



# Eco-friendly bio-based adhesive for plywood from natural rubber latex (NRL)-blended isocyanate cross-linked starch

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Received: 19 February 2022 / Accepted: 9 May 2022 / Published online: 24 May 2022  
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## Abstract

Synthetic adhesives used in the production of plywood are a matter of concern because of the emission of carcinogenic gas formaldehyde, increased environmental pollution, and the depletion of fossil fuels. In this study, a bioadhesive composed of natural rubber latex (NRL) and rice starch was developed. However, rice starch has low moisture resistance, resulting in low adhesion. Thus, to enhance the effectiveness of NRL-blended rice starch-based bioadhesive, rice starch was cross-linked with polymeric 4,4"-diphenylmethane diisocyanate (pMDI) resin, which is an environment-friendly, formaldehyde free, and moisture resistant that is highly compatible with starch. The chemical interaction, viscosity, solid content, and gel time of the developed NRL-isocyanate cross-linked rice starch-based bioadhesive was investigated. The efficacy of the formulated bioadhesive was demonstrated by the fabrication of plywood. The presence of isocyanate and urethane capabilities in the bioadhesive formulations was confirmed by Fourier transform infrared spectroscopy (FTIR). The bioadhesive type Iso-A was discovered to have the highest viscosity of 8270 mPa.s, whereas Iso-B has the shortest gel time of 3.46 min and the highest solid content of 44%; the higher solid content accelerates the gel time. In terms of physical and mechanical properties of plywood, Iso-B has the lowest thickness swelling (TS) value of 13%, lowest water absorption (WA) value of 52% and shear strength value of 1.92 MPa, which corresponds to the ISO 12466–2-2007 standard requirements. Based on the results, NRL-blended isocyanate starch-based bioadhesive could be a good potential raw material for eco-friendly plywood industries with adequate accuracy.

**Keywords** pMDI · Cross-linker · Rice starch · NRL · Bioadhesive · Plywood

## Introduction

In recent years, wood panel industries have made a pledge to use materials from sustainable sources. Wood-based panels, like particleboard, plywood, and fibreboard, are among the most widely used in the furniture and decorative industries around the world (Zhang et al. 2019). The most common synthetic adhesives used in the manufacturing of wood-based panels are urea–formaldehyde (UF), phenol–formaldehyde (PF), and melamine-urea–formaldehyde (MUF). Furthermore, more than half of the total global

formaldehyde output is being used to manufacture UF, PF, and MUF adhesives for wood-based panels. However, several disadvantages related to the production of wood-based panels have been identified, primarily because of the use of synthetic adhesives during the manufacturing process. The International Agency for Research on Cancer has identified formaldehyde as a carcinogenic substance for humans (Popović et al. 2020). Thus, the impact of using formaldehyde as a coupling agent in the wood composite industry, particularly in indoor applications, as well as the unreliability of fossil fuel resources and environmental concerns, have provided the industrial and scientific interest in transitioning from formaldehyde-based synthetic adhesives to bio-based adhesives to produce eco-friendly products (Valyova et al. 2017). Bio-based adhesives are found naturally and mainly organic substances that can take on a range of configurations and distinct features. For these distinguishing characteristics, bioadhesives are being used in a wide range of industrial applications, especially in the field of wood adhesives.

