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EFFECT OF TANNIC ACID (TA) ON INCREASING UREA-FORMALDEHYDE (UF) ADHESIVE PERFORMANCE

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ABSTRACT

The increasing awareness of environmental issues, including fossil fuel depletion and global warming, has positioned wood and novel wood products at the forefront due to their advantageous effects on reducing greenhouse gas (GHG) emissions and carbon footprints. The potential for the wood-based panels to be utilised in innovative and challenging constructions mostly relies on adhesives, which are often manufactured from oil-derived basic materials such as petroleum and natural gas. The crucial solution must encompass the transition to eco-friendly adhesives, the reduction of carbon dioxide emissions, and the adoption of more sustainable solutions. In a bid to create a more sustainable and conscious society, environmental regulations have also drawn attention to the use of green design principles and the production of bio-based adhesives from raw materials. Therefore, the key to satisfying the wood industry's current green expectations is creating an environmentally friendly adhesive using renewable resources. On the other side, making the adhesive bond as strong as or stronger than the wood itself is crucial for structural applications.

Tannic acid (TA), as a natural polyphenolic polymer, has shown big potential to be used as an eco-friendly bio-adhesive and alternative to petroleum-based adhesives, offering sustainability and reduced toxicity. In wood processing, tannic acid adhesives are valued for their strong bonding capabilities and natural origin, reducing dependence on synthetic resins. This study aimed to evaluate the potential of tannic acid application in conventional urea-formaldehyde (UF) wood adhesive formulations. Tannic acid-based UF (TA-UF) resins, with three different concentrations of tannic acid (1, 3, and 5% wt), were prepared, and adhesive properties were tested and compared with properties of pure UF resin. Testing of tensile shear strength showed that the addition of a higher concentration of tannic acid in UF adhesive formulation increases its adhesive and mechanical performances compared to pure UF adhesive, which implies that TA-UF resins could be successfully applied as an environmentally friendly, bio-based wood adhesive.

Keywords: tannic acid, UF resin, wood adhesive, biomaterials, bio-adhesive.

1. INTRODUCTION

Urea-formaldehyde (UF) resins are among the most widely used adhesives in the wood-based panel industry due to their low cost, high reactivity, and good bonding performance. However, their primary drawback lies in the emission of free formaldehyde during and after curing, which poses significant health and environmental concerns. In response to increasingly stringent regulations on formaldehyde emissions, recent research efforts have focused on modifying UF resins using bio-based additives that can improve environmental performance without compromising adhesive quality.

One such promising additive is tannic acid, a naturally occurring polyphenolic compound found in various plant sources. Due to its abundance of phenolic hydroxyl and carboxyl groups, tannic acid can potentially react with free formaldehyde and participate in cross-linking reactions within the resin matrix, thereby reducing emissions while enhancing or maintaining mechanical strength. Evaluating the performance of bio-based adhesive systems requires a comprehensive understanding of their interaction with the wood substrate. In addition to assessing tensile shear strength and formaldehyde emission levels, it is essential to analyse the chemical structure of the modified resins (e.g., via FTIR spectroscopy) and to characterise the failure mode after mechanical testing. Whether the failure is cohesive, adhesive, mixed, or substrate-related, it offers crucial information about the efficiency of the modification and the quality of the adhesive bond.

Tannic acid (TA) is a colourless to pale yellow solid with an astringent taste [1]. Due to its wide range of special chemical properties and health benefits, it became one of the most researched substances that serves as a commercial, raw additive in the coating, adhesive, health, pharmaceutical and food industries. TA is also known for its antimicrobial, anticancer, antiviral, and anti-inflammatory properties [2].

TA has a promising application as a non-toxic and inexpensive green crosslinking agent for multifunctional biomaterials, especially in the industry of wood adhesives [3]. Formaldehyde-based resins are used as binders in the production of wood-based panels thanks to their low cost and high reactivity advantages [4]. Urea formaldehyde (UF) is the dominant type of adhesive in the production of wood-based panels, due to its favourable characteristics, but these resins also have certain disadvantages related, first of all, to poor water resistance and formaldehyde emission (FE) [5]. Tannic acid can be used to fabricate bioadhesives by partial or complete replacement of phenolic substances in conventional wood adhesives [6].

