IOP Conference Series: Earth and Environmental Science

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To cite this article: E M Razinkov *et al* 2019 *IOP Conf. Ser.: Earth Environ. Sci.* **226** 012026

IOP Conf. Series: Earth and Environmental Science 226 (2019) 012026 doi:10.1088/1755-1315/226/1/012026

Production of environmentally friendly wood boards from the wastes of the forest complex enterprises

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Abstract. The substances of complex action are used to obtain environmentally safe wood boards in the technology of wood-based panels, which are simultaneously acceptors of formaldehyde, antiseptic or resin hardener. Such substances include: sodium pentachlorophenolate, chromium-copper boron compound and silicon fluorine ammonium. We have received copyright certificates for inventions or patents for all these substances. The developed technology of environmentally friendly wood-based panels was tested in industrial conditions with positive results. Plates are obtained meeting the environmental safety of the best classes of formaldehyde emissions and meeting the technical requirements for the plates on the strength properties.

1. Introduction

According to the technical requirements, particle boards produced by the Russian industry are intended for the production of furniture depending on the class of formaldehyde emission (gas, which is harmful for humans) - (E0.5; E1; E2): for children, for educational institutions, for preschool institutions and other furniture (E0.5); household furniture for public premises and products intended for the use inside residential and public buildings and premises (E2); for the production of other products, except furniture (E3). Boards for the construction purposes (floor constructions, paneling of low-rise wooden houses, etc.) do not meet the operational requirements, although boards are used for construction in foreign practice. According to the data of, the global demand for particle boards has increased since 2012 to 2016 from 75 to 88 million m^3 per year. In Russia, the volume of particle board production amounted to 7.4 million m^3 in 2015[1].

The production of wood-particle boards is promising in terms of the use of fine-grained raw materials and waste of the forest complex enterprises in the technology. It should be noted that the main drawback of particle boards is toxicity associated with the release of harmful gas (formaldehyde) in the production and operation of boards. In accordance with Russian technical requirements [2-4] commercially available boards should (on ecological safety) conform to formaldehyde emission classes E0.5, E1, E2 (Table1).

Table 1. Maximum permissible norms of formaldehyde release into the air depending on the class of emission of non-laminated particleboards.

Class of formaldehyde emission	Maximum permissible norms of formaldehyde content in the panel, established by perforator method, mg/100 g abs.	Maximum permissible rate of formaldehyde release from the panel into the air, established by the test method in the climatic chamber, mg/m^3 of air
E0.5	Up to 4.0	Up to 0.08
E1	From 4.0 up to 8.0	From 0.08 up to 0.124
E2	From 8.0 up to 20.0	From 0.124 up to 0.5

Note to Table 1: the maximum permissible rate of formaldehyde content in the plate, set by the perforator method in mg/100 g of absolutely dry board, is determined by the perforator method corresponding to European EN 120: 1992 [5]. The maximum permissible rate of formaldehyde emission from the plate into the air are established by the method of testing in the climatic chamber in mg/m^3 of air (simulating the conditions of operation of the plates) EN717-1:2004 [6], EN 717-2:1994 [7].

According to the data of the accredited testing laboratory of Lessertica LLC, the release of formaldehyde from ready-made industrial non-laminated plates according to the chamber test method (EN 717-1) was by years (mg/m^3 of air): 2012 - 0.27; 2013 - 0.034; 2014 - 0.24; 2015 - 0.15. According to the results of particle board tests (from needle chips and from large chips) made at 28 enterprises in 2015-2016, it was found that the release of formaldehyde from particle board made of needle chips was averaged at 0.17 mg/m3 of air, which exceeds the permissible level (0, 01 mg/m3 of air) by 17 times, and the release from particle board made of large chips is 0.15 mg/m3 of air, which exceeds the permissible level by 15 times. All this significantly hinders the export delivery of particle boards.

Formaldehyde (formic aldehyde, methanol, oxomethane) is a colorless gas with a sharp specific smell; it is soluble in water. Molecule of formaldehyde contains one carbon atom, two hydrogen atoms and one oxygen atom. The density is 0.815 g/l, the boiling point is -2 °C; the melting point is -92 ºC. Limit of explosion with air is -70.0-72.0 vol. % [8]. Formaldehyde irritates mucous membranes of the respiratory tract and eye membranes of humans. It may also cause inflammation of skin mucous membranes and its allergic reactions.

