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Hybrid Adhesive Made from Citric Acid and Sucrose for Particleboard Composed of Sengon (*Paraserianthes falcataria* (L.) *Nielsen*) Veener By-Products and Sorghum Stalk

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Abstract

Particleboard is a product of wood technology engineering made of wood particles and other lignocellulosic materials which are bonded with synthetic adhesives. As an information, utilization of natural adhesive for particleboard is still limited. This study aimed to determine the influence of hybrid adhesive made from citric acid and sucrose for particleboard properties composed of Sengon veener by-products and sorghum stalk. The hybrid adhesive content used was 15% of citric acid and 25% of sucrose that calculated from the dry weight of particles. The mixture ratio of hybrid adhesive for both citric acid and sucrose were 100:0, 75:25, 50:50, 25:75, and 0:100 (% w/w). The manufacture of particleboard was set on target density of 0.8 g/cm^3 under a press temperature of 200° C for 10 minutes. Mostly, the properties of particleboard were shown to be affected by the adhesive content and composition ratio between citric acid and sucrose and met the Japan Industrial Standard A 5908:2003. The value of modulus of elasticity, modulus of rupture, internal bond, and screw holding were ranged about 1.21-3.49 GPa, 12.77-20.24 MPa, 0.18-0.90 MPa and 167.71-321.27 N. Particleboard with 15% of citric acid content and the mixture ratio both citric acid and sucrose of 50:50 has the best physical and mechanical properties and met the requirement of Japan Industrial Standard A 5908:2003.

INTRODUCTION

Adhesive is one of the important factors in the manufacture of particleboard. This is in line with the fact that the type and properties of particleboard are largely determined by the type, composition, and other characteristics of the adhesive used. Sutiawan et al. (2020) stated that in the particle board production, adhesive as a binder between the particles so that it is able to produce particleboard that meet the standards.

Walker (2006) explained that adhesives occupy the largest portion of the cost of producing particleboard in the particleboard industry, which is around 30-34% of the total costs incurred. Particleboard is manufactured commercially using formaldehyde-based adhesives such as urea formaldehyde (UF) for the interior and phenol formaldehyde (PF) for the exterior panel applications. Formaldehyde-based adhesives are an option for the particleboard industry because they are relatively inexpensive and capable of producing particleboards with characteristics that meet the standards (Zhang et al., 2018). However, formaldehyde-based adhesives can cause health and environmental problems (Salem et al., 2013; Ferdosian et al., 2017).

Today, industrial adhesives are produced from petroleum based synthetic polymers, for example polyvinyl acetate, epoxy adhesive, phenol

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formaldehyde, urea formaldehyde, melamine formaldehyde and polyurethane rely on non-renewable and depleting petrochemical materials.

Concerns about the emission of formaldehyde and volatile organic compounds (VOC) from wood panel products, especially indoor applications such as particleboard, are currently the most important factors encouraging the reduction in the use of formaldehyde-based adhesives in the context of the toxic reduction program. Likewise, the synthetic adhesives industry encourages proper environmental and health safety for their products. Hazardous materials, VOC emissions, sustainability of raw materials, and final products as well as difficulties in recycling adhesives derived from petrochemical raw materials encourage the development of environmentally friendly adhesives based on natural materials as substitutes for synthetic adhesives.

The oil prices fluctuations and increases also stimulate commercial interest in the use of natural materials as a substitute for synthetic adhesives (Sarigiannis et al., 2011; Patel et al., 2012, Navarette et al., 2013, Hemmila et al., 2017). Several non-formaldehyde natural materials have been developed as particleboard adhesives, such as lignin (Ghaffar et al., 2014), lignosulfonates (Privas & Navard, 2013), protein (Khosravi et al., 2011) and citric acid (Umemura et al., 2015) with various advantages and disadvantages from each material. Therefore, studies on natural adhesives from renewable sources are still needed for the development of particleboard adhesives to replace synthetic adhesives.

