

Sustainable Development of Lignin-Based Biodegradable Coating from Palm Tree Waste for Aluminum Foil Applications

Shabib Al Rashdi

National University of Science and Technology

Nageswara Rao Lakkimsetty

`lakkimsetty.rao@aurak.ac.ae`

American University of Ras Al Khaimah

Reem Ahmed Al-Qassabi

National University of Science and Technology

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Abstract

The environmental challenges presented by synthetic, non-biodegradable coatings used in aluminum packaging have accelerated the search for sustainable alternatives. This research aimed to develop and optimize biodegradable lignin-based coatings derived from Date Palm Empty Fruit Bunches (DPEFB)-an abundant agricultural waste in Oman offering an eco-friendly solution that supports sustainable packaging and waste management. Five key objectives guided this work: (1) extract and purify lignin using environmentally friendly alkali treatment; (2) develop and optimize a lignin-based coating formulation to improve barrier performance; (3) apply the coating using scalable techniques such as dip-coating (with potential for roll- or spray-coating); (4) evaluate mechanical, thermal, and barrier properties; and (5) assess the environmental and economic feasibility of replacing synthetic coatings. The lignin extraction process yielded 24.92% purified lignin, confirmed through FTIR analysis (aromatic peaks at $\sim 1510\text{ cm}^{-1}$) and TGA, indicating thermal stability up to 250°C . The coating demonstrated strong adhesion and a 30% reduction in oxygen transmission rate (OTR), along with significantly improved water vapor resistance. SEM imaging verified uniform surface coverage with minimal defects. Economically, the proposed coating offers a 22–28% cost reduction compared to synthetic alternatives. With Oman generating over 3,000 tons of DPEFB annually, this research highlights a valuable pathway for waste valorization. A projected ROI of 1.6 within the first year of semi-industrial implementation confirms its commercial viability. This study presents a scalable, low-cost, and sustainable coating technology aligned with Oman Vision 2040, contributing to environmental protection, industrial innovation, and circular economy goals. It fills a critical gap in the development of biodegradable packaging materials using locally sourced biomass.

1.0 Introduction

The growing demand for sustainable and eco-friendly packaging solutions has placed significant pressure on industries to move away from synthetic, petroleum-based materials. Aluminum foil, widely used for food and industrial packaging, is often coated with synthetic polymers to enhance its barrier properties. These coatings, however, are non-biodegradable, contribute to plastic pollution, and rely heavily on fossil fuels, exacerbating environmental and climate challenges (Patel et al., 2022). To address these issues, natural polymers such as lignin abundantly available in agricultural waste have emerged as a promising alternative for developing biodegradable coatings.

This research focuses on utilizing lignin extracted from Date Palm Empty Fruit Bunches (DPEFB) to create sustainable, biodegradable coatings for aluminum foil. By transforming agricultural waste into value-added products, this research supports the circular economy while addressing the global need for sustainable packaging materials (Smith et al., 2021). There is an urgent need to reduce the environmental footprint of packaging materials. Synthetic coatings, while functional, have persisted in the environment for hundreds of years, contributing to pollution and ecosystem degradation (Liu et al., 2023). Furthermore, the production of these materials is energy-intensive, relying on non-renewable resources that emit significant amounts of greenhouse gases (GHGs). Transitioning to biodegradable

alternatives, particularly those derived from renewable resources such as agricultural waste, is critical to achieving global sustainability goals (Garcia et al., 2022).

The rapid growth of the packaging industry, particularly in food and consumer products, has led to an increasing reliance on synthetic materials such as plastics and chemically treated coatings. These materials pose significant environmental challenges, as they are derived from non-renewable resources and are difficult to degrade, contributing to plastic pollution and environmental harm. Aluminum foil, a widely used packaging material, is often coated with synthetic, non-biodegradable substances to enhance its barrier properties against moisture, oxygen, and other contaminants.