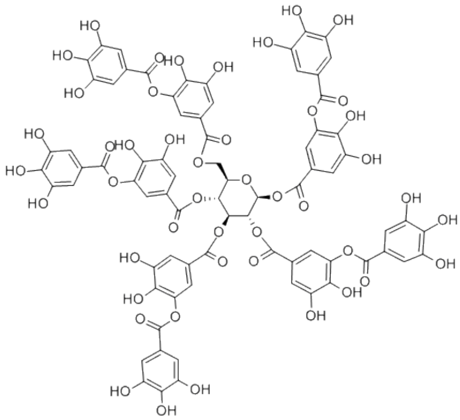

Figure 1. *Tannic acid structure.*

In this study, UF resins were modified with varying concentrations of tannic acid (1%, 3%, and 5%) to investigate its effect on the chemical structure, bonding performance, and failure behaviour of adhesive joints. The goal was to assess the feasibility of tannic acid as a formaldehyde-reducing, performance-preserving additive for UF adhesives used in wood composites. Tannic acid (TA) is a natural polyphenol and the most abundant natural compound after cellulose, hemicellulose and lignin [1]. This substance is widely found in plants, including hydrolysed and condensed tannins. Hydrolysable tannins consist of gallic or ellagic acid and sugar molecules (e.g., glucose), while tannic acid is composed of polymerised flavonoids with different degrees of polymerisation. Tannic acid is the most common hydrolysable tannin and is composed of a glucose ring esterified with five digallic acids [2,3]. Although the TA molecule carries no carboxylic acid group, it is called “acid” due to the presence of numerous phenol groups, which are responsible for its acidic character [3].

2. EXPERIMENTAL SECTION

2.1 Materials

A commercial UF resin (UF) was provided by a domestic company (Serbia). Tannic acid was purchased from Sigma Aldrich Chem (Steinheim, Germany). Ammonium chloride was purchased from Centohem (Serbia). Beech (*Fagus sylvatica*) logs were selected from a known locality and growth conditions (mountain Goč, Serbia). Afterward, primary boards with the desired orientation of growth rings were cut.

2.2. Tensile shear strength determination

2.2.1. Preparation of wood samples

The sawn timber was dried in a semi-industrial conventional kiln (Nigos MC 3000, capacity 0.8 m^3). The most homogeneous groups of testing samples were selected for further experiments.

2.2.2. Preparation of UF-tannic acid adhesive

Firstly, the adhesive solid content of UF resin was determined to be used for further adhesive formulations and analyses. Every formulation was prepared in a concentration calculated using solid weight relative to UF resin solid content. Ammonium chloride salt solution (concentration of 20%) was used as a hardener for all samples in the concentration of 1% w/w. Tannic acid was added to UF adhesive in concentrations of 1%, 3% and 5%, respectively, by weight. One group of samples, with pure UF resin, was used as a control group.

2.2.3. Characterisation of UF adhesive

Adhesive obtained in this research was performed according to the following standards: determination of pH (SRPS EN 1245:2012); determination of density (SRPS EN 542:2009); determination of conventional solids content and constant mass solids content (SRPS EN 827:2009); determination of tensile shear strength of lap joints for wood adhesives (SRPS EN 205:2017); and the adhesive was classified according to the standard for classification of thermosetting wood adhesives (SRPS EN 12765:2017).

The FTIR-ATR spectra were obtained with an FTIR spectrometer (iS20, Thermo Nicolet) with a resolution of 4 cm^{-1} in the wavelength region 4000–525cm^{-1}, using a diamond single reflection attenuated total reflectance (ATR). All spectra were obtained with 32 scans, and the background measurement was made using air.

2.2.4. Determination and optimisation of glue-line quality

The adhesive mixes were applied by a rubber roller onto one surface of the two wood specimens to be bonded (200 g/m^2). Assembling was always performed in parallel grain directions. The ply without direct application of the adhesive mix was always in the bottom position to improve the penetration into its structure. Again, a special effort was made to have the taper as low as possible, guaranteeing equal penetration conditions for all samples. Five joint samples were pressed in a hydraulic press at 120°C and 1.5 MPa for 15 minutes. Before testing, the single lap shear test specimens (150 mm × 20 mm × 5 mm) were conditioned at 20 ± 2°C and 65 ± 5% relative humidity for one week. The lap shear test will be conducted according to the SRPS EN 205 standard test on a hydraulic test machine (Wood tester WT4) with a measuring scale of 50 kN at a testing speed of 6 mm/min loading rate in tensile mode with the load direction always parallel to the grain in all tested specimens. The failure zone (shear area 20 mm × 10 mm) was examined using a light microscope to determine the proportion of wood failure and the thickness of the wood layer in the wood failure. Five replications were performed for each set of parameters. An analysis of variance (ANOVA) was applied to obtain centralised values and standard errors.