The use of formaldehyde-based adhesives also has been carried out by Astari et al. (2018), Sutiawan (2018), and Wang et al. (2018). The use of these adhesives causes problems for the environment and consumer health (Zhang et al., 2018). Therefore, natural adhesives are needed to overcome this problem. Biomass that potential can be used as binder are starch (Cahyana, 2014), sucrose (Marwanto et al., 2018) and citric acid (Prasetiyo et al., 2018). The use of sucrose and citric acid as adhesives for particleboard that has been done produces better properties than with starch. However, Mardhatillah (2018) stated that boards made of Sengon and sorghum particles with citric acid adhesive have low screw holding power value. According to Umemura et al. (2013), the addition of 75% sucrose to a board using pine wood as a raw material with citric acid and sucrose adhesives resulted in better physical and mechanical properties.

The development of natural sources as bio-adhesive can be used as alone or combination between several biomasses. The product made from combination of several materials as bio-adhesive can be named hybrid adhesive. Eventhough citric acid and sucrose has been studied as particleboard adhesive, yet its application for particleboard made of Sengon veener by-products and sorghum stalk has never been conducted before. Therefore, the aim of this study is necessary to determine the relationship between the composition of citric acid and sucrose as hybrid adhesives on the quality of particleboard made of Sengon and sorghum bagasse.

MATERIALS AND METHODS

Materials

Particles consisting of Sengon veener by-products and sorghum stalk were obtained from PT. GPL Tasikmalaya and a research field at BRIN Cibinong, West Java, Indonesia. Technical citric acid was purchased from Weifang Ensign Industry Co. Ltd and pure sucrose was obtained from Kanto Chemical Co. Inc.

Methods

The hybrid adhesive content consisting of citric acid and sucrose used were 15% and 25% based on the air-dried particles weight. The mixture ratio of hybrid adhesive both citric acid and sucrose were 100:0, 75:25, 50:50, 25:75, and 0:100 (% w/w). The manufacture of particleboard was set on target density of 0.8 g/cm^3 under a press temperature of 200° C for 10 minutes and press pressure of 2.5 MPa. The dimensions of particleboards were 30 x 30 x 1 cm (length, width, thickness).

Sengon veener by-products and sorghum stalk were processed into particles by a chipper and knife-ring flaker machines. The particles were screened using a sieving machine to obtain particles of uniform sizes. Particles that passed through 4 mesh and retain at 14 mesh sizes were used in this research. The particles used in this research were dried at 100-105 °C for 12 h in a technical oven until 5% moisture content to be reached. Test samples were conditioned, prepared, and tested using Shimadzu Universal Testing Machine (UTM) 50 kN for mechanical properties and properties based on JIS A 5908:2003 standard for particleboard. The number of samples test for each test is fives repetition samples.

The data were tested of particleboards eventually manufactured using ANOVA and SPSS 25 method. The level significance was set for all statistical test at 0.05 such that probability values than 0.05 were taken as indicative of statistically significant difference.

RESULTS AND DISCUSSION

Physical Properties

The target density of particleboard from this research was 0.80 g/cm^3. Overall, the value of target density from particleboard with all compositions has reached. The range of real density value from particleboard was 0.83-0.86 g/cm^3 as shown in Figure 1. The achievement of particleboard density target as shown in Figure 1 is influenced by the compression of particleboard raw materials in the mold during the hot pressing process.

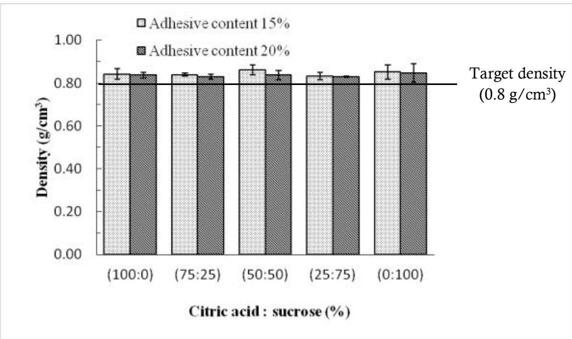

Figure 1. Real density of particleboard.

The JIS A 5908:2003 standard requires that the value of moisture content (MC) of particleboard is less than 13%. While, the results show that the MC value of particleboard ranged between 3.89 to 5.28% (Figure 2). The board with 20% adhesive content had an average MC value of 3.6% less than the board with 15% adhesive content. According to Fransiskus et al. (2015), the high mositure content value is caused by the high moisture content of raw materials. Not all of the water contained in the raw materials and adhesives can be removed during the hot pressing process.

