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EFFECT OF LIGNOSULFONATE CONTENT ON THE ADHESIVE STRENGTH AT VENEERING OF MEDIUM DENSITY FIBREBOARDS

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ABSTRACT

The transition to a circular bio-oriented industry requires the search for alternative raw materials, sources and binders in producing materials for the furniture industry.

Lignosulfonates (sulphite lignins) are by-products, residues, of cellulose production, and they are bio-based materials with adhesive properties.

This work presents a study evaluating lignosulfonates as binders in the veneering of Medium Density Fibreboards (MDF). An adhesive composition of urea-formaldehyde (UF) resin and ammonium lignosulfonate was used in the veneering. Preliminary experiments have been carried out that show a significant reduction in the adhesion strength when the lignosulfonate content in the adhesive system passes 60%. Therefore, the study used the substitution of UF resin with lignosulfonate from 0 to 60%, with a step of variation from 10%. It is carried out at the gelling time of the various adhesive systems.

An analysis of the adhesive strength in veneering at adhesive systems with different lignosulfonate content was performed. Based on this, conclusions and recommendations were derived.

Key words: veneering, Medium Density Fibreboards, adhesive system, ammonium lignosulfonate, urea-formaldehyde resin.

INTRODUCTION

In the general case, the veneering of wood-based panels, and mainly of Medium Density Fibreboards (MDF), is carried out using polyvinyl acetate (PVA) glue, urea-formaldehyde (UF), or melamine-urea-formaldehyde MUF) resins (Zdravković, V. et al. 2020; Bekhta, P. et al. 2022). All these binders contain substances harmful to human health (mainly formaldehyde). Therefore, an eco-friendly alternative should be sought (Papadopoulou, E. 2019).

On the other hand, the various types of technical lignin are waste products, mainly from the pulp and paper industry. As the amount of waste lignin worldwide exceeds 100 million tons. The main types of lignin products are Organosolv, Sulfite (Kraft), Hydrolysis lignin and lignosulfonates. Of these,

the most common is Kraft lignin. Still, a large part is burned in the factories where it is obtained for heat energy production and partial regeneration of chemical reagents. The quantities of organosolv and hydrolyses lignin are significantly limited. The most common product of the technical lignins is lignosulfonate (Savov, V. et al. 2022).

Lignosulfonates are water-soluble polymers. Depending on the substance with which they are extracted, they are divided into ammonium, calcium, magnesium etc. The sulfonation of lignin is one of the main reactions of the de-lignification process (Antov, P. et al. 2021 b).

Currently, lignosulfonates are mainly used in the woodworking industry as a binder, giving additional mechanical stability in the production of pellets. However, they

have considerable potential as bio-based adhesives for wood-based panels (Souza da Rosa, Th. et al. 2014).

There are successful studies on the fabrication of fibreboards (Antov, P. et al. 2020; Antov, P. et al. 2021 a; Savov, V. and Antov, P. 2020) and particleboards (Hemmilä, V. Et al. 2019; Bekhta P. et al. 2021) with lignosulfonates as a main or auxiliary binder. The primary data from these studies is that the action of lignosulfonates as a formaldehyde scavenger is established (Antov et al. 2021 c.; Bekhta P. et al. 2021). Also, these studies confirmed the need to increase the press factor when using adhesive systems with lignosulfonate in their composition (Savov, V. et al. 2020; Savov, V. et al. 2021). The increased press-factor can be explained by the slower polymerisation of lignosulfonates and by what was found in the study of Antov et al. (Antov et al. 2021 c) increase of the curing temperature with the participation of lignosulfonate in the adhesive system. Thus, in the cited study, an increase in the glass transition point from 105° C when using only UF resin to about 125° C is reported when a lignosulfonate content of 70% is reached in the adhesive system.

Also of interest is the need to use crosslinkers when using adhesive systems with lignosulfonates for particleboards and the lack of need for crosslinking when obtaining fibreboards. The significantly greater importance of hydrogen explains that and, in

general, lignin bonds in fabricating fibreboards.

