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SYNTHESIS, AND APPLICATION OF URETHANE AND BORATE-MODIFIED CASHEW NUT SHELL LIQUID (CNSL) DERIVATIVES IN FORMULATION OF SUSTAINABLE SURFACE COATINGS USING (CNSL + PROPANOL) AS SOLVENT.

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ABSTRACT

This research focuses on the synthesis, and application of cashew nut shell liquid (CNSL) derivatives—specifically urethane and borate-modified compounds—for the formulation of sustainable surface coatings using (CNSL + Propanol) as solvent. CNSL, a renewable, phenolic-rich resource, was chemically modified via urethanation and boration to enhance its performance characteristics for eco-friendly oil-based paints. Fourier Transform Infrared Spectroscopy (FTIR) confirmed successful derivatization, revealing characteristic functional groups such as –NHCOO– (urethane linkage) and B–O stretching vibrations, indicative of borate incorporation. Physicochemical analysis showed that the urethane derivative had lower density (0.96 g/ml), viscosity (2.6 Pa.s), and a refractive index of 1.48, along with a shorter drying time of 240 hours, making it suitable for rapid-curing coatings with minimal environmental impact. Conversely, the borate derivative exhibited higher density (1.3 g/ml), viscosity (3.2 Pa.s), and refractive index (1.67), with a prolonged drying time exceeding 528 hours, suggesting its potential for high-gloss, UV-resistant finishes. The study demonstrates that chemical modification of CNSL enhances its application potential in sustainable coatings, with urethane-CNSL derivatives being more suitable for fast-drying, user-friendly formulations, and borate derivatives offering functional advantages where long-term durability is essential. This work contributes to the development of bio-based alternatives in the coatings industry, aligning with green chemistry and circular economy goals.

Keywords: CNSL, borate, urethane, surface coatings, drying time.

INTRODUCTION

The rapid depletion of petroleum being a finite resource and the primary source of fundamental organic compounds has accelerated the shift towards renewable alternatives, such as cardanol, a phenolic derivative extracted from cashew nut shell liquid (CNSL), (Eneh, 2011). As an economical and eco-friendly resource, cardanol shows immense potential in diverse applications, including surface coatings, paints and varnishes (Telascrêa et al., 2014). Reserch is driven into the development of bio-based surface coatings owing to the increasing demands for sustainable and eco-friendly materials. Cashew nut shell liquid (CNSL), a renewable and non-edible biomass, has gained attention as a potential feedstock for the production of bio-based materials (Menon, et al., 2017). CNSL is a rich source

of phenolic compounds, particularly anacardic acid, which can be used to synthesis various polymer resins (Kumar et al., 2016). The use of CNSL-based resins in surface coatings has been explored in several studies, demonstrating their potentials as sustainable alternatives to petroleum base coatings (Siddique et al., 2019).

