

## Lignosulphonate and waste technical hydrolysis lignin as adhesives for eco-friendly fiberboard

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In this study is shown survey about possibility of utilization of Lignosulfonate and waste technical hydrolysis lignin within composition of medium density fiberboards (MDF) produced by dry method.

For the purpose of the study in the laboratory, they are produced MDF with different percentage participation of binder - phenol formaldehyde resin and input supplements lignosulfonate and Technical hydrolysis lignin in their composition. Identified are the main operational features of the boards and are compared with reference ones, without participation of the technical hydrolysis lignin and lignosulfonate. On this base is made analysis of results and are displayed adjacent conclusions and recommendations.

**Keywords:** Medium density fiberboards; MDF; lignosulfonate; technical hydrolysis lignin; phenol formaldehyde resin.

### INTRODUCTION

Wood-based panels are versatile products which made a number of requirements in terms of the environment. In recent years, increasing pressure to develop and produce environmentally friendly technologies for processing and wood products, create new demands on them.

Fiberboard, a structural and decorative material, is a fibrous-felted, homogeneous panel made from lignocellulosic fibers that are combined with a synthetic resin or other suitable bonding system and then bonded together under heat and pressure [1].

Consumption and production of the product MDF increased by continuous pace as in 2012 the consumption of MDF in Europe amount to 10.3 million m<sup>3</sup>, overtaking the need for chipboard [2].

The advantages of this type of boards as a building material and engineering possibilities are many, but there are drawbacks to natural wood. To improve the mechanical properties of the boards (MDF) wood pulp needs to be added adhesives such as emulsions. As adhesives are used water-emulsion synthetic resins having thermosetting properties and strong adhesion to wood. In addition are observed some disadvantages - high value materials contamination of manufacturing equipment, emissions in the environment and toxicity [3].

Wood composite boards are made based on phenol- or urea- formaldehyde resin and they attach to certain levels and amounts of formaldehyde. The process of the emission depends on the exogenous (temperature, relative humidity, air exchange) and endogenous (woody species, the amount of added

binders, type of binder, the production conditions, etc. factors [4].

Numbers of the board are made with phenol-formaldehyde resin. Phenol-formaldehyde resins (PFR) compared to other resins have many advantages such as low cost, ease of use when incorporated in the wood mass, rapid gel time in hot pressing, low temperature curing, resistance to microorganisms and mostly water repellent ability and strength characteristics of the finished boards. MDF boards produced by the PFR are suitable for use in both internal and external conditions. One major drawback of the PFR is the given free formaldehyde into the environment when used as a component in the production of MDF.

This determines the relevance of research on the possibility of reducing content of synthetic resins in the wood panels (MDF) through the use of products where there are no harmful emissions. And research related to the possibility of recovery of products dropped by other wood processing industries are included in the wood boards.

Many products are manufactured based on Phenol Formaldehyde resin that vapors of free formaldehyde, which can cause health problems or illnesses in humans. Emissions of free formaldehyde most common causes: irritation of the eyes and upper airways, when the human body is exposed to the emission of formaldehyde in high doses there is a risk of severe poisoning, and prolonged exposure may result in chronic toxicity and even cancer.

For these reasons, worldwide continuously is conducted research to reduce and eliminate the release of free formaldehyde from wood-composite

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plates. One of the effective methods to reduce the amount of free formaldehyde in fiberboard is the addition of lignosulfonate or technical hydrolysis lignin in the role of binders to appear partial or complete replacement of phenol-formaldehyde resins. On the other hand, it has a negative impact on the physical and mechanical properties of the resulting boards (MDF).

Globally experiments were performed in order to produce dry-formed fiberboards without a resin binder and they have generally been unsuccessful. In recent decades an old process has been re-investigated [1]. This process involves steam explosion of raw lignocellulosic material, thus hydrolyzing most of the hemicelluloses and plasticizing the lignin. The result of this pretreatment is a fiber that can be hot-pressed to produce fiberboard without the need for synthetic binders. Due to the important role of lignin in fiberboard manufacture, several studies have investigated the use of lignin as a natural adhesive and the possibility of replacing fibers with lignin [5].

There are studies in which are successfully produced medium density fiberboards (MDF) without the participation of synthetic resins. As a binder used is an enzymatic lignin [6-8].

