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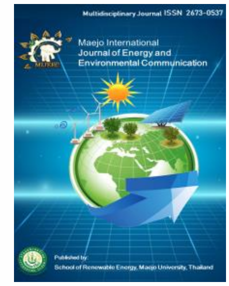

ARTICLE

Eco-friendly natural rubber latex and modified starch-based adhesive for wood-based panels application- A review

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A B S T R A C T

The use of formaldehyde-based synthetic adhesives for the development of wood-based panels has increased rapidly in the industry. Synthetic adhesives are regarded as a threat to human health and a pollutant because they emit formaldehyde's carcinogenic gas. The breakthrough has prompted to seek for a long-term solution to the formaldehyde threat with natural resources. There has been a significant rise in bio-based adhesive technology and development in the wood-based panel industry. The current review article is intended to present the potentials and the drawbacks of the development of bio-adhesives from sustainable resources such as natural rubber latex (NRL) and starch. This review also discussed the chemical modification and crosslinkers of starch to improve water resistance and adhesion properties. Moreover, this article discusses the compatibility of modified rice starch and NRL for their conclusive applications as wood-based panels adhesive. The findings suggested that bio-based adhesives could replace more synthetic-based adhesives with comparable performance in the near future.

1. Introduction

Wood-based panels adhesives are synthetic polymers intended to interact physically, chemically, or both with the surface of the wood in such a way that constraints are transmitted between bonded members, ideally without adhesive rupture or disconnection from the wood. Also, adhesives play a significant key role in determining the strength of the wood-based panels. Today, most of the adhesives used in the industries for bonding wood panels are synthetic adhesives, specifically phenol-formaldehyde, urea-formaldehyde and melamine-formaldehyde, which pose a risk to human health and polluted air quality by emitting carcinogenic gas, formaldehyde, during the industrial production and usage of wood-based panels (Moubarik et al., 2013). Formaldehyde has been classified as the source of nasopharyngeal cancer by the International Agency for Research on Cancer (IARC). Hence, California Air Resource Board (CARB) approved a regulation standard to lessen the formaldehyde emission from the wood-based panel's products. Therefore, renewable biopolymers such as starch (Li et al., 2014) and natural rubber latex (Hermiati et al., 2006) are developing sustainable-based adhesives to fasten the composite wood panels. The application of NRL as a wood-based panel adhesive had reported many years ago due to its high strength, flexibility and elasticity (Weeraratne et al., 1972). However, natural rubber latex-based adhesives' physical and mechanical strength is low compared to synthetic adhesives due to their less heat resistance and moderate water resistance (Marra, 1992).

To promote the adhesion properties of NRL, the incorporation of the bio-based polymer as filler had been found to

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improve the mechanical strength of NRL (Kim et al., 2016). As starch possess adhesion property naturally, reinforcing starch as an NRL filler has drawn concern on a large scale because of its abundance, inexpensiveness, and biodegradability (Liu et al., 2008). The starch can be found in roots, seeds and stalks of staple crops such as wheat, rice, potato, corn etc. Though, the molecular structure of starch must be chemically modified or crosslinked with synthetic polymers to enhance its hydrophilicity nature before incorporation into NRL to generate an excellent efficiency of wood adhesive (Lei et al., 2014, Tan et al., 2011). The inclusion of modified starch into NRL had improved the morphological and mechanical properties of the overall adhesive system.

The review focus on developing and applying the bio-adhesive based on natural rubber latex incorporated with modified rice starch for wood-based panels bonding.

2. Natural rubber latex and starch bio-based adhesive

Bio-based adhesives are natural adhesives derived from natural resources, such as starch, protein, lignin and tannin. It can provide an environmentally friendly and long-term substitute to the synthetic adhesives systems employed in the wood-based panel industry. The natural adhesive can reduce the formaldehyde threat and indoor air quality issues caused by the wood-based panels. Simultaneously, bio-based adhesives can help accelerate the wood-based panel industry more durable and reducing its reliance on fossil fuels. However, the complete replacement of synthetic adhesives with bio-based adhesives remains a significant challenge, owing to their relatively poor water resistance and large natural varieties due to various growing conditions. There are successful solutions to these drawbacks and discuss them in the present review article.