On the other hand, the board from this research has an average MC value of 35%, lower than the study conducted by Mardhatillah (2018) which used Sengon wood and sorghum as raw materials with 20% citric acid adhesive on the composition of particles is same (50:50 % w/w). Widyorini and Nugraha (2015) stated that the MC of particleboard made of Sengon wood glued bonded citric acid and sucrose adhesives with concentration of 15% was 4.96%. These results indicate that the addition of sucrose to citric acid with a certain concentration can reduce the value of the MC of the particleboard. Umemura et al. (2015) argued that the reaction between citric acid and sucrose at the right temperature will form bonds from the hydroxyl group so that the board will be more stable and also keep water entering the particleboard.

The results of statistical analysis at a significant level of 5% showed that the composition of the adhesive and both interaction significantly affected the MC of particleboard. The boards that used the adhesive content of 25% and 100% sucrose had a different effect compared to other compositions, but both had the same effect. The addition of 25% sucrose at adhesive content 15% had a different effect compared to 20%, but had the same effect in the addition of 100% sucrose at 15 and 20% adhesive contents. According to Fransiskus (2015), the high MC value is caused by the high MC of raw materials. Not all of the water contained in the raw materials and adhesives can be removed by the hot compression process.

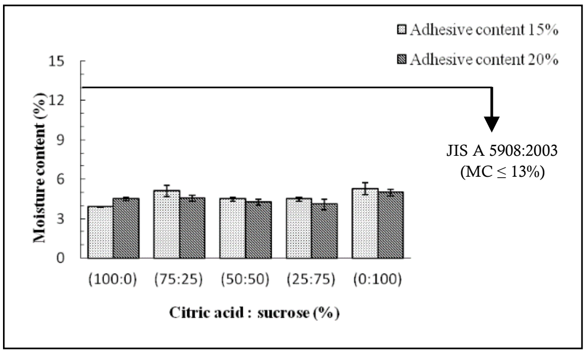

Figure 2. Moisture content value of particleboard.

The results showed that thickness swelling (TS) values are ranged from 6.19 to 29.01% (Figure 3). The JIS A 5908:2003 standard requires the TS value to be less than 12%. The TS increased in line with the addition of more sucrose. It means that particle is more compatible with citric acid adhesives than sucrose. Overall, the TS value of most boards does meet the standard. This is in accordance with the research conducted by Kusumah et al. (2017) regarding particleboard made of sorghum with the same adhesive composition and 20% adhesive content, which stated that the TS value are increased in line with the addition of higher concentrations of sucrose.

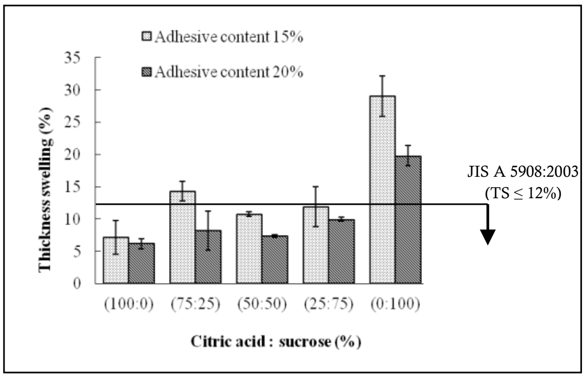

Figure 3. Thickness swelling value of particleboard.

The board with adhesive content of 20% had an average TS value of 13%, lower than the board with adhesive content of 15%. The addition of 50% sucrose to the board can increase the TS value until 19%. The boards using 100% citric acid in this study had an average TS of 11%, lower than boards made of Sengon and sorghum in the same composition and the same adhesive content (Mardhatillah, 2018).

Statistical analysis showed that the composition of adhesive, the content of adhesive, and the interaction of both had a significant effect on the value of TS boards. The addition of 100% sucrose has a different effect with the addition of other sucrose concentrations on the TS value. These results are in line with research conducted by Widyorini et al. (2015) that the use of 100% sucrose concentration on boards made of Sengon particles glued citric acid and sucrose adhesives has the highest TS value.