Responsible Editor: George Z. Kyzas

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There are several bioadhesives based on soy proteins, starch, lignin, and tannin for use in the fabrication of wood-based panels. Plant-based proteins, particularly soy protein, are extremely promising biomacromolecules that have been effectively used in the commercial fabrication of composite wood adhesives (Li et al. 2017). Following cellulose, lignin is nature's second-largest polymer and has emerged as a promising option for the development of adhesives for wood-based panels (Antov et al. 2020). Tannin from leaves, bark, and wood are also a promising material for the development of bioadhesives for wood-based panels due to their phenolic structure and the existence of aromatic rings of resorcinol, which make tannins highly reactive molecules (Chen et al. 2021). Among the numerous bio-based adhesives, the use of natural rubber latex (NRL) as a wood adhesive has been reported for many years, and this raw material has been referenced, but it has not been thoroughly studied (Hermiati et al. 2009). NRL derived from *Hevea brasiliensis* is a promising natural adhesive with good moisture and mould resistance and no toxicity (Thuraisingam et al. 2016). The incorporation of bio-based polymers as a blending agent has been documented to enhance the mechanical properties of NRL in terms of modulus, lowered elongation of failure, elevated hardness, and tear resistance (Kim et al. 2016). As an alternative to improve the adhesion properties of NRL, the incorporation of bio-based polymers has been approached. The development of starch as a rubber-blending agent has drawn interest on larger scales in recent years (Juliet et al. 2018; Soubam and Gupta 2021). It is mainly due to its low cost, and biodegradable plant product that has been used as adhesives for wood-based panels (Gu et al. 2019). However, starch must be modified before being incorporated into NRL because starch chains contain numerous soluble hydroxyl groups that easily associate with water molecules to form hydrogen bonds, leading to poor water resistance and steady drying (Sun et al. 2018). Regarding the modification of starch, cross-linking is one of the broad-based kinds of chemical modification of rice starch. To improve the properties of starch, polymeric 4,4"-pMDI was used as a cross-linker because it can easily react with a wide range of other functional groups containing active hydrogen atoms, such as amino, hydroxyl, carboxyl, carbamate, carbonamide, etc. Furthermore, due to the emission of carcinogenic gas from synthetic adhesives, the use of isocyanate adhesive for the binding of wood-based panels has increased in the industries. The modification of starch polymers with isocyanate functional groups has received little attention. Cross-linking starch with isocyanates, on the other hand, resulted in excellent thermal stability, bonding strength, and water resistance. It could be used as a substitute for petroleum-based counterparts in the approach of wood-based panel adhesives (Heinrich 2019; Hosseinpourpia et al. 2018; Qiao et al. 2015; Tan et al. 2011). There is no documentation that pMDI can cause

cancer in humans. According to experimental ecotoxicological records, its decomposition products are medium to low toxic to marine organisms and to land-based biodiversity such as plants and earthworms (Kapp 2005; U.S. Environmental Protection Agency 2011). The transition from synthetic adhesives to bioadhesives would add advantage to the biomass sectors and address the growing environmental concerns associated with synthetic adhesives. Thus, the present work focused on the development of bioadhesives, i.e. NRL-isocyanate cross-linked rice starch, via blending technique for plywood bonding. The compatibility and performance of bioadhesive formulations were investigated and compared. The characterisation of the bioadhesive involved FTIR, viscosity, solid content, and gel time. The physical and mechanical properties of plywood such as water absorption, thickness swelling, and shear strength were assessed.

## Materials and methods

### Materials

A rice starch and natural rubber latex (0.7 wt. % ammonia) were purchased from Gardener Global Enterprise, Kuantan. The extender, modification agent, and cross-linker of the rice starch, namely sodium hydroxide (48–50%), hydrochloric acid (32%), and 4,4"-pMDI (99.6%) were also purchased from Gardener Global Enterprise, Kuantan. The Meranti wood veneer was supplied by Mentakab Veneer & Plywood Sdn Bhd, Pahang, Malaysia.

### Methods

#### Modification of starch with a cross-linker

The starch solution was prepared by blending 50 g of rice starch powder in 100 mL of 1 M hydrochloric acid (HCl) at 300 rpm, 60 °C for 30 min. The pH of the starch solution was set to 8 by adding 1 M sodium hydroxide (NaOH), and the temperature was elevated to 90 °C. In this regard, the volume of NaOH to maintain the pH value of the starch solution has been recorded to be 80 mL. Similarly, after 30 min of gelatinisation, the temperature of the mixture was cooled to 60 °C, and 0.5 wt. % of isocyanate was added. Subsequently, the reaction temperature was increased to 80 °C for 30 min, and the starch solution was cooled to room temperature.

#### Bioadhesive preparation

To prepare a bioadhesive, the natural rubber latex (0.7 wt. % ammonia) and isocyanate crosslinked rice starch were mixed

into a beaker homogeneously based on the different weight proportions as shown in Table 1.

