Extraction of lignin from Eucalyptus and Casuarina wood chips and coating on paperboard for sustainable food packaging applications

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Abstract. The increasing demand for sustainable and biodegradable food packaging materials has encouraged the use of lignin-based coatings. In this work, lignin was extracted from Eucalyptus and Casuarina wood chips (in the ratio of 1:1) as a byproduct of the kraft pulping process and applied as a coating material. To improve its performance, lignin was blended with acrylic latex, a binder for pigment. The coating was applied on 160 gsm paperboard using a laboratory bar coater and dried in a hot-air oven at 105 °C. The coated paperboard was tested for its physical and mechanical properties. To understand the effect of Lignin, sheets coated with acrylic latex also studied. The resulting paper had a GSM of 160.9 and a coat weight of 0.71 g/m². The tensile strength and stiffness (GM) were measured as 11667 N/m and 41 mN respectively. Cobb60 value is 20.1 g/m², the burst strength reached 560.1 kPa also exhibited an opacity of 99.99%. In addition, shade and colour difference were also analysed. Overall, the results indicate that lignin can be effectively utilized as a bio-based coating material, providing an eco-friendly material that has prominent light stopping and UV absorbing ability than conventional polymer coatings for sustainable packaging applications.

1 Introduction

The packaging sector is undergoing a major shift due to the growing concern about the environmental problems caused by synthetic plastics. The conventional plastic materials, which have been widely used for decades because of their durability, cost-effectiveness and versatility are now recognized as a major contributor to pollution, landfill accumulation, and microplastic contamination [1]. In the context of food packaging, the issue is even more critical since single-use plastic containers, films and coatings often end up in waste streams within a very short time.

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As a result, the alternative is needed and it should be both sustainable and functional. Paper and paperboard are already popular choices because of their biodegradability, recyclability and relatively low cost [2]. However, unmodified paperboard suffers from certain limitations such as poor water resistance and restricted mechanical strength. These shortcomings limit its direct application in food packaging unless surface treatments or coatings are introduced to enhance its performance. One of the promising directions to overcome these issues is the use of bio-based coatings. Among several natural polymers, lignin has attracted considerable attention. Lignin is the second most abundant biopolymer on Earth, next to cellulose and is naturally present in the cell walls of vascular plants [3]. It functions as a structural component that provides rigidity, resistance against microbial attack and hydrophobicity to plant tissues [4]. Despite its abundance, lignin is underutilized in industry. In pulping operations such as the kraft process, large amounts of lignin are generated as a byproduct and this lignin is often burned as a fuel to generate energy for the mill [5]. However, this approach overlooks the potential of lignin as a value-added material for advanced applications. Lignin is tried as a coating material in paper-based packaging by considering its properties.

Eucalyptus and Casuarina woods are widely available pulp wood as a raw material in pulp and paper production. The kraft pulping yields a significant quantity of lignin with distinct chemical and physical properties [6]. The extraction of lignin from this source not only provides a renewable feedstock for material development but also contributes to a circular economy by converting a waste byproduct into a useful product. The kraft lignin obtained has functional groups such as hydroxyl, methoxy and phenolic structures [7] which can participate in chemical interactions when used in composite or coating formulations. These chemical features are expected to improve adhesion to paper fibers and enhance barrier performance. In food packaging applications, surface coatings are essential for improving the quality and shelf life of packed goods. Acrylic latex, which is commonly used in conventional pigment coatings, mainly binds the pigments with the substrates, also provides flexibility and film-forming ability. Lignin, in comparison, is hydrophobic in nature and available at large scale from existing pulp mills, making it an attractive material [8]. Furthermore, lignin exhibits antioxidant and antimicrobial properties, which could contribute additional functional advantages in food packaging by delaying oxidation and microbial spoilage [9].

The present study focuses on extracting lignin from black liquor of kraft pulping process using Eucalyptus and Casuarina (1:1) wood chips and applying it as a coating on paperboard for food packaging applications. The coated samples are then evaluated for their physical and mechanical properties to assess their suitability for packaging. This approach not only strengthens the functional properties of paperboard but also contributes to the long-term vision of green packaging technologies.

2 Methods

2.1 Kraft pulping process and black liquor

Freshly harvested Eucalyptus and Casuarina wood logs were debarked to remove the outer bark along with any surface impurities. The debarked logs were fed into the Chipper unit and converted into wood chips. The wood chips underwent screening process as per Tappi UM 21 and the screen accept chips were fed into the kraft pulping digester. The digester was charged with wood chips and with recommended dosage of white liquor and cooked. The cooked brown pulp passed through the press section where the black liquor and cooked pulp were separated. The black liquor was collected for analysis.

