

THE IMPACT OF SODIUM AND MAGNESIUM LIGNOSULFONATES ON AMINO RESIN ADHESIVES FOR WOOD- BASED PANELS PRODUCTION

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ABSTRACT

In the study, the effects of sodium lignosulfonate (NaLS) and magnesium lignosulfonate (MgLS) on urea-formaldehyde (UF) and melamine-urea-formaldehyde (MUF) adhesives were evaluated. Lignosulfonates were incorporated at 2.5%, 5.0%, and 7.5% (based on resin solids), and their influence on solids content, viscosity, gel time, pH, and free formaldehyde content was analysed. Both NaLS and MgLS reduced solids content and viscosity. Gel time in UF adhesives increased moderately from 53.7 s to 57–60 s (+6–12%), indicating slight curing retardation, whereas MUF systems showed a small decrease to ~81 s (–6.6%) with MgLS and minimal change with NaLS. Lignosulfonates also affected pH and formaldehyde content. Sodium lignosulfonate primarily enhanced adhesive flow and reduced viscosity due to weaker ionic interactions, whereas magnesium lignosulfonate influenced curing kinetics and pH through stronger electrostatic and coordination interactions. Free formaldehyde content decreased by approximately 10% in UF systems. In MUF adhesives, MgLS led to a slight reduction, whereas NaLS increased free formaldehyde by up to 30.8–32.1%. These results highlight a clear counterion-dependent behavior and demonstrate the potential of lignosulfonates as multifunctional, bio-based modifiers for wood adhesive systems.

Keywords: urea-formaldehyde resin; melamine-urea-formaldehyde resin; sodium and magnesium lignosulfonates; viscosity; curing kinetics; formaldehyde reduction; wood-based panel.

INTRODUCTION

Wood-based composite materials, including particleboard and medium-density fiberboard (MDF), play a crucial role in the modern wood-processing industry by enabling the efficient utilization of wood resources and the production of value-added products. The performance of these composites largely depends on the adhesive system used during manufacturing. With the continuous growth in global production of wood-based panels, the demand for industrial adhesives has also increased significantly. Although the resin content in panels typically accounts for only 2–14% of the oven-dry wood mass, adhesives account for a major share of total material costs, ranging from 30–50% (Solt *et al.*, 2019). Even minor variations in resin consumption or price can substantially affect the overall production cost of wood-based panels (Maloney, 1993). This enhances the importance of optimizing adhesive systems from both economic and technological perspectives.

For several decades, wood-based panels have been produced primarily using thermosetting adhesives from three main classes: aminoplasts, phenolics, and isocyanates (Pizzi, 2014). About 95% of these adhesives are formaldehyde-based (Kumar and Pizzi, 2019), with urea-formaldehyde (UF) resins dominating the market (~85%; ~11 million tons annually) (Pizzi *et al.*, 2020). Their widespread use is driven by low cost, high reactivity, strong adhesion to wood, short curing times, low pressing temperatures, water solubility, and a colorless bond line (Dunky, 2003). Consequently, UF resins are widely used in particleboard and MDF for interior applications.

However, UF adhesives have a major limitation: the emission of volatile organic compounds, particularly formaldehyde. Formaldehyde exposure can cause irritation of the eyes and respiratory tract and poses long-term health risks (Mantanis *et al.*, 2018). This has led to strict emission regulations in Europe, the United States, and Japan. In parallel, reliance on petroleum-derived raw materials raises concerns about cost stability and resource sustainability, driving the search for bio-based alternatives. Formaldehyde emissions are primarily determined by the free formaldehyde content of the resin rather than its total amount. Emissions can be reduced through process optimization and chemical or physical modification of adhesive systems. As a result, increasing attention is paid to renewable modifiers, e.g., underutilized by-products of the pulp and paper industry, such as technical lignin (including Kraft lignin) and lignosulfonates (LS).

Lignin-based materials are promising bio-based alternatives to conventional formaldehyde-containing resins. Lignin is the second most abundant natural biopolymer after cellulose, with an estimated total content of 3×10^{11} tons in the biosphere and an annual biosynthesis of about 2×10^{10} tons (Hu *et al.*, 2011). The pulp and paper industry generates 50–75 million tons of technical lignin annually, mainly as Kraft lignin and lignosulfonates (Mandlekar *et al.*, 2018), most of which remains underutilized and is typically used for energy recovery.

The polyphenolic structure of lignin enables its application as a partial substitute for phenol in phenol-formaldehyde (PF) resins. Substitution levels of 20–30% phenol have been achieved without significant loss of panel performance (Çetin and Özmen, 2002; Kouisni *et al.*, 2011), and in some cases, up to 80% replacement has been reported (da Silva *et al.*, 2017). Chemically modified lignin, such as phenolated or glyoxalated derivatives, further improves adhesive performance (El Mansouri *et al.*, 2007; Lei *et al.*, 2008). Among lignin derivatives, lignosulfonates, which account for up to 90% of commercially available lignin products, are attractive due to their industrial availability and functional versatility.