3. RESULTS AND DISCUSSION

For any bio-based wood adhesive, a comprehensive evaluation of its performance in wood composite applications is essential. Understanding the interaction between the adhesive and the wood substrate provides valuable scientific insight that informs the development of adhesives with improved mechanical strength, water resistance, thermal stability, rheological behaviour, and penetration capability.

In interpreting the measured bonding strength, it is also critical to consider the failure mode observed during mechanical testing. Four principal failure modes are typically recognised in adhesive-bonded wood composites:

(a) cohesive failure within the adhesive layer,
(b) adhesive failure at the interface,
(c) mixed failure, which combines both cohesive and adhesive elements, and
(d) wood cohesive failure, where the failure occurs within the wood substrate itself.

The identification of failure type provides key information about the bond quality and the relative strength of the adhesive versus the substrate and should be considered alongside strength data when assessing adhesive performance. UF adhesive is the most important adhesive in the wood industry, especially in the production of wood-based panels, primarily because of its relatively good characteristics and low price. However, considering that it has poor water resistance and high formaldehyde emission, it is necessary to make some modifications to the chemistry of the glue itself in order to make it more environmentally friendly. Here, we created modified adhesive formulations based on commercial UF resins (UF), cured both alone (UF) and with tannic acid (UFTA1, UFTA3, and UFTA5), and assessed their adhesive properties when the shear strength of samples with varying TA contents changed.

Table 1. *The technical properties of UF resin.*

Sample	Property					
	Solid content (wt%)	Density (g/cm^3)	Gel time (s)	pH	Free-F (wt%)	Viscosity, F^4_{20} (s)
UF	68	1.290	71	8.0	0.14	84

3.1. FTIR analysis

Fourier Transform Infrared (FTIR) spectroscopy was used to analyse the chemical structures of the adhesive resins. The corresponding FTIR spectra are presented in Figure 1, where Figure 1a displays the spectra of the commercial UF adhesive emulsion and its cured form (cUF), while Figure 1b shows the spectra of cured UF adhesives modified with 1%, 3%, and 5% tannic acid (labelled as UFTA1, UFTA3, and UFTA5, respectively).