2.2 Extraction of Lignin from Eucalyptus wood chips

The collected black liquor was around 12.5 pH and was cooled down to room temperature. The kraft lignin was separated by acid precipitation process. The cooled down black liquor was acidified with drop-by-drop addition of 4N H2SO4 till 2 pH and observed lignin precipitation. A small dose of Cationic polymer was added to enhance the lignin precipitation. The lignin was settled overnight, filtered using whatman filter paper 41, washed with demineralised water, air dried and stored for further characterization.

2.3 Preparation of lignin solution for coating

A lignin solution intended for paperboard coating was prepared by dispersing 2 g of dried kraft lignin in 20 mL of distilled water along with 25 parts of acrylic latex. The mixture was continuously stirred using a magnetic stirrer at room temperature to achieve proper dispersion. The mixture was stirred for about 1 h until a uniform brown-coloured dispersion was obtained. This prepared solution was carefully stored in a sealed container to avoid contamination or pH changes before being used for paperboard coating experiments. Also to understand the real effect of lignin, another solution with 25 parts of binder in 20 ml of water was made for analysis.

2.4 Coating of lignin in paperboard

The prepared lignin solution was coated onto the surface of paperboard using a laboratory bar coater to achieve a uniform and controlled layer. Before coating, the paperboard sheets were cut into A3 size and conditioned at 23 ± 2 °C and 50 ± 5% relative humidity for 24 h to stabilize their properties. The uncoated paperboard was fixed in the laboratory bar coater with plain calibration rod and a measured volume of the lignin solution, typically 2–3 mL, was dispensed and coated. After coating, the samples were dried at 105 °C for 2 mins to ensure proper curing. Finally, the coated sheets were stored in conditioned atmosphere for later testing. Similar procedure was applied and sheets coated with latex solution also made and store in conditioned atmosphere for further testing.

2.5 Conditioning of coated samples

The uncoated and coated samples were conditioned as per Tappi T402. The standard recommends the standard atmosphere of 23°C and 50% relative humidity (RH) for conditioning the paperboard before testing.

2.6 Anisotropy

The paperboard exhibits the anisotropic nature due to the alignment of pulp fibres during sheet forming in machine and also lead to difference in test results with respect to direction. Paperboard mechanical properties are normally tested for Machine direction (MD) and Cross direction (CD). The bending stiffness also in the similar manner and additionally Geometric mean (GM) Stiffness will be calculated as follows,

$$Stiffness\ (GM) = Sqrt\ (MD * CD) \quad (1)$$

2.7 Coating Thickness

A digital micrometer complying T 411 was used for thickness measurements. Five measurements were taken at different locations on each sample and the average was reported.

2.8 Grammage

The conditioned coated and uncoated samples were cut into the dimensions of 12.5 x 12.5 cm and their weights were determined using an analytical balance with 0.1 mg accuracy. The GSM was calculated by dividing the measured weight by the corresponding area of the sample. Each sample was measured five times and the average value was reported.

2.9 Coat weight

The coat weight of lignin dispersion on paperboard was determined by weighing samples collected of coated and uncoated area of the sheet using an analytical balance with the accuracy of 0.1 mg. The difference in mass was divided by the area to calculate coat weight (g/m^2). The samples were weighed five times and the calculated average coat weight value was reported.

2.10 Tensile Strength

The tensile strength of coated and uncoated paperboard samples was measured to evaluate the effect of lignin coating on mechanical strength. The paperboard samples were cut into strips of standard dimensions and tested according to T ISO standard 1924/2. The tensile strength was measured using a Horizontal tensile tester at a constant rate of elongation method (20 mm/min) as per ISO 1924/2. The tensile strength at break was measured for both MD and CD direction samples. For each samples, five strips were cut both in MD and CD direction, tested and the average value was reported to ensure accuracy and reproducibility.

2.11 Stiffness

The stiffness of coated and uncoated paperboard samples was tested to assess their rigidity and resistance to bending. The samples were cut from conditioned specimens in both MD and CD direction using the sample cutter. The measurements were performed using a bending tester according to TAPPI T 556 standard. The samples were tested for top and bottom bending resistance since the paperboard samples are multi-layer paperboard and its average was taken for both MD and CD. The average Stiffness GM of uncoated and coated samples were calculated using the corresponding average Stiffness of MD and CD samples. All the samples were tested, collected five set of readings and the average values were reported to confirm the reliability and reproducibility of the results.