However, lignin reactivity remains limited, often requiring modification or the use of crosslinking agents (Pizzi, 2006; Hu *et al.*, 2011; Hemmilä *et al.*, 2019; Bekhta *et al.*, 2021). In UF and MUF systems, lignosulfonates have been shown to reduce viscosity, act as plasticizers, and partially scavenge formaldehyde (Hemmilä *et al.*, 2017, 2019; Antov *et al.*, 2020, 2021; Bekhta *et al.*, 2021). Nevertheless, the influence of counterion type on these effects remains insufficiently studied, although it may significantly affect interactions with water, resin components, and formaldehyde.

While most research has focused on lignin in PF resins, its application in UF and MUF adhesives remains less explored despite their dominant industrial use in particleboard production. In this context, lignosulfonates represent promising multifunctional additives capable of partially replacing synthetic resin components, reducing formaldehyde emissions, and maintaining adhesive performance.

The novelty of the present study lies in the first systematic investigation of sodium and magnesium lignosulfonates as additives to UF and MUF resins for particleboard production. In particular, their effects on adhesive viscosity, acidity of the adhesive system, curing

kinetics, and free formaldehyde content were examined. By integrating performance-related and environmental aspects, this approach contributes to the development of more sustainable adhesive formulations.

The objective of this study is to evaluate the feasibility of using sodium (NaLS) and magnesium (MgLS) lignosulfonates as additives in UF and MUF resins. Specifically, the study aims to determine how different levels of resin substitution affect the properties of the modified adhesives and to provide mechanistic insights into the role of counterions in lignosulfonate-modified adhesive systems. In addition, the potential of these additives to improve adhesive performance and reduce formaldehyde emissions is assessed. The results are expected to provide new insights into the mechanisms of interaction between lignosulfonates and aminoplast resins, thereby supporting the development of more environmentally friendly adhesive systems for the production of wood-based panels. The present study is limited to adhesive system characterization, while the evaluation of the mechanical properties of wood-based panels will be the subject of future work.

MATERIALS AND METHODS

Materials

Adhesive formulations were prepared according to the recipes summarized in Table 1. First, the required amount of lignosulfonate solution was added to the UF or MUF resin under continuous mechanical stirring. The mixture was then homogenized using a mechanical stirrer for a specified time to ensure uniform distribution of lignosulfonate within the adhesive system. All preparations were performed at room temperature.

Tab. 1 Adhesive composition.

Adhesive type	UF resin content (%)	MUF resin content (%)	NaLS content (%)	MgLS content (%)
UF	100	-	0	0
UF + MgLS	97.5	-	2.5	0
	95.0	-	5.0	0
	92.5	-	7.5	0
UF + NaLS	97.5	-	0	2.5
	95.0	-	0	5.0
	92.5	-	0	7.5
MUF	-	100	0	0
MUF + MgLS	-	97.5	2.5	0
	-	95.0	5.0	0
	-	92.5	7.5	0
MUF + NaLS	-	97.5	0	2.5
	-	95.0	0	5.0
	-	92.5	0	7.5

The UF (solid content $66.1 \pm 0.06\%$, dynamic viscosity 440.7 ± 0.6 mPa.s) and MUF (solid content $64.9 \pm 0.20\%$, dynamic viscosity 232.7 ± 3.5 mPa.s) resins were used to prepare the adhesive systems. Magnesium lignosulfonate (MgLS; Borregaard, Germany) and sodium lignosulfonate (NaLS; Domsjö Lignin, Sweden) were applied as lignin-based additives. The lignosulfonates were used as aqueous solutions with a working concentration of 50%. The lignosulfonate addition levels corresponded to the replacement of 2.5%, 5.0%, and 7.5% of the UF or MUF resin in the adhesive formulations. MgLS had the following characteristics: total solids content $> 90\%$; pH (10% solution) 4.0 ± 1.0 ; insoluble matter \leq

0.8%; Mg content 3%; Cl \leq 0.1%; sucrose 6%; and density 450 – 600 kg/m³. NaLS exhibited the following properties: total solids content > 95%; pH (10% solution) 6 \pm 1; Na 9%; S 8.5%; Ca 0.12%; Cl 0.01%; insoluble substances < 0.1%; sulphate 7.5% (in the form of sulfate ions); and sucrose 2.0%.

