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STRUCTURE-PROPERTY ENGINEERING OF LOW-EMISSION STARCH-GELATIN BIO-ADHESIVES VIA POLYLACTIC ACID MODIFICATION

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ABSTRACT

The increasing demand for sustainable, low-emission adhesives has heightened interest in bio-derived polymer systems that can replace petroleum-based formulations without losing performance. In this study, the structure–property relationships of starch- and gelatin-based bio-adhesives were systematically engineered through modifications with polylactic acid (PLA) and polyvinyl acetate (PVAc). Adhesives were formulated using starches extracted from Ipomoea batatas, Manihot esculenta, and Eleusine coracana, combined with hide-derived gelatin, with controlled variations in modifier content. Comprehensive physicochemical characterization was performed, including measurements of viscosity, density, pH, isoelectric point, moisture and ash content, total volatile organic compounds (TVOC), and peel strength on leather substrates. The results show that incremental incorporation of PLA/PVAc (PVA) causes significant changes in rheology, molecular packing, and emission behavior, indicating polymer–polymer interactions and network restructuring within the adhesive matrix. Moderate levels of modifiers resulted in optimized viscosity, improved peel strength, and enhanced formulation stability, while excessive loading disrupted biopolymer entanglement and reduced mechanical performance. Importantly, all bio-adhesive formulations had TVOC levels well below international regulatory limits, confirming their low-emission profile. Among the systems examined, Eleusine coracana starch modified with 20% PVAc achieved peel strength comparable to a commercial adhesive standard. Overall, this work provides a mechanistic framework for designing high-performance, environmentally friendly starch–gelatin bio-adhesives and supports their potential for industrial use as sustainable alternatives to conventional synthetic adhesives.

Keywords: Bio-Based Adhesives, Starch–Gelatin Blends, Polylactic Acid Modification, Structure–Property Relationships, Low-VOC Adhesives, Physicochemical Characterization, Peel Strength, Sustainable Polymer Engineering

INTRODUCTION

Adhesives play a critical role across diverse industrial sectors, including wood composites, packaging, construction, textiles, and biomedical devices. However, conventional petroleum-based adhesives are increasingly scrutinized due to concerns related to volatile organic compound (VOC) emissions, environmental persistence, and reliance on non-renewable feedstocks. These challenges have intensified global research efforts toward the development of sustainable, low-emission, and bio-derived adhesive systems that can meet performance demands while complying with increasingly stringent environmental regulations (Agbogo et al., 2025; García-Badillo et al., 2021)

Starch- and gelatin-based adhesives have emerged as promising alternatives owing to their abundance, renewability, biodegradability, and low toxicity. Native starch adhesives, derived from botanical sources such as cassava, sweet potato, and millet, offer cost advantages and favorable film-forming properties but are often limited by poor water resistance, inadequate mechanical strength, and unstable rheological behavior. Similarly, gelatin-based adhesives exhibit excellent tack and adhesion due to their proteinaceous structure but suffer from moisture sensitivity and limited thermal stability. Consequently, unmodified biopolymer adhesives rarely achieve performance parity with synthetic counterparts, necessitating targeted structural modification and polymer blending strategies (Agbogo et al., 2025a; Amini Moghaddam et al., 2021).

(Dallaev et al., 2025; Linde et al., 2020; Nikhade & Jaiswal, 2025) Polymer–polymer modification has proven to be an effective route for enhancing the functional properties of bio-adhesives. In particular, the incorporation of synthetic or semi-synthetic polymers into starch matrices can significantly improve viscosity control, mechanical integrity, and application performance. Polylactic acid (PLA) and polyvinyl acetate (PVAc) derivatives are especially attractive modifiers due to their compatibility with biopolymers, tunable chemistry, and established industrial use. Previous studies on starch–PLA and starch–PVAc systems have demonstrated that controlled polymer blending can alter hydrogen bonding density, network architecture, and molecular packing, thereby improving adhesive strength and processability (Dallaev et al., 2025; Linde et al., 2020; Nikhade & Jaiswal, 2025)

Despite these advances, a systematic understanding of how polylactic-acid-based modification influences the structure–property relationships of hybrid starch–gelatin adhesive systems remains limited. In particular, the combined effects of PLA modification on viscosity, pH behavior, density, and emission characteristics are rarely examined across multiple starch sources and protein-based matrices under comparable formulation conditions. Such knowledge is essential for rational adhesive design, as physicochemical parameters directly govern application behavior, curing characteristics, storage stability, and environmental compliance. Recent reviews emphasize that source-dependent starch chemistry, polymer compatibility, and additive concentration must be jointly optimized to achieve reproducible, industrially viable bio-adhesives (Khouri et al., 2024; Ranakoti et al., 2022)

