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Enhancing the Quality of Cassava Starch-Based Wood Bioadhesive using Polyvinyl Alcohol

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Abstract. Many studies tried to innovate wood bioadhesives using renewable resources. This research attempted to synthesize wood bioadhesive using cassava starch and polyvinyl alcohol (PVA). As a new cross-linking agent, citric acid was added to the bioadhesive to ensure the bonding between cassava starch and PVA. PVA and citric acid contain hydroxyl groups that can initiate polymerization with starch. The synthesis of bioadhesive has three steps, namely, acid hydrolysis, oxidation, and cross-linking reactions. The operating condition was set at 70°C under atmospheric pressure for 90 minutes. The amount of added PVA varied at 0%, 10%, 20%, 30%, and 40% relative to the weight of cassava starch. Physical properties such as viscosity, solid content, shelf life, and dry and wet shear strength were analyzed to determine the best composition of starch and PVA. Results showed that the addition of PVA could enhance the bioadhesive's properties, especially its dry and wet shear strength.

Keywords: Cassava starch; Citric acid; Polymerization; Polyvinyl alcohol; Wood bioadhesive

1. Introduction

Wood adhesive is one of the essential materials in the wood industry (Dunky, 2017; Kaboorani and Riedl, 2011; Frihart, 2011) because of its easy and fast application in joining a pair of wood pieces. Synthetic adhesives such as phenol-formaldehyde and urea-formaldehyde are the popular types of adhesives (Ćehić, Talić, and Omer, 2023; Gonçalves *et al.*, 2021; Pizzi and Mittal, 2003). The industry has become comfortable with using this adhesive during manufacturing because of its good performance in gluing a couple of wood parts. However, this type of adhesive has a dangerous influence on human health. Synthetic adhesives contain highly volatile compounds that could cause irritation and respiratory problems in humans after prolonged contact (David and Niculescu, 2021; Kumar and Pizzi, 2019; Łebkowska, 2017).

Bioadhesive is an alternative to synthetic adhesives and has sufficient capability to glue a pair of wood pieces. Moreover, it offers several advantages, such as renewability, low cost, easy processing, reusability, and environmental friendliness (Borges *et al.*, 2023; Raydan *et al.*, 2021; Balgude, Sabnis, and Ghosh, 2017). However, this adhesive has weak strength and lousy durability compared with synthetic adhesives. Many researchers have

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tried to innovate and develop bioadhesives from many renewable resources to obtain excellent adhesive characteristics. Cassava starch is an abundant resource that shows potential as the raw material for adhesives (Admase, Mersha, and Kebede, 2024; Yu *et al.,* 2016; Ozemoya, Ajisegiri, and Idah, 2007). Due to its easy gelatinization and modification suitable for developing new wood bioadhesives (Wang *et al.,* 2017; Zhang *et al.,* 2015; Wang *et al.,* 2012). Many studies were conducted to improve the quality of cassava starch as a bioadhesive. Modifications, such as hydrolysis, oxidation, and gelatinization, were carried out to obtain high-quality starch bioadhesives. However, starch's retrogradation limits its performance as a bioadhesive. Several studies attempted to add an acid catalyst (Gadhave *et al.,* 2020; Monroy, Rivero, and García, 2019) or a surfactant (Li *et al.,* 2014) to prevent this retrogradation.

Starch retrogradation can be reduced by binding the starch with other monomers or polymers to form a new polymer model. Polyvinyl alcohol (PVA) is an example of a polymer that can be added to starch to improve the 'latter's adhesive properties (Elgharbawy *et al.,* 2024; Farshi *et al.,* 2024; Jaffe and Rosenblum, 1990).

PVA is a synthetic polymer that is cheap, non-toxic, and has high water solubility and elasticity (Chousidis, 2024; Barleany *et al.,* 2020; Gaaz *et al.,* 2015). PVA can be applied as a nanofiber membrane. The addition of 5% ramie cellulose nanocrystal (CNC) as reinforcement can increase the tensile strength by 2 times more than before it was added (Xu *et al.,* 2024; Nguyen *et al.,* 2023; Rochardjo *et al.,* 2021). Another study stated that adding a copolymer in the form of PVA and 2-ethylhexyl acrylate (2-EHA) grafted with ammonium persulfate (APS) in mortar could increase physical properties such as compressive strength and reduce water absorption (Bekbayeva *et al.*, 2020). The hydroxyl groups of PVA provide an opportunity to form a new polymer between PVA and starch (Sridach, Jonjankiat, and Wittaya, 2013; Reddy and Yang, 2010). As a new natural cross-linking agent, citric acid can initiate polymerization to ensure PVA–starch bonding. In this research, we tried to synthesize a new bioadhesive polymer from cassava starch and PVA using citric acid as the cross-linking agent. Physical properties such as viscosity, durability, and shear strength were analyzed to evaluate the adhesive quality of the bioadhesive polymer.

2. Methods

2.1. Materials

The following are the materials used in this research: cassava starch (PT. Budi Acid Jaya, Tbk), ammonium persulfate ($(NH_4)_2S_2O_8$) (LPPT UGM, Indonesia), polyvinyl alcohol (PVA) (99% hydrolyzed, Mw 89,000 – 99,800, Merck KgaA Company, Germany), sodium dodecyl sulfate (SDS) (CV. Gudang Kimia, Indonesia), 37% hydrochloric acid (HCl) (Merck KgaA Company, Germany), and wooden board (*Tectona grandis*) (UD. Makmur Jaya, Indonesia).