The pressing need for more sustainable packaging solutions, this research explores the potential of using lignin, a natural polymer derived from agricultural waste, as a biodegradable coating for aluminum foil. Lignin is abundant, renewable, and biodegradable, making it an ideal candidate to replace synthetic coatings. Date Palm Empty Fruit Bunches (DPEFB), a byproduct of palm agriculture, are particularly rich in lignin and are often discarded as waste. By extracting lignin from DPEFB and developing it into functional coating, this research aims to reduce reliance on petroleum-based materials, promote waste-to-value initiatives, and contribute to environmental sustainability. However, lignin's complex structure and relatively low solubility present challenges in converting it into suitable coating material. This research seeks to overcome these challenges through innovative extraction, modification, and application techniques. By designing a scalable process for lignin-based coatings, the research aims to demonstrate the feasibility of using natural polymers from agricultural waste to enhance the sustainability of packaging materials, reducing both environmental impact and material waste. To develop and optimize biodegradable lignin-based coatings derived from agricultural waste for aluminum foil applications, reducing environmental and climate impacts while promoting sustainable waste management.

The reviewed literature clearly demonstrates the growing potential of lignin as a sustainable alternative to synthetic polymer coatings, particularly for aluminum foil and packaging applications.

2.0 Methodology

The research methodology was systematically designed to develop biodegradable lignin-based coatings for aluminum foil using lignin extracted from Date Palm Empty Fruit Bunches (DPEFB). The experimental work involved raw material preparation, lignin extraction, physicochemical characterization, coating formulation, coating application, and performance evaluation. Characterization studies were conducted at Sultan Qaboos University (SQU), Oman, and Universiti Kuala Lumpur – Malaysian Institute of Chemical and Bioengineering Technology (UniKL-MICET), Malaysia.

DPEFB was collected from local agricultural farms and date-processing centers in Oman. The raw biomass was initially washed with distilled water to remove adhering impurities, followed by air-drying for 48 hours. The material was then oven-dried at 60°C for 24 hours to eliminate residual moisture. The dried biomass was milled using a ball mill to obtain particles smaller than 500 µm, thereby enhancing

the efficiency of the subsequent lignin extraction process. Lignin extraction was carried out using alkaline treatment followed by acid precipitation. In this process, 100 g of DPEFB powder was treated with 1 L of 10% (w/v) sodium hydroxide solution and refluxed at 90°C for 3 hours under constant stirring. The resulting black liquor was filtered, and the filtrate was acidified with 2 M sulfuric acid to pH 2.0 to precipitate lignin. The precipitated lignin was repeatedly washed with distilled water and oven-dried at 50°C for 24 hours. An average lignin yield of 170 g per 1000 g of DPEFB was obtained, corresponding to a recovery efficiency of approximately 85%.

The extracted lignin was characterized to evaluate its functional groups, thermal stability, and molecular weight distribution. Fourier Transform Infrared Spectroscopy (FTIR) analysis was conducted at UniKL-MICET, Malaysia, and confirmed the presence of characteristic lignin functional groups, including aromatic ring vibrations ($\sim 1510\text{ cm}^{-1}$), hydroxyl groups ($\sim 3400\text{ cm}^{-1}$), and carbonyl groups ($\sim 1720\text{ cm}^{-1}$). Thermogravimetric Analysis (TGA) was performed at Sultan Qaboos University under a nitrogen atmosphere over a temperature range of 25–600°C. The results showed initial weight loss below 150°C due to moisture evaporation and major thermal degradation between 200 and 400°C, indicating the thermal suitability of the lignin for packaging applications. Gel Permeation Chromatography (GPC) analysis was carried out at UniKL-MICET using tetrahydrofuran as the mobile phase with polystyrene standards for calibration. The lignin exhibited a polydispersity index ranging from 2.1 to 2.6, which is suitable for coating applications. Lignin-based coating formulations were prepared by dissolving lignin in an ethanol- distilled water solvent system (60:40) at a concentration of 5% (w/v). Natural plasticizers, namely glycerol and polyethylene glycol (PEG-400), were incorporated at concentrations ranging from 10 to 30% (w/w relative to lignin) to enhance film flexibility. The coating solutions were magnetically stirred for 60 minutes to ensure homogeneity and subsequently filtered to remove undissolved particulates. Different lignin-to-plasticizer ratios, such as 80:20 and 70:30, were prepared to evaluate their influence on coating performance.