In this study, we report a comprehensive investigation into the structure–property engineering of low-emission starch–gelatin bio-adhesives via polylactic acid modification. Adhesives were formulated from *Ipomoea batatas*, *Manihot esculenta*, and *Eleusine coracana* starches, as well as hide-derived gelatin, with systematic variation of polylactic-acid-based modifier content. The resulting formulations were evaluated in terms of viscosity, pH, density, isoelectric behavior, moisture and ash content, and total volatile organic compounds (TVOC). By correlating formulation chemistry with physicochemical performance, this work establishes mechanistic insights into polymer–polymer interactions and identifies optimal modification windows for achieving balanced rheology, stability, and low emissions. The findings contribute to the growing body of sustainable adhesive research and provide practical guidance for translating bio-based adhesive systems toward industrial application.

MATERIALS AND METHODS

Sample Collection

Fresh cow hide (approximately 6.0 kg) was obtained from the Biu abattoir, after which standardized dehairing and degreasing processes were carried out to obtain clean hide material suitable for subsequent processing. Plant-based raw materials comprising *Ipomoea batatas* roots (IBS), *Manihot esculenta* roots (MES), and *Eleusine coracana* seeds (ECS) were sourced directly from a local farm settlement in Biu, Borno State, Nigeria.

Method of Extraction

Isolation of Starch Dextrins from Vegetable Sources

Starch dextrins were isolated from *Ipomoea batatas* and *Manihot esculenta* roots using a wet-milling extraction technique adapted from Akpa (2012) and Agbogo (2023). Fresh root samples were first washed thoroughly with distilled water to remove surface contaminants and soil particles, after which they were manually peeled to eliminate the outer cortex. The peeled roots were then cut into smaller portions and blended with distilled water to produce a uniform slurry. This slurry was filtered through a clean muslin cloth to separate fibrous residues from the starch-rich filtrate. The resulting filtrate was allowed to stand undisturbed to enable gravitational settling of the starch. After complete sedimentation, the supernatant liquid was carefully decanted, and the starch cake was dewatered by gentle pressing in a clean fabric bag. The recovered wet starch was subsequently dried at ambient temperature until a constant weight was achieved, ensuring minimal thermal degradation. The dried material was then milled to obtain fine starch powder suitable for subsequent formulation and characterization of the adhesive systems. The same extraction protocol was applied to *Manihot esculenta* roots under identical processing conditions to ensure consistency and comparability across starch sources. The overall sequence of operations, root preparation, wet milling, filtration, sedimentation, dewatering, drying, and milling, constitutes the standardized workflow employed for starch production in this study.

Isolation of Starch Dextrin from Eleusine coracana

Starch dextrin was isolated from *Eleusine coracana* grains following a modified wet extraction method adapted from Muazu (2007) and Agbogo (2023). Approximately 6 kg of mature *E. coracana* grains were obtained from Biu, Borno State, Nigeria. The grains were manually sorted, thoroughly cleaned, and washed repeatedly with distilled water to remove adhering dust, chaff, and extraneous materials. After cleaning, about 5 kg of the grains were steeped in 200 mL of distilled water and allowed to soak for 24 h at ambient temperature to facilitate grain softening and starch release. The steeped grains were subsequently wet-milled using an electric grinder to obtain a homogeneous slurry. The resulting pulp was diluted with an additional 200 mL of distilled water and filtered through a calico cloth with an effective mesh size of approximately 250 μm to separate fibrous residues from the starch-rich filtrate. The filtrate was allowed to stand to permit starch sedimentation, after which 100 mL of 0.1 M sodium hydroxide solution was added to promote the separation of starch from proteinaceous components and to neutralize residual acidity. Excess alkali was removed by washing the sedimented starch three times with 200 mL portions of distilled water. The supernatant was carefully decanted after each wash, and the starch sediment was collected, spread on clean trays, and air-dried to constant weight. The dried starch aggregates were gently pulverized using a mortar and pestle and subsequently sieved through a 100 μm mesh to obtain fine starch powder suitable for subsequent formulation and characterization.