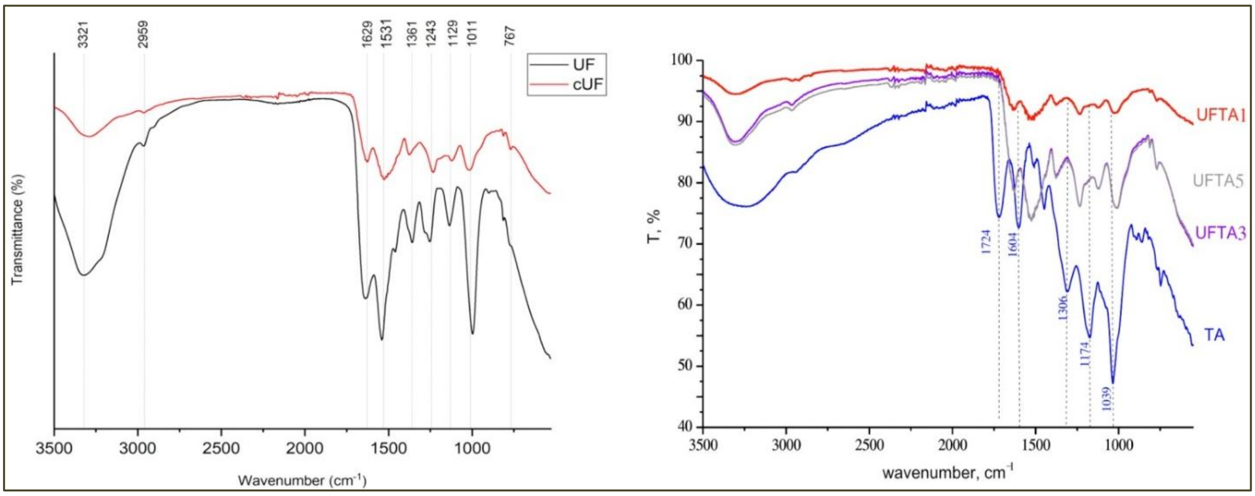

Figure 2. *FTIR spectra of: 1a) pure, commercial UF adhesive emulsion and cured resin (cUF); 1b) cured UF adhesive with different tannic acid concentrations (1%, 3%, 5%).*

As shown in Figure 1a, both the UF adhesive and its cured form (cUF) exhibit characteristic absorption bands. The broad peak around 3320 cm^{-1} corresponds to O–H and N–H stretching vibrations, while the band near 2969 cm^{-1} is attributed to C–H stretching of methyl and methylene groups. Peaks at 1659 cm^{-1} and 1531 cm^{-1} are assigned to C=O stretching and N–H bending of amide groups, respectively, indicative of urea-formaldehyde network formation. The cured resin (cUF) shows slight shifts and reduced intensity in these regions, suggesting partial reaction and cross-linking during curing.

In Figure 1b, the FTIR spectra of tannic acid-modified resins (UFTA1, UFTA3, and UFTA5) show notable differences compared to the unmodified cured resin. With increasing tannic acid content, a progressive reduction in the intensity of amide-related bands (1650–1530 cm^{-1}) is observed, indicating potential interaction between tannic acid and the resin matrix. Furthermore, enhanced absorbance around 1020–1240 cm^{-1} suggests increased C–O and C–N bond formation, likely due to the incorporation of polyphenolic structures from tannic acid. These spectral changes confirm chemical modification of the UF resin network, which may influence both the reactivity and emission behaviour of the adhesive.

The observed FTIR spectral changes are consistent with the expected chemical interactions between tannic acid and the UF resin network. Tannic acid, being rich in phenolic hydroxyl and carboxyl groups, can react with free formaldehyde or participate in the cross-linking process, thereby reducing the amount of unreacted formaldehyde available in the system. This is supported by the diminished intensity of amide bands and the appearance of new C–O and C–N stretching vibrations in the modified samples, particularly in UFTA3 and UFTA5.

3.2. Tensile shear strength determination

The tensile shear strengths of examined adhesives are shown in Figure 2. The tensile test results of the samples demonstrated that the resin mixed with tannic acid (1, 3 and 5% by weight) had very good performance compared to pure UF resin. The effect of tannic acid on the properties of UF adhesive is mainly related to its addition concentration and the crosslinking of tannic acid with polymer. Shear strength was slightly lower in the case with 1% and 3% tannic acid addition but increased with a concentration of 5% of tannic acid.

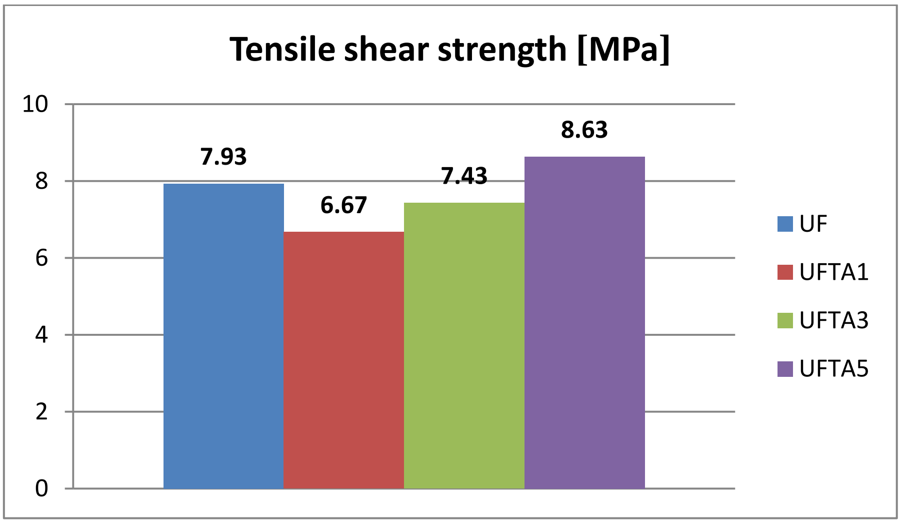

Figure 3. *Shear strength of beech wood samples with cured pure UF adhesive (UF) and samples with tannic acid addition in different concentrations (UFTA).*