In our country, there are strict requirements for the formaldehyde content in the air and at present its permissible level (PL) should be no more than 0.01 mg/m^3 of air.

This value of PL is included in the requirements for furniture made of board materials and included in the decision of the Customs Union Commission from 28.05.10, No. 229 for furniture and wood boards in the List of materials subject to sanitary and epidemiological supervision of 07.04.2011, No. 620. It is asubject to mandatory conformity assessment.

In addition, the above value of the PL of formaldehyde was included in the technical regulation On safety of furniture products, approved by the Russian Government on December 22, 2011.

The purpose of this work was to determine the maximum saturation of the air during the operation of structures made of particleboard in residential premises with the condition of ecological safety, as well as the search for effective formaldehyde acceptors of complex action reducing formaldehyde emission from plates and simultaneously improving other properties.

2. Experimental part

The emission of formaldehyde from the panels is mainly due to the presence of synthetic formaldehyde-containing resins. Such resins are urea formaldehyde (grades KF-MT-15, KF-NFP, etc.) and phenol-formaldehyde (SBJ-3014, SBJ-3024, etc.).These resins are low-toxic, which contain about

0.10 (SFJ-3014) and 0.05% (SFJ-3024) free formaldehyde. CFS contain a greater amount of free formaldehyde (up to 0.15%). The presence of free formaldehyde in these resins is mandatory, otherwise their adhesive ability is low and the resin is not suitable for use [9].

It is also known formaldehyde can be released from wood particles during their drying that in particle board technology. Thus, when drying spruce, pine and beech wood particles in the nozzle dryers at temperatures at the inlet and outlet of which is 280 and 140 ºC, respectively, the formaldehyde content in the exhaust air reaches 6.5 mg/m³. There are ways of formaldehyde formation from lignin, and also from polysaccharides by oxidation of starch and dialdehyde starch.

All of the above resins are the product of polycondensation of urea with formaldehyde or phenol with formaldehyde. Part of formaldehyde, for example, in the macromolecule of urea formaldehyde resin, is associated with urea through methylene (- CH2 -) or ether (- CH2-O-CH2 -) bridges. Depending on the conditions of polycondensation of the linear chain of the resin have one or two metrolinie end groups (-CH2OH-). The rest of the formaldehyde is in the resin in an unbound form and is easily released in the form of gas which is free formaldehyde.

Spatial structure of the resin occurs in the process of curing at an elevated temperature. Fragile essential bridges and end metallocene group move to a stable methylene with simultaneous separation of formaldehyde.

One part of free formaldehyde, together with water vapor, partially comes out of the boards during hot pressing. Another part penetrates into the voids in the board and remains there in the form of polioxymetheleneglycole or in the free form. The third part of formaldehyde is released into the environment from the finished boards during their operation.

The chemistry of formaldehyde release from resins is well studied by a number of researchers, we will not dwell on the results they obtained. Let us focus only on those that are not yet known, but which we have obtained experimentally and are of considerable interest for the studied question.

These resins are low-toxic, which contain about 0.10 (SFJ-3014) and 0.05% (SFJ-3024) free formaldehyde. CFS contain a greater amount of free formaldehyde (up to 0.15%).We have conducted microtechnologies experimental studies on model samples of chipboard and binder by scanning electron microscopy on the microscope "Karl Zeiss Jena" for the purpose of determining the state of the cured binder in the board. The results has showed that the cured mass of the binder in the particle board has a bumpy surface with a large number of voids (Figure 1), as well as cracks and bubbles. These bubbles can remain not destroyed (not burst), but can burst (Figure 2). At the same time, from the burst bubbles, the vapor-gas mixture can be additionally released together with formaldehyde.

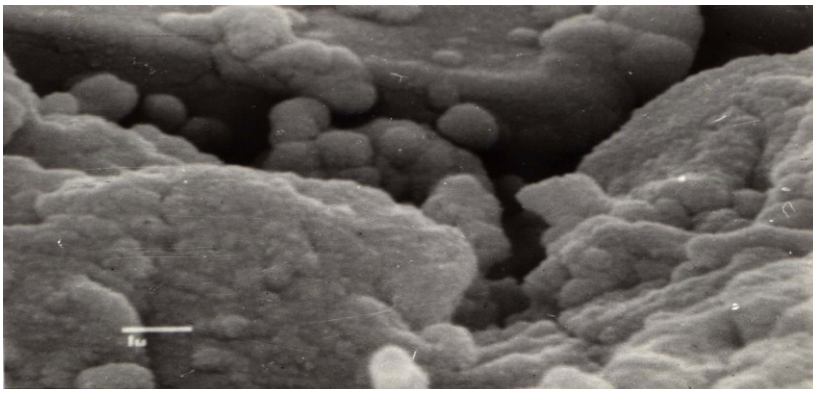

Figure 1. The view of bumpy surface area of the cured binder.