The addition of 25% and 100% sucrose with 15% adhesive content in this study had a different effect with 20% adhesive content. Liao et al. (2016) argued that the board made of cone bagasse bonded using citric acid and sucrose adhesives had TS value that decreased in line with the addition of adhesive content from 0 to 40%.

Mechanical Properties

The mechanical properties of particleboard such as modulus of rupture (MOR), modulus of elasticity (MOE), internal bond (IB) and screw holding power (SHP) bonded using hybrid adhesive made from citric acid and sucrose are presented in Figure 4 to 7.

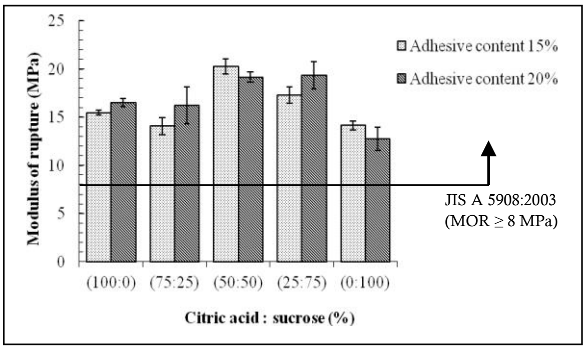

Figure 4. Modulus of rupture from particleboard.

The Figure 4 showed modulus of rupture (MOR) value of particleboard that ranged from 12.77-20.24 MPa. Overall, MOR values met the JIS A 5908:2003 standard which mention the minimum value of 8 MPa (type 8), moreover until requirement type 13 (value of 13 MPa) and type 18 (value of 18 MPa) for base particleboard standard. The value of MOR tends to increase with the increase in the amount of sucrose, but the board with a composition of 100% sucrose has a lower MOR than the other boards. This is in accordance with the research conducted by Kusumah et al. (2017) on particleboard using sorghum particles bonded with 20% citric acid and sucrose adhesive. Santoso et al. (2016) stated that the MOR value of particleboard made of nipah fronds glued with 20% citric acid and sucrose adhesive decreased with increasing sucrose content more than 75%.

The results of statistical analysis showed that the adhesive content had no significant effect on the MOR from particleboard, while the hybrid adhesive composition and the interaction between both had a significant effect. The addition of 25% sucrose in this study had the same effect with the board using 100% citric acid, and the addition of 50% sucrose had the same effect with the board using 75% sucrose. The composition of hybrid adhesive with 100% sucrose in this study had a different effect compared to other compositions. The use of 100% sucrose with 15% adhesive content has the same effect with 20% adhesive content and the use of 25% sucrose with 15% adhesive concentration. It can be said, the hybrid adhesive made from citric acid and sucrose proven to improve and enhance MOR properties of board. According to Bowyer et al. (2007), board properties are influenced by density, adhesive concentration, and particle geometry.

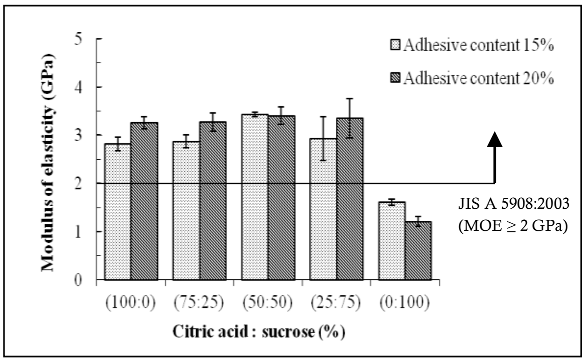

Figure 5. Modulus of elasticity values from particleboard.

The modulus of elasticity (MOE) value of board that ranged from 1.21 to 3.43 GPa (Figure 5). Nearly, the value of MOE was exceeding and met the JIS standard which mention the minimum MOE value of 2.0 GPa, except on board with 100% of sucroce that did not met the JIS standard. These results are in line with research conducted by Kusumah et al. (2017), the use of 100% sucrose produced the board made of sorghum particles with the lowest MOE value compared than other compositions. The properties of sucrose as binder material for particleboard adhesive are weaker than citric acid. The highest average value of MOE is a board with an adhesive content of 20 % and the lowest value of MOE was the board with the composition of citric acid: sucrose 0:100. Most of the boards in this study that used an adhesive content of 20% also met type 18, and boards that used an adhesive content of 15% met type 13. It means, the hybrid adhesive consisting of citric acid and sucrose is effective in repairing and increasing the MOE value of particleboard.