The tensile shear strength of the unmodified and tannic acid-modified UF adhesives is presented in Figure 3. The reference UF resin exhibited a strength of 7.93 MPa, serving as the baseline for comparison. The addition of 1% tannic acid (UFTA1) resulted in a decrease to 6.67 MPa, suggesting that low tannic acid content may interfere with optimal polymer network formation or introduce brittleness at the adhesive interface. An increase to 3% tannic acid (UFTA3) led to partial recovery of shear strength (7.43 MPa), while the 5% formulation (UFTA5) outperformed the reference resin, reaching 8.63 MPa. This improvement is likely due to enhanced cross-linking and interaction between the phenolic groups of tannic acid and the resin matrix, which strengthens the adhesive bond.

These findings confirm that while small additions may temporarily weaken bonding, higher tannic acid content improves tensile shear strength, likely through chemical integration into the resin network. Combined with reduced formaldehyde emissions (as indicated by FTIR), the UFTA5 formulation demonstrates potential for more sustainable and high-performance UF adhesives.

The different failure modes observed after the adhesion tests—namely cohesive, adhesive, and substrate failure—were identified through surface analysis of the bonded specimens using high-resolution imaging. Cohesive failure was defined as adhesive remaining on both overlapping surfaces, indicating internal failure within the adhesive layer. Adhesive failure occurred when adhesive was present on only one surface, reflecting poor bonding at the interface. Substrate failure was characterised by the fracture of the wood substrate itself, with the adhesive bond remaining intact. All tested specimens exhibited 100% substrate failure, regardless of tannic acid concentration. This consistent failure mode strongly suggests that the adhesive bond strength exceeded the internal strength of the wood substrate and that the bond was not the weakest point in the assembly. From a technical and industrial perspective, this outcome is highly desirable, as it confirms the structural integrity and reliability of the adhesive joint under load.

Moreover, the occurrence of substrate failure even in samples modified with tannic acid (UFTA1, UFTA3, UFTA5) indicates that the incorporation of bio-based additives did not compromise bonding performance. In fact, the enhanced tensile shear strength observed for UFTA5, combined with substrate failure as the dominant fracture mode, highlights the potential of tannic acid-modified UF resins for structural applications where high bond strength and environmental performance are both critical.

Figure 4. *The joint samples underwent complete (100%) wood fracture following tensile shear testing.*

4. CONCLUSION

Wood adhesives based on UF resin and tannic acid were prepared and characterised, and their adhesive and mechanical properties were investigated to determine their potential as wood adhesives in order to reduce formaldehyde emission. This study showed how adding tannic acid, a bio-based phenolic chemical, to urea-formaldehyde (UF) adhesives could enhance their mechanical and environmental performance. FTIR spectroscopy confirmed chemical interactions between tannic acid and the UF resin matrix, evidenced by changes in functional group intensities and the emergence of new bands corresponding to C–O and C–N bonds. These findings indicate partial integration of tannic acid into the polymer network.

The modified adhesives exhibited variable tensile shear strength depending on tannic acid content. While a 1% addition (UFTA1) resulted in decreased strength, higher concentrations, particularly 5% (UFTA5), improved mechanical performance beyond that of the unmodified UF resin. Notably, all adhesive formulations - regardless of tannic acid content - resulted in 100% substrate failure, confirming that the bonding strength exceeded the internal strength of the wood substrate.

In addition to mechanical improvements, the presence of tannic acid is known to reduce formaldehyde emissions due to its phenolic and acidic functional groups, which can chemically bind or adsorb free formaldehyde. This dual effect — enhanced bond strength and reduced emissions — positions tannic acid-modified UF resins as a promising and more sustainable alternative for wood-based composite production.

Further research may focus on optimising the concentration and distribution of tannic acid within industrial formulations, as well as evaluating long-term durability and emission behaviour under variable environmental conditions.

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