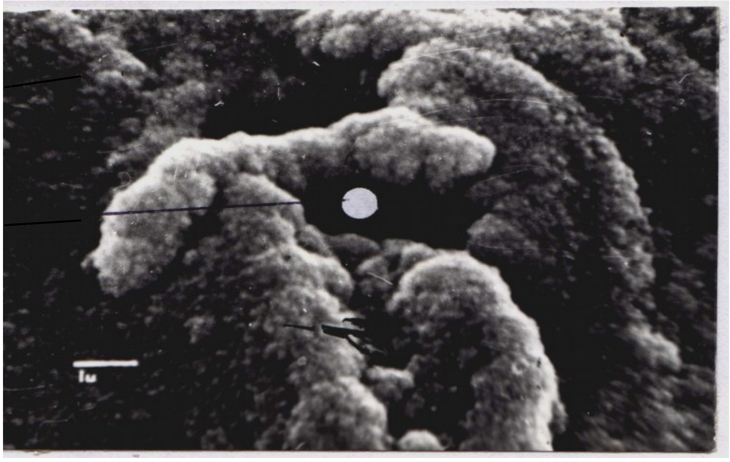

Figure 2. The surface of the chips in chipboard coated with the cured binder with a view to the not destroyed vapor-gas bubbles in the binder.

The separation of formaldehyde from the boards may be due to a change in the structure or condition of the cured adhesive layer between the wood particles in the board. The change in the structure of the layer is due to the deformation of the finished boards while manufacturing products out of them. There is a potential for additional formaldehyde release from the boards due to this change. It is difficult to quantify the amount of such emission, but it is possible, what we are doing now.

Thus, the cured adhesive layer between the wood particles is not monolithic one, but it is saturated, as shown above, with voids ("bubbles") in which the vapor-gas mixture together with formaldehyde is encapsulated. At mechanical deformation of plates (at cutting, transportation, assembly, etc.) these "bubbles" are destroyed and formaldehyde can be allocated from them in addition.

The allocation of formaldehyde from the boards is influenced by a large number of factors.

Molar ratio of components in the production of resins is one of the main factors influencing the content of free formaldehyde in finished resins. At present, low-toxic resins with a molar ratio of urea to formaldehyde from 1:1 to 1:1.28: KF-02M; KF-02; SK-75; KF-MT-15KP; KF-MT-15; KF-01; KF-NFP with a free formaldehyde content of 0.10-0.15 % are obtained. A significant number of works [10-12] indicate that the higher the content of free formaldehyde in the resin, the stronger its emission.

To reduce the release of formaldehyde from resins, a method of their modification is used which is adding melamine, terpene and other acids to the resins. The duration of tar storage influences the content of free formaldehyde in resins. The reason for this is the deepening of the process of polycondensation of resins. Consumption of resin or its content in particle boards has a great influence on the release of formaldehyde from finished boards. The amount of formaldehyde released from the boards is not proportional to the amount of resin added to them, but the more resins in particle boards, the higher the toxicity of the boards. The concentration and completeness of the curing also has an effect on the release of formaldehyde from the boards. Increasing the concentration of resin helps to reduce the release of formaldehyde from the boards. The temperature and duration of pressing have a great influence on the selection of formaldehyde from the panels Increasing the temperature and duration of pressing increases the release of formaldehyde from the panels during their pressing, and therefore reduces its amount in the finished panels. Chip drying mode also affects the separation of formaldehyde from the boards. During drying of wood particles formaldehyde is released due to thermal and hydrolytic reactions of wood decomposition. The ambient air temperature has a significant effect on the selection of formaldehyde plates. With the increase in temperature, the isolation of formaldehyde from the panels also grows. Relative humidity affects the concentration of formaldehyde in the air of the room in which the boards are operated. Increasing the relative humidity of the air above 60 % leads to an increase in the concentration of formaldehyde in the air, and lowering it below 60 % to a decrease. The value and multiplicity of air exchange affects the concentration of formaldehyde in the air. With the increase of air exchange, the concentration of formaldehyde in the air decreases, and the intensity of its release increases. The use of methods of processing and finishing of the boards is one way of reducing the emissions of formaldehyde from boards.