The possibility of using lignosulfonates as an adhesive for the veneering of wood-based panels has been poorly studied.

The presented study aimed to evaluate lignosulfonate as a binder in the veneering of Medium Density Fibreboards.

MATERIALS AND METHODS

Materials

The adhesive systems used for the study were composed of UF resin and ammonium lignosulfonate. Ammonium lignosulfonate (ALS) was with the following properties: total solids content: 48.6%, ammonium content: 4.1%, sodium content: 0.1%, total sulfur content: 6.8%, sugars: 20%, viscosity (cps): 400 at 25°C, pH: 4.5%, specific gravity: 1.220 g.cm⁻³, and boiling point: 104°C.

The UF resin, kindly supplied by Kronospan-Bulgaria, had a 1.0 molar ratio and 50% dry solid content.

For veneering it was used an oak (*Quercus robur* L.) veneer with a thickness of 0.55 mm was used.

The adhesive consumption rate was 160 g.m⁻². A laboratory press RMS ST 100, Italy, was used for veneering, and the pressure was 0.4 MPa.

The adhesion strength test was carried out by EN ISO 4624:2016, Fig. 1.



Figure 1: Applying the adhesive and testing the samples

Preliminary results

To be established more precisely the range of variation of the lignosulfonate content in the adhesive system, preliminary experiments were carried out with a complete

replacement of UF resin with ALS at a variation step of 20%. The hot-pressing temperature was 180°C, and the hot-pressing time was 60 s.

The data from the conducted preliminary studies are presented in Fig.2.

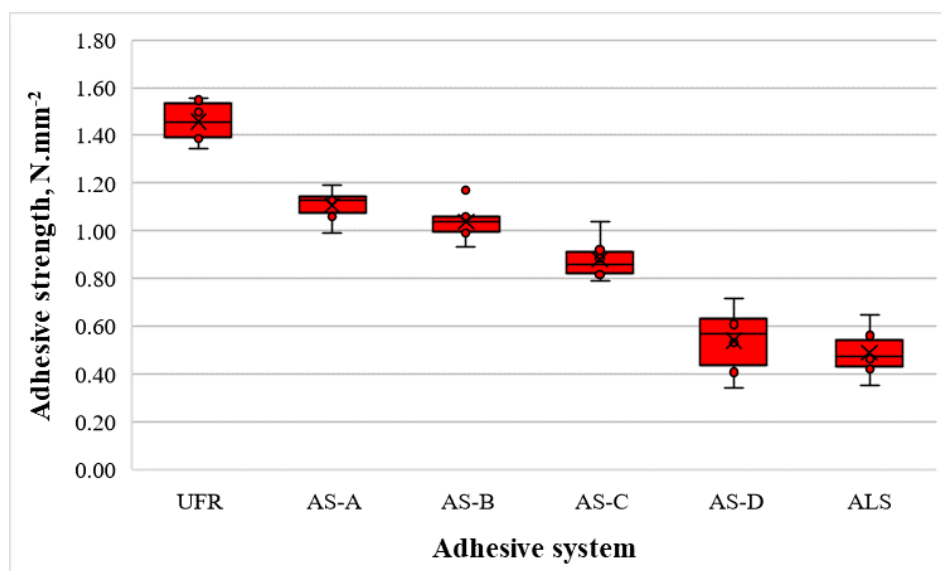


Figure 2: The adhesion strength at a hot-pressing temperature of 180°C and a press time of 60 s (UFR – UF resin; AS-A – 80% UF resin/ 20% ALS; AS-B – 60% UF resin/ 40% ALS; AS-C – 40% UF resin/ 60% ALS; AS-D – 20% UF resin/ 80% ALS; ALS – ammonium lignosulfonate)

The preliminary results show a significant decrease of 2.98 times in the adhesion strength with complete replacement of UF resin with ALS. In practice, there are two main declines in adhesion strength. The first is when adding 20% ALS to the adhesive system. Thus, the adhesion strength when using an adhesive system of 80% UF resin and 20% ALS is 1.32 times lower than the adhesion strength when using only UF resin. The other

significant drop was when the ALS content increased from 60 to 80% in the adhesion system. In this case, the reduction in strength is 1.32 times.

