Feasibility of Bio-Based Molasses and Citric Acid for The Manufacturing of Oil Palm Frond Particleboard

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The oil palm frond (OPF) particleboard can be manufactured using bio-adhesive-based molasses and citric acid (MOCA) at different ratios. Before particleboard manufacture, each bio-adhesive was diluted in distilled water with a solid content of 59 wt% at three different mixture ratios of MO and CA (100:0, 75:25, and 50:50). Subsequently, the OPF particles were mixed with MOCA adhesives, oven-dried at 80 °C for 12 h, and then hot pressed at 200 °C for 10 min. In general, the basic properties and thermal behaviour of the MO adhesive changed with the increasing amount of CA. The MOCA adhesives had a lower gelation time, viscosity, pH, and a comparable solids content to that of the pure MO adhesive. The thermal behaviour of the MOCA adhesive showed an alteration in the melting point with slightly lower weight loss during thermal degradation. Applying MOCA adhesive in OPF particleboard manufacturing significantly increased its physical properties, including dimensional stabilization and mechanical properties. The OPF particleboard bonded with the MOCA adhesive at a 50:50 mixture ratio generated a product with higher dimensional stabilization and the best mechanical properties. The latter product fulfilled the JIS A 5908:2003 standard, except for the MOR and SHS parameters.

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INTRODUCTION

Shifting perspectives and perceptions of consumer needs for wooden materials are causing an upward trend in the global production and consumption of particleboard products. Globally, the total quantity of particleboard production is estimated to reach 96.01 million m^3 in 2020, with the highest producers held by Asia (43.8%) and Europe (42.2%), followed by America (11.6%), Africa (1.32%), and Oceania (1.13%) (Lee *et al.* 2022). Nonetheless, this phenomenon reflects the positive economic capability of the particleboard industries in each region. However, this particular trend is directly driving the classical issues of demand vs supply for raw wood materials to support the continuation of the manufacturing process, leading to further issues environmental and legal concerns

(Baharuddin *et al.* 2023). Initiative approaches to overcome these issues can be sought by optimizing the availability of renewable agricultural biomass, such as straw (Mo *et al.* 2003), stalk (Bekhta *et al.* 2014), bagasse (Kusumah *et al.* 2016, 2017a,b; Salatein *et al.* 2024; Syahfitri *et al.* 2022), husks (Zeleniuc *et al.* 2019), leaves (Pirayesh *et al.* 2015), and palm (Ghani *et al.* 2024; Zakaria *et al.* 2021) as an alternative raw material for particleboard industries. Among these prospective sources, oil palm is preferred due to its abundant availability and the large amount of under-utilized residues generated from oil palm plantations.

Indonesia's palm oil plantation area is estimated to reach 15.34 million hectares in 2022 (BPS 2023), with most of the area concentrated in Kalimantan and Sumatera (Nabila *et al.* 2023). During the lifecycle of oil palm trees, they create massive oil palm biomass (OPB) residue in the solid state, particularly oil palm fronds (OPF). This type of waste is usually available throughout the year from plantation sites that yield during pruning and replanting activities (Ooi *et al.* 2017). OPF generated from those activities is approximately 10.4 to 14.4 t/ha, respectively, and obtained around 28.9 Mt/year in 2020 (Nabila *et al.* 2023; Ooi *et al.* 2017). Apart from its abundant availability, OPF is a lignocellulose biomass that contains large amounts of cellulose (54.4%) and hemicellulose (20.7%), as well as lignin (9.0%) and extractives (16.0%) under dry conditions (Shrivastava *et al.* 2020). Utilization of such material is still limited for mulching, ruminant pellet, and pulp purposes (Nabila *et al.* 2023). The advanced utilization of this kind of residue may benefit the environment and socio-economic development and increase its value significantly.

