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# Formulation-Dependent Properties of Rice Starch-Based Adhesives for Wet-Strength Paper Labels

**ABSTRACT**

*The development of sustainable adhesives from renewable resources represents an important step toward reducing the environmental impact of packaging materials. This study investigated four rice starch-based adhesive formulations (L1–L4), including systems modified with chitosan and alginate, for potential application in paper labels on glass packaging. The physicochemical properties were characterized by measuring pH, apparent viscosity, and non-volatile solids content, while mechanical performance was evaluated using tensile tests on three paper substrates (V1–V3) in the machine direction (MD) and cross-machine direction (CD). The adhesive formulations exhibited pH values between 5 and 7, apparent viscosities ranging from 2,500 to 5,000 mPa·s, and solids contents of 10–20%. L1 (starch–chitosan–alginate) demonstrated the highest viscosity (5,000 mPa·s) despite having the lowest solids content (10%), indicating that rheological behavior was strongly influenced by formulation composition rather than solids concentration alone. Mechanical testing revealed that adhesive with rice starch, chitosan and alginate exhibited the highest average maximum tensile force (3.98 N), whereas rice starch and alginate adhesive achieved the highest average elongation (30.3 mm). The maximum recorded tensile force was 5.1 N, while elongation reached 66.1 mm, demonstrating considerable differences among adhesive formulations, paper substrates, and testing directions. These findings highlight the importance of balancing rheological characteristics, mechanical resistance, and flexibility when designing bio-based adhesives. Overall, rice starch-based formulations showed promising potential for sustainable paper labeling applications. Further investigation of adhesive durability, washing resistance, and industrial processing performance is required to establish their suitability for wet-strength and removable labels on glass packaging.*

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**KEY WORDS**
Rice starch-based adhesives, bio-based adhesives, chitosan, alginate, tensile properties, paper labels, sustainable packaging

## 1. Introduction

The transition to resource-efficient packaging has stimulated interest in adhesives derived from renewable feedstocks. Adhesives form a small but functionally important component of paper-based packaging: they must wet the substrate, develop sufficient interfacial contact, withstand conversion and distribution stresses, and remain compatible with the intended end-of-life processing. Consequently, replacing a conventional formulation with a bio-based adhesive requires an application-specific assessment of processing rheology, bonding performance and durability rather than an evaluation of renewable content alone (Vrabič-Brodnjak & Vidmar, 2025). Glass-bottle and jar labels represent a particularly demanding case because the adhesive joins a fibrous and often wet-strength-treated paper to a smooth, non-porous glass surface. Labels may encounter condensation, immersion, mechanical handling and cleaning operations. As a conventional technology benchmark,

Márquez, Alarcia and Velasco (2020) developed water-based acrylic pressure-sensitive adhesives for glass-bottle paper labels and assessed their behaviour in cold and wet conditions. Their findings demonstrate that moisture resistance and label adhesion are important practical criteria, although the acrylic chemistry and application process differ from those of starch-based systems. Wash-off behaviour during container cleaning introduces an additional design challenge: resistance during service and ease of removal during recovery may require a controlled balance, which should be verified using defined test criteria. Starch is an attractive candidate for waterborne adhesives because it is widely available, renewable and rich in hydroxyl groups capable of hydrogen bonding with cellulose-rich paper surfaces. Gelatinised starch can provide viscosity and film formation, but its response depends strongly on concentration, processing history and moisture. Such hydrophilic formulations may undergo changes in cohesion and water sensitivity, making the use of other polysaccharides a plausible route to modification. Work on starch–chitosan materials has identified opportunities to tailor intermolecular interactions and physical behaviour through blending, although observations made on standalone films cannot automatically be equated with bond strength at paper–glass interfaces (Wan Yusof, Sabar & Zailani, 2024; Vrabič-Brodnjak & Vidmar, 2025; Klenovšek & Vrabič-Brodnjak, 2026). Chitosan is a polysaccharide containing amino and hydroxyl functionalities and can participate in hydrogen bonding and, depending on pH and protonation, electrostatic interactions. Recent reviews describe starch–chitosan blends as tunable polymer systems for films and coatings, with properties dependent on preparation and component ratios (Wan Yusof, Sabar & Zailani, 2024). For an aqueous adhesive, these interactions may influence dispersion, apparent viscosity, setting behaviour and adhesive-layer integrity. However, any specific advantage for bonded wet-strength paper needs to be substantiated by measurements of peel or shear behaviour, fracture locus and performance following wet exposure; compatibility alone does not establish improved adhesion. Alginate offers a complementary modification strategy through its hydrophilic, carboxylate-bearing chains and pronounced ability to modify aqueous solution rheology. Kiattijiranon, Auras and Sane (2024) reported hydrogen bonding, interfacial adhesion and improved functional performance in sodium-alginate/starch multilayer packaging films. This supports the broader materials rationale for combining these two polysaccharides, but those results relate to films rather than to liquid adhesive deposition and paper-to-glass bonding. In practice, higher viscosity can improve resistance to flow or penetration in some coating operations while making pumping, transfer, levelling or spreading more difficult in others. Thus, viscosity must be considered together with non-volatile solids, coat weight and the specific application equipment.