Adhesive analysis

For each adhesive formulation prepared according to the specified recipe (Table 1), the following properties were determined: solids content, dynamic viscosity, curing time, hydrogen ion concentration (pH), and free formaldehyde content. The solids content of the prepared adhesive mixtures was determined gravimetrically by the weight method. Dynamic viscosity was measured using a rotary viscometer “ROTAVISC hi-vi I” (IKA Viscometers, Germany), while pH was determined with an I-315 pH meter. The curing time (gel time) of the adhesive mixtures was determined at 100 °C. The free formaldehyde content of the resin was determined by titration. All measurements were performed in triplicate. Due to the limited number of replicates, only descriptive statistics (mean and standard deviation) were applied in this study.

RESULTS AND DISCUSSION

The properties of UF and MUF adhesives with varying levels of NaLS and MgLS are summarized in Table 2. The results presented in Table 2 show consistent trends in adhesive properties with increasing liginosulfonate content. Although the absolute changes in some parameters are relatively moderate, the direction of change is systematic across both UF and MUF systems. The gradual decrease in solids content and viscosity with increasing liginosulfonate concentration suggests that the observed effects are not random but are directly related to the incorporation of liginosulfonate solutions into the adhesive matrix.

Tab. 2 Properties of adhesives.

Adhesive type	LS content (%)	Solid content (%)	Dynamic viscosity (mPa.s)	Gel time (s)	pH	Free formaldehyde content (%)
UF	0	66.1 (0.06)	394.7 (2.5)	53.7 (0.58)	7.5 (0.06)	0.64 (0.02)
UF + MgLS	2.5	65.0 (1.39)	389.7 (2.3)	53.7 (0.58)	6.8 (0.06)	0.61 (0.01)
	5.0	64.6 (0.10)	380.0 (2.7)	57.7 (0.58)	6.4 (0.06)	0.58 (0.01)
	7.5	64.0 (0.10)	373.0 (1.7)	57.3 (0.58)	6.3 (0.06)	-*
UF + NaLS	2.5	65.8 (0.06)	391.3 (1.5)	54.3 (0.58)	7.0 (0.06)	0.59 (0.01)
	5.0	65.3 (0.06)	378.0 (1.0)	56.7 (0.58)	6.9 (0.06)	0.54 (0.01)
	7.5	64.7 (0.58)	358.7 (1.2)	59.7 (0.58)	6.8 (0.06)	-*
MUF	0	64.9 (0.20)	251.0 (1.7)	86.7 (0.58)	9.4 (0.06)	0.078 (0.001)
MUF + MgLS	2.5	63.3 (0.17)	244.3 (2.1)	84.0 (0.58)	8.3 (0.06)	0.077 (0.002)
	5.0	63.2 (0.12)	235.3 (2.5)	83.1 (0.58)	8.0 (0.06)	0.075 (0.002)
	7.5	62.8 (0.10)	227.3 (0.6)	81.2 (0.58)	7.6 (0.06)	-*
MUF + NaLS	2.5	64.7 (0.06)	245.7 (1.2)	85.3 (0.58)	8.8 (0.06)	0.103 (0.001)
	5.0	64.3 (0.12)	233.0 (3.0)	84.0 (1.00)	8.7 (0.06)	0.102 (0.001)
	7.5	64.1 (0.06)	227.0 (1.0)	84.3 (0.58)	8.1 (0.06)	-*

Notes: LS = liginosulfonate; UF = urea-formaldehyde resin; MUF = melamine-urea-formaldehyde resin; MgLS = magnesium liginosulfonate; NaLS = sodium liginosulfonate. Values are presented as mean \pm standard deviation (n = 3). *Free formaldehyde content could not be determined by titration due to interference from the solution color.

The relatively small standard deviations obtained for viscosity, gel time, and pH indicate good reproducibility of the measurements and confirm the stability of the prepared adhesive formulations. The monotonic reduction in viscosity particularly in NaLS-modified systems suggests that the dispersing effect of lignosulfonates increases proportionally with concentration. In contrast, MgLS-modified adhesives show slightly smaller changes in viscosity but more noticeable changes in curing behavior and pH, indicating that the divalent magnesium cation influences chemical interactions within the system rather than only its rheological behavior.

Such systematic trends support the interpretation that the observed differences between NaLS and MgLS adhesives are primarily related to the different ionic characteristics of Na⁺ and Mg²⁺ counterions and their interactions with lignosulfonate functional groups and resin components.

Rheological properties and solids content

The incorporation of lignosulfonates affected both the solids content and the rheological behavior of the UF and MUF adhesive systems. In all formulations, an increase in lignosulfonate content resulted in a gradual decrease in solids content. For UF adhesives modified with MgLS, the solids content decreased from 66.1% in the reference adhesive to 64.0% at a 7.5% substitution level. A comparable, but slightly less pronounced, reduction was observed for UF adhesives containing NaLS, with the solids content decreasing from 66.5% to 64.7%. Similar trends were observed in MUF systems, where MgLS reduced from 64.9% to 62.8%, while NaLS decreased to 64.1%.

