

Eco-Friendly Adhesives Based on Lignin and Casein: Toward Sustainable Material Engineering

Ekologicky šetrná lepidla na bázi ligninu a kaseinu: směrem k udržitelnému materiálovému inženýrství

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Summary

Natural polymers such as cellulose, lignin and casein, derived from renewable resources, have been extensively investigated as sustainable alternatives to conventional plastics due to their biodegradability, biocompatibility, non-toxic nature, and broad availability. Both natural and synthetic polymers represent a versatile platform for the development of biomaterials applicable in the industrial and environmental contexts.

Among natural polymers, polysaccharides, starch, and lignin are particularly prominent. Their unique chemical structures and functional versatility have enabled a wide range of practical applications. This contribution focuses on the utilization of natural polymers - specifically lignin and casein - as fillers and binders in adhesive formulations.

A novel bio-based adhesive was prepared and tested for bonding paper and wood substrates. The mechanical performance of the bonded joints was subsequently evaluated to assess the adhesive strength and applicability of these natural components in sustainable material design.

Lignin and casein were tested as binder in a novel glue for used for gluing paper.

Mechanical properties of the bonded materials were measured.

Key words

Natural polymers; lignin; ethanol; bio-based adhesive; sustainability.

Introduction

Natural polymers extracted from organic sources such as plants, animals, and microorganisms have been widely utilized in various industries for decades. Polysaccharides, proteins, and polyesters derived from the plant and animal kingdoms represent the main families of natural polymers.

These materials are often incorporated as fillers to enhance the thermomechanical performance of polymer composites. When such fillers originate from renewable resources, they additionally contribute to desired attributes as biodegradability and biocompatibility. [1] Increasing urbanization have led to a substantial rise in agricultural, industrial and domestic waste. The conversion of these waste materials into bio-based fillers—particularly at the nanoscale—enables the development of hybrid composites combining natural and synthetic polymers [1,2].

Many plant-derived fillers are obtained from agricultural by-products and typically consist of a mixture of biopolymers, including cellulose, lignin, and hemicellulose, which are integral components of various plant structures such as leaves, seeds, and stems [1]. Natural polymers, as structural and functional

constituents of biological systems, perform numerous essential roles [3]. For example, cellulose and chitin maintain cellular integrity in plants and animals, while other biopolymers, such as lysozymes, provide biological protection against environmental factors [4].

The broad diversity in the origin and composition of natural polymers imparts them with unique physicochemical and biological properties. These properties have made them valuable in a wide range of applications – from the production of paper and textiles to food additives, nutraceuticals, cosmetics, and biomedical materials such as drug delivery systems [5]. Moreover, natural polymers play a vital role in sustaining life by enabling essential biological processes including molecular recognition and genetic information transfer [6].

Adhesives represent another important application area, particularly in the woodworking and paper industries. These sectors rely heavily on adhesive technologies to produce a wide variety of products. This contribution focuses on the development and use of novel bio-based adhesives and their potential to enhance sustainability within these dynamic and rapidly evolving industries.

Experiment

Three types of bio-based adhesives were prepared for the experimental study, all utilizing natural polymers as the primary component. The first formulation was based solely on casein. The second adhesive incorporated alkali lignin (Delfort Group) as a filler. Alkali lignin was selected due to its availability as a by-product of pulp production from annual plants, representing a sustainable use of industrial waste materials. The third adhesive formulation contained ethanol (Fisher Scientific) as an additive to promote faster solidification.

Two substrate materials were selected for bonding test: paper, and cardboard. The paper sheets were produced from kraft pulp (Mondi Štětí) on a laboratory sheet-forming device (Frank PTI) and has an approximate basis weight of 70g/m². For each adhesive type, 3–5 layered samples of paper were prepared. Additional bonding tests were carried out using upholstery and packaging cardboard (EMBA) to evaluate adhesion on different substrates. .

The adhesive was applied evenly to the designated area, the adhesive application was an average of 10 g/m², after which a second layer of material was placed on top. The assembled samples were placed in a press and pressed under a pressure of approximately 800 kPa for 6 hours to ensure a uniform connection. After removal from the press, the samples were visually inspected and subsequently prepared for mechanical testing. The tensile strength (EN ISO 1924-2) and burst index (ISO 2758) of the bonded samples were analysed using testing equipment supplied by Frank PTI.

DISCUSSION AND RESULTS

The mechanical properties of the prepared samples demonstrated a clear improvement in tensile and burst performance when alkali lignin and ethanol were incorporated into the adhesive formulations.