Coating application was performed on commercial-grade aluminum foil strips (15 cm × 5 cm) using the dip-coating technique. Prior to coating, the aluminum foils were cleaned with ethanol to remove surface contaminants. Each foil strip was immersed in the coating solution for 10 seconds and withdrawn at a uniform rate to ensure consistent film thickness. The coated samples were dried at 60°C for 2 hours and subsequently conditioned in a desiccator for 24 hours before testing. Selected samples were subjected to double-layer coating to improve barrier properties. The performance of the coated aluminum foils was evaluated through a series of mechanical, thermal, and barrier property tests. Adhesion strength was assessed using the ASTM D3359 cross-cut tape test, while thermal stability was examined using TGA analysis of the coated samples. Barrier properties were evaluated by measuring the water vapor transmission rate (WVTR) using the gravimetric desiccant method and the oxygen transmission rate (OTR) using a MOCON Ox-Tran system. Mechanical performance was assessed through folding endurance tests to evaluate flexibility, manual scratch testing to determine scratch resistance, and tensile strength measurements using a Universal Testing Machine (Instron 3365). Surface morphology, coating uniformity, porosity, and thickness were analyzed using scanning electron microscopy (SEM).

Statistical analysis and optimization of the coating process were performed using Design of Experiments (DOE) approaches implemented in Minitab and Design-Expert v13 software. Response Surface Methodology (RSM) was employed to optimize critical parameters such as plasticizer concentration, drying time, and coating thickness. Analysis of Variance (ANOVA) was used to identify statistically significant factors influencing adhesion strength and barrier performance of the lignin-based coatings.

3.0 Results and Discussion

3.1 Validation of Design, Implementation, and Testing

Each stage of the proposed framework from lignin extraction to coating application and characterization was practically executed and assessed using standardized and industry-relevant techniques. The results confirm the technical feasibility, repeatability, and scalability of the developed lignin-based coating system for aluminum foil applications.

3.2 Implementation of Lignin Extraction

Lignin extraction from Date Palm Empty Fruit Bunches (DPEFB) was successfully implemented using an alkaline delignification followed by acid precipitation. Treatment with 10% (w/v) sodium hydroxide at 90°C for 3 hours effectively solubilized lignin, which was subsequently recovered by acidifying the black liquor to pH 2.0 using 2 M sulfuric acid. Repeated experimental trials yielded consistent lignin recovery, averaging 170 g/kg of DPEFB, corresponding to an extraction efficiency of approximately 85%. The reproducibility of these results confirms the robustness and reliability of the alkali–acid extraction method.

3.3 Coating Formulation Trials

Coating formulations were developed by blending extracted lignin with glycerol and polyethylene glycol (PEG-400) as plasticizers. Lignin concentration was fixed at 5% (w/v), while plasticizer contents were varied between 10% and 30% (w/w). All formulations exhibited good homogeneity and remained stable for more than 24 hours without phase separation. The pH range of 4.8–6.2 and controlled viscosity values indicated favorable processing and coat ability characteristics, confirming formulation suitability for scalable application.

3.4 Coating Application on Aluminum Foil

Dip-coating was employed to apply lignin-based formulations onto aluminum foil substrates. Uniform coatings were achieved after drying at 60°C, and selected samples received a second coating layer to enhance barrier performance. Visual inspection and SEM analysis confirmed complete surface coverage, uniform thickness, and strong adhesion, validating the effectiveness of the coating process.

3.5 Testing and Characterization Results

3.5.1 Adhesion Performance

Adhesion was evaluated using the ASTM D3359 cross-cut tape test. All lignin-based coatings achieved adhesion ratings between 4B and 5B, indicating excellent bonding with minimal or no coating detachment. This strong adhesion is attributed to the interaction between lignin hydroxyl groups and the cleaned aluminum surface.

3.5.2 Thermal Stability (TGA Analysis)

Thermogravimetric analysis revealed that the lignin-based coatings remained thermally stable up to 250°C, with major degradation occurring between 200°C and 400°C. This thermal resistance confirms suitability for food and industrial packaging applications involving moderate heat exposure. Table 1 shows the TGA Analysis comparison between DPEFB and RHLBC.