Cashew is the common name of a tropical evergreen tree *Anacardium occidentale*, in the family Anacardiaceae. It is native to South America and is the source of the cashew nut and the cashew apple, an accessory fruit (Cashew, 2025). The tree can grow as tall as 14 meters (46 feet), but the dwarf cultivars, growing up to 6 m (20 ft), prove more profitable, with earlier maturity and greater yields (Wikipedia, n.d.). The cashew nut is edible and is eaten on its own as a snack, used in recipes, or processed into cashew cheese or cashew butter. The nut is often simply called a 'cashew'. The cashew apple is a light reddish to yellow fruit, whose pulp and juice can be processed into a sweet, astringent fruit drink or fermented and distilled into liquor. In 2023, 3.9 million tons of cashew nuts were harvested globally, led by the Ivory Coast and India (Wikipedia, n.d.). In addition to the nut and fruit, the shell yields derivatives used in lubricants, waterproofing, and paints (Jostock, 2008).
The cashew tree (*Anacardium occidentale L*.) believed to have first originated in Brazil, (Nair, 2021) is an evergreen tropical tree with a commercially significant nut. Despite the Cashew nutshell being considered an agro waste, (Mele, 2010) the liquid within the honeycomb shell structure has drawn significant attention due to essential chemical compounds present (Voirin, 2014). These compounds includes: anacardic acid, cardanol, cardol, and methyl cardol (Mubofu, 2018). Extraction methods, influenced by the temperature (Mubofu, 2018 and the geographical distributions affect the percentage composition of these components. Common extraction methods such as (1) solvent, (2) thermal, and (3) mechanical extractions result in different compositions. Low temperatures extractions (below 100 ^{0}C) yield higher anacardic acid levels (60-65%), while high temperatures lead to elevate cardanol levels (60-65%) due to the decarboxylation of anacardic acid in CNSL into cardanol (Garkal, 2014). Raw CNSL with a high anacardic acid percentage is acidic and corrosive (Mahanwar et al., 1996). The applications of raw CNSL are limited due to its instability at elevated temperatures. Raw CNSL is subjected to thermal treatments for converting the anacardic acid into cardanol to produce technical CNSL (t-CNSL). According to Manhanwar *et al*.(1996), the optimal temperature for this reaction is 140 ^{0}C, at which the decarboxylation reaction prevails. In contrast, higher temperatures lead to thermal polymerization. Cardanol, the major component in t-CNSL serves as a crucial green starting material in organic syntheses offering versatility for various transformations through hydrogenation, halogenation, epoxidation, and cross-linking owing to the functionality of its hydrocarbon chain and the aromatic ring (Voirin, 2014). Though having worldwide availability, there are some challenges in CNSL. For example, the cashew tree is usually grown from seeds placed directly in the field. Seed nuts should be thoroughly dry, clean, and free from insect or fungal attack. Cashew seeds should be sown or planted during the rainy season. Once the rainy season is over, seeds should be stored properly until the next rainy season before they are planted in the field. If not, they may lose their germination capacity. As plantation of cashew seeds is seasonal, harvesting is another challenge in ensuring the continuous production of cashew trees throughout the year. There are a number of other challenges like land preparation, spacing, fertilizer use, entomological/pathological problems, etc., that need to be taken care of in order to have maximum production. Also, during processing, CNSL is difficult to remove from the shell with high yields due to the hard outer shell, the intricate honeycombed features of the pericarp and the thermally

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sensitive nature of the CNSL. Methods for removing CNSL from the shell include roasting, hot-oil bath, steam processing at 27^0C, quick roasting at 30^0C, cold methods, and the solvent extraction method (Rector, 1936). Which is commercially used, these methods have some constraints like low yield, polymerization of CNSL at processing temperature employed, long extraction times (22–366h), large amounts of solvent, harsh mechanical pretreatment, etc. (Tyman et al., 1989).

Besides having processing constraints, CNSL and their derivatives possess a number of technical benefits over other renewable oils. Unlike oils, the extracted CNSL contains a number of useful phenolic derivatives with meta-substituted long chain saturated/unsaturated hydrocarbons which makes them suitable for a number of polymerization reactions through addition as well as condensation mechanisms. Also, the combination of aromatic ring and long chain hydrocarbon helps to maintain the good balance between flexibility and hardness properties of the coatings (Sanger *et al*, 2011). On this basis they are used in a number of industrial applications, including brake linings materials, laminating resins, adhesives, ion-exchange resins, paint and coating resins, foundry chemicals, lacquers, fine chemicals, hybrid materials, water proofing agents, surface active agents, synthetic rubber, wax compounding, etc.(Velmurugan and Loganathan, 2012)) . Due to reactive phenolic hydroxyl, one-step synthesis of epoxy resin with 100% conversion can be possible, unlike epoxies derived from oil. Also, CNSL-based epoxy synthesis does not involve the use of hazardous chemicals like peroxides which are used in epoxidation of oils. Thus there are no handling or health-related issues in CNSL-based resin synthesis. Also, chemically unmodified CNSL has been reported to reduce the corrosion rate of carbon steel by over 90% due to phenolic hydroxyl which gets adsorbed on metal surface (Mutasingwa, 2004). In the 21st century, cashew cultivation increased in several African countries to meet the manufacturing demands for Cashew milk, a plant milk alternative to diary milk. (Osborn, 2015). In Mozambique, *bolo polana* is a cake prepared using powdered cashews and mashed potatoes as the main ingredients. This desert is common to South African (Philippa, 2009).