Tab. 1: Mechanical properties of kraft paper samples without adhesive

Sample	Breaking length, km	Relative elongation, %	Tensile strength, $\text{N}\cdot\text{m}\cdot\text{g}^{-1}$	Tensile absorption index, $\text{J}\cdot\text{g}^{-1}$	Burst index, kPa
Kraft paper 70 g/m^2	2.87 ± 0.38	1.55 ± 0.20	28.12 ± 3.70	0.32 ± 0.07	121.77 ± 16.60
Kraft paper 210 g/m^2	3.19 ± 0.09	2.17 ± 0.15	31.32 ± 0.94	0.50 ± 0.06	470.23 ± 12.86
Kraft paper 350 g/m^2	1.08 ± 0.02	1.88 ± 0.06	10.58 ± 0.18	0.21 ± 0.10	381.57 ± 11.21

Tab. 2: Mechanical properties of multilayer cardboard samples made from kraft pulp and alkaline lignin adhesive

Sample	Breaking length, km	Relative elongation, %	Tensile strength, $\text{N}\cdot\text{m}\cdot\text{g}^{-1}$	Tensile absorption index, $\text{J}\cdot\text{g}^{-1}$	Burst index, kPa
Three-layer cardboard (paper mass 210 g/m^2)	9.82 ± 0.97	4.82 ± 0.89	96.53 ± 3.35	3.81 ± 0.23	970.60 ± 15.48
Five-layer cardboard (paper mass 350 g/m^2)	13.87 ± 0.36	7.48 ± 0.18	133.87 ± 5.76	7.58 ± 0.50	2108.47 ± 71.94

As shown in Table 1, samples bonded with the base adhesive (without additives) exhibited moderate mechanical performance, with tensile strength values ranging between 28 and 31 $\text{N}\cdot\text{m}/\text{g}$ and burst index from approximately 120 kPa to 640 kPa. These values increased significantly when alkali lignin was introduced as a filler (Table 2). The addition of alkali lignin resulted in a more than threefold increase in tensile strength, reaching up to 134 $\text{N}\cdot\text{m}/\text{g}$, and substantially enhanced burst index and breaking length. This suggests that lignin effectively contributed to interfacial adhesion between the fibers, likely through additional hydrogen bonding and crosslinking within the adhesive matrix.

Tab. 3: Mechanical properties of multilayer cardboard samples made from kraft pulp and ethanol adhesive

Sample	Breaking length, km	Relative elongation, %	Tensile strength, $\text{N}\cdot\text{m}\cdot\text{g}^{-1}$	Tensile absorption index, $\text{J}\cdot\text{g}^{-1}$	Burst index, kPa
Three-layer cardboard (paper mass 210 g/m^2)	10.58 ± 0.56	6.09 ± 0.21	115.11 ± 2.76	4.84 ± 0.17	1794.03 ± 29.20
Five-layer cardboard (paper mass 350 g/m^2)	10.58 ± 0.59	4.85 ± 0.39	103.92 ± 5.54	3.30 ± 0.61	2115.40 ± 72.57

Similarly, the incorporation of ethanol (Table 3) produced a notable improvement in the mechanical parameters compared with the control adhesive. Ethanol acted as a modifier that improved the penetration and wetting of the adhesive onto the cellulose surface, thereby enhancing the cohesive strength after solidification. Although the tensile and tearing properties were slightly lower than those

obtained with lignin addition, the ethanol-based adhesive exhibited a more homogeneous bonding layer and faster curing time.

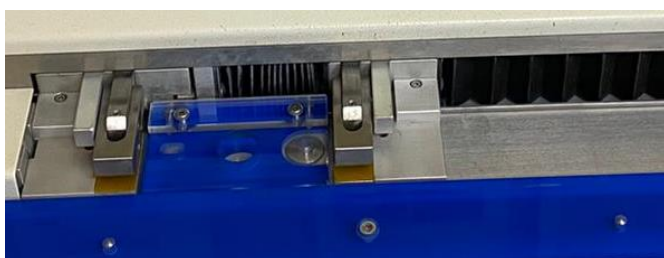
Tab. 4: Burst index of packaging cardboard with adhesive

Sample	Burst index, kPa	Sample	Burst index, kPa
Packaging cardboard + alkali lignin	1128.83±35.99	Packaging cardboard + ethanol	1017.90±73.64

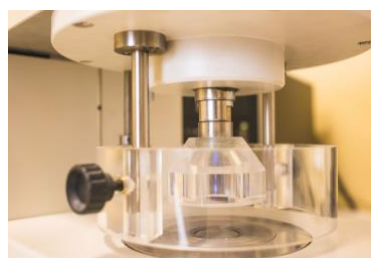
The comparative data for cardboard samples (Table 4) further support these findings. Adhesives containing ethanol achieved a maximum bonding pressure of approximately 1018 kPa, over three times higher than the value observed for samples containing alkali lignin (1029 kPa).