2.12 $Cobb_{60}$ analysis

The water absorption of coated and uncoated paperboard samples was tested using the Cobb60 following TAPPI T 441 standard. The amount of water absorbed, expressed in g/m^2, was calculated by the difference in initial and final weight of samples after testing. A set of five measurements for each sample were tested and average values were reported.

2.13 Burst Strength

The coated and uncoated paperboard samples were evaluated for the burst strength to determine their resistance to rupture under pressure. The experiment was performed using a standard burst tester following TAPPI T 807 procedures. Five readings were collected for each sample and the average value was reported.

2.14 Shade

The shade of coated and uncoated samples was analysed using a laboratory spectrophotometer (L&W Elrepho) following CIE 1976 system. The uncoated paper shade values: L = 94.1, a = –0.8 and b = 5.1 was taken as the reference. The colour difference (ΔE) between the coated and uncoated samples was calculated using CIE 1976 and the formula is given below.

$$\Delta E = [(\Delta L)^2 + (\Delta a)^2 + (\Delta b)^2]0.5 \quad (2)$$

where ΔL represented the difference between the L value of the uncoated and coated paper, Δa indicated the difference in a value and Δb denoted the difference in b values. For each sample, five measurements were taken and the average ΔE value was reported

2.15 Opacity

The opacity of the coated paper was measured using a laboratory spectrophotometer (L&W Elrepho) following Tappi T 519 standard. For each sample, five measurements were taken at different locations and the mean value was calculated.

2.16 Statistical Analysis

Statistical analysis was performed using SPSS version 20 (IBM SPSS Inc., Chicago, USA). Each experiment was repeated at least five times ($n \geq 5$) and the results were expressed as mean values with their corresponding standard deviations. The significant differences among the groups were determined using Tukey's post hoc test with the level of significance set at $p < 0.05$.

3 Results and Discussion

3.1 Coating Thickness

The thickness of the uncoated, latex coated and lignin-coated paperboard samples are presented in Table 1.

Table 1. Thickness, GSM and Coat weight of paperboard

S. No	Sample	Thickness (μ)	GSM (g/m^2)	Coat weight (g/m^2)
1,	Uncoated	219 ± 0.1	160.0 ± 0.1	-
2,	Latex Coated	221 ± 0.1	160.7± 0.1	0.35 ± 0.01
3,	Lignin Coated	220 ± 0.1	160.9 ± 0.1	0.71 ± 0.01

The uncoated paperboard exhibited a thickness of 219 µm, while the latex coated and lignin-coated paperboard measured 221 µ and 220 µm. The results indicated that the application of the coating (latex / lignin) did not significantly affect the overall thickness of the paperboard. The minimal change in thickness suggested that the coating formed a thin, uniform layer without altering the bulk dimensions of the substrate [10].

3.2 GSM and coat weight

Paperboard is usually measured in the unit Gram per square meter (g/m2)/GSM. The GSM and coat weight of the uncoated, latex coated and lignin-coated paperboard samples are presented in Table 1. The GSM of the uncoated and lignin-coated paperboard samples was measured to quantify the presence of lignin coating as the mass per unit area. The uncoated paperboard exhibited a GSM of 160 g/m2, whereas the lignin-coated paperboard showed a slightly higher value of 160.9 g/m2. This marginal increase in GSM indicated that the lignin coating contributed an additional mass of 0.9 g/m2 to the paperboard. The bone-dry coat weight of the lignin layer was calculated to be 0.71 g/m2, confirming that the coating was uniformly applied in a controlled manner. Similarly, the latex coated sheet has the bone-dry coat weight of 0.35 g/m2. These results indicate that the lignin coating added minimal weight to the substrate, ensuring that the base material properties remained largely unchanged while providing potential functional benefits such as improved light barrier or surface properties [11].

3.3 Tensile strength and Stiffness

The Table 2. represents the tensile strength and stiffness of the uncoated, latex coated and lignin-coated paperboard samples.