2.2. Synthesis of Bioadhesive

In this experiment, approximately 50 g of cassava starch was dissolved in 100 mL of 0.5 M HCl. The resulting solution was placed in a three-neck flask and heated to 70°C with stirring at 500 rpm using a mercury stirrer. After 10 minutes, 0.4 g of $(NH_4)_2S_2O_8$ was added to the solution, followed by the addition of 1 g of SDS after 30 minutes. Separately, 5 g of PVA was dissolved in 50 mL of Aquadest and added to the three-neck flask. Then, 0.4 g of citric acid was added to the solution and stirred for 1 hour. The experiment was repeated with different amounts of PVA (0, 10, 15, and 20 g) using the same procedure.

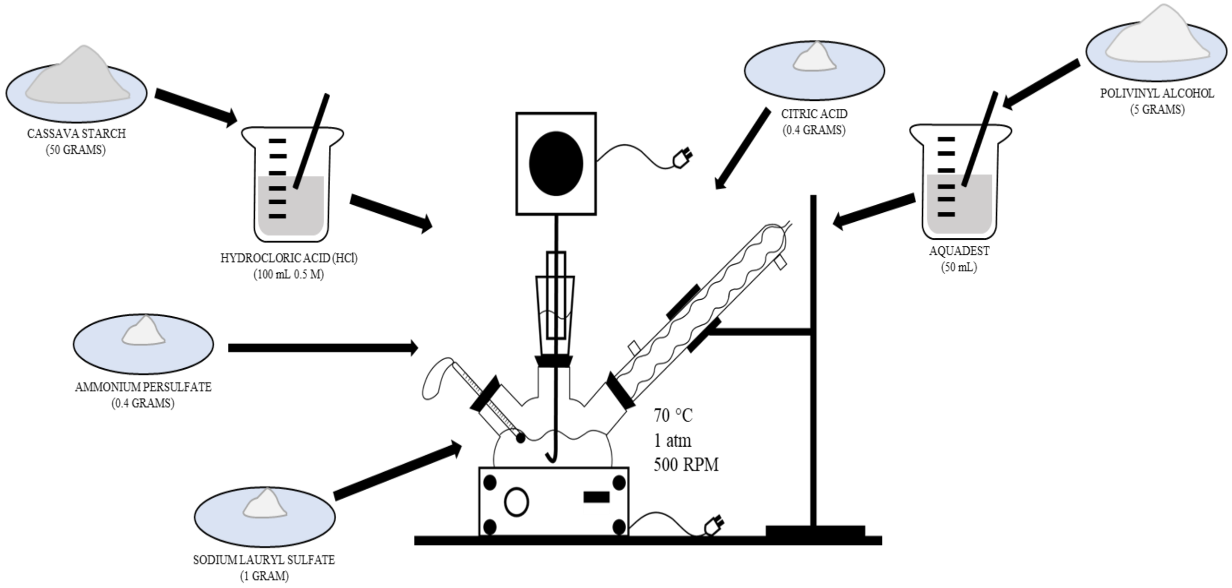


Figure 1 The Illustration of Bioadhesive Synthesis in the Temperature of 70 °C and Pressure 1 atm

2.3. Viscosity Analysis

The 'adhesive's viscosity was evaluated in accordance with ASTM D1084. The adhesive was transferred to a beaker glass, and a suitable spindle was installed by considering the thickness of the adhesive. The size and rotating speed (rpm) of the spindle in the DV-E Brookfield Viscometer were controlled. Viscosity was recorded as the value consistently shown on the monitor. The allowed error for viscosity was in the range of 10%–90%.

2.4. Shelf-Life Analysis

The shelf life of the adhesive was measured in accordance with ASTM D1337. The sample was kept at room temperature with 50% relative humidity, and its viscosity was tested every 7 days for 31 days. The thickness was evaluated in accordance with ASTM D1084.

2.5. Solid Content Analysis

The solid content of the adhesive was evaluated in accordance with ASTM-D2369. In brief, 1 g of the adhesive was placed in a petri dish and dried in the oven for 60 minutes at 110 ± 2.5°C. The sample was then cooled and stabilized in a desiccator for 10 minutes and finally weighed. The solid content of the adhesive was calculated using Equation 1:

$$S = \frac{m_2 - m_0}{m_1 - m_0} \times 100\%, \tag{1}$$

where S is solid content (%), m_0 is the weight of the petri dish (g), m_1 is the weight of the petri dish and sample before heating (g), and m_2 is the weight of the petri dish and sample after heating (g). The analysis was repeated three times, and the results were averaged.

2.6. Dry Shear Strength Analysis

Dry shear strength was evaluated in accordance with ASTM D906. Wood pieces with 30 × 25 × 2 mm dimensions were glued using the adhesive and pressed under 3.27×10^{-2} N/mm^2 for 24 hours. The wood samples were stored at room temperature with 50% RH for 7 days, and their maximum load was measured using a universal testing machine at 2 mm/min of testing speed. The shear strength was calculated using Equation 2:

$$\sigma_t = \frac{W}{A \times 1000}, \tag{2}$$

where σ_t is the shear strength of adhesive (MPa), W is the maximum value load (N) that was shown in the monitor of the universal testing machine, and A is the glued area of the wood (mm^2). The measurement was repeated five times, and the obtained values were averaged.