2.1. Natural rubber latex as bio-adhesive

Natural rubber latex is a biopolymer of cis-1,4-polyisoprene. It is executed from the rubber tree scientifically named Hevea braziliensis. This biopolymer is well known for its impressive properties that even synthetic rubber latex could not win over as it is highly flexible at low temperature, high elasticity, resistant against corrosion, impact and abrasion, provides greater adhesion strength due to the impermeability nature, good insulator and heat dispersing agent (Rippel and Galemback, 2009). Natural rubber latex is mould and moisture resistant with no toxicity that boosts green adhesive formulations' application (Nasir et al., 2013). The particleboard formed with NRL based adhesive had enhanced the mechanical properties of the board (Nakanishi et al., 2018). However, Hermiati et al. (2013) stated that the application of virgin NRL alone for the development of bio-adhesive is not advisable. The biopolymer must be chemically modified or blend with conventional adhesives to enhance the adhesion strength. Bras and Piccini (1951) were the first to report the improvement in adhesion property when the latex is blended directly with synthetic resin. The performance of NRL resin blend adhesive had been studied on the different types of timber species and found that bond strength of NRL based adhesives was increased significantly when blended with urea-formaldehyde and phenol-formaldehyde resins. When NRL is a graft with diacetone acrylamide (DAAM) using adipic acid dihydrazide (ADH) as the crosslinking agent, the lap shear strength is improved considerably with the content of DAAM in the latex adhesives (Thongnuanchan, et al., 2007). Thus, utilization of natural rubber latex as an adhesive material is only practicable upon chemical modification or incorporation with bio-polymers.

Table 1 Chemical composition of natural rubber latex (Baker and Fulton (1997).

Aspects	Details
pH	6.5 to 7.0
Dry rubber content	30 to 45%
Non-rubber content	3-5%
Aqueous serum	53%
Molecular weight	105 to 107

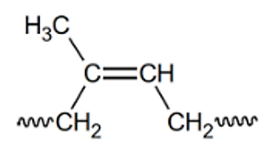

Figure 1 Structure of cis-1,4-polyisoprene *Hevea brasiliensis* (Miller 2018).

2.2. Starch as bio-adhesive

Starch is a white substance widely found in plant tissues, roots and obtained from rice, corn, wheat, tapioca and potatoes, which are insoluble in cold water or alcohol and one of the inexpensive, biodegradable and sustainable polymers (Gadhave et al., 2017a). It is a mixture of two polysaccharide fractions- amylose and amylopectin- both of distinct shapes and sizes. The proportion of these two polysaccharides differ on the genesis of the starch. Starch is a favourable material for the evolution of bio-adhesives due to its good adherence, inexpensive, easy accessibility and excellent film formation properties (Tan et al., 2011 and Onusseit, H. 1992). A starch from rice, wheat, and potato has been used in manufacturing wood-based panels. However, the adhesive properties of starch vary with its genesis, and the adhesion strength of starch is not durable to fasten wooden products (Salleh et al., 2015).

Table 2 Amylose and amylopectin content of starches (Gadhave et al. 2017a).

Starch type	Amylose content (%)	Amylopectin content (%)
Dent corn	25	75
Waxy corn	<1	>99
Tapioca	17	83
Potato	20	80
High-amylose corn	55 - 70 (or higher)	45 - 30 (or lower)
Wheat	25	75
Rice	19	81

Due to starch's high viscosity, the entanglement of molecular weight macromolecules makes it hard to use them for industrial applications. The viscosity of starch can be reduced by decreasing the number of entanglements per chain by adding molecules to swell the polymer network. Hence, starch needs to be chemically modified or crosslinked to utilize in the wood industry (Moubarik et., 2010).