The reduction in solids content is primarily due to the addition of lignosulfonates in aqueous form, which increases the water fraction of the adhesive. The observed differences between MgLS and NaLS systems may suggest that the counterion influences hydration and intermolecular interactions. In particular, Mg²⁺ could form stronger electrostatic and coordination interactions with sulfonate groups than Na⁺, potentially promoting more strongly hydrated structures and contributing to the slightly greater reduction in solids content in MgLS-based adhesives. Although lignosulfonates introduce additional dissolved and colloidal matter, their contribution appears to depend on ion type. NaLS (9% Na), with higher solubility and ionic content, may integrate more uniformly into the aqueous phase, contributing more effectively to the measured solids. In contrast, MgLS (3% Mg), possibly due to stronger ionic associations and a higher insoluble fraction, may partly behave as a dispersed phase, resulting in a somewhat lower effective contribution to solids content.

The addition of lignosulfonates influenced the viscosity of the adhesive mixtures. In UF systems, viscosity decreased from 394.7 to 373.0 mPa.s (4.5%) with MgLS and to 358.7 mPa.s (9.1%) with NaLS. A similar trend was observed for MUF adhesives, where viscosity decreased from 251.0 to 227.3 mPa.s (9.4%) with MgLS and to 227.0 mPa.s (9.6%) with NaLS. This reduction is commonly attributed to the amphiphilic nature of lignosulfonates, which can act as dispersing and plasticizing agents, improving resin distribution and reducing intermolecular interactions. Similar behavior was reported in previous studies (Hemmilä *et al.*, 2019; Li *et al.*, 2023; Ait Benhamou *et al.*, 2025; Nafisah *et al.*, 2025; Paez and Fatehi, 2025).

The slightly stronger effect observed in NaLS-modified adhesives may be related to differences in ionic characteristics. Na⁺, as a monovalent ion, may form weaker associations with sulfonate groups, allowing greater molecular mobility and more effective dispersion. In contrast, Mg²⁺ could promote limited ionic bridging between lignosulfonate chains, potentially leading to some degree of association that moderates the viscosity reduction. Although lignosulfonates introduce additional macromolecular content and can interact with

the resin through hydrogen bonding and ionic interactions, their overall effect in these systems appears to be dominated by their dispersing and plasticizing action. Differences between NaLS and MgLS may therefore reflect a balance between dispersion efficiency and intermolecular association.

The influence of the ionic form of lignosulfonates has also been highlighted in recent studies on lignin-modified UF and MUF adhesives. It has been shown that counterions can affect molecular aggregation, electrostatic interactions, and water retention within adhesive systems (Wu *et al.*, 2021; Li *et al.*, 2023). Antov *et al.* (2020) further reported that lignosulfonates containing different metal cations can modify adhesive viscosity and curing behavior due to differences in ionic interactions and coordination effects.

Gel time and curing behavior

The influence of lignosulfonates on curing behavior differed between UF and MUF adhesive systems. In UF-based adhesives, lignosulfonate addition led to a moderate increase in gel time, from 53.7 s to about 57–60 s (≈ 6 –12%), indicating slight retardation of curing. This effect may be associated with the presence of phenolic hydroxyl groups, which can react with formaldehyde, temporarily reducing its availability for polycondensation. In addition, the relatively large lignosulfonate macromolecules may introduce steric constraints that slow the formation of methylene linkages. Similar effects have been reported in previous studies (Hemmilä *et al.*, 2019; Li *et al.*, 2023; Ait Benhamou *et al.*, 2025; Nafisah *et al.*, 2025; Paez and Fatehi, 2025).

In contrast, MUF adhesives showed a different response depending on the counterion. MgLS-modified systems exhibited a slight decrease in gel time, from 86.7 s to ~ 81 s ($\approx 6.6\%$), suggesting a mild acceleration of curing. This may indicate a catalytic contribution of Mg^{2+} ions, which could facilitate condensation reactions or interactions with oxygen-containing functional groups. For NaLS-modified MUF adhesives, gel time remained largely unchanged, suggesting a weaker influence on curing kinetics.

Overall, these trends point to a counterion-dependent effect. Lignosulfonates may influence curing through interactions with formaldehyde and reactive intermediates, as well as by modifying the acidity of the system. NaLS, typically associated with higher alkalinity, may slightly retard acid-catalyzed curing, whereas MgLS, being more acidic, may reduce this effect or even promote curing to a limited extent.

pH of adhesive systems

The incorporation of lignosulfonates resulted in a systematic decrease in pH across all adhesive formulations. In UF adhesives modified with MgLS, the pH decreased from 7.5 to 6.3 (16% reduction) as the lignosulfonate content increased to 7.5%. Similarly, “UF + NaLS” adhesives showed a decrease from 7.7 to 6.8 (11.7% reduction). A comparable trend was observed in MUF adhesives, where the pH decreased from approximately 9.4 to 7.6 (19.1% reduction) in MgLS-containing systems and to 8.1 (13.8% reduction) in NaLS-modified adhesives.