2.7. Wet Shear Strength Analysis

Wet shear strength was evaluated in accordance with ASTM D1183. Dry shear strength was measured by determining the exact dimension of the wood that was glued and pressed under 3.27×10^{-2} N/mm^2. After 24 hours, the sample was immersed in water for another 24 hours. The sample was dried for 24 hours and stored at 50% RH and at room temperature for 7 days. Afterward, the maximum load of the samples was measured using a universal testing machine at 2 mm/min of testing speed. The wet shear strength was calculated using Equation 2. The measurement was repeated five times, and the obtained values were averaged.

2.8. Fourier Transform Infrared (FTIR) Spectroscopy

FTIR spectroscopy was applied to identify the functional groups in the samples using spectrophotometer FT-IR Mb 3000 (LPPT UGM, Yogyakarta, Indonesia). The spectra ranged between 650 and 4000 cm^{-1}, and 32 scans were performed.

2.9. DinoLite Microscopy and Scanning Electron Microscopy (SEM)

The surfaces and morphology of the bioadhesive were analyzed using a DinoLite microscope and a SEM from LPPT UGM, Yogyakarta, Indonesia.

3. Results and Discussion

Various PVA and starch compositions were examined to determine the best combination that would provide the best performance for the bioadhesive. The PVA: starch ratios of 1:10, 2:10, 3:10, 4:10, and 0:10 (wt./wt.) were labeled as SP1, SP2, SP3, SP4, and SP0, respectively (the blank sample: regular bioadhesive without PVA). The experiment was carried out using a batch reactor, which is easy to control and can provide the best conversion in the reaction.

3.1. Reaction Mechanisms of Adhesive Synthesis

Several mechanisms are involved in bioadhesive synthesis. The first process involved hydrolysis and gelatinization using HCl solution as the catalyst at 70°C under atmospheric pressure. In this step, the glycosidic bonds in cassava starch were destroyed, resulting in acid-thinned starch (ATOS). The starch's primary alcohol or primary hydroxyl groups were then oxidized using $(NH_4)_2S_2O_8$ (Imanzadeh *et al.,* 2007).

This process aimed to form the carboxyl groups (-COOH) in the structure to support polymerization. The other hydroxyl groups or secondary alcohol could be oxidized to carbonyl groups (-CO). SDS and PVA were dissolved in aquadest and added to the mixture. SDS was added to prevent retrogradation during cassava starch gelatinization. After 10 minutes, citric acid was added to the mixture as the cross-linking agent. The two possible reaction paths in cross-linking reactions are polymerization between starch molecules and polymerization between starch and PVA. These synthesis processes are shown in Figure 3.

This solution polymerization reaction requires a non-monomeric solvent, aquadest (H_2O) (Ahmed and Rashid, 2022). PVA is dissolved in aquadest (liquid phase), so the opportunity to in-contact with oxidized starch, PVA, and citric acid is greater. Moreover, the heat of the polymerization reaction can be removed by water. The hydroxyl group of PVA interacts with the carboxylic group of citric acid, thus forming an ester. The molecules formed become larger, so the average molecular weight increases.

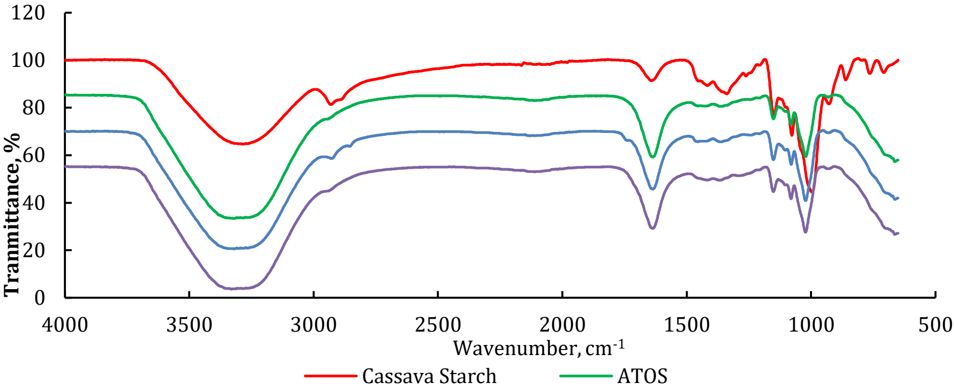


Figure 2 Infrared of cassava starch, acid-thinned and oxidized starch (ATOS), SP0, and SP3