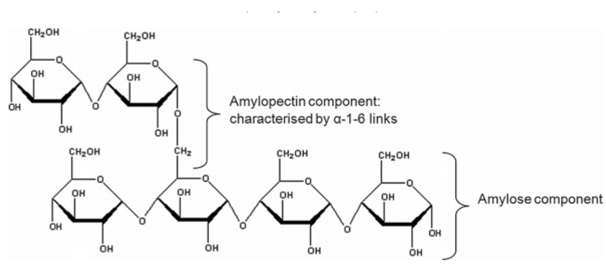

Figure 2 Starch co-polymer with amylose and amylopectin component (Masina et al., 2017).

2.3. Starch as filler for natural rubber latex-based adhesive

Earlier, starch has been utilized as a main component for adhesive development and as a bio-based filler for many elastomeric components, either for composites or adhesives development. From the mechanical point of view, the use of native starch as a reinforcing filler for rubber-based composites had been investigated long ago. Infusion of native starch into rubber latex matrix is the reason behind the weakening of mechanical strength of composite wood due to incapable of homogeneous mixing with the base component. This is due to the hydrophilicity of starch granules that led to the incompatibility of the non-polar, hydrophobic nature of the rubber matrix. When (Kiing et al., 2013) employed native sago starch as filler to reinforce natural rubber latex; it led to the depletion of mechanical properties of composite panels. Progressively, to overcome the incompatibility and impact caused by the incorporation of starch molecules in natural rubber particles, (Wu et al., 2004) employed a process to achieve uniform starch in NRL matrix dispersion called the latex compounding method. The starch granules were put for gelatinization at temperature 90°C, and the gelatinized starch undergoes for compounding with NRL followed by immediate coagulation using calcium chloride. Based on the latex compounding theory, the size of starch particles was shrunk considerably, and this novel method enhanced the starch dispersion. The rubber-starch based adhesives prepared through the latex compounding method exhibit optimized performance compared to the synthetic adhesives. Consequently, to improve the reinforcing ability of starch on the NRL matrix, various surface treatment methods have been proposed and investigated by researchers.

2.4. Chemical modification and crosslinking of starch-based adhesive

Chemical modification or hydrolysis is a chemical process in which starch is added to the aqueous solution of hydrochloric acid or sulphuric acid and in aqueous sodium hydroxide or potassium hydroxide at a definite temperature (Okunlola and Akingbala, 2013). In an acid modification, starch's water resistance and adhesion properties are altered without disturbing its granular structure to enhance the solubility and gel strength. Generally, starch is exposed to mineral acid below the gelatinization temperature (Pratiwi et al., 2018). The modification of starch increases the short linear chain (amylose), which favour enhanced retrogradation (Dundar and Gocmen, 2013). The process of acidic hydrolysis forms a crystalline structure of starch with an adequate length of amylose for the desirable formation of highly resistant and active starch molecules (Ferrini et al., 2008 and Ozturk et al., 2011). It also reduces the viscosity of the starch, which is easier for incorporation with other biopolymers (Odeku et al., 2009).

Similarly, the surface treatment of starch with sodium hydroxidsuccessfully removing the subsidiaries components found in starch granules that may affect its dispersion with other bio-polymers (Uthumporn et al., 2012). Alkaline hydrolysis favours the uncoiling, separation of double helices and obstruction of the crystalline structure of starch. Thus, alkaline hydrolysis on starch expands starch's amorphous area and enhances the corresponding reaction with other biopolymers. Also, the bonding strength and water resistance of starch-based adhesive can be synthesized via the graft copolymerization of oxidized starch with an olefin monomer and silane coupling agent (Zhang et al., 2015). A nature-friendly starch adhesive with zero formaldehyde-emission was developed from urea and oxidized starch, where the adhesive shows an exceptional comprehensive performance and the stability of adhesive was enhanced by adding titanium dioxide nanoparticles (Zhao et al., 2018).