A team of researchers from China have produced fiberboards with 10% enzymatic hydrolysis lignin activated at a temperature of pressing than 210° C and a cycle time of 60 s.mm<sup>-1</sup>. The water content of the fiber mass is at relatively high levels by 10 to 18 percent, in order to assist pseudo-phase transition of lignin. These plates with physical and mechanical indicators are close to the standard required and are completely non-toxic, as they are made of biological, natural, ingredients. Studies are still at the stage of laboratory research [6].

The object of this study is to investigate the use of lignosulphonate product and waste technical hydrolysis lignin as adhesives for eco-friendly fibreboard.

## MATERIALS AND METHODS

### Materials

**Wood fiber mass.** For the production of MDF in the laboratory were used wood fibers of mixed species - hardwood, 80% (*Fagus sylvatica*= 60% and *Populus Alba* = 40%) and softwood 20% (*Pinus sylvestris*).

**Bonding agent and other chemicals.** Lignosulfonate - Obtained from spruce wood with low hygroscopicity and average sugar content is used as an additive to replace a part of used Phenol Formaldehyde resin.

Chemical data: Calcium = 6%; Sugars = 7%; Ash = 16%; Solids = 93%; pH = 4.3 ± 0.8; Bulk density 550 kg/m<sup>3</sup>.

Technical hydrolysis lignin (THL) - Waste hydrolyzed lignin produced in the manufacture of fodder yeast. Chemical analyses of technical hydrolysis lignin were determined according to the follow methods: cellulose (Kurshner); lignin (Klason), ash (TAPPI T-15m) [9] and Elemental Analyzer Euro EA 3000 used for analysis of C, N, S and H.

Chemical data: C - 44.6%; S - 0.7%; H - 8.6%; N - 0.16%; Solids - 92.1%; Lignin - 78%; Cellulose - 12.8%; Ash - 9.1%.

Phenol formaldehyde resin: Solids = 48%; Molecular ratio = 1: 1.3  
Density = 1,1780 g.cm<sup>-3</sup>; pH 7.1-7.9; Viscosity at 25°C = 1,400 cps.

For the purpose of the study it was carried out a varying of the amount of the phenol formaldehyde resin from 5 to 15% used for producing of fiberboards (MDF). To determine the influence of the content of the lignosulfonate on the performance of the MDF panels are produced in the laboratory in modifying the content of lignosulfonate from 0 to 20% in increments of 5%. Control boards were made containing only the PFR of 5, 10 and 15%. At these concentrations were conducted experiments with the addition of THL and lignosulfonate.

**MDF panel manufacture.** Pre-prepared wood fiber mass (garnetted stock to the point of 11° Schopper-Riegler method and dried to 8% moisture content) was mixed with Phenol Formaldehyde resin. In some of the experiments the use of additives THL and lignosulphonate was conducted after preliminary gluing and other event together with resin. After subsequent drying of the wood fiber mass, to prepare a carpet timber, which is compressed in a hot press at a temperature of 200° C and 90 s/mm of the boards' thickness. Predefined density is 800 ± 20 kg.m<sup>-3</sup>.

### Methods

**Producing of MDF.** The experiment was designed to study the effect of two factors (percentage of added waste technical hydrolysis lignin and lignosulfonate) on four levels, and to compare fiberboards only with phenol formaldehyde resin. These levels were chosen on the basis of the literature review and on the research group's previous experiences in producing fiberboards (MDF) with externally added technical hydrolysis lignin. The test results of physical and mechanical properties of boards are displayed through specialized testing machine (Zwick Roell) and software to it (TestXpert® II).

Bonding of lignocelluloses material is essential for the manufacture of particleboard, fiberboard, OSB, laminated wood products and plywood. bonding of wood fibers or particles can be achieved by high pressure and temperatures, a phenomenon known as auto-adhesion [10]. The effect can be used for making binderless boards and panels, but compared to boards made with synthetic adhesives the mechanical properties are inferior.

Technical hydrolysis lignin was injected to the wood fibers by two methods - dry and wet method. The maximum concentration is 10%. PFR's concentration varies from 5 up to 15%. Lignosulphonate has concentration of 5 or 10%.