Therefore, as a result of the preliminary studies, it was concluded that there is a need to increase the hot-pressing time. It was found that it is not appropriate for the ALS content in the adhesion system to be above 60%. However, increasing the press factor

could lead to the pre-curing of UF resin. Therefore, the temperature for hot-pressing was reduced.

Experimental plan

It was decided in the experiment to replace UF resin with ALS at a replacement

step of 10% until reaching a content of 60% of ALS in the adhesion system. Also, the hot-pressing time increased to 150 s, and the pressing temperature was lowered to 150°C, Table 1.

Table 1: Experimental plan

Type of adhesive system	Content of UF resin in the adhesive system, %	Content of ALS in the adhesive system, %
UFR	100	0
AS1	90	10
AS2	80	20
AS3	70	30
AS4	60	40
AS5	50	50
AS6	40	60
ALS	0	100

RESULTS AND ANALYSES

The gelation time data of the different adhesion systems with UF resin and ALS are presented in Fig.3.

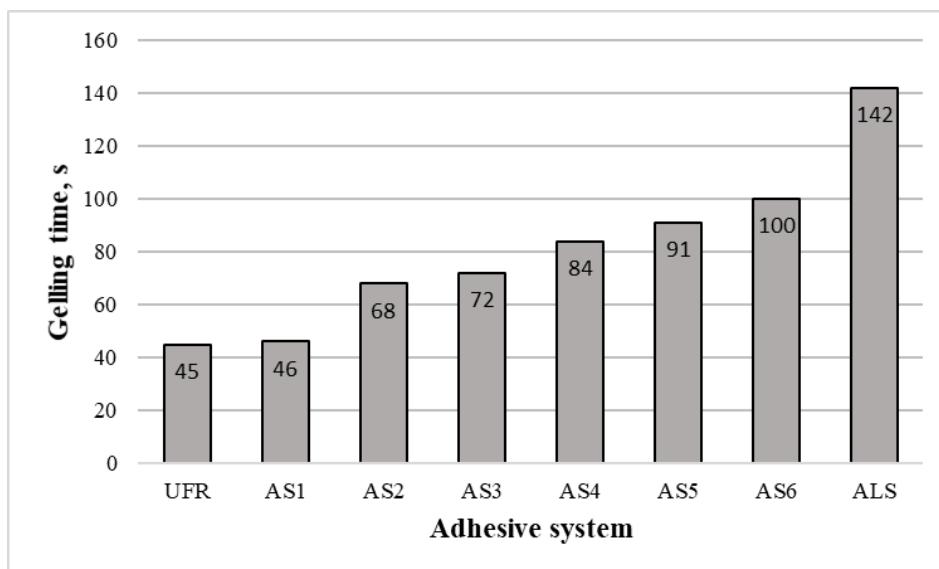


Figure 3: Gelling time of different adhesive systems with UF resin and ALS

UFR – Urea-formaldehyde resin; AS1 – 90% UF resin, 10% ALS; AS2 – 80% UF resin, 20% ALS; AS3 – 70% UF resin, 30% ALS; AS4 – 60% UF resin, 40% ALS; AS5 – 50% UF resin, 50% ALS; AS6 – 40% UF resin, 60% ALS; ALS – Ammonium lignosulfonate

The data show an increase in gelling time of 2.22 times when the ALS content in the adhesion system increases from 0 to 60%. The difference in gelling time between UF resin and ALS is 3.16 times.

In practice, there is no difference between the gelling time of UF resin and an adhesion system with 10% UF resin and 90% ALS. That can be explained by the small amount of ALS in the adhesion system and

by its action as a hardener of UF resin. Then, a relatively smooth increase of about 1.1-time was observed for each 10% increase in ALS content in the adhesive system.

Data on adhesive strength are presented in Fig. 4.