Gelatin Extraction from Hide Material

Gelatin was extracted from cow hide trimmings and splits following an alkaline pretreatment and hydrolytic extraction procedure adapted from Mittal and Pizzi (2003). Approximately 6 kg of hide trimmings obtained from the abattoir were initially washed thoroughly with tap water to remove adhering blood, dirt, and surface impurities. The cleaned hides were then soaked in an aqueous lime solution prepared from calcium hydroxide and distilled water and maintained for a period of 72 h at ambient temperature. The alkaline soaking treatment facilitated the removal of non-collagenous proteins and other extraneous materials while simultaneously conditioning the collagenous matrix for subsequent gelatin extraction. Following lime treatment, the hides were rinsed repeatedly with distilled water to remove residual alkali prior to gelatin recovery. Gelatin extraction was subsequently carried out by controlled hydrolysis of the pretreated collagen using the same extraction protocol employed for bone-derived gelatin, as described by Mittal and Pizzi (2003).

Formulation of Adhesives from Natural Polymers

Adhesive formulations were prepared using starch- and gelatin-based binders through a controlled thermal and chemical modification process. In a typical formulation, 150 mL of deionized water was transferred into a clean beaker, and 1.0 g of polylactic acid (PVAc) was gradually added under continuous magnetic stirring to ensure complete dispersion. The mixture was heated to 60 °C using a heating mantle and maintained at this temperature for 30 min to obtain a homogeneous polymer solution. This too applied to the polyvinyl acetate solution, after which the two polymers were mixed (Solution A).

In a separate vessel, 10.0 g of starch and 0.5 g of borax were introduced into 100 mL of sodium hydroxide solution and stirred continuously at 60 °C for 30 min to promote starch gelatinization and crosslink activation, yielding a uniform alkaline dispersion (Solution B).

While maintaining the temperature at 60 °C, Solution B was rapidly added to Solution A under continuous stirring. Immediate gelation was observed upon mixing, indicating the formation of a crosslinked polymer network. The reaction mixture was held at 60 °C for an additional 5 min and stirred thoroughly to ensure uniformity and to allow the adhesive to attain its maximum viscosity.

Following completion of the reaction, the beaker was removed from the heating mantle and allowed to cool slightly, after which 2 mL of triethylamine and 0.2 g of citric acid were added sequentially as modifiers to enhance adhesive performance and stability. Adhesive modification was achieved by systematically varying the concentration of polylactic acid+polyvinyl acetate to investigate the effect of esterification on the physicochemical properties of the formulations.

The same formulation protocol was applied to gelatin-based adhesive systems, with starch replaced by gelatin as the primary binder, following the method reported by Li et al. (2012).

Determination of Physicochemical Properties

The series of physicochemical tests, namely, viscosity, moisture content, ash content, pH values, isoelectric point, density, and Total volatile organic compounds (TVOC), was carried out on the formulated adhesives. The same test was also performed on the commercial adhesive used as a standard. The research procedures for each of the parameters were adapted from Agbogo et al., 2023.

Determination of peeling strength

Muva peeling machine (Model 5038) was used to monitor the polymer adhesion of leather-leather substrate (7.5 mm). 180 ° peel tests were employed to analyze the fracture behaviour of the formulated adhesives. The current research examines the behaviour of adhesively bonded joints under various loading rates (500 g to 2000g) by conducting a series of peel tests. In all tests, bonded thin sheets of leather substrates using the formulated adhesives were each subjected to different loadings to determine the peeling rate of the various specimens (Georgiou *et al.*, 2003).

RESULTS AND DISCUSSION

This section presents and critically analyzes the physicochemical and mechanical performance of starch- and gelatin-based adhesive formulations modified with polylactic acid. The results are discussed in relation to formulation composition, polymer–polymer interactions, and network architecture, with particular emphasis on viscosity, pH behavior, density, moisture and ash content, isoelectric point, total volatile organic compound (TVOC) emissions, and peel strength. By correlating these parameters, the discussion elucidates structure–property–performance relationships that govern adhesive spreadability, bonding efficiency, environmental compliance, and end-use suitability. Comparative evaluation across multiple starch sources and gelatin matrices provides mechanistic insight into formulation optimization for sustainable, low-emission bio-adhesive applications.