The board in this study has a fairly high value of MOE. Mardhatillah (2018) stated that boards made from Sengon and sorghum particles with mixture ratio of 50:50 using 20% citric acid adhesive had MOE of 3.02 GPa. This value is 8% lower than this study which used the same composition of raw materials and adhesives. This is because the slenderness ratio (SR) value of Sengon particles from this study is greater than the previous study (22.93). A larger SR value means the particles are easier to orientate and less adhesive is needed to bind the particles in each area. Maloney (1993) argued that the ideal SR value for particles as raw materials for particleboard is 150.

The results of statistical analysis showed that the adhesive content had no significant effect on the value of MOE from particleboard, while the hybrid adhesive composition and the interaction of both factors had a significant effect. The addition of 100% sucrose in this study had a different effect with the addition of other sucrose concentrations. The use of 100% citric acid has a different effect than the use of 50% sucrose. The use of 100% sucrose with 15% adhesive content in this study had a different effect with 20% adhesive content. According to Haygreen and Bowyer (1996), the value of MOE of the board is influenced by the content and type of adhesive, board density, adhesive bonding power, and particle geometry. Based on the results of this study, the addition of sucrose at a certain concentration can increase the value of MOE from particleboard. It can be assumed that the mixed solution of citric acid and sucrose as a hybrid adhesive has a stronger adhesive bonding power than only the sucrose solution.

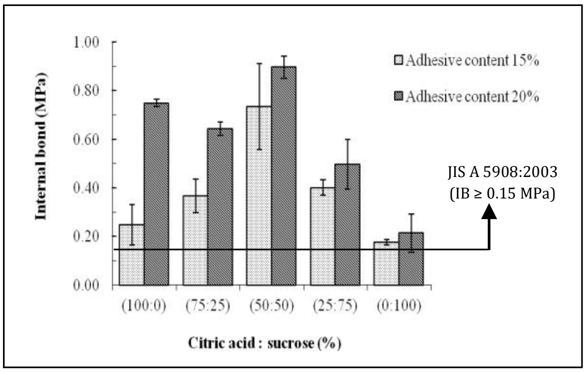

Figure 6. Internal bond values of particleboard.

The value of internal bond (IB) obtained in this study ranged from 0.18 to 0.90 MPa (Figure 6). The value of IB from the board has complied with the JIS A 5908:2003 standard of type 8 which requires an IB value greater than 0.15 MPa. Most of the boards met the standard type 18. The addition of 5% adhesive content in this study (from 15% to 20%) proven increased the IB of the board until 55%. Boards bonded with 100% citric acid have IB values that differ greatly between 15% and 20% adhesive content. This is presumably because the amount of adhesive used at 20% adhesive content is higher so that the adhesive distribution becomes better. According to Ariyani (2009), the value of IB from particleboard influenced by the distribution of the adhesive, that the distribution of the adhesive used is even, so the IB of boards will be higher. Prasetiyo et al. (2020) argued that compatibility properties between adhesive and particles enhance the IB value of the board.

The addition of 50% sucrose concentration on hybrid adhesive with 20% adhesive content for particleboard has the highest IB value. The value of IB from the boards in this study is almost the same trends with the research study by Kusumah et al. (2017) on boards made of sorghum particles with 20% citric acid and sucrose adhesive. Widyorini and Nugraha (2015) stated that a board used Sengon particles with 15% citric acid and sucrose adhesive, has an optimum IB value in the composition of 50:50 citric acid:sucrose adhesive. However, the value of IB is 47% smaller than this study at the same concentration and composition of the hybrid adhesive. It is suspected that the use of sorghum as raw material for particleboard proven to increase the IB properties. According to Mardhatillah (2018), the boards made of Sengon : sorghum particles (25:75 % w/w) with 20% citric acid adhesive have a higher IB value than boards made of 100% Sengon particles.