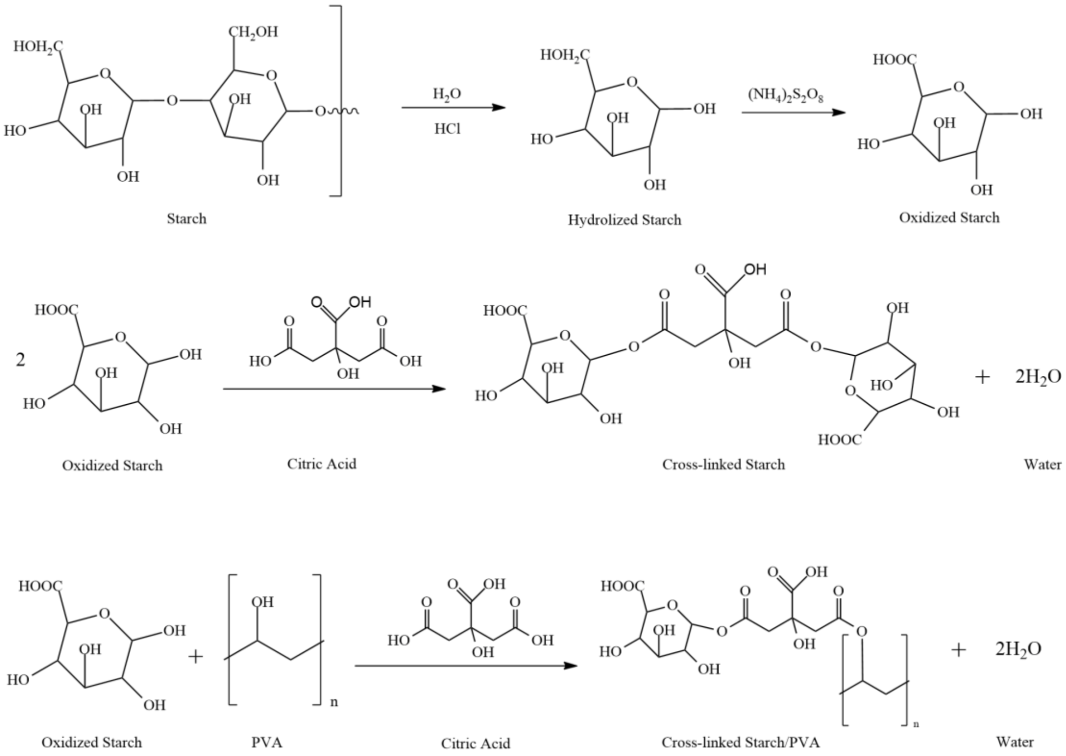


Figure 3 Sequence of reaction in starch/polyvinyl alcohol adhesive formation with citric acid addition

3.2. Viscosity of Adhesives

Viscosity was analyzed following ASTM D1084 to measure the thickness of the adhesive using a spindle with the lowest rotating speed. This setting was selected because the adhesive is a non-Newtonian thixotropic fluid; hence, its viscosity will decrease over time (Bird, Stewart, and Lightfoot, 2002). The viscosity was calculated from the average of measured viscosities on the Brookfield monitor.

The viscosity of adhesives is shown in Figure 4. First, the viscosity decreased from 2118 cP to 106.2 cP and then gradually increased to 984, 2312, and 5127 cP. When the PVA content was 10%, the viscosity decreased due to the excess water in SP1. Meanwhile, the viscosity increased with PVA content due to the increase in the average molecular weight caused by the cross-linking reaction. PVA also introduced some solid particles into the adhesive, thus increasing its viscosity.

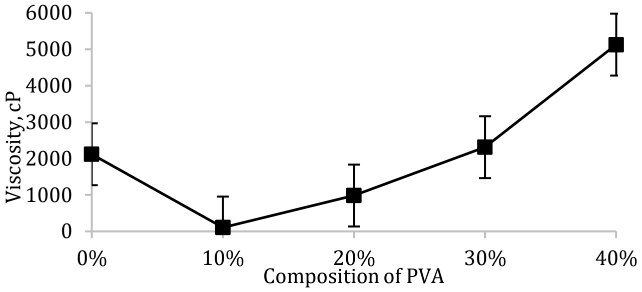


Figure 4 Viscosity of the adhesives with various polyvinyl alcohol additions

Viscosity is related to the ability of the adhesive to cover the surface of the wood easily. Therefore, adhesives with high viscosity are not desirable. SP1 had the lowest value of viscosity. The addition of water and PVA in this sample rendered it easy to apply. However, this addition affected the composition of solids and liquids in the adhesive. The polymerization between starch or starch/PVA also produced a large amount of water. This reaction involved condensation because of the interactions between the hydroxyl group on PVA and carboxylic acid on citric acid and between the hydroxyl group on starch and carboxylic acid on citric acid (Figure 3).

3.3. Shelf Life of Adhesives

The shelf life of the adhesive was evaluated to estimate its durability. The samples' viscosity was measured per ASTM D1084 every week for 31 days. The results are shown in Figure 5.

The viscosity of each sample increased over time. After 24 days, SP4 substantially thickened, so its viscosity could not be measured using the Brookfield Viscometer. The viscosity increased due to the contact between the adhesive and air trapped in the container. An adhesive with high viscosity is difficult to apply to any wood 'specimen's surface.

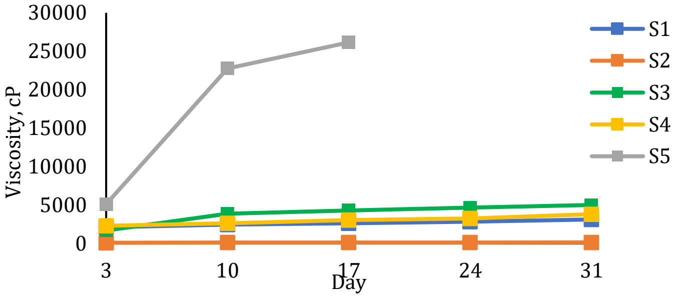


Figure 5 Viscosity of bioadhesives in a certain period

The significantly increased viscosity of SP4 shows that excessive PVA addition can have a negative impact on the quality of the adhesive. Additionally, clumping occurred in the adhesive that had been stored for a long period of time. This phenomenon increased the viscosity level of the adhesive and decreased its adhesion strength, thus making it nonreusable.

Solidification is the other reason for the increased viscosity of the adhesive. Excessive PVA addition will increase the number of solid particles in the adhesive, allowing PVA and starch to solidify. This phenomenon is plausible because no other additives have been incorporated into the adhesives.

3.4. Solid Content of Adhesives

The adhesives' solid content was measured per ASTM D2369 to identify their volatility level. High content of solid particles will decrease the volatility of the adhesive.