Physicochemical Analysis

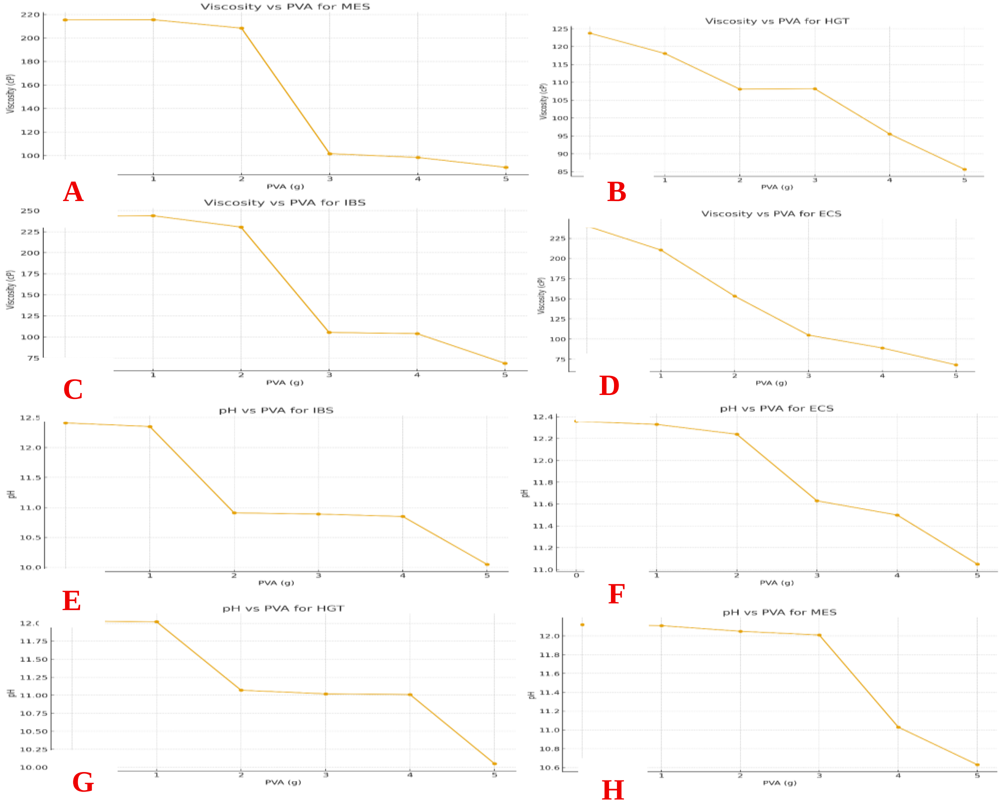

Figure 1: Variation of key Physicochemical Properties of Starch-based and Gelatin-based Adhesives as a Function of Polylactic Acid + polyvinyl Acetate (PVA) Concentration

Variation of Key Physicochemical Properties of Starch-based and Gelatin-based Adhesives as a Function of Polylactic acid+polyvinyl Acetate (PVA) Concentration.

The physicochemical properties of the formulated starch-based and gelatin-based adhesives were systematically influenced by the incremental incorporation of polylactic acid (PVA) (0–5 g), as illustrated in Figure 1. Variations in viscosity, pH, and density collectively reflect changes in polymer–polymer interactions, network integrity, and molecular packing within the adhesive matrix, parameters that are critical for adhesive processability, bonding performance, and industrial applicability. These trends provide a rational framework for tailoring bio-adhesive formulations toward specific end-use requirements, including controlled rheology, chemical stability, and compliance with low-emission regulations. Recent studies have similarly emphasized that polymer blending and starch modification are essential strategies for achieving performance parity between bio-based and petrochemical adhesives. (kumar et al., 2025; Olomo, 2022).

Viscosity exhibited a non-linear dependence on PVA concentration, with marked reductions at higher loadings in several starch-based systems. At low to moderate PVA levels, enhanced hydrogen bonding and improved miscibility promote transient intermolecular interactions between starch or gelatin chains and the added polymer, resulting in relatively stable rheological behaviour. However, excessive PVA incorporation disrupts the native starch/gelatin gel network through plasticization and partial phase dilution, leading to reduced polymer entanglement density and lower viscosity. This behaviour is consistent with reported trends in starch polyester and starch synthetic polymer adhesive systems, where viscosity initially stabilizes or increases before declining at higher modifier contents. Additionally, source-dependent variations in starch structure, particularly differences in amylose/amylopectin ratio and granule morphology among *Ipomoea batatas*, *Eleusine coracana*, and *Manihot esculenta*, further modulate the rheological response. Viscosity is a critical parameter governing adhesive mobility, spreadability, and substrate penetration (Qi & Sun, 2011; Watcharakitti et al., 2022). Measurements conducted after 24 h at room temperature revealed that formulations without PVA modifier exhibited excessively high viscosity, limiting practical applicability. Incorporation of a 20% modifier resulted in a substantial viscosity reduction, enabling uniform spreading and effective substrate wetting. However, PVA contents above 30% produced overly fluid systems with inadequate penetration and bonding efficiency in Figure 1(A-D). This behaviour aligns with prior studies emphasizing the need for balanced viscosity in adhesive applications (Wang et al., 2017). The viscosities of the formulated adhesives were comparable to commercial cyanoacrylate adhesives (28–292 cP) and significantly lower than those reported for polyketone-based wood adhesives (1000–175,000 cP) (Cui et al., 2017; Robertson et al., 2021) The observed viscosity reduction with increasing PVA content is primarily attributed to decreased total solids concentration rather than temperature effects, as supported by earlier studies (Barash et al., 2010; Yamada et al., 2000; Yu et al., 2009) Kinematic viscosity trends further corroborate these findings (Figure 1D), indicating competitive bond strength for finger millet-based adhesives, consistent with the assertion that finger millet starch exhibits strong functional synergy with tapioca starch (Balasubramanian, 2011). These findings indicate the existence of an optimal PVA window for each formulation, within which workability, spreading, and bond strength are balanced for industrial application.