An additional gap concerns the paper substrate itself. Wet-strength grades are designed to retain integrity under aqueous exposure, but grammage and paper anisotropy can modify deformation and failure in bonded assemblies. Samples taken parallel to the machine direction (MD) and the cross direction (CD) can exhibit different tensile responses owing to fibre orientation. Moreover, a tensile or peel-type loading configuration probes a different stress state from an in-plane shear configuration. Directly comparing several adhesive compositions across paper grades and directions can therefore clarify whether an apparent improvement reflects adhesive properties, substrate deformation or the test geometry. Against this background, the present work compares four rice starch-based formulations: starch–chitosan–alginate (L1), starch–alginate (L2), starch–chitosan (L3) and starch only (L4). Three wet-strength papers with grammages of 65, 80 and 75 g/m² were tested in MD and CD, and the study includes adhesive pH, Brookfield apparent viscosity, non-volatile solids and reported mechanical and washing evaluations. Beyond the primary measurements, we examine the descriptive ratio of viscosity to solids content to highlight formulation-dependent thickening at different concentrations. The objective of our research was to link formulation, processing-relevant physicochemical behaviour and the requirements of paper labels for glass packaging, while distinguishing measured outcomes from mechanisms that remained to be confirmed experimentally.

# 2. Methods and materials

## 2.1. Adhesives and paper substrates

Four formulations were designated L1–L4 as summarized in Table 1. The carbonate component reported-$CO_3$ in the description after was used for pH regulation in L1 and L3. Three wet-strength papers were examined: V1 (65 g/m²), V2 (80 g/m²) and V3 (75 g/m²). Test strips were cut parallel to the machine direction (MD) or cross direction (CD).

**Table 1**

Materials and formulation design

| Sample name | Adhesive formulation | Substrate conditions |
|---|---|---|
| L1 | Rice starch + chitosan + alginate | V1, V2, V3; MD and CD |
| L2 | Rice starch + alginate | V1, V2, V3; MD and CD |
| L3 | Rice starch + chitosan | V1, V2, V3; MD and CD |
| L4 | Rice starch only | V1, V2, V3; MD and CD |

## 2.2. Physical-chemical analysis

The pH of each formulation was measured with a calibrated pH meter following EN 1245:2011. Samples were equilibrated at 23 ± 2 °C, and measurements were performed in duplicate. Apparent viscosity was measured with a Brookfield rotational viscometer at 23 ± 0.5 °C, using the same spindle and rotational speed for all samples. Because polymer-based adhesives may be shear-dependent, the reported viscosities are compared as apparent values under shared test settings. Non-volatile solids content was determined gravimetrically, using 2 g of formulation dried at 105 ± 2 °C to constant mass; the reported method cites ASTM D1489. Solids content (%) = 100 × (dry residue mass / initial sample mass).

## 2.3. Mechanical analysis and washing analysis of durability of labels

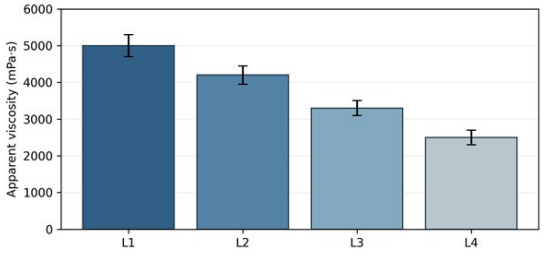
Bonded paper specimens underwent tests designated T-tests on dynamometer Instron tensile testing machine, with comparisons between MD and CD direction. The precise specimen geometry was 10 x 1.5 cm, test speed was 25 mm/min, length between the testing clamps was 10 cm and 10 replicates were made at each sample.