The application of OPF as a raw material for particleboard manufacturing has been conducted by other researchers using various methods with and without adhesive. Istana *et al.* (2023) studied the performance of OPF-reinforced composite particleboard bonded with urea-formaldehyde (UF) resin for sound absorption purposes, which resulted in superior performance with an α_n value greater than 0.95, and effectively absorbed medium-to-high-frequency sound. Wahida and Najmuldeen (2015) fabricated hybrid particleboard from OPF particles and Empty Fruit Bunch (EFB) fibers (30:70 mixture ratios) with UF resin at 200 °C for 300 s, resulting in good mechanical properties. Zakaria *et al.* (2021) manufactured OPF particleboard bonded with UF resin at 180 °C for 10 min, which exhibited better performance than EFB particleboard in terms of mechanical strength and dimensional stability. On the other hand, binderless particleboard can also be made using OPF particles directly (without any treatment) at 180 °C for 20 min (Hashim *et al.* 2012) or using the stem-exploded method (Laemsak and Okuma 2000; Suzuki *et al.* 1998). Although particleboard can be made without a binder (binderless), its properties are generally inferior to particleboard bonded with an adhesive (Hashim *et al.* 2011). Therefore, the application of formaldehyde resin as a binder, such as UF and phenol formaldehyde (PF), is preferred, resulting in satisfying properties that meet the minimum standard. Unfortunately, those types of resin can cause broad environmental issues and health risks during the manufacturing process and the lifespan of particleboards (Sutiawan *et al.* 2023). An innovative path should be developed to find a bio-adhesive with comparable properties to formaldehyde adhesive.

Citric acid (CA), a natural polycarboxylic acid with three carboxyl groups, has been extensively utilized as a potential material for bio-adhesive component in particleboard fabrication with comparable properties (Kusumah *et al.* 2016, 2017a,b; Umemura *et al.* 2012, 2013, 2015). Umemura *et al.* (2013) demonstrated the bonding mechanism of CA and wood, mainly due to ester linkages between the carboxyl group of CA and the hydroxyl group of wood components. Even though the particleboard bonded with CA resulted in

comparable properties, the adhesion area of the particles during manufacture was still concentrated in a few areas within the total specific area of the particleboard, as well as possibly reduced mechanical properties owing to the high level of acidic compounds. Combining such compounds with sucrose-based adhesives (mixture ratio of 25 to 75%) can provide excellent bonding performance (Umemura *et al.* 2013, 2015). Molasses is one of the potential sucrose-based adhesives obtained from sugar factory waste, which contains a high sucrose content of approximately 47 to 48% (Mustafa *et al.* 2023). Composites bonded with molasses exhibited desirable physical and mechanical properties as the molasses content increased (Syahfitri *et al.* 2022). Therefore, the main focus of this research is to assess the feasibility of bio-adhesive-based molasses and citric acid (MOCA) on the physical and mechanical properties of particleboards made from OPF particles.

MATERIALS AND METHODS

Preparation of Oil Palm Fronds (OPF) and Bio-adhesive Based Molasses and Citric Acid (MOCA)

Oil palm fronds (OPF) were selected as raw materials for this study of particleboard manufacturing. OPF was acquired from an oil palm plantation in the Cikabayan Experimental Garden, IPB University, Indonesia. The leaves were cut off to obtain the specific OPF without any mixture materials and then processed with a chipper and a knife-ring flaker machine to produce small-sized particles. The particles were screened using a sieving machine to obtain a uniform size, which passed through a sieving device having sizes of 4 and 20 mesh. The obtained uniform particles were oven-dried at 80 °C until the moisture content was less than 8%. In addition, the main bio-adhesive used in this study was produced by different distributors. Molasses (MO) was obtained from PTPN XII, East Java, Indonesia, while citric acid (CA) was produced by Merck distributors. The raw materials of OPF and MOCA adhesive in this study are shown in Fig. 1.