The pH of the adhesive systems in Figure 1(E-H) showed a gradual decline with increasing PVA content, indicating modification of the chemical microenvironment within the adhesive matrix. This trend is attributable to ester functionalities and residual acidic end groups associated with lactic-acid-derived or partially hydrolyzed polymer additives, which shift the acid-base equilibrium of the system. Differences in buffering capacity between starch- and gelatin-based matrices further influence this response; gelatin, as a protein-based polymer, exhibits pH behaviour governed by amino acid composition and crosslink density, whereas starch lacks intrinsic buffering capacity. Consequently, pH changes are more pronounced in starch-dominated systems. These variations are significant because pH influences polymerization kinetics, bond formation, microbial stability, and substrate compatibility. The accompanying isoelectric point (PI) measurements indicate that most formulations operate near charge-neutral conditions, corresponding to minimized viscosity and favourable flow behavior during application. The pH of the formulated adhesives ranged from 10 to 12, exhibiting only marginal variation (Figure 1). This consistency is attributed to the uniform use of sodium hydroxide concentration and distilled water volume across all formulations. However, a gradual decline in pH with increasing PVA content was observed, consistent with reports that lower pH adhesive systems may adversely affect polymerization and bond strength. Figure 1 further indicates that formulations with lower pH values exhibited reduced bond strength. Isoelectric point (PI) analysis revealed that the PI values were close to the initial pH before ion exchange, suggesting that the adhesives operate near their minimum viscosity state, which is favourable for application performance (Lim et al., 2015; Schuurmans Stekhoven et al., 2008). At the PI, adhesive molecules possess no net charge, minimizing solubility and stabilizing flow behaviour.

Density measurements in Figure 2B revealed a modest but consistent increase with increasing PVA concentration across most formulations, reflecting enhanced molecular packing and increased effective solids content due to copolymerization between natural biopolymers and the synthetic modifier. Because all samples were synthesized under identical thermal conditions, density variations primarily arise from compositional rather than processing effects. Gelatin-based formulations exhibited comparatively lower densities, consistent with the greater chain flexibility and distinct network architecture of protein matrices relative to polysaccharides. Density is a practical indicator of formulation compactness, coat weight, and adhesive film thickness, and the observed trends suggest that controlled PVA addition can be used to fine-tune application coverage and material efficiency. Adhesive density, defined as mass per unit volume, is governed by formulation composition and processing temperature. Elevated production temperatures can simultaneously induce adhesive swelling (increasing volume) and moisture loss (reducing mass), resulting in competing effects on density (Sha & Adachi, 2015; Tokunaga et al., 2024). In this study, minimal density variations were observed across formulations (Figure 1B), largely because all samples were synthesized at a constant temperature of 60 °C. A consistent increase in density was observed with increasing PVA concentration, reflecting the copolymerization between the natural biopolymers and the synthetic polymer, which enhanced molecular packing and solids content.

Ash content, which represents the total inorganic fraction of an adhesive, ranged from 3.30 to 4.99% across the formulated samples (Figure 3), exceeding the 0.5% limit reported for unmodified starch adhesives. Elevated ash levels are primarily attributed to phosphate groups inherent to starch granules and added inorganic fillers (Ma et al., 2023). These inorganic components are commonly incorporated to enhance density, smoothness, and adhesive interactions with the substrate. It has been demonstrated that inorganic fillers enhance intermolecular bonding and peel strength by strengthening adhesive-substrate adhesion. Notably, the commercial adhesive exhibited a higher ash content (>5%) than all formulated samples, with the highest value among the bio-adhesives (4.99%) recorded for the *Eleusine coracana* starch formulation containing 40% PVA, indicating that the inorganic content of the formulated adhesives remains within industrially acceptable limits.