A separate washing challenge exposed bonded labels to a commercial washing-machine cycle for 1 h at 55 °C and drying using commercial, washing tablets.

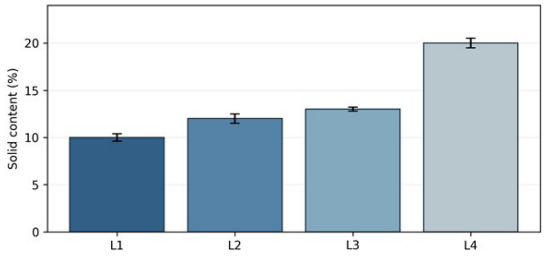
# 3. Results and discussion

## 3.1. pH, viscosity and solids content

According to results, pH varied from 5 (L3), 6 (L2) to 7 (L1 and L4). Apparent viscosity decreased in the order L1 > L2 > L3 > L4, whereas non-volatile solids decreased in the opposite order (L4 > L3 > L2 > L1). Thus, the observed processing-relevant viscosity cannot be explained by solids loading alone. Figure 1 presents the measured viscosities and Figure 2 the solid contents. The physicochemical characterization of the four adhesive formulations (L1–L4) revealed notable differences in pH, apparent viscosity, and non-volatile solids content, indicating that adhesive composition strongly influences their processing behavior. The pH values ranged from 5 to 7, with L3 exhibiting the lowest pH (5), followed by L2 (6), while L1 and L4 showed neutral conditions (pH 7). These differences may influence the ionization of functional groups, intermolecular interactions, and the structural organization of the adhesive systems. As shown in Figure 1, apparent viscosity progressively decreased from approximately 5000 mPa·s for L1 to 4200 mPa·s for L2, 3300 mPa·s for L3, and 2500 mPa·s for L4. This represents an overall viscosity reduction of approximately 50% between L1 and L4. Such differences are particularly relevant for industrial adhesive applications, as viscosity directly affects adhesive flow, spreadability, coating uniformity, and application efficiency. From a practical perspective, L4 appears particularly promising for applications requiring relatively high solids content combined with low viscosity. Such a combination may facilitate adhesive application while potentially reducing the amount of water that must be removed during drying. Conversely, the higher viscosity of L1 may be advantageous in applications requiring greater resistance to flow, although it could present challenges during coating or spreading.




» **Figure 1:** *Apparent viscosity of the four adhesives L1-L4; error bars reproduce the supplied ± values*

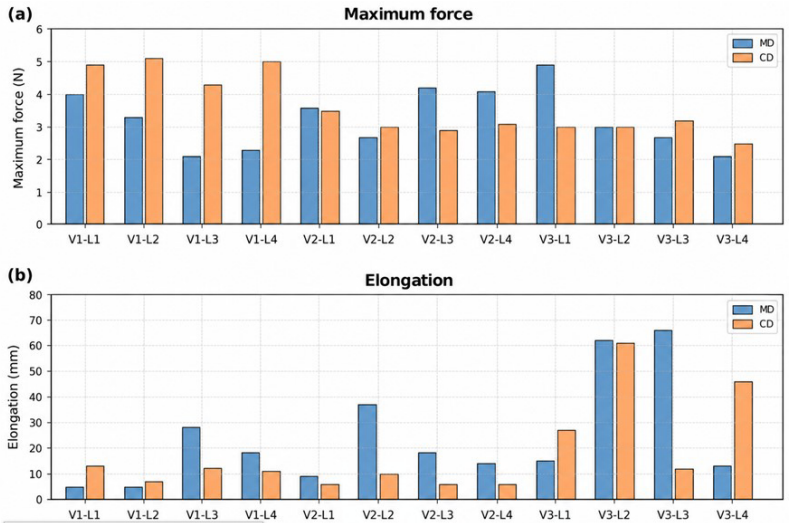



» **Figure 2:** *Non-volatile solids content of the four adhesives L1-L4; error bars reproduce the supplied ± values*

## 3.2. Mechanical properties

The mechanical properties of the adhesive formulations (L1–L4) were evaluated using tensile tests (T-test) on three paper substrates (V1–V3) in the machine direction (MD) and cross-machine direction (CD). Maximum tensile force and elongation at maximum force are presented in Figure 3a and 3b.