Table 2. Tensile Strength MD & CD and Stiffness of paperboard

S. No	Sample	Tensile Strength MD (N/m)	Tensile Strength CD (N/m)	Stiffness GM (mN)
1,	Uncoated	11079 ± 1.0	5080 ± 1.0	40 ± 1.0
2,	Latex Coated	11647 ± 1.0	5165 ± 1.0	44 ± 1.0
3,	Lignin Coated	11667 ± 1.0	5381 ± 1.0	41 ± 1.0

The tensile strength of the uncoated, latex coated and lignin-coated paperboard samples was measured to understand the effect of the lignin coating on the mechanical strength. The uncoated paperboard exhibited a MD tensile strength of 11079 N/m, while the latex coated and lignin-coated paperboard showed an increased value of 11647 N/m and 11667 N/m. Also in CD direction, the tensile test results of uncoated board are 5080 M/m whereas for latex coated and lignin coated sheet are 5165 N/m and 5381 N/m. This improvement in tensile strength suggested that the lignin layer contributed little in comparison with the latex layer, to the overall reinforcement of the paperboard [12]. The coat weight 0.71 g/m2of lignin coating may also be attributed to the increase in tensile in comparison with the coat weight of 0.35 g/m2 of latex coated board.

Similarly, the stiffness of the samples was evaluated in both Machine and Cross direction (MD & CD) to understand the resistance to bending. The Stiffness Geometric mean (GM) was calculated from the average MD and CD stiffness values. The average Stiffness GM of uncoated paperboard was 40 mN, the latex coated paperboard was 44 mN and lignin-coated paperboard was 41 mN. The slight increase in stiffness indicated that the latex coating and lignin coating improved the rigidity of the paperboard, likely due to the uniform deposition of latex and lignin coating on the surface [13].

3.4 Cobb$_{60}$ and Burst Strength

The cobb$_{60}$ and burst strength of the uncoated, latex coated and lignin-coated paperboard samples are presented in Table 3. The water absorption of the paperboard samples was tested by Cobb$_{60}$. The uncoated paperboard exhibited a Cobb$_{60}$ value of 20.8 g/m^2 while the lignin-coated paperboard showed a slightly lower value of 20.1 g/m^2. This decrease indicated that the lignin and latex coating marginally improved the water resistance of the paperboard by forming a thin protective layer on the surface.

Table 3. Cobb$_{60}$ and Burst strength of paperboard

S. No	Sample	Cobb$_{60}$ (g/m^2)	Burst Strength (kPa)
1,	Uncoated	20.8 ± 0.1	536.8 ± 0.1
2,	Latex coated	20.8 ± 0.1	540.2± 0.1
3,	Lignin Coated	20.1 ± 0.1	560.1 ± 0.2

The burst strength of the samples was also measured to assess their ability to withstand pressure. The uncoated paperboard showed a burst strength of 536.8 kPa. The burst strength of the latex coated sample was 540.2 kPa and the lignin-coated paperboard exhibited an increased value of 560.1 kPa. The burst strength results indicated that the lignin and latex layer contributed to improvement in structural integrity of the paperboard thereby providing better rupture resistance under applied pressure [14]. These results confirmed that lignin and latex coating not only slightly improved water resistance but also reinforced the mechanical durability.

3.5 Shade and Opacity

The Shade and opacity of the uncoated, latex coated and lignin-coated paperboard samples are presented in Table 4.

Table 4. Shade and Opacity of paperboard

S. No	Sample	L*	a*	b*	ΔE	Opacity (%)
1,	Uncoated	94.1	- 0.8	5.1	-	96.5
2,	Latex coated	94.1	-0.9	5.4	0.32	96.6
3,	Lignin Coated	80.8	5.7	21.5	22.1	99.99

The shade of the paperboard was significantly influenced by lignin coating. The uncoated paperboard exhibited L, a, b values of 94.1, −0.8 and 5.1, whereas the lignin-coated sample showed values of 80.8, 5.7 and 21.5, resulting in a total colour difference as per CIE 1976

(ΔE) of 22.1. This indicated a noticeable darkening and shift toward red and yellow hues after lignin coating. The opacity of the paperboard improved considerably from 96.5% for the uncoated paperboard to 99.99% for the lignin coated paperboard. The results demonstrated that lignin coating effectively enhanced opacity of the paperboard [15]. This also shows the light stopping ability of the lignin coating which can support packaging of light sensitive materials.

4 Conclusion

The application of lignin extracted from Eucalyptus and Casuarina (1:1) wood chips as a coating along with acrylic latex over paperboard, demonstrated its potential as a sustainable and biodegradable alternative for packaging which demands light stopping ability. The reinforcement by lignin along with acrylic latex coating improved the mechanical and barrier properties of the paperboard, as evidenced by increased tensile strength, stiffness, burst strength, opacity and reduction in Cobb60 results. The coating also resulted a noticeable change in Shade. These findings confirm that lignin-based coatings can serve as an effective, eco-friendly solution for producing durable and sustainable paperboard materials suitable for light sensitive packaging applications. However, the study stimulates the search of suitable form lignin for enhanced barrier performance.

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