The objective of this research is to synthesize and apply two derivatives of cashew nut shell liquid (CNSL), for its potential use in the formulation of sustainable surface coatings. The study aims to develop environmentally friendly coatings materials by modifying CNSL through chemical processes to enhance its performance properties such as adhesion, viscocity, refractive index and drying time. By replacing conventional petroleum-based binders with bio-based alternatives, this research seeks to contribute to green chemistry initiatives and promote the use of renewable resources in industrial coatings applications.

MATERIALS AND METHOD

A second wash with distilled water ensured the removal of any potential surface contaminants. The cleaned shells were sun dried for three days to eliminate moisture. The dried shells were then crushed using a mechanical grinder and sieved to obtain uniform particle sizes (approx. 1–2 mm) for efficient extraction.

Sample collection for this study was conducted within the Elu-Ohafia area of Abia State, Nigeria, as shown in the Figure 1. The sampling locations were strategically selected to capture spatial variability across key parts of the community, including areas of high human activity and major transport routes. Three main sample points, represented by red dots on the map, were identified near significant landmarks such as the Ohafia Central Motor Park, Blesspring Hotel, and along Prof. Kalu Ezera Road. These sites were chosen based on their accessibility, proximity to potential sources of environmental contamination, and relevance to the study objectives. The sampling area is

well-defined by the community boundary, which includes major roads like the expressway cutting through Ebem and Ama-Uke, and minor roads linking residential and commercial zones. This spatial coverage ensured representative sampling within the Elu-Ohafia region for subsequent laboratory analysis.

Extraction Procedure

The ground CNS was subjected to Soxhlet extraction using Acetone. Approximately 250g of finely ground shell material was placed in a thimble inside the Soxhlet apparatus. Acetone was used as the primary solvent due to its high polarity and extraction efficiency. The setup was maintained at 60–70°C for 6–8 hours. The resulting dark brown CNSL was collected and concentrated using a rotary evaporator at 50°C to remove the acetone.

The concentrated extract was stored in amber bottles at 4°C for further use.

Synthesis of CNSL + Borate (borate resin)

The resin was synthesized using a modified protocols developed by Akaranta and Aloko, (199) and Keetasombat and Soykeabkaew, (2014). 50ml of CNSL was poured into a 250 ml flat bottom flask. 20 ml of Borate was added into the flask. The reaction proceeded in the presence of NaOH as catalyst and was heated to a temperature100^0C-130^0C on a magnetic bath and was stirred continually for a period of 50 min.

Synthesis of (CNSL + Urethane) Resin

The resin was synthesized using a modified protocols developed by Akaranta and Aloko, (199) and Keetasombat and Soykeabkaew, (2014). 50ml of CNSL was poured into a 250 ml flat bottom flask. 20 ml of urethane was added into the flask. The reaction proceeded in the presence of NaOH as catalyst and was heated to a temperature 100^0C- 130^0C on a magnetic bath and was stirred continually for a period of 50 min.

Synthesis of (CNSL +Propanol) Solvent

Thirdly, 50ml of CNSL was poured into a 250 ml of flat bottom flask. 20 ml of propanol was added into the flask. Synthesis of CNSL + Propanol (as a Solvent or Diluent). This is not a resin-forming reaction but a solubilization or mild esterification (if catalyzed). Propanol is simply mixed with CNSL to reduce viscosity. No reaction occurs; it's just a solvent/diluent system.

Characterization of Synthesized Resins and Solvent

The cashew nut shell liquid (CNSL+ Borate), (CNSL+ Urethane), and (CNSL + Propanol) was subjected to Fourier transform infrared spectroscopic (FTIR) analysis to ascertain the various functional groups using a Bruker Alpha Platinum ATR in the wavelength range of 500–4000 cm^{-1}. The diamond was cleaned with isopropanol and a background scan was taken. The sample was then placed directly on the crystal and the pressure gauge was applied to ensure maximum contact. The sample was then scanned and the spectrum generated.

Formulation of Surface Coatings

Sample A: Surface coatings (oil paint) with synthesis Borates Resin and synthesis Solvent (Propanol).

Prepared borate resin was mixed with the prepared solvent with continuous stirring for about 10mins, TiO_2 (pigment) was added with continuous stirring for homogeneity for about 30 mins. The viscosity of the mixture was adjusted by the addition of more solvent. The coatings was covered and kept for evaluation.