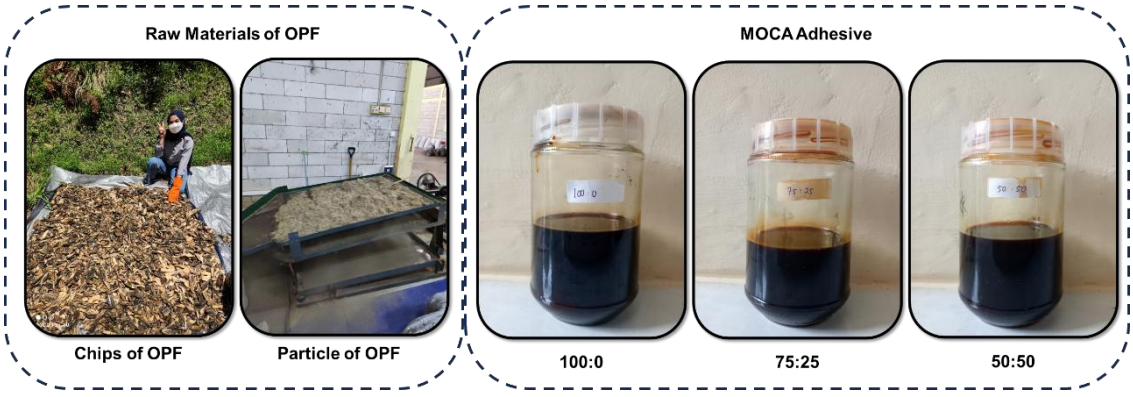

Fig. 1. Raw materials of OPF and MOCA adhesive in this study

Before particleboard manufacture, each bio-adhesive was diluted in distilled water to a 59 wt% concentration with three different weight ratio mixtures of MO and CA, namely 100:0, 75:25, and 50:50. The solution was mixed properly under a magnetic stirrer with a speed of 900 rpm for 10 min. The basic properties of the MOCA adhesive in different weight ratio mixtures were examined to evaluate each characteristic that was deemed likely to affect particleboard manufacture. The properties encompassed were pH, solid content

(SC), viscosity, and gelation time. The non-volatile solid content for each resin was ascertained by oven-drying 2 g of the samples at 105 °C for 3 h and then dividing the oven-dried weight by the initial weight. Adhesive viscosity was analyzed using a rotational Rheometer (RheolabQC, AntonPaar, Austria). The viscosity of the resins was examined using a cone-plate viscometer with a No. 2 spindle at 60 rpm and 25 °C. The gelation time of the resin was measured in boiling water by using a gelation-time meter (Techne GT6, Coleparmer, Washington, DC, USA), and the time of each resin was recorded from the start until the beep sound was heard. Each test had three repetitions to evaluate the MOCA adhesive's characteristics. In addition to these basic properties, the thermal behaviour of the MOCA adhesive in different weight ratio mixtures was examined using thermo-gravimetric analysis (TGA) 4000 instrument (PerkinElmer 4000, Waltham, MA, USA) and differential scanning calorimetry (DSC) DSC 4000 instrument (PerkinElmer 4000, United States) according to Sutiawan *et al.* (2023). The adhesive samples were freeze-dried for one hour and then pulverized to less than 60 mesh (powder). For the TGA test, the samples were loaded in the ceramic pan and scanned from 25 °C to 500 °C at a 10 °C/min rate under nitrogen purging. The samples were sealed under an aluminium pan and scanned from 25 to 400 °C at 10 °C/min for the DSC test.

Manufacture of Particleboard

The parameters of particleboard manufacture that should be considered cautiously are resin content, pressing time, and temperature. For this study, all of these parameters were based on the previous research conducted by Kusumah *et al.* (2016), where 20 wt%, 10 min, and 200 °C, respectively. Before the manufacturing process, the OPF particles were sprayed with the MOCA adhesive in different weight ratio mixtures (100:0, 75:25, and 50:50). Then the mixed particles were oven-dried at 80 °C for 12 h to a moisture content of less than 10%. Particleboard manufacture was continued by forming the mixed particles into mats with dimensions of 300 mm × 300 mm, and the thickness was controlled by a 9 mm metal bar, then hot pressed under the abovementioned conditions. The targeted dimensions of the particleboards were 300 mm × 300 mm × 9 mm. The particleboards were conditioned at room temperature for 1 week before testing. The manufacturing of particleboard in this study is shown in Fig. 2.

Fig. 2. Manufacturing of particleboard

Assessment of Particleboard Properties

The particleboard properties observed in this study were physical and mechanical, which were conducted according to the JIS A 5908:2003 standard. Physical properties consist of moisture content (MC), density, water absorption (WA), and thickness swelling (TS), with a sample size of 50 mm × 50 mm × 9 mm for each treated board. All samples underwent measurements to determine their oven-dried volume and weight and obtain MC and density values. TS and WA assessments were conducted by immersing the samples in distilled water at 20 °C for 24 h, followed by oven drying at 103 ± 2 °C for 24 h. The dimensions of the samples were then measured, including the length, width, and thickness at marked positions on the samples.