The solid content of the adhesives is shown in Figure 6. First, the value decreased from 30.62% to 25.16% and then increased gradually to 28.00%, 30.19%, and 32.70%. When the PVA content was 10%, the solid content decreased due to the low amount of solid particles in SP1. The solid content increased with the amount of PVA addition because many nonvolatile molecules were formed after the cross-linking reaction.

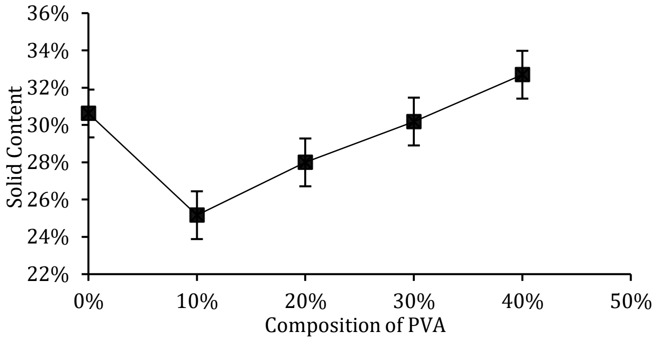


Figure 6 Solid content of the adhesives with various polyvinyl alcohol additions

Compared with SP0, the other samples had lower solid content, except for SP4. The reaction between PVA and starch increased the water content of the samples. Meanwhile, PVA addition also increased water accumulation in the adhesive. The excess water will reduce the solid content of the adhesive because the water will evaporate at oven temperature.

3.5. Dry Shear Strength of Adhesives

Shear strength is an essential parameter because it estimates the bonding strength of the adhesive. The shear strength measured in the dry condition is shown in Figure 7. The dry shear strength increased with the amount of PVA addition. In the dry state, the shear strength increased from 0.55 MPa to 2.54 MPa. Compared with SP0, SP4 had increased initial shear strength by 316.82% in the dry state and 678.13% in the wet state. These results indicated that the addition of PVA increases the shear strength of the adhesives. The cross-linking reaction between starch and PVA enhances the starch-based adhesive's bonding strength.

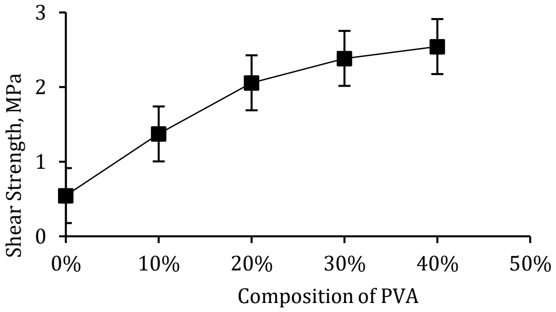


Figure 7 Dry shear strength of the adhesives with various polyvinyl alcohol additions

PVA addition can improve the chemical structure of the cassava starch adhesive. The bond of cassava starch–citric acid–cassava starch or cassava starch–citric acid–PVA rendered the resulting polymer highly stable. The interaction between hydroxyl groups (-OH) and carboxyl groups (-COOH) formed crosslinked molecules. The amount of these groups decreased, and the adhesive shear strength increased due to the absence of affinity effects on the water in the air.

3.6. Wet Shear Strength of Adhesives

Wet shear strength was analyzed per ASTM D1183 to determine theadhesive's resistance under moist or wet conditions. The adhesive's strength under various conditions must be evaluated to determine whether it can be used in the wet state. The results of the wet shear strength analysis are shown in Figure 8.

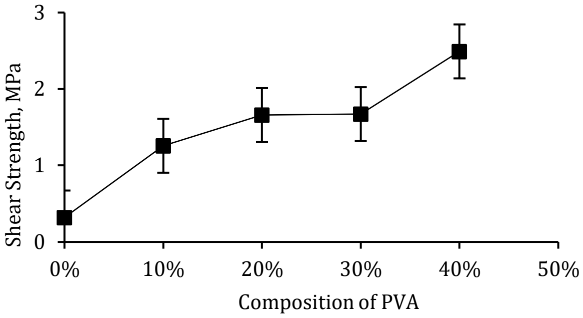


Figure 8 Wet shear strength of the adhesives with various polyvinyl alcohol additions

The wet shear strength increased with PVA addition. The shear strength increased from 0.32 MPa to 2.49 MPa. All adhesives had lower shear strength in the wet state than in the dry state. The shear strength decreased due to the high affinity between water and cassava starch. The hydroxyl groups (-OH) in cassava starch have an affinity for water, allowing cassava starch to be used as an adhesive that can dissolve in water. The dissolution of the adhesive into the water will affect the adhesive used during the gluing process. When a large amount of adhesive dissolves, the adhesion strength becomes weak. This affinity to water is caused by the similarity of polarity between the hydroxyl groups (-OH) and water. In some samples, the added PVA can prevent the cassava starch adhesive from dissolving during soaking. The PVA addition produced ester bioadhesives (crosslinked starch) and increased the water accumulation in the adhesive. The high-water content in the adhesive reduced the water resistance and broke the bonding of the wood specimens. Thus, the solubility of the adhesive increased, leading to a low bonding strength under moist conditions.

Compared with the dry shear strength, the wet shear strength decreased significantly. Table 1 shows the decrease in adhesive shear strength in each sample. In particular, SP4 experienced a reasonably small decrease in shear strength. Therefore, PVA addition can reduce the solubility of cassava starch in water. This effect can be attributed to the reduction in the hydroxyl groups (-OH) because of polymerization. Reducing the number of hydroxyl groups will reduce the solubility and increase the water resistance of the adhesive.