Sample B: Surface coatings (oil paint) with synthesis Urethane Resin and synthesis Solvent (Propanol).

Prepared Urethane resin was mixed with the prepared solvent with continuous stirring for about 10mins, TiO_2 (pigment) was added with continuous stirring for homogeneity for about 30 mins. The viscosity was adjusted with the addition of more solvent. The coatings was covered and kept for evaluation.

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RESULT AND DISCUSSION

Physiochemical parameters of the coatings

Table 1: physiochemical properties and drying time of urethane bio-based and Borate bio-based coatings

S/N	Parameters	Sample A	Sample B
1.	Density (g/ml)	0.96	1.3
2.	Viscosity (Pa.s)	2.6	3.2
3.	Reflective Index	1.48	1.67
4.	Dry Time (hrs)	240	> 528

Where

Sample A: Urethane bio-based surface coatings

Sample B: Borate bio-based surface coatings

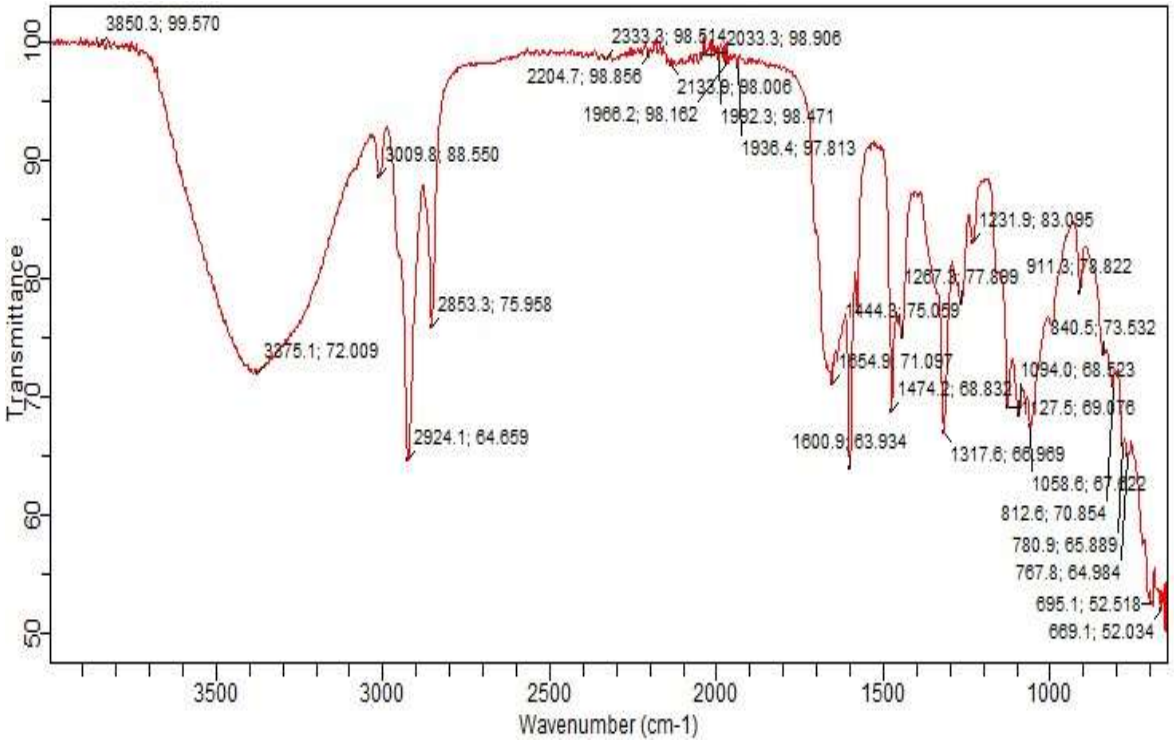

Figure 1: FTIR of Synthesized Borate Resin

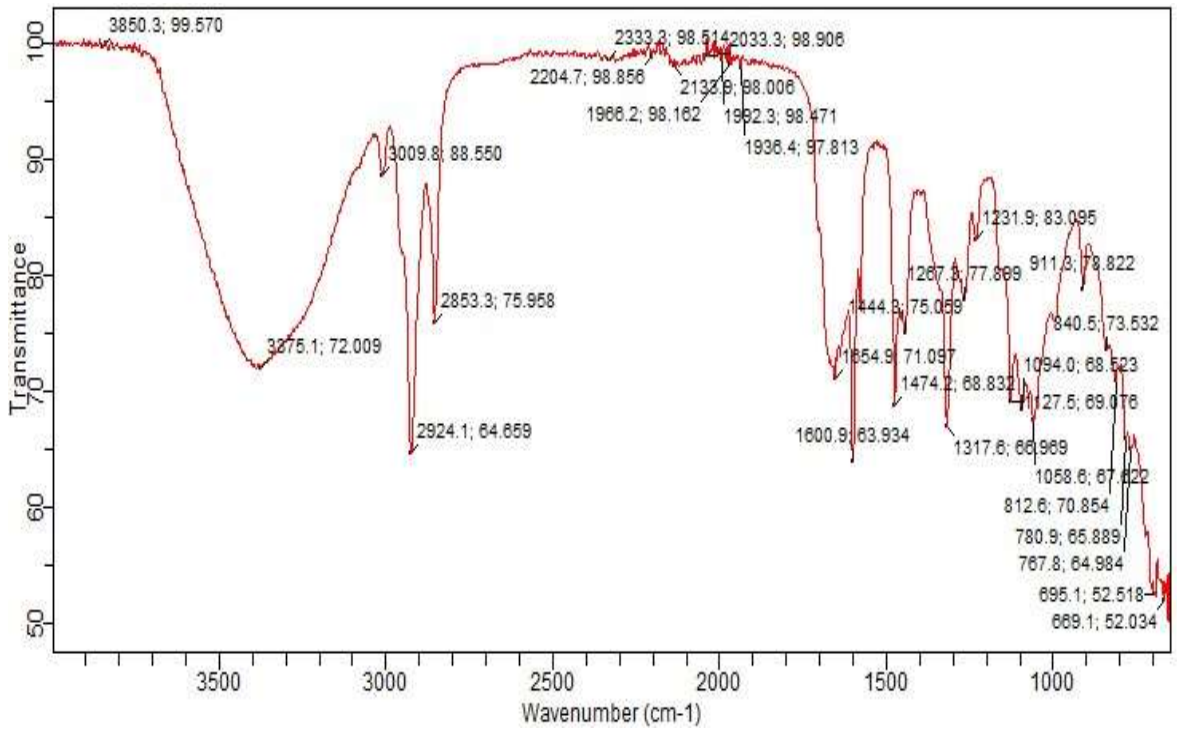

Figure 2: FTIR of Synthesize Urethane Resin

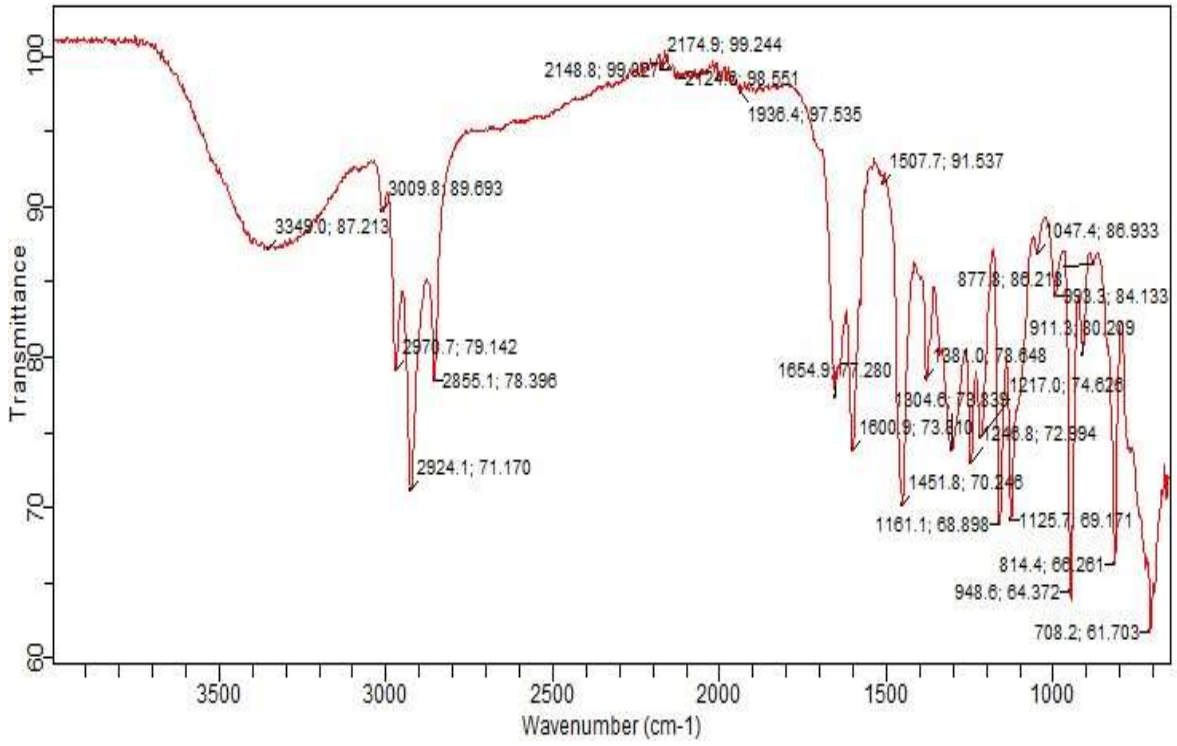

Figure 3: FTIR of Synthesize Propanol Solvent.

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DISCUSSION

Density

Lower density (Sample B) suggests a lighter material, which may be easier to apply and can improve spread ability over surfaces. Higher density (Sample A) could imply better pigment load and durability, but might make the paint thicker and harder to apply uniformly without additives. Sustainability Implication: Lower density may reduce the carbon footprint in transport, while higher density may indicate more solids, which could enhance coating longevity (fewer recoats = less environmental impact).