This pH reduction can be attributed to the acidic character of lignosulfonates, which contain sulfonic acid groups that partially dissociate in aqueous solution. The more pronounced pH decrease in MgLS-modified systems may be related to differences in ionization behavior and complexation ability between Mg^{2+} and Na^+ ions. Magnesium ions may interact with oxygen-containing functional groups through coordination or ion-association effects, which could influence the acid–base equilibrium of the system and contribute to a greater shift toward more acidic conditions. In contrast, NaLS, with higher

sodium content (Na^+ 9%), may provide greater buffering capacity, helping to moderate the pH decrease compared to MgLS systems with lower magnesium content (Mg^{2+} 3%).

Changes in pH are particularly relevant for amino resin adhesives, since the curing reactions of UF and MUF resins are strongly influenced by acidity. Therefore, the presence of lignosulfonates may indirectly modify curing kinetics by altering the pH of the adhesive mixture.

Free formaldehyde content

The incorporation of lignosulfonates resulted in a reduction in free formaldehyde content in both UF and MUF adhesive systems. In UF adhesives modified with MgLS, the free formaldehyde content decreased from 0.64% in the reference formulation to 0.58% (9.4% reduction) at a lignosulfonate content of 5%. Similarly, in UF adhesives containing NaLS, the free formaldehyde content decreased from 0.60% to 0.54% (10% reduction).

In MUF adhesives, the effect was less consistent. In the “MUF + MgLS” systems, a slight reduction was observed (1.3–3.8%), whereas in the “MUF + NaLS” systems, the free formaldehyde content increased by 30.8–32.1%. This behavior may be attributed to the higher sodium content (9%) and the more alkaline character of NaLS, which can shift the system toward higher pH values, thereby reducing the efficiency of formaldehyde scavenging reactions and slowing the incorporation of free formaldehyde into the polymer network. In contrast, MgLS, with a lower metal content (3%) and a more acidic character, is less disruptive to curing conditions and thus retains a limited formaldehyde-reducing effect.

The reduction in free formaldehyde can be explained by the inherent reactivity of lignosulfonates. Phenolic hydroxyl groups present in lignin structures may react with formaldehyde via electrophilic substitution, forming methylolated derivatives and methylene bridges. As a result, lignosulfonates can act as partial formaldehyde scavengers, reducing the amount of unreacted formaldehyde while becoming incorporated into the adhesive network. Similar effects have been widely reported for lignin-based additives in amino resin systems (Antov *et al.*, 2021; Bekhta *et al.*, 2021; Li *et al.*, 2023; Pizzi, 2024; Ait Benhamou *et al.*, 2025; Nafisah *et al.*, 2025; Paez and Fatehi, 2025). For example, Pizzi (2024) demonstrated that lignin-derived materials participate in condensation reactions with formaldehyde through phenolic hydroxyl groups, forming methylene bridges within the polymer network. Likewise, Hemmilä *et al.* (2019) reported that lignin incorporation reduces formaldehyde emissions and alters curing behavior due to interactions with formaldehyde intermediates during polymerization.

The presence of metal cations may also influence this process. Magnesium ions can form coordination complexes with lignosulfonate functional groups, thereby partially restricting the accessibility of reactive phenolic sites. In contrast, sodium ions form weaker ionic associations, allowing greater molecular mobility; however, in MUF systems, their stronger alkalizing effect appears to dominate, reducing the efficiency of formaldehyde scavenging.

Mechanistic interpretation: influence of Na^+ and Mg^{2+} cations

The observed differences between NaLS- and MgLS-modified UF and MUF systems can be attributed not only to the nature of the counterions but also to their different ionic contents and physicochemical behavior in aqueous media. NaLS, characterized by higher sodium content (Na : 9%) and greater alkalinity, more strongly affects system pH, viscosity, and gel time, while simultaneously enhancing dispersion and accessibility of reactive functional groups. In contrast, MgLS, with a lower magnesium content (Mg : 3%) and a more acidic character, exhibits a weaker buffering capacity and a more limited influence on system

reactivity. These differences collectively influence solid content, rheological behavior, curing kinetics, and formaldehyde scavenging efficiency of the adhesive systems.

From a rheological perspective, NaLS exhibited a stronger viscosity-reducing effect compared with MgLS. This behavior may be explained by differences in the valence and association behavior of the two cations. Sodium ions (Na^+) are monovalent and are likely to interact weakly with negatively charged sulfonate groups. As a result, NaLS molecules remain relatively dispersed in aqueous systems and exhibit higher molecular mobility. This may promote better dispersion of resin components and reduce intermolecular interactions, thereby contributing to lower viscosity of the adhesive mixture. In contrast, magnesium ions (Mg^{2+}) are divalent and may form stronger electrostatic interactions with sulfonate groups on lignosulfonate chains. The presence of Mg^{2+} may therefore promote ionic bridging between lignosulfonate molecules, leading to partial aggregation or the formation of weak supramolecular networks. These interactions may reduce the dispersing efficiency of lignosulfonates and result in slightly higher viscosities compared with NaLS-modified systems.