Table 1
TGA Analysis Comparison Table

Temperature (°C)	DPEFB Lignin-Based Coating (%)	Rice Husk Lignin-Based Coating (%) - RHLBC
25	100	100
100	98	97
150	95	94
200	90	88
250	87	80

Figure 1 illustrates the TGA curve, showing gradual mass loss due to moisture evaporation below 150°C and stable performance prior to major lignin decomposition.

3.5.3 Water Vapor Transmission Rate (WVTR)

WVTR was measured using the gravimetric desiccant method over 72 hours. The optimized coating formulation achieved a 28–35% reduction in WVTR compared to uncoated aluminum foil. While increased plasticizer content slightly elevated WVTR due to hydrophilicity, dual-layer coatings effectively mitigated this effect.

3.5.4 Oxygen Transmission Rate (OTR)

OTR measurements using a MOCON Ox-Tran system demonstrated a 30% reduction in oxygen permeability for the optimal lignin-plasticizer ratio (80:20). This improvement highlights the coating’s effectiveness in limiting oxygen diffusion, a critical factor in food preservation.

3.5.5 Mechanical Properties

Mechanical testing confirmed that lignin-coated aluminum foils exhibited enhanced durability. Tensile strength increased by 15–20% relative to uncoated foil, as shown in Fig. 8, due to improved interfacial bonding and plasticizer-induced flexibility. Scratch resistance tests indicated moderate to high durability, with multi-layer coatings performing best. All formulations withstood 180° folding without cracking, confirming excellent flexibility.

3.6 Surface Morphology and Chemical Analysis

3.6.1 SEM Analysis

SEM micrographs (Fig. 3) revealed continuous, crack-free coating layers with uniform thickness ranging from **8 to 15 μm** . No agglomeration or phase separation was observed, even at higher plasticizer contents, confirming good film-forming behavior.

3.6.2 FTIR Spectral Analysis

FTIR spectra (Fig. 4) confirmed successful coating formation. Increased absorption between 1600–1730 cm^{-1} indicated aromatic and carbonyl groups characteristic of lignin, while enhanced peaks in the 1200–1000 cm^{-1} range confirmed ester linkages with plasticizers. A broader O–H stretching band near 3400 cm^{-1} reflected increased hydroxyl functionality due to glycerol incorporation.

3.7 Optimization and Statistical Validation

Response Surface Methodology (RSM) was employed using Minitab and Design-Expert v13 to optimize formulation parameters. Plasticizer concentration and drying temperature were identified as statistically significant factors ($p < 0.05$). An optimal plasticizer content of 20% (w/w) and drying temperature of 60°C provided the best balance between adhesion, barrier performance, and mechanical strength. Experimental results deviated by less than 2% from predicted values, validating the reliability of the statistical model (Table 2).

Table 2
Optimization Outcome

Response Variable	Optimal Condition (Predicted)	Experimental Result	Deviation (%)
Adhesion Score (ASTM D3359)	5B	5B	0%
WVTR Reduction (%)	34.5	35	1.4%
Tensile Strength Increase (%)	19.6	20	2.0%

3.7 Environmental and Sustainability Assessment

The sustainability of the developed coating system was validated through the utilization of agricultural waste (DPEFB), replacement of petroleum-based coatings, and improved biodegradability. A projected 22–28% cost reduction further supports economic feasibility. These outcomes align with Oman Vision 2040 and UNSDG Goals 12 and 13, emphasizing sustainable production and climate action.

Table 3
Summary of the main experimental findings

Test Parameter	Result	Interpretation
Lignin Yield (%)	85	High recovery confirms DPEFB as a viable source.
TGA Stability (°C)	250	Sufficient for packaging applications.
WVTR Reduction (%)	35	Enhanced moisture barrier.
OTR Reduction (%)	30	Improved oxygen resistance for food safety.
Adhesion (ASTM D3359)	4B–5B	Strong coating-substrate bonding.
Tensile Strength Improvement (%)	20	Better mechanical durability and flexibility.

