even increase).

4 Conclusion

Having analyzed modern approaches to reducing the toxicity of wood board materials made on the basis of urea-formaldehyde resins, it can be concluded that the methods used are very diverse and have varying degrees of effectiveness. Using some methods allows you to significantly reduce the toxicity of finished products, but at the same time its cost increases significantly. When using others, difficulties arise with the organization of the technological process, etc. The use of low-toxic resins often leads to a decrease in the physical and mechanical properties of slab materials. In our opinion, the most promising approach to solving this problem is the use of cheap, affordable and environmentally friendly modifiers and fillers of carbamide-formaldehyde resins capable of absorbing formaldehyde and firmly retaining it in the production and operation of wood glued materials. The main task facing the developers is to reduce the toxicity of finished wood slab materials at low cost, so as not to increase the cost of finished products.

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