Mechanical properties include the modulus of elasticity (MOE) and modulus of rupture (MOR), internal bonding (IB), and screw-holding strength (SHS). The bending strengths of particleboards (MOE and MOR) and IB were evaluated by conducting a three-point bending test using a universal testing machine (UTM type AG-IS 50 kN, Shimadzu, Japan) on 200 mm × 50 mm × 9 mm and 50 mm × 50 mm × 9 mm samples of each treated board, respectively. The loading speed and effective span for the bending test were 10 $mm.min^{-1}$ and 150 mm, respectively, whereas the loading speed for the IB test was 2 $mm.min^{-1}$. SHS samples with dimensions of 50 mm × 100 mm were drilled with a 3 mm deep hole in the two centre positions of the cross-section. Each hole was 25 mm from the edge side. The screws used for the SHS test were 2.7 mm in diameter and 16 mm in length. Screws 2.7 mm in diameter and 16 mm in length were inserted into the hole to a depth of 9 mm, leaving 7 mm of shank free for loading grip. The pulling-out load speed was approximately 2 $mm.min^{-1}$.

Fourier Transform Infrared (FTIR) Spectroscopy

FTIR spectroscopy with a Universal Attenuated Total Reflectance (UATR) accessory (PerkinElmer Inc., Waltham, USA) was used to determine the functional groups of the particleboards. For this analysis, 2 mg of each sample in powder form (30 to 40 mesh) was scanned from 400 to 4000 cm^{-1} with a resolution of 4 cm^{-1}, with three scans per treatment at 24 °C room temperature.

Data Analysis

The experimental design was randomized with different weight ratio mixtures (100:0, 75:25, and 50:50) of adhesive and particles as a factor. The data are presented as the average and standard deviation. An analysis of variance (ANOVA) was conducted to evaluate the effect of different weight ratio mixtures on particleboards' physical and mechanical properties. If there were significant differences between each factor and the interaction, Duncan's multiple distance test was used.

RESULTS AND DISCUSSION

Basic Properties of Bio-adhesive Based Molasses and Citric Acid (MOCA)

A bio-based citric acid and molasses (MOCA) adhesive was successfully synthesized at different mixture ratios. The basic properties of the prepared MOCA adhesive are listed in Table 1. It can be observed that the basic properties of the prepared MOCA adhesive, such as gelation time, pH, solid content (SC), and viscosity, were lower than those of previous research using each type of adhesive, namely MO and CA separately

(Syahfitri *et al.* 2022; Sutiawan *et al.* 2023, 2024). It was reported that the basic properties of MO and CA adhesives are as follows: gelation times are 5.9 and 4.6 min, pH ranges from 4.79 and 2.3, solid contents are 58.80 and 56.20%, and viscosities are 152 and 7.3 mPa.s, respectively (Syahfitri *et al.* 2022; Sutiawan *et al.* 2023). These values were lower than those of the pure MO (100:0) and MOCA adhesives prepared in this study (Table 1). Similar results were also reported by Sutiawan *et al.* (2024), who used a combination of molasses and citric acid (MOCA) at different mixture ratios for plywood production.

Table 1. Basic Properties of MOCA Adhesive at Different Mixture Ratios

Composition MO:CA (w/w%)	Gelation time (min)	pH	Solid content (%)	Viscosity (mPa·s)
100:0	5.90	4.40	55.48	152.69
75:25	2.40	2.08	56.44	116.13
50:50	2.00	1.49	55.60	56.47

Owing to the different mixture ratios of the MOCA adhesive, the addition of CA to the MO adhesive resulted in a shorter gelation time and a decrease in pH and viscosity values. As shown in Table 1, the viscosity and gelation time gradually decreased as the CA ratios within the MO adhesive increased. These phenomena are similar to those reported by Sutiawan *et al.* (2024). This could be due to the molecular mobility of CA, which can directly affect its viscosity (Sutiawan *et al.* 2023). Lower viscosity with shorter gelation time of adhesive properties is more desirable in manufacturing composite products due to higher distribution, penetration, wettability, and manufacturing processes (Syahfitri *et al.* 2022). These values are also much lower than those of commercial formaldehyde-based adhesives, particularly urea-formaldehyde (UF), which has a viscosity and gelation time of 214 to 444 mPa·s and 2 to 5 min, respectively (Jeong and Park 2019; Nuryawan *et al.* 2020; Syahfitri *et al.* 2022). In addition, the pH value of the MO adhesive also decreased as the CA ratio increased, from 4.40 to 2.08 and 1.49, at 75:25 and 50:50 MOCA adhesive mixture ratios, respectively. However, these values were slightly higher than those of the CA-sucrose adhesive at the same mixture ratio studied by Umemura *et al.* (2013). The lower pH obtained after increasing the CA ratios was expected because of the highly acidic compound. Sutiawan *et al.* (2023) and Kusumah *et al.* (2017a) reported that CA is categorized as a high-acidity adhesive with pH values ranging from 0.30 to 2.3. Furthermore, the solid content slightly increased in the MOCA adhesive at different mixture ratios compared with that of the pure MO adhesive (100:0). However, these values were not too far from the range of the solid content target (59 wt%).