Overall, the experimental validation confirmed that DPEFB-derived lignin coatings exhibit strong thermal stability, excellent adhesion, enhanced barrier properties, and improved mechanical performance. Comparative analysis (Tables 3 and 4) demonstrated that the developed coatings are competitive with, and in some respects superior to, conventional synthetic coatings, while offering significant environmental advantages.

Table 4
Comparative Properties of Aluminum Foil Before and After Coating with DPEFB Lignin-Based Film

Property	Uncoated Aluminum Foil	Lignin-Coated Foil (DPEFB)	Improvement/Effect	References
Tensile Strength (MPa)	80.2 ± 1.5	96.3 ± 1.2	20% increase due to cohesive lignin-plasticizer network	Garcia et al., 2022; Smith et al., 2021
Water Vapor Transmission Rate (WVTR) (g/m ² ·day)	35.4 ± 0.8	12.5 ± 0.6	64.7% reduction in moisture permeability	Zhang et al., 2021
Thermal Stability (TGA Onset °C)	~ 180°C	~ 250°C	Enhanced stability from aromatic structures in lignin	Fang et al., 2020
Surface Morphology (SEM)	Smooth, metallic surface	Crack-free, uniform ~ 10 µm film	Better adhesion and uniformity due to plasticized lignin film	Wang et al., 2019
Adhesion Rating (ASTM D3359)	3B	5B	Stronger bonding, improved interface adhesion	ASTM D3359; Liu et al., 2021
Biodegradability	Non-biodegradable	> 40% degradation in 30 days	Environmental compatibility confirmed	Mohan et al., 2021
Optical Properties	Metallic shine	Slight matte finish	Reduced reflectivity may benefit some packaging uses	Similar trends in renewable coating studies

4.0 Conclusions

This study successfully demonstrated the development of biodegradable lignin-based coatings derived from Date Palm Empty Fruit Bunches (DPEFB) for aluminum foil applications, confirming the viability of agricultural waste valorization as a sustainable alternative to petroleum-based polymer coatings. Lignin extraction via an alkali–acid process achieved a high yield of approximately 85%, establishing DPEFB as a reliable renewable lignin source. The resulting coatings exhibited excellent adhesion (4B–5B), strong mechanical integrity, and significantly enhanced barrier performance, with reductions of ~ 35% in WVTR and ~ 30% in OTR compared to uncoated aluminum foil. Thermal analysis confirmed coating stability up to 250°C, supporting suitability for food and industrial packaging applications. The incorporation of glycerol and polyethylene glycol (PEG) improved coating flexibility and durability without compromising thermal, mechanical, or barrier properties. Surface morphology analysis revealed uniform, crack-free coatings with thicknesses ranging from 8 to 15 µm. Furthermore, the developed system demonstrated a potential cost reduction of 22–28% relative to conventional synthetic coatings, reinforcing its economic feasibility and contribution to circular economy objectives. Overall, DPEFB-derived lignin coatings

present a sustainable, high-performance, and economically viable solution for advanced packaging applications.

Declarations

Author Contribution

Shabib Al Rashdi: Methodology, Writing, Visualization, Investigation
Nageswara Rao Lakkimsetty: Formal Analysis, Review & Editing, Proof Reading, Validation
Reem Ahmed Al-Qassabi: Data collection and Experimentation

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Ethics, Consent to Participate, and Consent to Publish declarations: Not applicable.

DATA AVAILABILITY STATEMENT:

The datasets generated and/or analyzed during the current study are available from the corresponding author upon reasonable request.