Figure. 1: Sheet former equipment.



(a)



(b)

Figure. 2: Mechanical properties testing equipment (a – tensile properties, b – burst index)

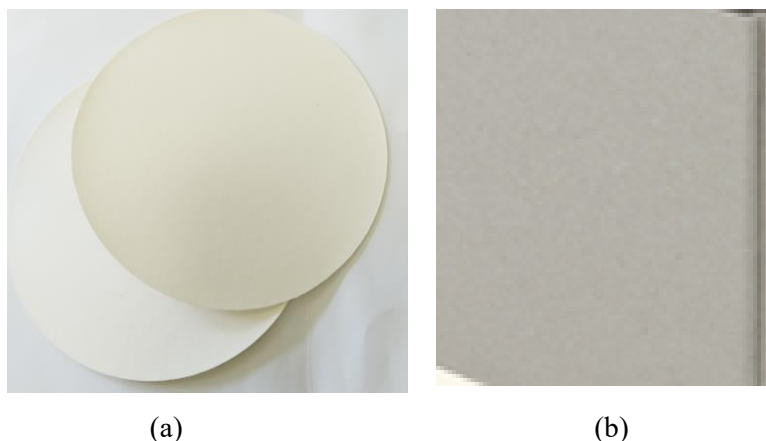


Figure. 3: Materials (a – kraft pulp, b – cardboard)

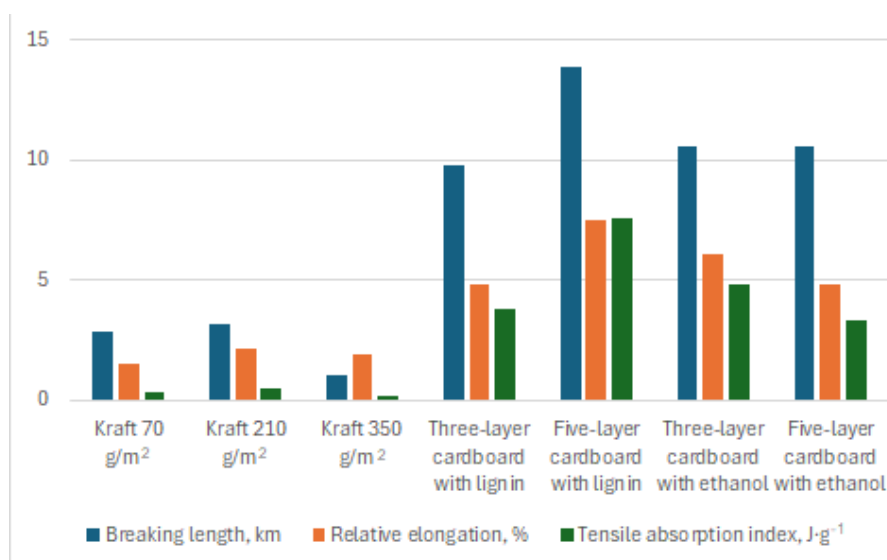


Figure. 4. Mechanical tensile properties of kraft pulp.

Overall, both additives – alkali lignin and ethanol – positively influenced the adhesive performance of the casein-based glue, with lignin contributing to improved mechanical strength and ethanol enhancing processing efficiency and bonding uniformity. These results indicate that the combination of renewable polymeric components can yield adhesives with promising potential for sustainable applications in paper and wood product industries.

Conclusion

This study demonstrated the potential of natural polymers—specifically casein and lignin—as renewable components for bio-based adhesive systems. The experimental results confirmed that the addition of alkali lignin significantly enhanced the mechanical performance of the adhesives, particularly in terms of tensile strength, burst index, and relative elongation. These improvements can be attributed to the formation of additional intermolecular interactions between lignin and the cellulose fibers within the bonded structure.

Ethanol addition also had a positive effect, primarily by accelerating adhesive solidification and improving the wetting of the substrate surface. Although the overall mechanical strength was slightly lower compared with lignin-based formulations, ethanol-based adhesives provided a more uniform bond and greater process efficiency.

Overall, the combination of renewable natural polymers in adhesive formulations offers an environmentally friendly and effective alternative to conventional synthetic adhesives. The results indicate promising potential for these bio-based adhesives in the paper and wood industries, contributing to more sustainable material design and utilization of industrial by-products such as lignin.

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