**Test methods of physical and mechanical properties of MDF.** There were held 50 attempts and produced 50 pieces samples of MDF. All samples were cut before testing by saw and prepared according to the international standard ISO EN 325[11]. The MDF boards were annealed under conditions of 65% humidity and 25 ° C for a period of 72 h. until it reaches the equilibrium humidity and the adhesion in all processes finish before being conducted to a physical-mechanical test. The mechanical properties such as bending strength,

modulus of elasticity, water absorption and swelling are determined in accordance with international standards EN 310 [12], EN 317 [13], EN 622-5 [14]. The physical properties such as thickness swelling and water absorption are determined after 24 hours immersion in water requirements of EN 317.

## RESULTS AND DISCUSSION

### *Mechanical properties of the MDF*

Figure 1 and 2 shows the change in the indicator values Bending strength and Modulus of elasticity in the production of MDF with PFR and Lignosulphonate.

When added lignosulphonate as a binder to 5% were not observed significant changes in indicators of the boards produced with the PFR. By increasing the percentage of Lignosulphonata between 10 - 15% clearly stands increasing of bending strength of the samples. Respectively the trend is to carry on indicators modulus of elasticity for the same samples. Compared to the control samples the amendment is within 62% [12].

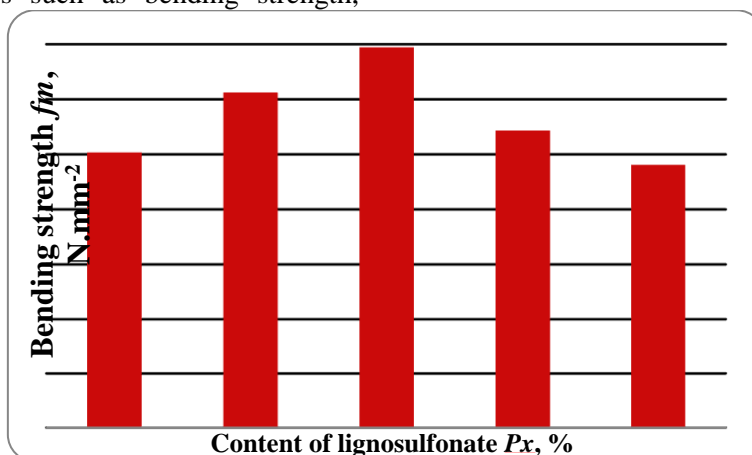


Figure 1. Bending strength of MDF, depending on the content of the lignosulphonate.

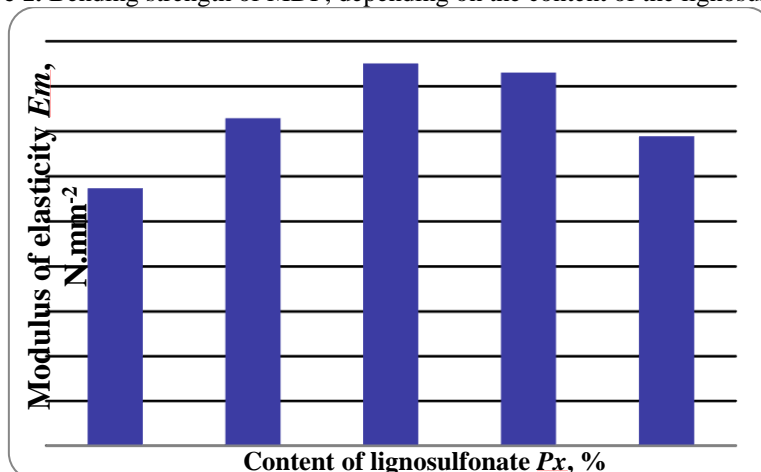


Figure 2. Modulus of elasticity in bending of MDF, depending on the content of the lignosulphonate.

In the indicator for thickness swelling, boards without lignisulfonat are approximately 50%, while the addition of 15 to 20% sensitively is reduced to 20% of swelling (Figure 3). The improvement is 40% on the sampled magnitude [13]. Concerning the

water absorption without lignosulfonate MDF value is above 85%, but with the addition of lignosulfonate from 15 to 20% of the indicator is reduced to 60% water absorption (Figure 4). The improvement is 73% on research magnitude [13].