The type and brand of resin play a large role in the preparation of low-toxic particle boards. In domestic practice CFS are used in the vast majority due to the availability and cheapness of raw materials for their production. However, as it is indicated above, the resulting boards have increased toxicity, which allows enterprises to produce plates of E1 and E2 classes. Despite the significant increase in the cost of particle board, the production of low-toxic E0.5 class boards has begun in Russia. The process is pushed by the Swedish IKEA concern , placing orders in Russian factories, and increased requirements for export products. The production technology of E0.5 plates is mastered and introduced at 7 plants: Volgodonsk plant of wood-based panels; The first TLD; Dyadkovo-DOS; EggerDrevprodukt Gagarin LLC; Ltd. IKEA Industry Novgorod; Kronoshpan; Syktyvkar Plywood Factory. These plants use expensive melamine-containing urea resin with melamine content of up to 30%. However, the cost of such plates is high due to the high cost of resin.

Saturation of air volume by boards. Saturation, in common terminology, is the ratio of the surface area or mass of the material to the volume of the room. The increase in the saturation of the air volume by boards leads to an increase in the concentration of formaldehyde in the room. For example, if the boards with the phenol-formaldehyde resin STJ – 3014 three months after manufacture and the air temperature in the chamber of 56 °C released 0.010 mg/m³ of formaldehyde of air at a saturation of 0.4 m²/m³ of air (only floor is made of boards), then at a saturation of 1.9 m²/m³ it is 12 times more- 0.0120 mg/m³.

During the operation of particle board (mainly in the construction of furniture) in residential premises one of the main factors of their use is limiting (allowable) saturation of the volume of air in the room (ratio of the area of the plates to the volume of air in the room m^2/m^3 of air) until the concentration of formaldehyde in the air reaches permissible level (0.01 mg/m^3 of air).

We have obtained a formula for determining such saturation for which we have used a real example. Analytical calculations to determine the maximum saturation with the plates of the air volume of residential premises is made under the following conditions of operation of the boards:

- the company produces E1 class emission boards;
- living area -20 m^2 (S_{la}, m^2);
- room ceiling height -2.5 m^2 (h_c, m);

Where does the volume of the room – 50 m^3 (V_r, m^3).

When testing the release of formaldehyde from the plates by the chamber method, the saturation with the plates of the air volume (we show this saturation as S_{test}, m^2/m^3 of air) is (with technical requirements) provided for:

- cabinet furniture, tables, beds of shield construction (0,95 – 1,05 m^2/m^3 of air);
- furniture for sitting and lying, beds with soft backs and elements (0.285 – 0.315 m^2/m^3 of air);
- floor coverings and wall panels (0.38 – 0.42 m^2/m^3 of air).

According to the Russian technical requirements, the rate of emission from the plates of the formaldehyde of E1 emission class should not exceed 0.124 mg/m3 of air (we show it as E1 MPC) at saturation S_{test} =1 m^2/m^3 of air.

If we take into account the maximum permissible concentration (MPC) or the permissible level (PL) of formaldehyde in the air of residential premises 0.01 mg / m3 of air according to sanitary standards (we denote it as MPC_{tox}), and the maximum saturation with the plates of the room air volume (H_{E1}, m^2/m^3 of air) then in its final form the limiting saturation with the plates of the volume of the room (P_s in m^2/room) will look as follows:

P_s =[($S_{test} \times MPC_{tox}$): MPC_{E1}] × V_r, m^2 /room, or

P_s =[($S_{test} \times MPC_{tox}$): MPC_{E1}] × S_{la}xh_s, m^2 / room

Where P_s - limit saturation of room volume with boards, m^2 particle board/room;

S_{test} - specified saturation by plates with air volume when tested using chamber method, m^2/m^3 of air;

MPC_{tox} - MPC (PL) formaldehyde in the air of residential premises (0.01 mg/m^3 of air) according to sanitary standards, mg/m^3 of air

MPC_{E1} - permissible formaldehyde emission rate from boards of a certain formaldehyde emission class, mg/m^3 of air

V_r - volume of the room, m^3;

S_{la} - living area, m^2;

$H_{s,}$ - room ceiling height, m.