Viscosity

Higher viscosity (Sample A) means it is thicker and may require more effort to apply, but may offer better sag resistance and coverage per coat. Lower viscosity (Sample B) implies easier application, especially useful for DIY or spray-based applications. Sustainability Implication: Optimal viscosity reduces the need for thinning agents (often VOC-based), making Sample B slightly more user-friendly and sustainable if no solvents are needed.

Refractive Index

Higher refractive index (Sample A) results in more gloss and better light reflection—desirable for aesthetic purposes. (Sample A) lower index may provide a matte or satin finish, suitable for non-reflective coatings. Sustainability Implication: Higher refractive index often correlates with better UV resistance, potentially increasing product lifespan.

Physical parameter

Dry Time (hrs)

Significantly faster drying time for Sample B, a major practical advantage for application and turnaround. Sample A very slow dry time may lead to dust contamination, longer curing schedules, and delayed usability. Sustainability Implication: Longer dry times mean more energy/resource consumption for curing (especially in industrial settings). Sample B is clearly more efficient here.

There were no references throughout this section confirming your claims with respect to drying time

FTIR Synthesize Bio-based Resin (sample A)

The FTIR spectrum of the borate-synthesized compound from fig.2 characteristic absorption bands corresponding to multiple functional groups. Vibrational bands observed at 669 cm^{-1}, 695 cm^{-1}, and 911 cm^{-1} are indicative of out-of-plane C–H bending in alkenes (Pavia et al, 2015). A prominent cluster of peaks between 767 cm^{-1} and 1317 cm^{-1} corresponds to C–X stretching vibrations, typically associated with alkyl halides, though these may also overlap with B–O–C or B–O–B linkages characteristic of borate esters. The absorption bands at 1444 cm^{-1} and 1474 cm^{-1} are attributed to bending vibrations of alkyl and methylene groups. Notable peaks at 1601 cm^{-1} and 1655 cm^{-1} suggest the presence of N–H bending (amines) and amide C=O stretching, respectively (Silverstein et al, 2014). High-intensity bands in the 1936 –1992 cm^{-1} region, though assigned as carbonyls, likely represent overtone or combination bands rather than fundamental vibrations. Signals at 2134 cm^{-1}, 2205 cm^{-1}, and 2333 cm^{-1} fall within the C≡C and C≡N stretching region, indicating possible alkyne or nitrile functionalities (Smith, 2011). The aliphatic C–H stretching vibrations are confirmed by bands at 2853 cm^{-1} and 2924 cm^{-1}. Broad absorptions at 3009 cm^{-1}, 3375 cm^{-1}, and 3850 cm^{-1} are consistent with O–H stretching modes, suggesting the presence of hydroxyl and carboxylic acid groups (Smith, 2011). Collectively, the spectral data confirm the incorporation of alkenes, alkyl halides, amines, carbonyls, alkynes, and hydroxyl-containing moieties in the borate-based compound.