### Preparation of three-layer plywood

To assemble three-layer plywood panels, 1.5-mm-thick Meranti wood veneers were cut into 270 mm × 270 mm sheets. The Meranti veneer sheets were oven-dried at 50 °C for about 24 h before plywood assembling. Subsequently, a uniform amount of 20-g bioadhesive slurry was applied on the back and face of the veneer sheets manually by a roller brush. The glued veneer was then sandwiched between two uncoated veneers, with the grain orientations of two adjoining veneers perpendicular to each other, to form the three-layer plywood with aspects of 270 mm × 270 mm × 4.5 mm. The assembled plywood sample was kept for 2 min for the adhesive to penetrate and then subjected to a hot press at 120 °C temperature and 3 MPa pressure for 5 min. The plywood was conditioned at room temperature for at least 24 h after hot pressing before being tested.

### Characterisation of bioadhesive

The physical properties of bioadhesive were studied, including gel time, solid content, and viscosity. The gel time was determined by placing 100 g of the ready bioadhesive in a vial and dipping it in a steam water bath. The gel duration for a given adhesive specimen is the amount of time taken to form a gel at a given temperature. The findings were presented as the mean of three replicates. To determine solid content, the bioadhesives were oven-dried as per the ASTM D-4426–93 standard. The method applied to measure viscosity was according to ASTM D-1084–91. The viscosity of the bioadhesive was measured using a digital Brookfield viscometer at 25 °C with a rotor set at 60 rpm and was determined in the form of centipoise (Cp). Also, the functional groups of the adhesive was characterized by attenuated total reflection-Fourier-transform infrared spectroscopy (ATR-FTIR). For this purpose, the different bioadhesive formulations Iso-A, Iso-B, and Iso-C were recorded at ambient temperature in the range of 4000–400 cm<sup>-1</sup> wavelength on the PerkinElmer express spectrum version 1.03.02 with 4-cm A'1 resolution, averaging 5 scans per sample.

**Table 1** Different formulations of natural rubber latex-cross-linked rice starch

Formulation of bioadhesive	Wt. of NR latex (g)	Wt. of cross-linked rice starch (g)
Iso-A	5	15
Iso-B	10	10
Iso-C	15	5

### Investigation of plywood properties

The effect of different bioadhesive proportions on the physical and mechanical properties of plywood panels was investigated. Water absorption (WA) and thickness swelling (TS) of the panels were measured in accordance with ASTM D1037-12. At ambient temperature, five samples of each bioadhesive composition with sizes of 50 mm × 50 mm × 6 mm were soaked in distilled water. The water absorption was measured two times, at 2 h and 24 h. The thickness swelling was measured after 24 h immersed in distilled water. The sample's mass and width were measured using an electronic balance and a virtual calliper before and after they were removed from the water. A blotting paper was used to remove excess water from the specimens that had been removed from the water. WA and TS data are averages of five replicates. The WA was calculated using the equation below:

$$\text{Waterabsorption\%} = \frac{W2 - W1}{W1} \times 100$$

where W1 is the sample's weight before soaking and W2 is the sample's weight after soaking; the following equation was used to calculate the TS:

$$\text{Thicknessswelling\%} = \frac{T2 - T1}{T1} \times 100$$

where T1 denotes the thickness before immersion in water, and T2 denotes the thickness after immersion in water.

The ISO 12466–1-2007 standard was used to determine the shear strength of plywood panels. Ten plywood samples were cut to determine shear strength and then submerged in water at ambient temperature for 24 h to determine adhesion strength. The experiment was conducted using a Shimadzu UTM AG–X plug series at a crosshead rate of 5 mm/min. The average of ten samples was used to calculate the results. Adhesion strength was calculated by using the formula below:

$$\text{Shearstrength} = \frac{\text{Maximumforce}(F)}{\text{Surfacearea}(A)}$$

## Results

### Physical characterisation of bioadhesive

Viscosity is an important physical property that must be determined when selecting the best formulation of bioadhesive because it governs the adhesion strength and flow