Volatile organic compounds (VOCs) are regulated due to their environmental and health impacts. Regulatory limits introduced by agencies in the United States, Canada, the European Union, and Hong Kong typically range from 50 to 250 g L^{-1} for adhesives and sealants. All formulations in this study exhibited TVOC values between 4.06 and 19.60 g L^{-1} (Figure 3), well below established regulatory thresholds. These results confirm that the starch- and gelatin-based adhesives developed in this work are environmentally benign and compliant with international VOC regulations, supporting their suitability for sustainable adhesive applications.

Taken together, the physicochemical trends observed in Figure 3 confirm that adhesive performance is governed by the interplay between polymer compatibility, molecular architecture, and additive concentration. Moderate PVA incorporation enhances processability and structural cohesion, whereas excessive loading compromises rheological stability and network integrity. Importantly, the relatively low TVOC levels measured across all formulations align with current regulatory requirements for environmentally sustainable adhesives, reinforcing the viability of these starch- and gelatin-based systems as low-emission alternatives to conventional products. Overall, this study establishes a clear structure-property–performance relationship that can guide the rational design and optimization of bio-based adhesive formulations for industrial use.

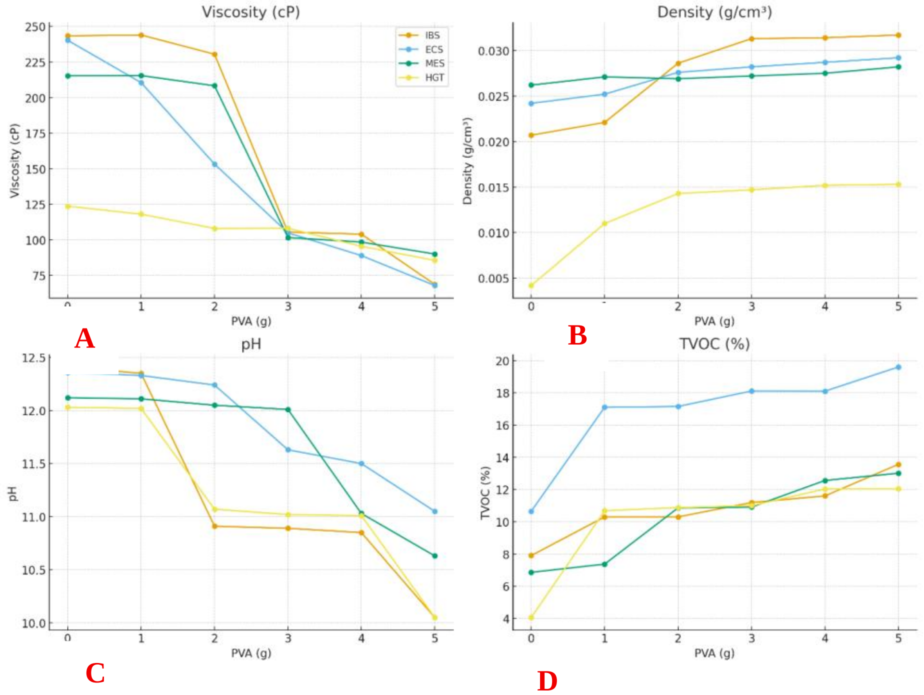

Figure 2: Influence of polylactic Acid (PVA) Concentration on Key Physicochemical Properties of Starch-based and Gelatin-based bio-adhesives. Panels (A–D) show the Effect of Increasing PVA Loading (0–5 g) on Viscosity, Density, pH, and Total Volatile Organic Compounds (TVOC), Respectively. Each Coloured Line Represents one Adhesive System: Orange = Ipomoea Batatas Starch (IBS), Blue = Eleusine Coracana Starch (ECS), Green = Manihot Esculenta Starch (MES), and Yellow = Hide Gelatine (HGT)

Building on the mechanistic trends identified in Figure 1, Figure 2 extends the analysis to formulation stability and environmental performance by incorporating TVOC behaviour alongside rheological and physicochemical parameters. The pronounced viscosity reduction at higher PVA loadings reinforces the conclusion that excessive modifier disrupts the biopolymer network, while density increases confirm progressive matrix compaction. Importantly, the inclusion of TVOC data reveals starch-source-dependent volatilization pathways, with ECS exhibiting the highest emissions, highlighting the role of botanical origin, granule morphology, and additive compatibility in governing volatile release. Such variability aligns with prior reports demonstrating that starch chemistry and processing history significantly influence emission profiles in bio-adhesive systems. The combined trends underscore a critical formulation trade-off: while PVA improves processability and structural compactness, excessive loading may compromise rheological stability and emission performance. Consequently, Figure 2 provides an applied perspective that complements Figure 1, emphasizing the need for optimized additive windows to achieve industrially viable, low-emission bio-adhesives.