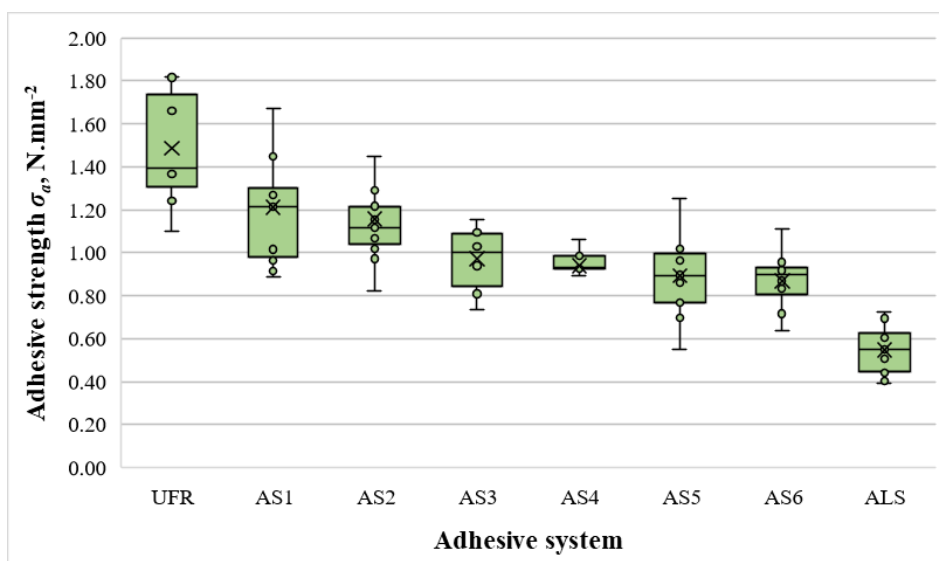


Figure 4: The adhesive strength at a hot-pressing temperature of 150°C and a press time of 150 s
 UFR – Urea-formaldehyde resin; AS1 – 90% UF resin, 10% ALS; AS2 – 80% UF resin, 20% ALS; AS3 – 70% UF resin, 30% ALS; AS4 – 60% UF resin, 40% ALS; AS5 – 50% UF resin, 50% ALS; AS6 – 40% UF resin, 60% ALS; ALS – Ammonium lignosulfonate

The analyse of the experimental data shows that despite the increased time for hot-pressing, the adhesive strength when using ALS is 2.71 times lower than the adhesive strength when using UF resin for the veneering of MDF.

Even when replacing 10% UF resin with ALS, a drop in strength of 1.22 times was observed. In practice, at the selected duration of pressing, there is no difference between the adhesion strength at an ALS content of 10 and 20% (the p-value from the t-test is 0.62).

The subsequent significant decrease is when the lignosulfonate content in the adhesion system increases from 20 to 30%. In this case, the drop in adhesive strength is 1.19 times.

The subsequent increase of the lignosulfonate content in the adhesive system to 60% leads to a very smooth decrease in the adhesive strength. The drop in adhesive strength

when increasing the lignosulfonate content from 30 to 60% is only 1.12 times.

CONCLUSIONS

In conclusion, using lignosulfonates as a bio-based adhesive in MDF veneering is a possible eco-friendly solution to eliminate synthetic binders with free formaldehyde in their composition.

However, the research found some disadvantages of using lignosulfonate as a veneering adhesive. When only lignosulfonate was used for veneering, a significant drop in the adhesive strength was observed, even as press time was increased. That, in turn, suggests using lignosulfonate as a partial substitute or eco-friendly additive in the adhesive system. Even in this case, given the adhesion strength and hot-pressing time, the lignosulfonate content in the adhesive system is not recommended to be above 60%, and it is even suggested to be up to 20%.

When the optimal content of lignosulfonate in veneering is determined, the effect of lignosulfonate as a formaldehyde scavenger should be considered. The influence of lignosulfonate and UF resin crosslinking on adhesive strength at veneering would also be of interest in future studies.

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Conflicts of Interest: The authors declare no conflict of interest.

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