Table 1 Reduction in the shear strength of the adhesives after being immersed in water

Sample	Shear Strength Reduction
SP0	41.45%
SP1	8.29%
SP2	19.37%
SP3	29.91%
SP4	1.99%

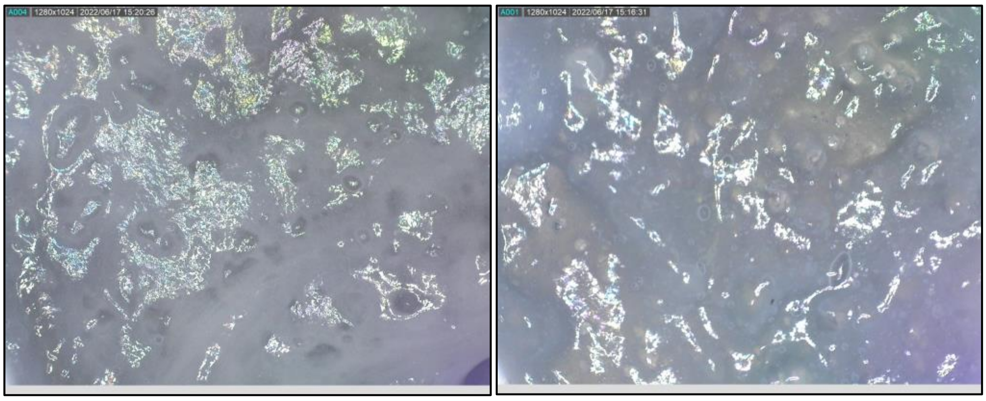
3.7. FTIR of Adhesives

FTIR spectroscopy was carried out to observe the structural changes of the starch-based adhesives during the reaction. The molecular structures were compared among cassava starch, ATOS, SP0, and SP3. The SP4 was not used because it had dried out. The analysis result is shown in Figure 2. At wavenumber 1022.43 cm^{-1}, the peak intensity of C–O bonds decreases from cassava starch to ATOS. Meanwhile, the peak intensity of O–H groups increases at wavenumber 3330.34 cm^{-1}. This finding indicated the occurrence of hydrolysis that destroyed the C-O bonds in amylose and formed the hydrolyzed starch. In addition, O-H bonds in cassava starch increase because hydroxyl groups are formed more in each single molecular hydrolyzed starch.

SP0 has a higher peak intensity at 1022.43 cm^{-1} and 1636.4 cm^{-1}. This is due to the addition of citric acid to SP0 from oxidized starch. The number of carboxyl groups increases and ester groups may start to form. Overall, no significant change was observed between SP0 and SP3 due to the compensatory role of the new O-H bonds generated from the polymerization between starch/PVA and citric acid. However, SP3 has a broader peak than SP0 in the 3000-3500 cm^{-1}. The hydroxyl group in PVA reacts with a carboxylic group from citric acid, so it was resulting in shifts, changes, and broadening of the absorption band SP3 to form ester groups

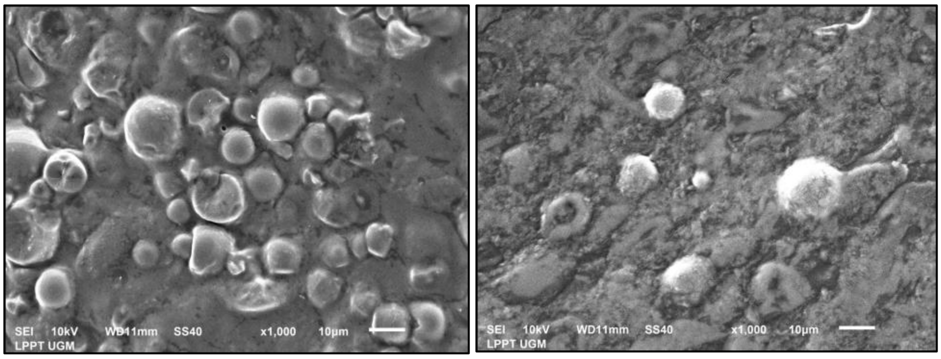
3.8. Morphology of Adhesive

The morphology of adhesives was analyzed using a DinoLite microscope and a SEM. The bioadhesive films with citric acid addition (Figure 9b and 10b) were smoother than those of the adhesive without the cross-linking agent (Figure 9a and 10a). This finding indicated that citric acid provides good homogeneity to the adhesive solution and increases the capability of the adhesive to bond the wood pieces and create a strong bonding.



(a) (b)

Figure 9 Morphology of the adhesive without citric acid addition (a) and with citric acid addition (b) visualized using a DinoLite microscope



(a) (b)

Figure 10 Morphology of the adhesive without citric acid addition (a) and with citric acid addition (b) visualized using scanning electron micrographs

4. Conclusions

PVA addition improves the properties of cassava starch-based bioadhesive. The viscosity and solid content of bioadhesives tend to increase with the PVA content. The dry and wet shear strength are enhanced when the PVA amount is increased. The dry and wet shear strengths of SP4 are 316.82% and 678.13% higher than those of the adhesive without PVA, respectively. However, an excessive amount of PVA addition could drastically increase the viscosity of the adhesive and accelerate the thickening process, leading to the poor durability and reusability of the bioadhesive. Therefore, the suitable composition of PVA addition to the starch adhesive was in sample S1 (10% of PVA content) because it offered a low viscosity value. On the other hand, this sample had enough value of shear strength to apply as wood bioadhesive directly. For future research, it is crucial to analyze the characteristics of adhesive toxicity and vary the use of starch as raw material.