The results of statistical analysis showed that composition of hybrid adhesive, adhesive content, and both interaction had a significant effect on the value of the IB from the boards. The addition of 50% and 100% sucrose in this study had a different effect from the other compositions on the value of IB, and both had different effects. The use of 100% sucrose with 15% adhesive content in this study, had the same effect with 20% adhesive content. The use of 50% sucrose in hybrid adhesive had the same effect on the concentration of the adhesive used. The addition of sucrose proven to increase the value of IB from particleboard with a certain concentration. Wiwit et al. (2019) stated that the lowest IB value of particleboard made from sago by-products bonded with citric acid and sucrose adhesive was on board with using 100% sucrose. According to Umemura et al. (2015), the IB of particleboard using pine wood particles glued with citric acid and sucrose adhesives is influenced by the board density and compression temperature. The optimal value of IB was obtained from the board which has a density of 0.8 g/cm^3 and a compression temperature of 200 °C. Umemura et al. (2013) also argued that the addition of adhesive concentration on boards using pine wood particles with citric acid and sucrose as adhesives can increase the value of IB properties. This is because the reaction between citric acid and sucrose can form a strong bond.

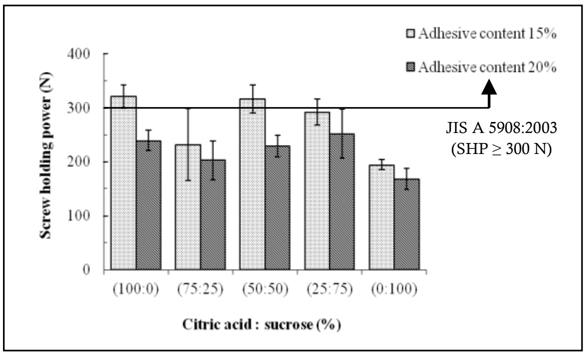

Figure 7. Screw holding power from particleboard.

The measurements of screw holding power (SHP) show the maximum force exerted on the particleboard in a given area until the screw is released. The value of SHP from particleboard is shown in Figure 7. The SHP data range from 167.71 to 321.27 N. The minimum requirements of SHP strength in the JIS A 5908:2003 standard is 300 N. Boards that met the standard are boards with mixture ratio of hybrid adhesive consisting of citric acid : sucrose is 100:0 and 50:50 (5 w/w) with an adhesive content of 15%. The average SHP values of a board with an adhesive content of 15% has a higher value than a board with a content of 20%. This is related to the use of citric acid adhesive which is acidic so that the particles become more brittle. According to Gambhir and Jamwal (2014), the more brittle particles require very little energy so they have lower SHP.

The value of SHP from particleboards tends to decrease in line with the addition of 50-100% sucrose on the hybrid adhesive. This is in accordance with the research conducted by Wiwit et al. (2019), stated that the highest SHP was obtained from boards made from Sago pulp bonded with citric acid and sucrose adhesive (50:50 % w/w) and decreased with increasing sucrose concentration. The board with the composition of citric acid : sucrose (50:50 % w/w) with adhesive content of 15% and 20% in this study, had SHP value 14% greater than the study conducted by Mardhatillah (2018), on particleboard using Sengon and sorghum particles with citric acid adhesive on the same raw material composition.

Statistical analysis presented that the composition of hybrid adhesive and the adhesive content had a significant effect on the value of SHP. The addition of 25% and 100% sucrose had a different effect than the other additions on the SHP values. Jatmiko (2006) stated that the particles of raw materials used affect the value of the SHP.

Generally, larger particles have a greater SHP value.

CONCLUSION

The results showed that the mixture ratio from hybrid adhesive consisting of citric acid and sucrose proven to enhance and improve physical and mechanical properties of particleboard. Overall, the boards composed Sengon by-products and sorghum stalk particles bonded with hybrid adhesive made of citric acid and sucrose met the JIS A 5908:2003 standard. The best board with superior value of physical and mechanical properties produced with the mixture ratio of hybrid adhesive consisting of citric acid : sucrose is 50:50 at an adhesive content of 15%. The results can be concluded that hybrid adhesive consisting of citric acid and sucrose for particleboard made of Sengon by-products and sorghum stalk particles suitable to development as adhesive for particleboard manufacturing.

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