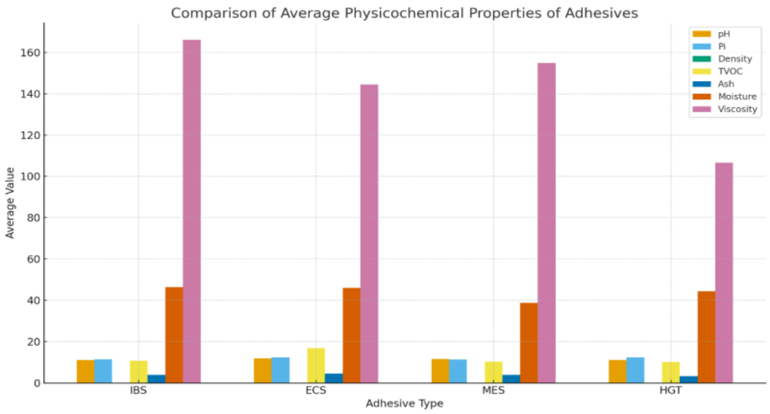

Figure 3: Comparison Of Average Physicochemical Properties Of Starch- And Gelatin-Based Adhesive Formulations

Qualitative Comparative Statistical Analysis of Physicochemical Performance of Bio-Based and Commercial Adhesives

Figure 3 presents a comparative bar chart of the average physicochemical properties of the formulated starch- and gelatin-based adhesives alongside a commercial benchmark adhesive. The parameters evaluated: pH, isoelectric point (Pi), density, total volatile organic compounds (TVOC), ash content, moisture content, and viscosity collectively provide insight into formulation chemistry, structural characteristics, and functional performance.

The pH and Pi values (Figure 3) reveal a clear distinction between the bio-adhesives and the commercial adhesive. All formulated adhesives exhibit strongly alkaline pH and elevated Pi values, clustering within a narrow range. This behaviour is characteristic of starch- and protein-based systems, where abundant hydroxyl and amide functional groups, coupled with borax-induced crosslinking, promote alkaline environments. In contrast, the commercial adhesive displays markedly lower pH and Pi values, indicating a fundamentally different chemical architecture, likely incorporating acidic stabilisers or synthetic polymer emulsions. This separation highlights the intrinsic chemical differences between natural polymer networks and industrial adhesive formulations.

Density measurements further reinforce this distinction. The starch-based adhesives (IBS, ECS, and MES) show closely comparable densities, while the gelatin-based adhesive (HGT) exhibits a moderately reduced value. The commercial adhesive, however, demonstrates a substantially lower density, positioning it as a clear outlier. This suggests a lighter formulation, potentially attributable to higher solvent content, aeration, or foam-like microstructures commonly employed in industrial adhesive products to enhance spreadability and application efficiency.

TVOC analysis (Figure 2) indicates formulation-dependent volatility behaviour. ECS and the commercial adhesive form a high-TVOC group, whereas IBS, MES, and HGT cluster at lower values. The relatively reduced TVOC levels of most bio-adhesives suggest lower volatile emissions, which is advantageous from an environmental and occupational health perspective. The elevated TVOC observed in ECS may reflect compositional differences arising from starch source variability or modifier interactions.

Ash content provides one of the most striking contrasts in the comparative analysis. All bio-adhesives exhibit low and narrowly distributed ash contents, indicating minimal inorganic residue and reinforcing their organic, biodegradable nature. In contrast, the commercial adhesive displays an exceptionally high ash content, signifying substantial inorganic filler incorporation. Such filler loading is commonly used in industrial adhesives to enhance mechanical strength, thermal resistance, and dimensional stability, but it also differentiates the commercial product sharply from the bio-derived formulations.

Moisture content trends (Figure 3) align with the hydrophilic nature of starch and gelatin matrices. IBS, ECS, and HGT show high moisture retention, while MES exhibits moderately reduced moisture, suggesting improved drying behaviour. The commercial adhesive demonstrates extremely low moisture content, indicative of a highly concentrated or solvent-based system designed for rapid curing and moisture resistance. This contrast underscores a key performance trade-off between natural polymer adhesives and synthetic commercial products.