Acknowledgments

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References

Admase, A.T., Mersha, D.A., Kebede, A.Y., 2024. Cassava Starch-Based Hot Melt Adhesive for Textile Industries. *Scientific Reports*, Volume 14, p. 20927, https://doi.org/10.1038/s41598-024-70268-y

Ahmed, B.Y., Rashid, S.O., 2022. Synthesis, Characterization, and Application of Metal-Free Sulfonamide-Vitamin C Adduct to Improve the Optical Properties of PVA Polymer. *Arabian Journal of Chemistry,* Volume 15(10), p. 104096, https://doi.org/10.1016/j.arabjc.2022.104096

Balgude, D., Sabnis, A., Ghosh, S.K., 2017. Synthesis and Characterization of Cardanol Based Reactive Polyamide for Epoxy Coating Application. *Progress in Organic Coatings,* Volume 104, pp. 250–262., https://doi.org/10.1016/j.porgcoat.2016.11.012

Barleany, D.R., Ananta, C.V., Maulina, F., Rochmat, A., Alwan, H., Erizal, 2020. Controlled Release of Metformin Hydrogen Chloride From Stimuli-Responsive Hydrogel Based On Poly(N-Isopropylacrylamide)/Chitosan/Polyvinyl Alcohol Composite. *International Journal of Technology,* Volume 11(3), pp. 511–521, https://doi.org/10.14716/ijtech.v11i3.2330

Bekbayeva, L., Negim, E., Gulzhakhan, Y., Ganjian, E., 2020. Utilization of Poly(Polyvinyl Alcohol-g-2-Ethylhexyl Acrylate) as Admixture for Mortar. *International Journal of Technology,* Volume 11(2), pp. 259–268, https://doi.org/10.14716/ijtech.v11i2.3927

Bird, R.B., Stewart, W.E., Lightfoot, E.N., 2002. Phenomena, Transport. Transport Phenomena, Wiley and Sons, pp. 582–605, ISBN 0-471-41077-2

Borges, C.S.P., Jalali, S., Tsokanas, P., Marques, E.A.S., Carbas, R.J.C., da Silva, L.F.M., 2023. Sustainable Development Approaches through Wooden Adhesive Joints Design. *Polymers,* Volume 15(1), p. 89, https://doi.org/10.3390/polym15010089

Ćehić, M., Talić, H., Omer, S.E., 2023. Eco-Friendly Adhesives for Wood-Based Composite Production. *IOP Conference Series: Materials Science and Engineering,* Volume 1298, p. 012013, https://doi.org/10.1088/1757-899X/1298/1/012013

Chousidis, N., 2024. Polyvinyl Alcohol (PVA)-Based Films: Insights from Crosslinking and Plasticizer Incorporation. *Engineering Research Express,* Volume 6, p. 025010, https://doi.org/10.1088/2631-8695/ad4cb4

David, E., Niculescu, V.C., 2021. Volatile Organic Compounds (VOCs) as Environmental Pollutants: Occurrence and Mitigation Using Nanomaterials. *International Journal of Environmental Research and Public Health,* Volume 18(24), p. 13147, https://doi.org/10.3390/ijerph182413147

Dunky, M., 2017. *Adhesives in the Wood Industry.* In: Pizzi, A., Mittal, K.L. (Eds.), Handbook of Adhesive Technology, 3rd Edition. Taylor and Francis, https://doi.org/10.1201/9781315120942

Elgharbawy, A.S., El Demerdash, A.G.M., Sadik, W.A., Kasaby, M.A., Lotfy, A.H., Osman, A.I., 2024. Synthetic Degradable Polyvinyl Alcohol Polymer and Its Blends with Starch and Cellulose—A Comprehensive Overview. *Polymers,* Volume 16(10), p. 1356, https://doi.org/10.3390/polym16101356

Farshi, P., Mirmohammadali, S.N., Rajpurohit, B., Smith, J.S., Li, Y., 2024. Pea Protein and Starch: Functional Properties and Applications in Edible Films. *Journal of Agriculture and Food Research,* Volume 15, p. 100927, https://doi.org/10.1016/j.jafr.2023.100927

Frihart, C.R., 2011. Wood Adhesives: Vital for Producing Most Wood Products. *Forest Products Journal,* Volume 61(1), pp. 4–12, https://doi.org/10.13073/0015-7473-61.1.4

Gaaz, T.S., Sulong, A.B., Akhtar, M.N., Kadhum, A.A.H., Mohamad, A.B., Al-Amiery, A.A., 2015. Properties and Applications of Polyvinyl Alcohol, Halloysite Nanotubes and Their Nanocomposites. Molecules, Volume 20(12), pp. 22833–22847, https://doi.org/10.3390/molecules201219884

Gonçalves, D., Bordado, J.M., Marques, A.C., dos Santos, R.G., 2021. Non-Formaldehyde, Bio-Based Adhesives for Use in Wood-Based Panel Manufacturing Industry—A Review. Polymers, Volume 13(23), p. 4086, https://doi.org/10.3390/polym13234086

Imanzadeh, G.H., Zamanloo, M.R., Mansoori, Y., Khodayari, A., 2007. Aqueous Media Oxidation of Alcohols with Ammonium Persulfate. Chinese Journal of Chemistry, Volume 25(6), pp. 836–838, https://doi.org/10.1002/cjoc.200790153