The starch crosslinked with sodium borate, epoxy chloropropane, hexamethoxymethylmelamine and isocyanates manifest high glueing properties and water resistance (Qiao et al., 2015). An epoxy resin is a general commercial adhesive that can be used as a crosslinker of starch and other bio-based adhesives (Nie et al., 2013). At the same time, epoxy resins are mainly tested for veneer glueing, imparting an adequate shear strength in arid and wet cases. The starch modified with isocyanate enhanced plyboard bonding properties and moisture resistance and can efficiently react with several functional groups, such as amino, carboxyl and hydroxyl groups with no formaldehyde emissions (Gu et al., 2010; Tan et al., 2011). In Indonesia, the application of isocyanate adhesive for the bonding of composite wood has been expanded due to the formaldehyde carcinogenic gas emission from the synthetic adhesive (Nuryawan and Alamsyah, 2017). A methyl methacrylate-grafted starch was modified with three different crosslinkers as dimethyloldihydroxyethyleneurea (DMDHEU), glutaraldehyde (G.A.) and N-methylol acrylamide (N.M.A.). It has been found that DMDHEU crosslinked composite exhibits minimum water absorption compared to G.A. and NMA-based crosslinked composites. , the interaction between crosslinked polymer and wood is exceptional in DMDHEU crosslinked composites (Baishya and Maji 2014). When starch-based adhesive was prepared using- Methacryloxypropyl trimethoxy silane (KH570) as a crosslinking agent, it improved the shear strength and promoted thermal stability. Furthermore, the KH570 enhanced the shear-thinning property and weakened the pseudoplastic behaviour of the adhesive (Chen et al., 2017). Therefore, to improve the incompatibility of starch on the NRL matrix, the starch must be chemically modified or blend with conventional adhesive/crosslinkers to overcome its limitation upon blending with NRL.

2.5. Natural rubber latex fuse modified starch-based adhesive for wood-based panels

Starch is widely used in binders, sizing materials, glues and paste. It also played a significant role as a bio-based filler for elastomeric components for composite wood adhesives. The ability of starch as filler for rubber-based adhesive had been looking over long ago from the mechanical point of study. However, the hydrophobicity of starch granules has led to the incompatibility of the non-polar, hydrophobic nature of rubber latex. Hence, to improve the reinforcing ability of starch on the NRL matrix, chemical modification of starch is viable to overcome its limitation upon blending with NRL (Wang et al. 2015).

Similarly, Misman et al. (2018) found that modification on either micro or nanostructure of starch effectively responds to the increment of the mechanical properties of NRL-based adhesives. Akbari et al. (2014) concluded that adding an equal amount of modified rice starch with NRL improved the thermal stability, which technically means starch can act as a perfect binder to NRL, and this combination can be an alternative to synthetic adhesives for the manufacturing of wood composites. The chemically modified starch filler enhanced the mechanical properties of NRL-based composites in terms of internal bonding and shear strength. The finding was attributed to expanding starch granular structures upon modification that enhanced the compatibility between hydrophobic NRL and hydrophilic starch (Qi et al., 2006).

3. Conclusion

Most of the adhesives used today in the wood composite industries depend on formaldehyde emitting sources. Therefore, there is a longing for formaldehyde-free and nature-friendly adhesives concerning the formaldehyde emissions from wood-based panels products. Subsequently, NRL and modified rice starch are perfect raw materials for manufacturing wood-based panels adhesives due to their tremendous economic, easy processing, and treatment. However, many technological voids must be overcome to bring this novel approach widely utilized in industries and catalyze the commercialization of the products. From the review, we can conclude that research on bio-adhesive can open a novel awareness into the design of environment-friendly and formaldehyde-free wood-based panels with high bonding strength and water resistance for various applications.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

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