behaviour of adhesives in the fabrication of plywood. In general, high viscosity adhesives may retain a significant amount of adhesive on the surface with little absorption into the pores; however, low-viscosity adhesives will easily penetrate deep into the wood through the veneer surface, leaving very few adhesives on the surface to bond the plywood. In either case, the bonding strength is low (Qi and Sun 2010). In this case, for adhesive applications, the solution must have great storage consistency, flowability, wettability ability, and viscosity, and the resulting adhesives bonding must be highly resistant to degradation under various conditions (He 2017). Starch particles surpass natural rubber latex-based particles in size and viscosity, with rice starch granules ranging in size from 3 to 5  $\mu\text{m}$  (Singh et al. 2016), whereas rubber latex particles have a size range from 0.08 to 2  $\mu\text{m}$  (Schmidt et al. 2010). Thus, starch cross-linking was performed to ensure proper starch particle blending and diffusion into latex emulsion, which can increase the viscosity and consistency of starch (Din et al. 2020). The viscosity of isocyanate rice starch and NRL compounds was examined, as shown in Fig. 1. According to the GB/T2794-1995, a suitable viscosity for plywood adhesive ranges from 5000 to 25,000 mPa.s (Fan et al. 2016). The highest viscosity value was found in Iso-A of 8270 mPa.s, and with increasing starch concentration, the effect of cross-linked rice starch is visible in the bioadhesive formulation Iso-A. This is due to the inclusion of rigid starch, which severely restricts the motion of natural textured rubber chains (Jang et al. 2013). Although the solid content of bioadhesives was not significantly different, for all the formulations Iso-A, Iso-B, and Iso-C, the solid content fell within 28 to 44%, which is within the minimum solid content requirement for industrial-based adhesive application (Islam et al. 2021b).

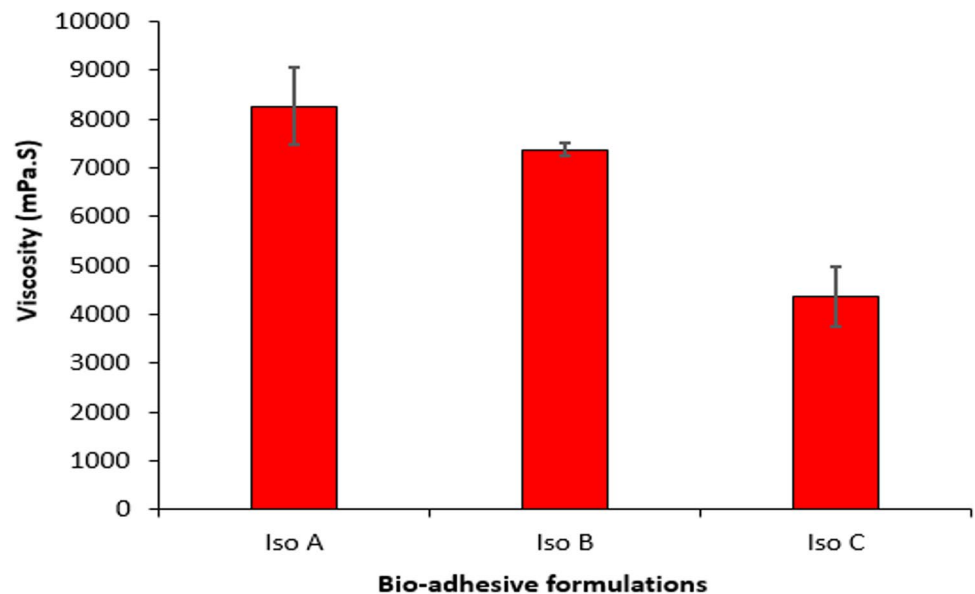
**Table 2** Solid content % of bioadhesives