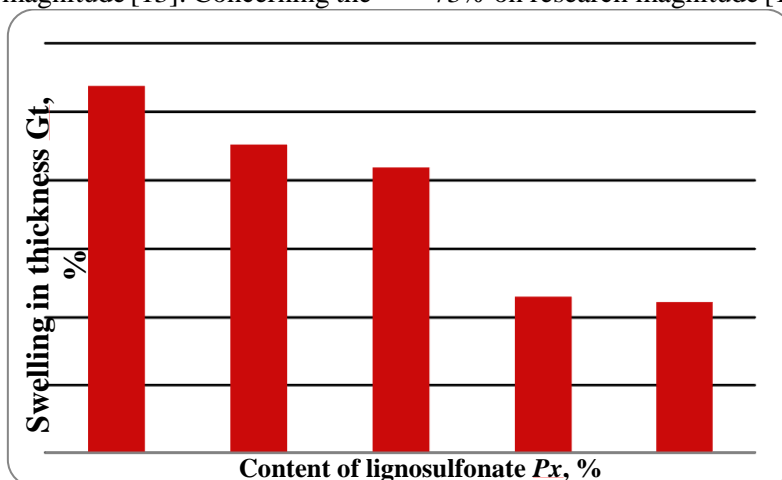


Figure 3. Thickness swelling of MDF, depending on the content of the lignosulfonate.

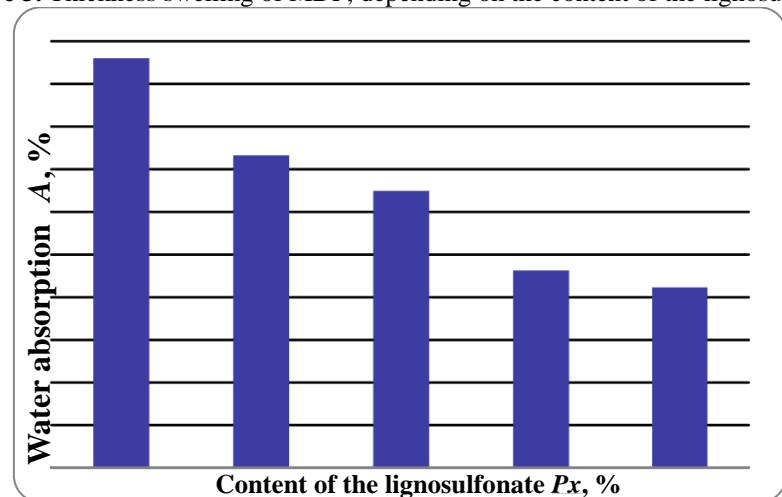


Figure 4. Water absorption of MDF, depending on the content of the lignosulfonate.

It is interesting that in the production of fiberboards (MDF) without the PFR, but only a 10% contribution from the lignosulfonate is obtained bending strength of 22 N.mm<sup>-2</sup>, which is a value very close to the required standard (Figure 5) [13].

Participation of 5% PFR and 5% Lignosulfonate follow the trend of increasing bending strength. The trend is maintained by increasing the content of the resin with 15% and 10% lignosulfonate. After exceeding the participation of the PFR with more than 15% effect is negative and the participation of lignosulfonata of 10% does not change the performance of research parameter (Figure 6).

Additional positive feature is the reduction of free formaldehyde, because it reduces the total quantity of Phenol Formaldehyde resin used in achieving the same, even more favorable physical and mechanical properties.

## CONCLUSIONS

Fibreboards obtained by the addition of lignosulphonate as a natural adhesive have good mechanical and water resistance properties that fully satisfy the relevant standard specifications.

Lignosulfonates can successfully be used as substitutes for conventional binders used in the production of fiberboards.

In the preparation of medium density fiberboard, with scientific purposes it is found that the waste technical hydrolysis lignin is more like active additive than a binder. By proper technique to use in the composition of wood fiber mass, ensuring proper mixing with the fibers and the resin it is possible to reduce the involvement of phenolic resins, and hence the free formaldehyde in the finished product.