FTIR of Synthesize Bio-based Resin (sample B)

"The FTIR spectral analysis of the synthesized material in Fig. 3 Confirms the presence of urethane linkages, as evidenced by the strong absorption band at 1716.4 cm^{-1} corresponding to the carbonyl (C=O) stretching vibration of the urethane group, and a broad band at 3365.8 cm^{-1} indicative of N–H and O–H stretching, characteristic of hydrogen-bonded urethane or residual hydroxyl groups (spectroscopy online). Additional peaks in the region of 1162.9 cm^{-1} to 1246.8 cm^{-1} correspond to C–O–C stretching vibrations, confirming the presence of ether linkages typically found in polyether or polyester urethane structures. The presence of aromatic rings is supported by absorption bands at 1086.5 $cm^{-1}$and 1606.5 cm^{-1}, while aliphatic and aromatic C–H stretching is observed between 2853.3 cm^{-1} and 3011.7 cm^{-1}(Chem LibreText). Minor absorption bands in the range of 2132.0 cm^{-1} to 2167.4 cm^{-1} suggest the possible presence of acetylenic or residual isocyanate groups, while weak bands around 1627.0 cm^{-1} and 1654.9 cm^{-1} indicate unsaturation, likely due to alkenes or aromatic C=C bonds. (Nature, polym j) Overall, the spectrum strongly supports the successful formation of a urethane-based polymer or compounds.

FTIR of Synthesize Solvent (Propanol).

The FTIR spectrum of the synthesized compound in Fig. 4 reveals prominent absorption bands corresponding to O–H stretching (~3349 cm^{-1}), C–H stretching (2855 cm^{-1}–3009 cm^{-1}), C–O stretching (1047 cm^{-1}–1304 cm^{-1}), and aromatic/aliphatic C=C vibrations (1450 cm^{-1}–1650 cm^{-1}), confirming the presence of alcohol, alkyl, ether, and aromatic functional groups. Notably, the absence of a strong absorption around 1700 cm^{-1}, typically associated with C=O stretching in carboxylic acids, indicates that no carboxylic acid group was formed during the synthesis. These results suggest that the reaction involving propanol led to the successful incorporation of hydroxyl and aromatic components, possibly forming an aromatic alcohol or ether, while eliminating the possibility of carboxylation or esterification.

Comparative Performance in Sustainable Coatings

Sample B appears more sustainable and user-friendly, especially for fast-drying, lightweight, and low-viscosity requirements. Sample A may be preferred where gloss, hardness, or UV resistance are critical, but it's long drying time limits efficiency. In eco-friendly oil paint formulations, urethane-modified oils may offer the best trade-off between performance and environmental impact.

There were no references throughout this section confirming your claims with respect to comparative studies

Implications and Future Directions

The successful synthesis and characterization of CNSL derivatives and surface coatings in this study demonstrate the potential of CNSL as a sustainable feedstock for the production of industrial coatings. Further studies are needed to explore the scalability and commercial viability of this technology.

Limitations

One limitation of this study is the use of a small-scale synthesis and coating formulation process. Further studies are needed to scale up the process and evaluate the coatings' performance under industrial conditions.

CONCLUSION

From the results, it implies that Urethane-CNSL derivatives are more viable for general purpose, sustainable applications, especially where ease of handling and faster drying time is required. Borate-CNSL derivatives though shows long curing time, may be preferred where UV stability, film hardness and durability is required. This work brings to light the potential of CNSL as a renewable resource in green chemistry and coatings

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formulation, bridging the gap between sustainability and performance.

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