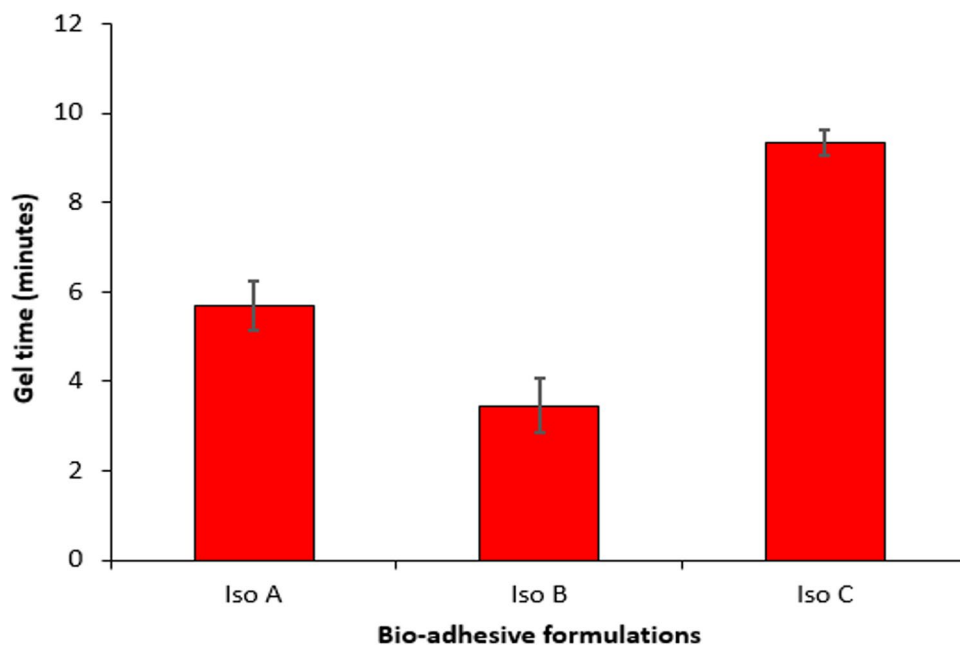
Bio-adhesive formulations	Solid content (%)
Iso-A	36 $\pm$ 3.2
Iso-B	44 $\pm$ 2
Iso-C	28 $\pm$ 4.8

Data are averages of three replicates  $\pm$  SD

Based on Table 2, it is obvious that the increase of isocyanate rice starch in terms of weight into latex-based adhesive emulsion created a subsequent incline in the solid content of the overall system. It is crucial for the development of bioadhesives with an accurate range of solid content value to improve the adhesion strength and its corresponding performance on plywood. Hence, the solid content cannot be too high as it can lead to a highly viscous solution with restricted flowability and difficulty to be applied to wood veneers. On the other hand, low solid content will lead to a low viscous solution that failed to disperse perfectly within the adherents and resulted in low drying time (Imam et al. 2001). Gel time can be technically defined as the duration taken for a resin to become more viscous and lose its liquidity and flowability when heated in the presence of a specific catalyst or hardener. The solid content value is the most key factor affecting its gel time further implying that there should be a connection between these two adhesives properties. The lowest gel time was seen in Iso-B with 3.46 min. Consequently, this discussion explained the reduction of gel time displayed by adhesive formulations with increasing solid content as depicted in Fig. 2 below. The adhesives with lower solid content values had lower concentrations, indicating the presence of more water in the adhesive system, which leads to a