The influence of cation type is also reflected in the curing behavior of the adhesive systems. In UF adhesives, lignosulfonate addition generally increased gel time, indicating a mild retardation of curing. However, MUF adhesives modified with MgLS showed a slight decrease in gel time, suggesting that magnesium ions may influence condensation reactions, potentially through coordination or acid–base effects. Divalent metal ions such as Mg^{2+} can act as Lewis acid centers, facilitating the formation of methylene bridges between methylol groups in amino resins. However, this effect should be considered as a possible contribution rather than a confirmed catalytic mechanism in the present study. In contrast, sodium ions are less likely to participate in such coordination interactions due to their lower charge density and weaker complexation ability. Consequently, NaLS is likely to influence the adhesive system primarily through physical effects, such as dilution, dispersion, and plasticization, rather than through catalytic interactions during curing.

Another important factor is the impact of the cation type on the acid–base properties of the adhesive system. MgLS solutions typically exhibit slightly lower pH compared with NaLS, which may contribute to a greater reduction in pH when incorporated into UF and MUF adhesives. Since the curing of amino resins is strongly pH-dependent, this pH shift may further influence reaction kinetics and network formation.

Taken together, these effects help explain the distinct roles of the two lignosulfonate types observed in the present study. Sodium lignosulfonate primarily acts as a rheological modifier and dispersing agent, leading to greater viscosity reduction and improved flow properties. In contrast, magnesium lignosulfonate appears to exhibit stronger ionic interactions, potentially influencing curing behavior and the chemical environment of the adhesive system.

Implications for wood composite manufacturing

From a technological perspective, the modification of UF and MUF adhesives with lignosulfonates may offer several advantages for wood-based panel production. The reduced viscosity observed in NaLS-modified adhesives may improve adhesive spreading and penetration into the porous wood structure. Improved flow properties are particularly beneficial in processes such as particleboard and fiberboard manufacturing, where uniform adhesive distribution is critical for achieving consistent bonding performance.

At the same time, the slight reduction in free formaldehyde content observed in lignosulfonate-modified systems may contribute to lower formaldehyde emissions from finished wood-based panels. The ability of lignin-derived materials to act as partial

formaldehyde scavengers has been widely recognized as an important strategy for developing more environmentally friendly wood adhesives.

The influence of MgLS on curing behavior may also be technologically relevant. The slight acceleration of curing observed in MUF adhesives containing MgLS suggests that divalent magnesium ions may facilitate condensation reactions in amino resin systems. Faster curing kinetics can potentially improve press productivity and reduce the pressing time required during panel production, provided that adhesive viscosity and penetration remain within acceptable limits.

Although the results demonstrate promising potential, the industrial applicability of lignosulfonate-modified adhesive systems has not been validated in this study and requires further investigation under pilot- and industrial-scale conditions.

Practical interpretation of NaLS vs MgLS performance

The differences observed between sodium and magnesium lignosulfonates indicate that the choice of lignosulfonate type should be carefully considered based on the targeted adhesive performance.

Sodium lignosulfonate appears to function primarily as a rheological modifier and dispersing agent, resulting in greater reductions in viscosity and improved flow properties of the adhesive system. These characteristics may be advantageous in applications requiring enhanced adhesive spreading and penetration.

In contrast, magnesium lignosulfonate appears to exert a stronger influence on curing kinetics and pH adjustment, likely due to its higher charge density and greater association capability with Mg^{2+} ions. The divalent magnesium cation may form ionic bridges between sulfonate groups and interact with oxygen-containing functional groups within the resin system, thereby influencing the development of the polymer network during curing.

Consequently, sodium lignosulfonates may be more suitable for applications requiring improved rheological behavior. In contrast, magnesium lignosulfonates may provide additional benefits in formulations where modification of curing behavior or resin reactivity is required. The results, therefore, highlight the importance of considering the counterion chemistry of lignosulfonates when designing lignin-modified adhesive systems.

Overall assessment

Overall, the results demonstrate that both sodium and magnesium lignosulfonates can be incorporated into UF and MUF adhesive systems without significantly compromising their fundamental physicochemical properties. However, the type of counterion appears to play a measurable role in determining the magnitude of the observed effects.

Sodium lignosulfonate tends to induce stronger reductions in viscosity, likely due to weaker ionic interactions between lignosulfonate molecules, thereby increasing molecular mobility within the adhesive system. In contrast, magnesium lignosulfonate may engage stronger electrostatic and coordination interactions due to the divalent nature of Mg^{2+} ions, which could more noticeably influence water retention, pH reduction, and curing behavior.