Our calculations has showed that in a room with an area of 20 m^2, only boards of the E0.5 class of emission with an area of only not more than 6.25 m^2 can be used safely , i.e. only one desk can be installed in a room. When using plates of E1 and E2 classes, such a restriction on the board area is even more significant.

When there is only furniture for sitting and lying in the room, beds with soft backs and elements from particle board of E1 class, the maximum saturation of the room volume with plates is also very low, it is only up to 1.27 m^2/room, and if there are floor coverings made of partocle board (of the same emission class) saturation is only up to 1.69 m^2/room.

It is shown by domestic and foreign experience that the way to reduce the emission of formaldehyde from the boards is the use of technology in the formaldehyde acceptors, i.e. chemicals that can react with formaldehyde during the manufacturing of boards [13].

The complexity of solving the problem of searching for such substances lies, first, in the fact that formaldehyde interacts with a limited number of chemical substances, many of which are difficult to access, expensive and require special preparation, according to well-known scientific publications. Secondly, not all of the known chemicals can be used in the technology of board production for various reasons – they may be volatile at raised temperature, explosive, etc.

At present, in order to reduce the amount of formaldehyde liberated from the boards, it is recommended to introduce ammonium water into them. At the raised temperature (105-180°C), ammonia water decomposes into ammonia and water. Ammonia reacts with formaldehyde to form hexamethylentetramine (urotropine) and water. The introduction of ammonium water in the amount of 1-3 % of the mass of liquid resin makes it possible to reduce the release of formaldehyde from the plates by 20-40 %. However, this method of reducing the release of formaldehyde on urea-formaldehyde resins from boards is undesirable. The introduction of ammonia water slows the hardening of the resin. Ammoniac water can be added only to the outer layers of the plates, it is not advisable for internal layers, since this reduces the productivity of the board production department.

Urea is also used, which is added to the resin or applied to the surface of the plates. Urea derivatives are also used – ethyl sulfide and other compounds. Addition of urea to urea-formaldehyde resin causes a decrease in the molar ratio of carbamide to formaldehyde, and thus the binding of free formaldehyde. However, physical and mechanical properties of the plates deteriorate. The addition of urea or ammonia to urea-formaldehyde resins imparts brittleness to the resins. In order to avoid brittleness of the resin, it is necessary to introduce specifically synthesized urea-formaldehyde oligomers into the resins.

The formaldehyde released from the particleboards reacts with the sulfur dioxide gas in the air in the presence of saturated steam. In addition, some sulphites and sodium silicates react with formaldehyde. Reduction of the release of formaldehyde materials is possible due to the acceleration of the condensation of formaldehyde with organic catalysts. Condensation of formaldehyde with itself leads in the presence of calcium hydroxide to sugar-like oxalaldehyde and oxyketones.

Despite a fairly large number of works, mostly foreign, concerning the search of chemicals that reduce considerably the release of formaldehyde from the boards, effective substances have not yet been found.

3. Results and Discussion

As our research has shown, the most effective method of reducing the release of formaldehyde from boards is the use of chemicals with complex action in the technology of plates. In this case the problems of both reducing formaldehyde from plates and giving them other important properties such as biostability, etc. are solved simultaneously.

Sodium pentachlorophenolate, as the most effective preparation, is an effective antiseptic at the same time. It is advisable to introduce it into boards together with a binder (mainly urea-formaldehyde). It dissolves in water, making it, however, to become alkaline one. Obtaining the desired gelling time of the resin at 100 °C, which characterizes the rate of hardening of the binder, is provided by adding higher (2.2-2.5 times) than usual amount of hardener – ammonium chloride. The hot pressing mode does not differ from the mode in the board production without the addition of sodium pentachlorophenolate. The content of sodium pentachlorophenolate in the slab should be 0.8 to 2.0 % of the mass of absolutely dry chips. The rate of dissolution of sodium pentachlorophenolate in water contained in the resin is significantly increased by feeding it to the resin for mixing with it in the form of a dense aqueous solution (in a weight ratio of sodium pentachlorophenolate: water equal to 1:1).