Jaffe, L., Rosenblum, M., 1990. Poly (Vinyl Alcohol) for Adhesives. Handbook of Adhesives, pp. 401–407, https://doi.org/10.1007/978-1-4613-0671-9_22

Kaboorani, A., Riedl, B., 2011. Effects of Adding Nano-Clay on Performance of Polyvinyl Acetate (PVA) as a Wood Adhesive. *Composites Part A: Applied Science and Manufacturing,* Volume 42(8), pp. 1031–1039, https://doi.org/10.1016/j.compositesa.2011.04.007

Kumar, R.N., Pizzi, A., 2019. Adhesives for Wood and Lignocellulosic Materials. In: Adhesion and Adhesives: Fundamental and Applied Aspects. Wiley-Scrivener, https://doi.org/10.1002/9781119605584

Li, Z., Wang, J., Cheng, L., Gu, Z., Hong, Y., Kowalczyk, A., 2014. Improving the Performance of Starch-Based Wood Adhesive by Using Sodium Dodecyl Sulfate. *Carbohydrate Polymers,* Volume 99, pp. 579–583, https://doi.org/10.1016/j.carbpol.2013.08.062

Łebkowska, M., Załęska-Radziwiłł, M., Tabernacka, A., 2017. Adhesives Based on Formaldehyde – Environmental Problems. *BioTechnologia,* Volume 1(1), pp. 53–65, https://doi.org/10.5114/bta.2017.66617

Monroy, Y., Rivero, S., García, M.A., 2019. Sustainable Panels Design Based on Modified Cassava Starch Bioadhesives and Wood Processing Byproducts. *Industrial Crops and Products,* Volume 137(2), pp. 171–179, https://doi.org/10.1016/j.indcrop.2019.04.062

Nguyen, Y.T.H., Ly, T.B., Bui, B.A.T., Le, P.K., 2023. Biodegradable Polyvinyl Alcohol Composite Film Reinforced by Crystalline Nanocellulose from Rice Straw. *Chemical Engineering Transactions,* Volume 106, p. 83, https://doi.org/10.3303/CET23106083

Ozemoya, P.O., Ajisegiri, E., Idah, P.A., 2007. Production of Adhesives from Cassava Starch. *Leonardo Electronic Journal of Practices and Technologies,* Volume 10 (1), pp. 93–98, https://doi.org/10.1088/1742-6596/1378/3/032079

Pizzi, A., Mittal, K.L., 2003. Urea-Formaldehyde Adhesives. In: Handbook of Adhesive Technology, 2nd Edition. Marcel Dekker, Inc., https://doi.org/10.1201/9780203912225

Raydan, N.D.V., Leroyer, L., Charrier, B., Robles, E., 2021. Recent Advances on the Development of Protein-Based Adhesives for Wood Composite Materials—A Review. *Molecules,* Volume 26(24), p. 7617, https://doi.org/10.3390/molecules26247617

Reddy, N., Yang, Y., 2010. Citric Acid Cross-Linking of Starch Films. Food Chemistry, Volume 118(3), pp. 702–711, https://doi.org/10.1016/j.foodchem.2009.05.050

Rochardjo, H.S., Fatkhurrohman, Kusumaatmaja, A., Yudhanto, F., 2021. Fabrication of Nanofiltration Membrane Based on Polyvinyl Alcohol Nanofibers Reinforced with Cellulose Nanocrystal Using Electrospinning Techniques. *International Journal of Technology,* Volume 12(2), pp. 329–338, https://doi.org/10.14716/ijtech.v12i2.4173

Sridach, W., Jonjankiat, S., Wittaya, T., 2013. Effect of Citric Acid, PVOH, and Starch Ratio on the Properties of Cross-Linked Poly(Vinyl Alcohol)/Starch Adhesives. *Journal of Adhesion Science and Technology,* Volume 27(15), pp. 1727–1738, https://doi.org/10.1080/01694243.2012.753394

Wang, Y., Xiong, H., Wang, Z., Zia-ud-Din, Chen, L., 2017. Effects of Different Durations of Acid Hydrolysis on the Properties of Starch-Based Wood Adhesive. *International Journal of Biological Macromolecules,* Volume 103, pp. 819–828, https://doi.org/10.1016/j.ijbiomac.2017.05.102

Wang, Z., Li, Z., Gu, Z., Hong, Y., Cheng, L., 2012. Preparation, Characterization and Properties of Starch-Based Wood Adhesive. Carbohydrate Polymers, Volume 88(2), pp. 699–706, https://doi.org/10.1016/j.carbpol.2012.01.023

Xu, Y., Wu, Z., Li, A., Chen, N., Rao, J., Zeng, Q., 2024. Nanocellulose Composite Films in Food Packaging Materials: A Review. *Polymers,* Volume 16(3), p. 423, https://doi.org/10.3390/polym16030423

Yu, H., Fang, Q., Cao, Y., Liu, Z., 2016. Effect of HCl on Starch Structure and Properties of Starch-Based Wood Adhesives. *BioResources,* Volume 11(1), pp. 1721–1728, https://10.15376/biores.11.1.1721-1728

Zhang, Y., Ding, L., Gu, J., Tan, H., Zhu, L., 2015. Preparation and Properties of a Starch-Based Wood Adhesive with High Bonding Strength and Water Resistance. *Carbohydrate Polymers,* Volume 115, pp. 32–37, https://doi.org/10.1016/j.carbpol.2014.08.063