The results revealed differences in mechanical performance depending on adhesive formulation, paper substrate, and testing direction. L1 exhibited the highest average maximum tensile force (3.98 N), followed by L2 (3.35 N), L3 (3.23 N), and L4 (3.18 N). The highest individual force (5.1 N) was recorded for L2 on V1 in the CD direction. In contrast, L2 demonstrated the highest average elongation (30.3 mm), followed by L3 (23.7 mm), L4 (18.0 mm), and L1 (12.5 mm). The highest individual elongation (66.1 mm) was observed for L3 on V3 in the MD direction. Differences between MD




» **Figure 3:** *a) Maximum force and b) elongation at break in MD in CD direction for all 4 adhesives (L1-L4) and 3 paper samples (V1-V3)*

and CD further indicated the influence of paper fiber orientation on mechanical behavior. Nevertheless, L1 provided the highest average tensile-force resistance, whereas L2 and L3 showed greater deformability. These findings suggest that adhesive performance depends on both formulation and substrate characteristics, highlighting the importance of balancing strength and flexibility for paper-based packaging applications.

## 3.3. Washing resistance and adhesive bond stability

In addition to the mechanical characterization, the adhesive formulations were subjected to a washing challenge at 55 °C for 1 h in the presence of washing tablets. This experiment was designed to evaluate adhesive-bond stability under conditions involving elevated temperature, water exposure, and detergent-induced chemical interactions. Washing resistance is an important characteristic for adhesive applications involving removable labels, reusable packaging systems, and materials that may undergo aqueous cleaning processes. Exposure to warm water may promote swelling and plasticization of hydrophilic adhesive components, particularly polysaccharide-based materials such as alginate and chitosan. Water penetration into the adhesive network can disrupt hydrogen bonding and reduce cohesive or interfacial strength. Furthermore, detergent components may alter the adhesive–substrate interface and facilitate bond weakening or label separation. The pH and viscosity differences observed among the formulations may also influence water penetration and washing behavior.

As shown in Figure 4, all formulations exhibited varying degrees of label detachment and paper degradation. L1 showed extensive label separation, with a large portion of the paper detached from the glass surface, suggesting reduced adhesion under washing conditions. In contrast, L2 demonstrated greater label retention, with most of the paper remaining attached to the glass. L3 exhibited pronounced paper tearing and surface deterioration, while L4 showed partial label detachment and fragmentation. These observations suggest that adhesive composition influences wash-off behavior and the integrity of paper labels during washing. From a reusable glass packaging perspective, easier label removal, as observed for L1, may be advantageous for cleaning and relabeling processes. However, further quantitative evaluation of adhesive residue and detachment efficiency is required to establish the suitability of the formulations for industrial washing applications.



*L1* *L2* *L3* *L4*

» **Figure 4:** *Representative photographs of paper labels bonded to glass containers after washing at 55 °C for 1 h using washing tablets: (a) L1, (b) L2, (c) L3, and (d) L4*

### 3.4. Implications for sustainable packaging

The findings demonstrate the potential of biopolymer-containing adhesive formulations for paper-based packaging applications. The most important observations were the superior overall T-test response of L1, the increased flexibility associated with alginate and/or chitosan incorporation, and the influence of paper orientation on mechanical performance. The opposite trends observed for viscosity and solids content further demonstrate that adhesive processing characteristics cannot be predicted solely from non-volatile solids concentration. These results suggest that formulation-dependent molecular interactions play an important role in determining adhesive behavior and that optimization should account for both processing requirements and the mechanical performance of bonded substrates. Therefore, L1 appears promising where mechanical resistance under the investigated T-test conditions is prioritized, whereas L4 offers potential processing advantages owing to its combination of lower viscosity and higher solids content. The final selection of an adhesive formulation should also consider its mechanical performance across different substrates and its resistance to, or controlled removal during, washing.

## 4. Conclusions

Four rice starch-based adhesive formulations exhibited pH values of 5–7, apparent viscosities of 2,500–5,000 mPa·s, and solids contents of 10–20%. L1 (starch–chitosan–alginate) demonstrated the highest viscosity despite having the lowest solids content, indicating that adhesive composition and intermolecular interactions strongly influence rheological behavior. Mechanical testing revealed that L1 provided the highest average maximum tensile force (3.98 N), while L2 exhibited the greatest average elongation (30.3 mm). Differences between machine (MD) and cross-machine (CD) directions further highlighted the influence of paper substrate orientation on bonding performance. Overall, the results demonstrate the potential of rice starch-based adhesives as sustainable alternatives for paper labels on glass packaging. Further optimization and washing-resistance evaluation are necessary to establish their suitability for industrial applications, particularly wet-strength and removable labeling systems.

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