Viscosity measurements further highlight formulation effects on rheological behaviour. The commercial adhesive exhibits the highest viscosity, reflecting a dense polymer filler network optimised for strong bonding performance. Among the bio-adhesives, IBS and MES show relatively high viscosities, suggesting effective gel network formation and good load-bearing potential. ECS presents intermediate viscosity, while HGT displays the lowest viscosity, consistent with protein-based systems forming less rigid networks compared to starch-rich matrices.

Overall, the qualitative comparative analysis depicted in Figure 3 demonstrates clear clustering of the bio-based adhesives across most physicochemical parameters, reflecting shared natural polymer architectures. The commercial adhesive consistently separates from this cluster, driven by its low density, low moisture content, high ash residue, and elevated viscosity. These differences underscore the distinct formulation strategies and performance profiles between bio-derived adhesives and industrial commercial products, while also highlighting the potential of starch- and gelatin-based systems as viable, lower-cost, and environmentally benign alternatives with tunable properties.

Peeling Test

Table 1: The Peeling Strength Of The Formulated Adhesive Systems From Ipomea Batatas Starch (IBS), Eleusine Coracana Starch (ECS), Manihot Esculenta Starch (MES), Hide Gelatine (HGT) And Bone Gelatin (BGT), And Commercial Adhesive

Natural Adhesives	% Modifier	Load (g)	Force (N)	Peeling Strength (MPa)	Average Velocity (cm/s)
IBS	0 % PVAc	750	7.50	1.01	0.15
	20 % PVAc	1250	12.50	1.67	0.19
ECS	0 % PVAc	1000	10.00	1.35	0.16
	20 % PVAc	1500	15.00	2.02	0.12
MES	0 %PVAc	750	7.50	1.01	0.13
	20 % PVAc	1250	12.50	1.68	0.12
HGT	0 % PVAc	500	5.00	0.67	0.11
	20 % PVAc	1000	10.00	1.35	0.15
COMM.		1500	15.00	2.02	0.18

Peeling strength is an important property to characterize the adhesion of materials. Cow Leather was used as the testing material, and an effective Muver peeling instrument (Model 5038) was set up to test peeling force. This peeling test is a debonding process; hence, it is an energy-driven process. In the peeling test, calibrated weights were hung at one edge of the leather test piece. With the gravity of the calibrated weights, the adhesive test piece slowly peeled off from the substrate.

The peeling off procedure was observed, so the peeling strength and peeling rate, as well as the peeling force, were calculated as presented in Table 1. The strength of the starch-based adhesive (from Eleusine *coracana*) with 20 % PVAc was found to have almost the same peeling strength value as that of the commercial adhesive (2.02 Mpa), with a different peeling rate (0.18 cm/s) found to be lower than that of the commercial adhesive used as control. This is suggestive of the fact that starch-based adhesive (from Eleusine *coracana*) with 20 % PVAc is a good and better adhesive than the counterparts obtained from other sources, such as Ipomea *batatas* (1.68 MPa), Manihot *esculenta* (1.67 MPa), Hide (0.67 MPa), and possesses almost equal strength with the commercial adhesive used for standard measurement. However, it can be observed that some of the adhesives from different sources seem to have the same values but vary in peeling rate. This means that increasing peeling strength leads to increasing peeling rate, and peeling rates are positively correlated to adhesive energy, as the adhesive energy required is larger when the cracking velocity increases.

CONCLUSION

This study demonstrates that targeted modification of starch- and gelatin-based adhesive systems with polylactic-acid-derived modifiers provides an effective pathway for engineering low-emission, high-performance bio-adhesives. Systematic variation of modifier content revealed clear structure–property–performance relationships, in which polymer–polymer interactions, hydrogen bonding, and network architecture govern rheological behavior, physicochemical stability, and adhesion strength. Moderate levels of polylactic acid and polyvinyl acetate enhanced viscosity control, density, and peel strength, while excessive incorporation disrupted biopolymer entanglement and diminished adhesive performance. Importantly, all formulated adhesives exhibited total volatile organic compound levels well below international regulatory limits, confirming their environmental compatibility. Among the evaluated systems, *Eleusine coracana* starch–based formulations achieved peel strength comparable to a commercial adhesive benchmark, highlighting the potential of source-specific starch selection in bio-adhesive design. Overall, this work establishes a mechanistic foundation for rational formulation of sustainable starch–gelatin adhesives and supports their translation toward industrial applications where performance, cost efficiency, and environmental compliance are critical.

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