Chrome-copper-boric compound ChCB-3324, including sodium dichromate, copper vitriol, borax, boric acid in a ratio of 3: 3: 2: 4 is also a good antiseptic. It is applied to the shavings in the form of an aqueous solution, followed by chip drying. Its content in the slab should be from 1.0 to 1.5 % of the mass of absolutely dry shavings.

Ammonium silicofluoride, being a good acceptor of formaldehyde in the manufacture of boards on urea-formaldehyde resins, additionally has the properties of an antiseptic and hardener of urea-formaldehyde resins. This makes it possible to exclude from the board technology the use of a chlorine-containing hardener, ammonium chloride. It, like sodium pentachlorophenolate, is introduced into the plates together with the resin. Moreover, CFA is the safest of these three complex preparations.

As it is shown by our experiments for the production of environmentally friendly particle boards, it is expedient to use phenol-formaldehyde resins as a binder, in particular, brands SFJ-3014 This resin contains 0.15% free formaldehyde, as currently used in the mass production of wooden boards' resins KF-MT-15 and KF-NFP. The plates on this resin have much less toxicity than the plates on CFS. However, the use of SFJ-3014 resin in the technology of plates requires an increase in the duration of hot pressing by 30-40 % of the length of the pressing of plates on the resin KF-MT-15.

The technology of low-toxic particle boards on urea-formaldehyde resin of the KF-MT-15 grade has been tested by us with positive results under the industrial conditions using the formaldehyde acceptor - ammonium fluorofluoride. Urea-formaldehyde resin of KF-MT-15 brand with a content of free formaldehyde of 0.15% has been used in the production of particle board. The resin consumption has amounted to 1.53 g/m^2 of particle board, and the consumption of CFA - 7.5 - 8.5 kg/m^3. Hardener of resin (ammonium chloride) has not been used. The thickness of the finished polished plates has been 16 mm, and the density was 720-787 kg/m^3. Several methods have been tested for introducing CFA into the chip-adhesive mixture. The method of administering the substance to the resin with its further dissolution in the water in the resin turned out to be the most expedient one. Mode of hot pressing of plates has remained to be unchanged.

Emmission of formaldehyde has been determined by the perforator method using photoelectrocolorimetric analysis of formaldehyde content. As a result, plates are obtained formaldehyde emission of which was 3.15 - 6.2 mg/100 g of absolutely dry plate, which corresponds to the formaldehyde emission of E0.5 and E1 classes. From the control plates, formaldehyde emission was 11.0 mg/100 g of absolutely dry plate.

The resulting plates meet Russian specifications. The flexural strength was 15.1 MPa, and the tensile strength perpendicular to the plate surface was 0.36 MPa (according to technical requirements, these indicators should not be lower, respectively, for the plates of P-1 brand - 10.0 and 0.24 MPa, and for P-2 brand- 11.0 and 0.35 MPa). Industrial production of such boards does not require large material costs from the enterprise. Calculations has showed the economic feasibility of plate production using ammonium silicofluoride as formaldehyde acceptor.

4. Conclusions

The particle boards produced by industry today, even on low-toxic resins, do not fully meet the requirements of the Ministry of Health of Russia for isolating harmful substances from humans. Such a substance is formaldehyde.

The problem of the practical use of chipboard produced by the domestic industry in the construction of furniture, floor coverings, etc. is low permissible (allowable) saturation of the room's air volume with plates. So, in a room with an area of 20 m^2, plates of E0.5 emission class plates with an area of no more than 6.25 m^2 can be safely used for humans. Practically only one desk can be installed in a room. When using plates of E1 and E2 classes, such a restriction on the area of plates is even more significant.

The most effective method for reducing the toxicity of particle boards is the use of complex action preparations, which are simultaneously formaldehyde acceptors, antiseptics and hardeners. Such substances can be: sodium pentachlorophenolate; chromium-copper-boric composite based on sodium dichromate, copper sulfate, borax and boric acid; ammonium silicofluoride. They are able to interact in the slab with formaldehyde, which is released mainly from resins, to reduce biodegradation of particle boards in building structures, and ammonium silicofluoride is also a good hardener of urea-formaldehyde resins, which makes it possible to exclude the use of ammonium chloride (containing human carcinogenic chlorine) from particle board technology.

Preparatory works for the introduction of formaldehyde acceptors into the particle board technology is not complicated and does not require considerable additional material costs.

Industrial approbation of obtaining environmentally friendly boards confirmed the possibility of using complex action preparations in the technological processes.

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