These findings highlight the importance of considering the ionic form of lignosulfonates when designing lignin-based modifiers for amino resin adhesives and demonstrate the potential of lignosulfonates as multifunctional additives that can influence rheological behavior, curing characteristics, and formaldehyde content.

CONCLUSION

Sodium (NaLS) and magnesium (MgLS) lignosulfonates exhibited distinct effects despite sharing the same lignosulfonate backbone, highlighting the role of counterions and their content (Na: 9%; Mg: 3%) in governing adhesive behavior. Both sodium and magnesium lignosulfonates reduced the solids content and dynamic viscosity of adhesives while maintaining acceptable gel time and pH for industrial applications. NaLS primarily acts as a rheological modifier, resulting in greater viscosity reduction and improved flow. In contrast, MgLS appears to influence curing behavior and pH, likely through ionic interactions and coordination effects. Lignosulfonates reduced the free formaldehyde content in most adhesive systems (except the “MUF + NaLS” systems), likely due to the reactivity of phenolic hydroxyl groups, demonstrating their potential as partial formaldehyde scavengers. The observed differences between NaLS and MgLS may be related to the distinct behavior of Na⁺ and Mg²⁺ ions: Mg²⁺ may promote stronger electrostatic interactions, whereas Na⁺ enables greater molecular mobility and dispersion within the adhesive system. Overall, modifying UF and MUF adhesives with lignosulfonates offers dual benefits: improved rheological behavior for enhanced adhesive spreading and reduced formaldehyde content. These effects may contribute to process optimization, such as improved coating uniformity and potentially adjusted pressing parameters in wood-based panel production. Lignosulfonates, as renewable, lignin-based additives, represent a promising approach for partial substitution of petrochemical resin components in wood adhesives, contributing to more sustainable adhesive systems.

REFERENCES

- Ait Benhamou, A., Abid, L., Calvez, I., Cloutier, A., Nejad, M., Stevanovic, T., Landry, V., 2025. Advances in Lignin Chemistry, Bonding Performance, and Formaldehyde Emission Reduction in Lignin-Based Urea-Formaldehyde Adhesives: A Review. *ChemSusChem* 18, e202500491. <https://doi.org/10.1002/cssc.202500491>
- Antov, P., Savov, V., Krišťák, L., Réh, R., Mantanis, G.I., 2021. Eco-friendly, high-density fiberboards bonded with urea-formaldehyde and ammonium lignosulfonate. *Polymers* 13(2), 220. <https://doi.org/10.3390/polym13020220>
- Antov, P., Savov, V., Neykov, N., 2020. Sustainable bio-based adhesives for eco-friendly wood composites. A review. *Wood Research* 65, 1, 51–62. <https://doi.org/10.37763/wr.1336-4561/65.1.051062>
- Bekhta, P., Noshchenko, G., Réh, R., Krišťák, L., Sedliačik, J., Antov, P., Mirski, R., Savov, V., 2021. Properties of eco-friendly particleboards bonded with lignosulfonate-urea-formaldehyde adhesives and pMDI as a crosslinker. *Materials* 14(17), 4875. <https://doi.org/10.3390/ma14174875>
- Çetin, N.S., Özmen, N., 2002. Use of organosolv lignin in phenol-formaldehyde resins for particleboard production: II. Particleboard production and properties. *International Journal of Adhesion and Adhesives* 22, 6, 481–486. [https://doi.org/10.1016/S0143-7496\(02\)00059-3](https://doi.org/10.1016/S0143-7496(02)00059-3)
- da Silva, M.A., dos Santos, P.V., Silva, G.C., Lelis, R.C.C., do Nascimento, A.M., Brito, E.O., 2017. Using lignosulfonate and Phenol-Formaldehyde adhesive in particleboard manufacturing. *Scientia Forestalis/Forest Sciences* 45, 115, 423–433. <https://doi.org/10.18671/scifor.v45n115.01>
- Dunky, M., 2003. Adhesives in the wood industry. In *Handbook of Adhesive Technology*, 2th ed.; Revised and Expanded. Pizzi, A., Mittal, K.L., Eds.; Marcel Dekker, Inc.: New York, NY, USA; Basel, Switzerland, 2003; 71p, <https://doi.org/10.1201/9780203912225.ch47>