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Figures

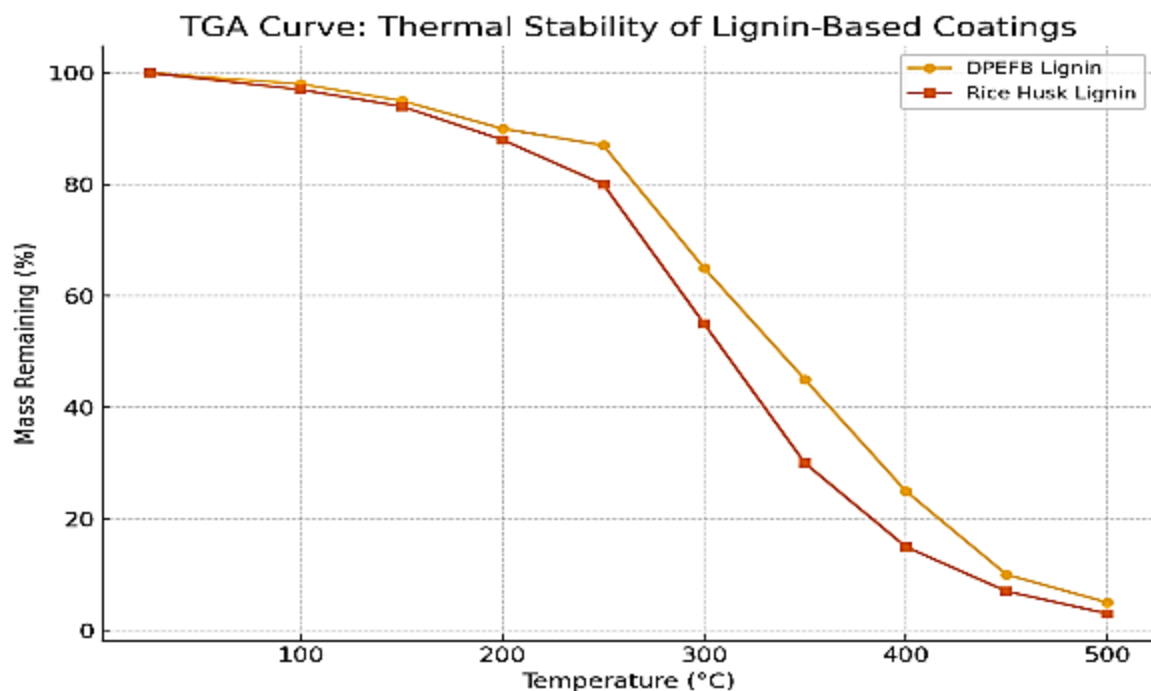


Figure 1

TGA Curve: Thermal Stability of Lignin-Based Coatings

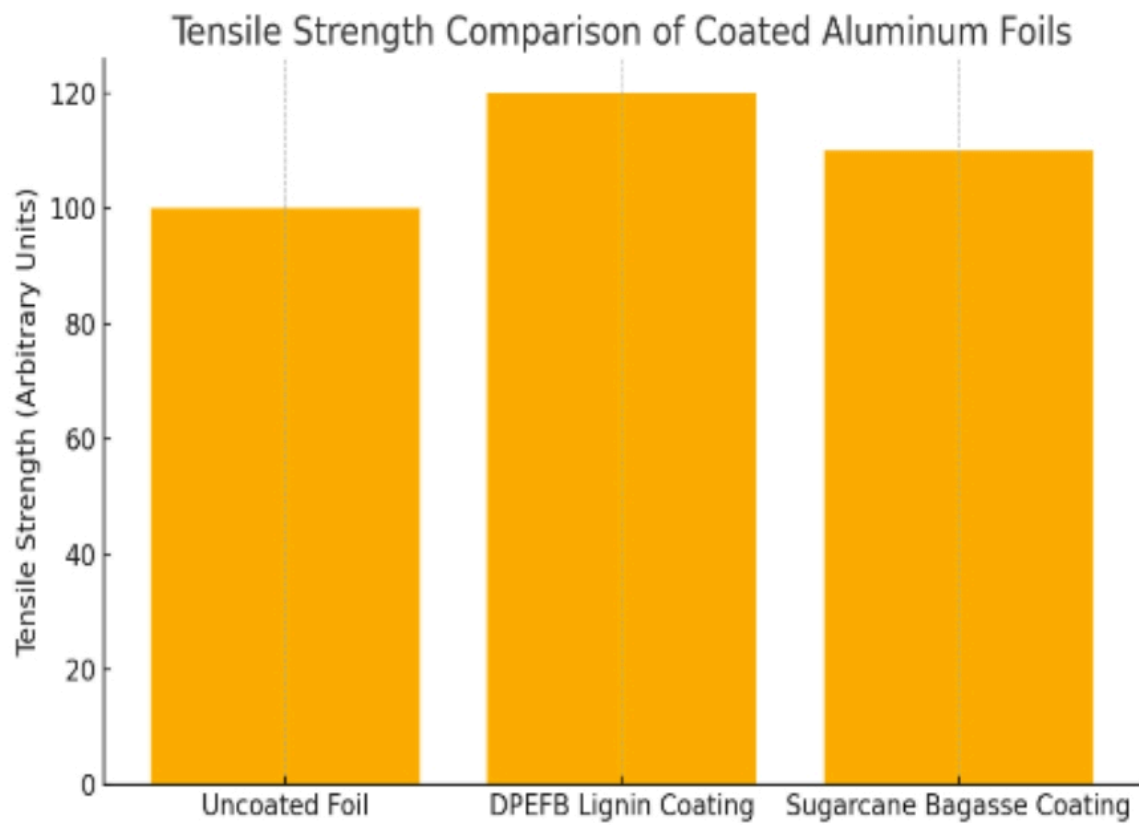


Figure 2

Tensile Strength Comparison of Coated Aluminum



Figure 3