**Fig. 1** Viscosity of bioadhesives



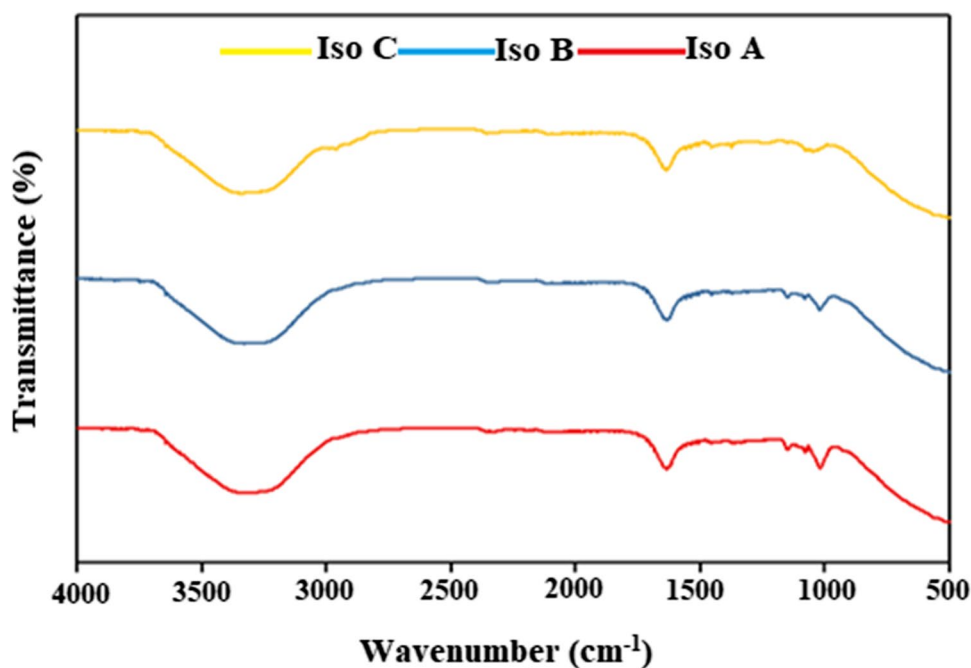
**Fig. 2** Gel time of bioadhesives

longer gel time. Thus, the curing reaction was diluted, and excessive moisture content acted as an energy barrier for the complete curing of adhesive (Gadhve et al. 2017).

### ATR Fourier transform infrared analysis

The absorption peak and functional groups of formulated bio-adhesives Iso-A, Iso-B, and Iso-C were monitored using ATR FTIR spectroscopy as shown in Fig. 3 and Table 3. The OH stretching absorption peak at  $3354\text{ cm}^{-1}$

was caused by the formation of a hydrogen bond from the affiliation of hydroxyl and carboxyl groups (Wang et al. 2015). The absorption peak between  $1000$  and  $1200\text{ cm}^{-1}$  was due to the polysaccharide skeleton's  $-\text{CO}$  stretching (Valodkar and Thakore 2010). On the other hand, the absorption peaks between  $1375$  and  $1448\text{ cm}^{-1}$  represent  $\text{N}=\text{O}$  bending of methane caused by the accumulation of ammonia-containing natural rubber latex to starch (Mohd Makhtar et al. 2013). The infrared absorption peak at  $2138\text{ cm}^{-1}$ , which can be contributed to  $-\text{NCO}$  groups,

**Fig. 3** IR spectra of different formulations of isocyanate starch-NRL-based bioadhesive

**Table 3** FTIR wavenumbers ( $\text{cm}^{-1}$ ) of isocyanate starch-NRL-based bioadhesive

Chemical assignment	Iso-A	Iso-B	Iso-C
OH	3354	3352.37	3334.27
–CO	1000–1200	1000–1200	1000–1200
N = O bending of methane	1375–1448	1375–1448	1375–1448
–NCO	2268	2344	2138
–CO rise in absorbance	1650–1750	1650–1750	1650–1750
NH bending amino and C–N stretch	1530	1533	1529

signifies that the cross-linking of starch with isocyanate was successful (Girouard et al. 2016). Furthermore, the existence of urethane bonds between isocyanate and starch was confirmed by a rise in the absorbance of –CO groups at  $1650\text{--}1750\text{ cm}^{-1}$ , as well as the bending vibration of –NH and the stretching vibration of C–N at  $1530\text{ cm}^{-1}$  (Hosseinpourpia et al. 2018).

### Physical properties of plywood

One of the most critical issues for the intermittent use of plywood in wet conditions is dimensional stability. The effect of the WA and TS properties of Meranti plywood bonded with different bioadhesive formulations Iso-A, Iso-B, and Iso-C was investigated. The results of WA samples were measured after two intervals of water immersion as shown in Table 4. After 2 h of water immersion, the average changes of WA samples bonded with Iso-A, Iso-B, and Iso-C were 46%, 44%, and 57%, respectively. The lowest and highest WA after 24-h water immersion was seen in samples bonded with Iso-B (54%) and Iso-C (72%), respectively. The results confirmed an equal amount of starch-NRL-based components in wood composites resulted in a lower water uptake capacity, lending support to the discovery of Agnantopoulou et al. (2012). Furthermore, the average TS value after 24 h of immersion in water ranged from 13 to 20%. As shown in Fig. 4, the sample bonded with Iso-B had the lowest TS (13%), whereas the highest TS was seen in Iso-C (20%). However, the TS value