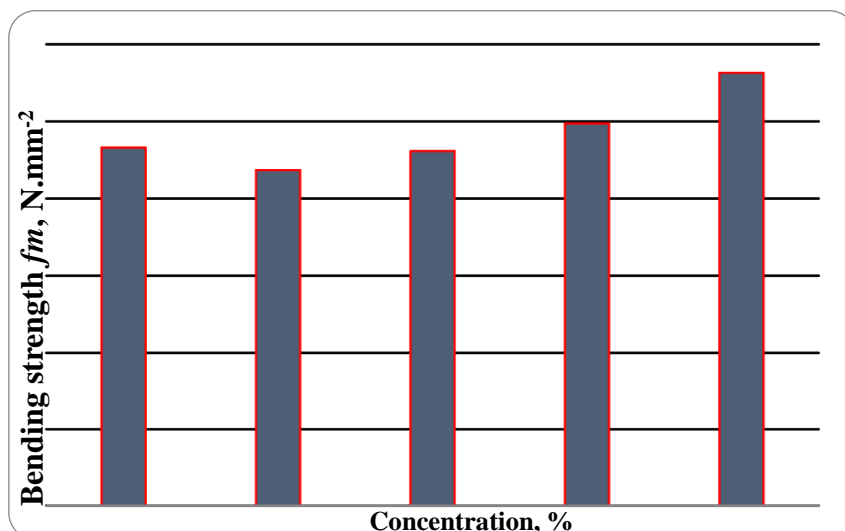


Figure 5. Bending strength of MDF depending on the content of the PFR and lignosulfonate.

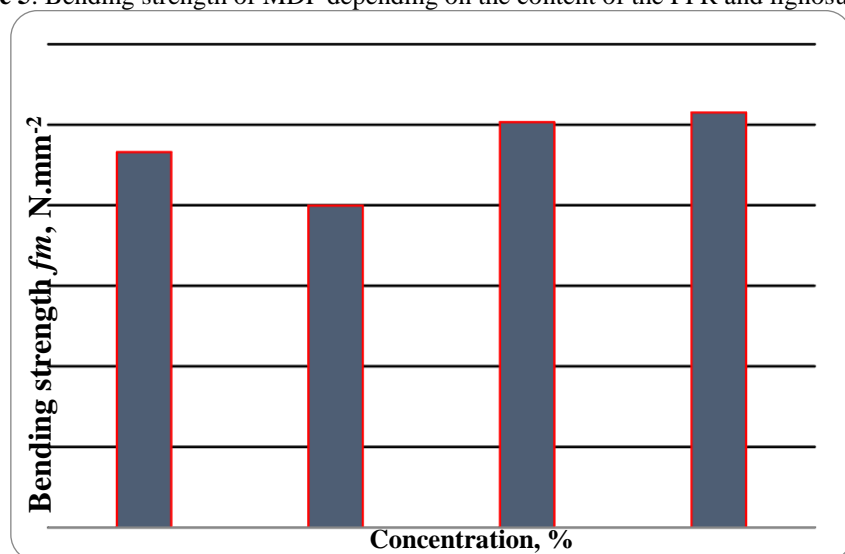


Figure 6. Bending strength of MDF with continuous content Technical hydrolysis lignin and various content of PFR.

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**ЛИГНОСУЛФОНАТ И ОТПАДЪЧЕН ТЕХНИЧЕСКИ ХИДРОЛИЗЕН ЛИГНИН В РОЛЯТА НА СВЪРЗВАЩО ВЕЩЕСТВО ПРИ ПРОИЗВОДСТВОТО НА ЕКОЛОГИЧНИ ПЛОЧИ ОТ ДЪРВЕСНИ ВЛАКНА**

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(Резюме)

В настоящият доклад е представено изследване относно възможността за оползотворяване на лигносулфонат и отпадъчен технически хидролизен лигнин в състава на плочи от дървесни влакна със средна плътност (MDF) произведени по сух метод. За целта на изследването, в лабораторни условия, са произведени плочи с различно процентно участие на свързващо вещество - фенолформалдехидна смола и влаганите добавки лигносулфонат и технически хидролизен лигнин в техния състав. Определени са основните експлоатационни показатели на плочите и са сравнени с тези на еталонни такива, без участие на технически хидролизен лигнин и лигносулфонат. На тази база е осъществен анализ на получените резултати и са изведени прилежащи изводи и препоръки.