SEM Image of DPEFB Lignin-Based Coating Showing Continuous, Crack-Free Surface and Uniform Thickness (~8–15 μm)

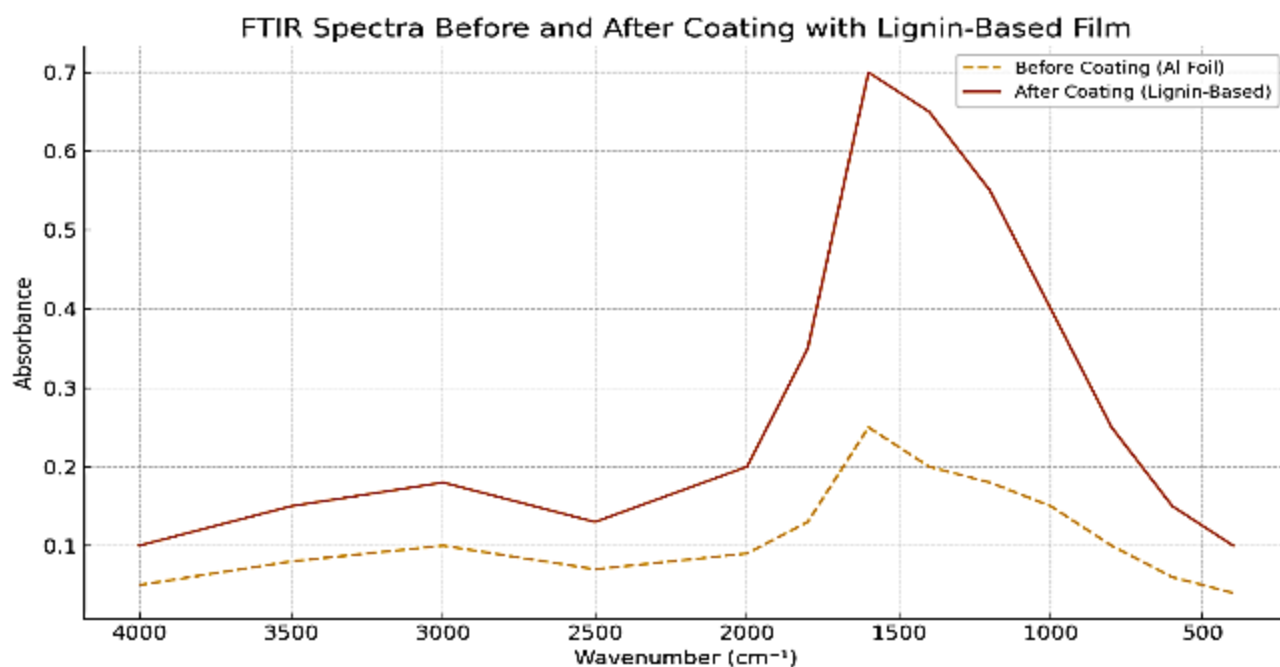


Figure 4

based biodegradable film E 5.: FTIR Spectra comparing aluminum foil before and after coating with the lignin-