**Table 4** The WA of plywood

Bioadhesive formulation	Water absorption (WA) 2 h (%)	Water absorption (WA) 24 h (%)
Iso-A	$46 \pm 0.4$	$58 \pm 0.7$
Iso-B	$44 \pm 0.1$	$52 \pm 1.2$
Iso-C	$59 \pm 0.5$	$72 \pm 0.8$

Data are averages of five replicates  $\pm$  SD

of Iso-A (17%) was not significantly different from Iso-C. Thickness swelling value is highly related to fibre bonding, where a lower TS value resembled good bonding between fibres (Izani et al. 2013). The results indicated that the dimensional stability of samples was improved after starch was added to the bioadhesive formulations. The linking of starch molecules and OH groups accelerate particle bonding while slowing moisture absorption in the plywood matrix. This explains why Meranti plywood panels fabricated with a bioadhesive containing an equal amount of NRL and isocyanate cross-linked rice starch have lower WA, TS, and less impact on dimensional stability.

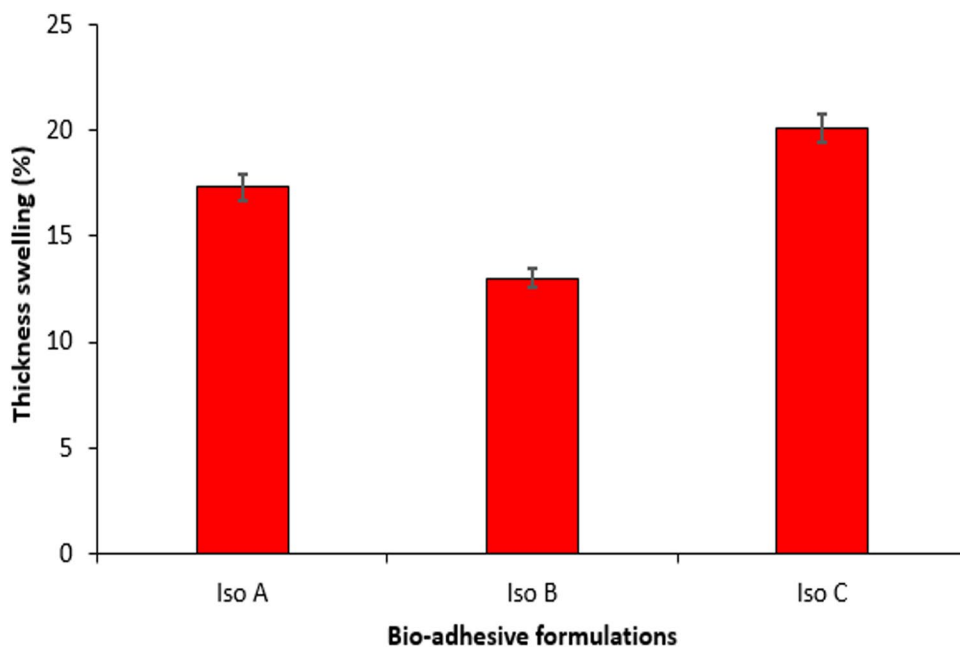
### Mechanical properties of plywood

The shear strength test is mainly used to assess the efficiency of adhesives in plywood. Figure 5 depicts the shear strength of plywood bonded with various formulations of isocyanate rice starch and NRL. The shear strength value rises as the amount of cross-linked rice starch in the bioadhesive increases. The plywood bonded with formulations Iso-B yielded a high shear strength value of 1.92 MPa, respectively, and the lower shear strength value of 1.11 MPa was seen in plywood bonded with formulation Iso-C. The findings were like those of (Islam et al. 2021a, b) but contradicted those of (Akbari et al. 2014). This effect is believed to be caused by starch's affinity for forming strong adhesion, as starch has good adhesion and polymerisation properties (Kiatkamjornwong et al. 2001; Shey et al. 2006). Besides that, as the starch intensity rises, the hydrogen bonding between the interparticle enhances, boosting the mechanical strength of the board. However, there was no substantial difference in shear strength values between the various bioadhesive formulations, the shear strength values obtained corresponded to the ASTM 12,466–2-2007 standard requirements, which require a value greater than 1 MPa. From Fig. 5, one can conclude that the best bioadhesive formulation was Iso-B based on the shear strength value.