- El Mansouri, N.E., Pizzi, A., Salvado, J., 2007. Lignin-based wood panel adhesives without formaldehyde. *Holz als Roh- und Werkstoff* 65, 65–70. <https://doi.org/10.1007/s00107-006-0130-z>
- Hemmilä, H., Adamopolus, St., Hosseinpourpia, R., Ahmed, Sh.A., 2019. Ammonium Lignosulfonate Adhesives for Particleboards with pMDI and Furfuryl Alcohol as Crosslinkers. *Polymers* 11, 10, 1633. <https://doi.org/10.3390/polym11101633>
- Hemmilä, V., Adamopoulos, S., Karlsson, O., Kumar, A., 2017. Development of sustainable bio-adhesives for engineered wood panels – A Review. *RSC Advances* 7, 61, 38604–38630. <https://doi.org/10.1039/C7RA06598A>
- Hu, L., Pan, H., Zhou, Y., Zhang, M., 2011. Methods to Improve Lignin's Reactivity as a Phenol Substitute and as Replacement for Other Phenolic Compounds: A Brief Review. *BioResources* 6, 3, 3515–3525. <https://doi.org/10.15376/biores.6.3.3515-3525>
- Kouisni, L., Fang, Y., Paleologou, M., Ahvazi, B., Hawari, J., Zhang, Y., Wang, X.-M., 2011. Kraft lignin recovery and its use in the preparation of lignin-based phenol formaldehyde resins for plywood. *Cellulose Chemistry and Technology* 45, 7-8, 515–520
- Kumar, R.N., Pizzi, A., 2019. Environmental Aspects of Adhesives–Emission of Formaldehyde. In: *Adhesives for Wood and Lignocellulosic Materials*, Wiley-Scrivener Publishing: Hoboken, NJ, USA, 2019; pp. 293–312
- Lei, H., Pizzi, A., Du, G., 2008. Environmentally friendly mixed tannin/lignin wood resins. *Journal of Applied Polymer Science* 107, 1, 203–209. <https://doi.org/10.1002/app.27011>
- Li, D., Yu, L., Li, L., Liang, J., Wu, Z., Xu, X., Zhong, X., Gong, F., 2023. Melamine–Urea–Formaldehyde Resin Adhesive Modified with Recycling Lignin: Preparation, Structures and Properties. *Forests* 14(8), 1625. <https://doi.org/10.3390/f14081625>
- Maloney, T.M., 1993. *Modern particleboard and dry process manufacturing*. San Francisco: Miller Freeman Inc.; 1993.
- Mandlekar, N., Cayla, A., Rault, F., Giraud, S., Salaün, F., Malucelli, G., Guan, J.-P., 2018. An overview on the use of lignin and its derivatives in fire retardant polymer systems, lignin-trends and applications, 2018, InTech.Matheus Poletto, IntechOpen. *Lignin Trends Appl.* <https://doi.org/10.5772/intechopen.72963>
- Mantanis, G.I., Athanassiadou, E.T., Barbu, M.C., Wijnendaele, K., 2018. Adhesive systems used in the European particleboard, MDF and OSB industries. *Wood Material Science and Engineering* 13(2), 104–116. <https://doi.org/10.1080/17480272.2017.1396622>
- Paez, J., Fatehi, P., 2025. Incorporation of lignin into adhesives: a review. *Green Chemistry* 27, 12499–12537. <https://doi.org/10.1039/D5GC02998H>
- Nafisah, P.M., Wibowo, E.S., Mubarak, M., Darmawan, W., Lubis, M.A.R., Santoso, A., Kusumah, S.S., Afif, S., Park, B.D., 2025. The role of lignin in enhancing adhesion performance and reducing formaldehyde emissions of phenol-resorcinol-formaldehyde resin adhesives. *International Journal of Biological Macromolecules* 323(2), 147251. <https://doi.org/10.1016/j.ijbiomac.2025.147251>
- Pizzi, A., 2006. Recent developments in eco-efficient bio-based adhesives for wood bonding: Opportunities and issues. *Journal of Adhesion Science Technology* 20, 8, 829–846. <https://doi.org/10.1163/15685610677638635>
- Pizzi, A., 2014. Synthetic Adhesives for Wood Panels: Chemistry and Technology - A Critical Review. *Reviews of Adhesion and Adhesives* 2(1), 85–126. <https://doi.org/10.7569/RAA.2013.097317>
- Pizzi, A., Papadopoulos, A.N., Policardi, F., 2020. Wood Composites and Their Polymer Binders. *Polymers* 12(5), 1115. <https://doi.org/10.3390/polym12051115>
- Solt, P., Konnerth, J., Gindl-Altmutter, W., Kantner, W., Moser, J., Mitter, R., 2019. Technological performance of formaldehyde-free adhesive alternatives for particleboard industry. *International Journal of Adhesion and Adhesives* 94, 99e131. <https://doi.org/10.1016/j.ijadhadh.2019.04.007>
- Wu, Z., Chen, S., Liang, J., Li, L., Xi, X., Deng, X., Zhang, B., Lei, H., 2021. Plasma treatment induced chemical changes of alkali lignin to enhance the performances of lignin-phenol-formaldehyde resin adhesive. *Journal of Renewable Materials* 9, 1959–1972. <https://doi.org/10.32604/jrm.2021.016786>

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