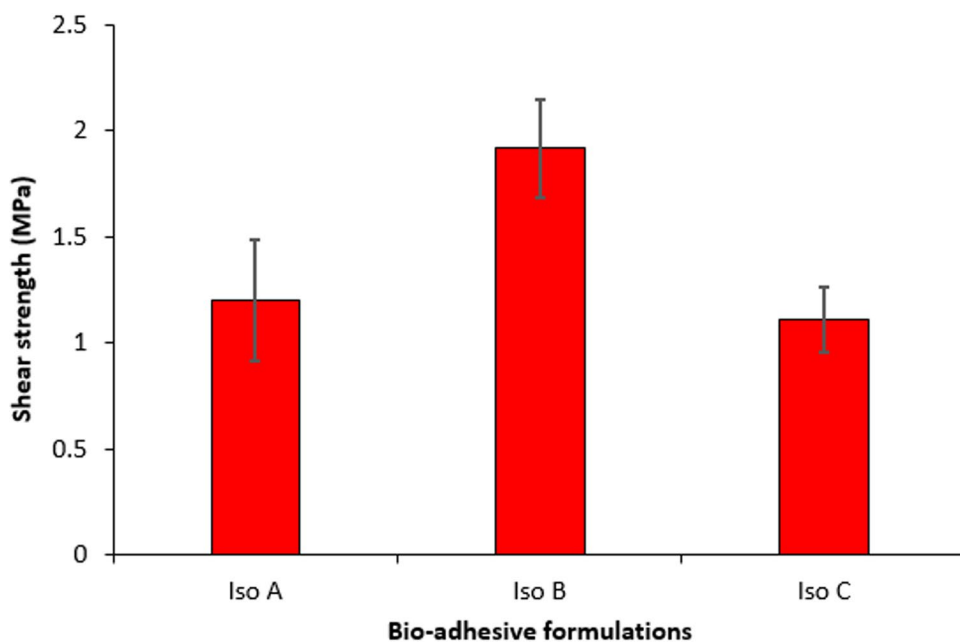
### Conclusions

Rice starch was successfully modified by polymeric 4,4"-pMDI resin to reduce its hydrophilicity and enhance its compatibility with NRL. The bioadhesive Iso-A had the highest viscosity of 8270 mPa.s., and the bioadhesive type Iso-B has the shortest gel time of 3.46 min and the highest solid content of 44%. ATR FTIR confirmed the presence of isocyanate and urethane functionalities in starch and NRL macromolecules, and thus the hypothesis was approved. The TS (13%) and WA (52%) of plywood bonded with formulation Iso-B demonstrated a remarkable improvement in dimensional stability due to the lower water uptake capacity, this effect is

**Fig. 4** Thickness swelling (%) of plyboard



**Fig. 5** Shear strength of bioadhesives



believed to be caused by a starch affinity for forming good adhesion when blended with NRL. Despite this, the shear strength of 1.92 MPa was found to be sufficient to replace the synthetic adhesives by exceeding the standard requirements, where the value must be greater than 1 MPa. As a result, it is possible to conclude that isocyanate cross-linked starch can improve the properties of NRL and could be a good potential raw material to use in plywood adhesive with adequate accuracy, contributing to the complete eradication of hazardous synthetic adhesives application.

**Author contribution** Triveni Soubam: conceptualization; formal analysis; investigation; methodology; original draft; visualization; review; and editing. Arun Gupta: supervision; review; and editing. Saidatul Shima Jamari: review and editing.

**Funding** The author would like to thank the Faculty of Chemical & Process Engineering Technology, Universiti Malaysia Pahang for the financial support through FRGS/1/2017/TK05/UMP/02/1, RDU170132 in conducting the research work.

**Data availability** The raw data supporting the conclusions of this article will be made available without restriction by the authors.

## Declarations

**Ethics approval** Not applicable.

**Consent to participate** The authors declare that they agree to publish this paper in Environmental Science and Pollution Research.

**Consent for publication** The authors declare that they agree with the publication of this paper in this journal.

**Competing interests** The authors declare no competing interests.

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