The thermal behaviours of the pure MO and MOCA adhesives at different mixture ratios can be observed using the TGA-DSC profiles. As can be seen in Fig. 3, the TGA profile of pure MO (100:0) has a lower thermal degradation, particularly at 125 °C, with higher weight loss than that of other treatments (Fig. 3). Adding CA to the MO adhesive at different mixture ratios increased the temperature during thermal degradation, which was 190 and 210 °C at 75:25 and 50:50 MOCA adhesive mixture ratios, respectively. However, this resulted in a much lower weight loss during thermal degradation. This indicates that the mixture of CA within the MO adhesive can prolong the temperature applied (190 to 210 °C) with slightly lower weight loss during thermal degradation. This result was relatively good and showed a stable MOCA adhesive formulation. Kusumah *et al.* (2017a) also studied the formulation of CA and sucrose for sorghum-bagasse particleboard

manufacture, and it was reported that moderate weight loss started to occur during thermal degradation using those mixed solutions at 150 °C.

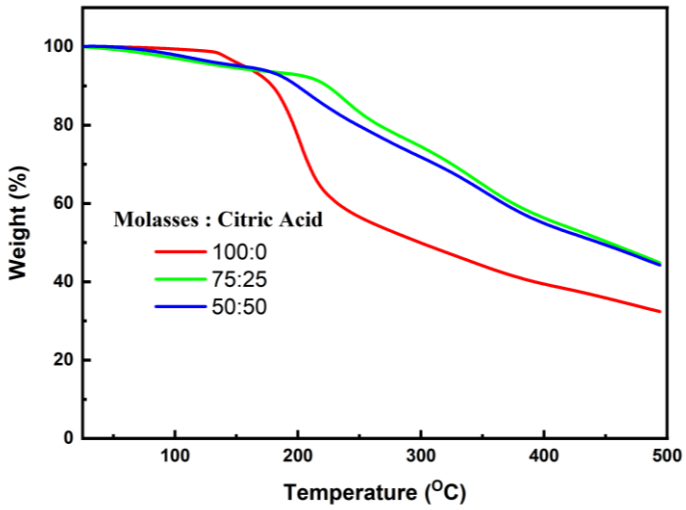

Fig. 3. TGA profile of pure MO and MOCA adhesive at different mixture ratios

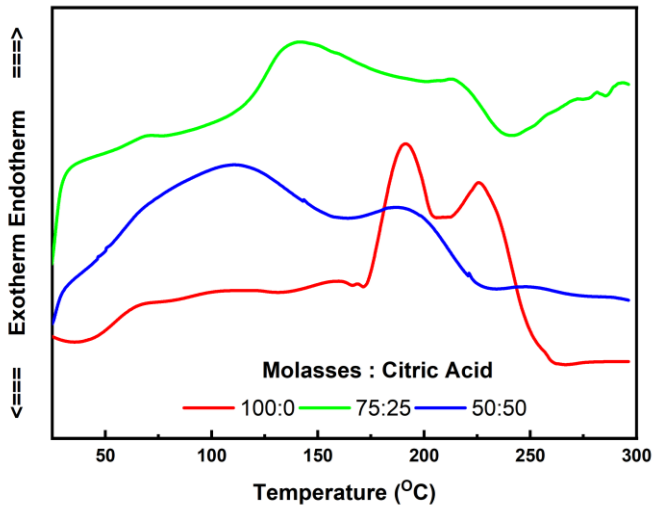

Fig. 4. DSC profile of pure MO and MOCA adhesive at different mixture ratios

Meanwhile, the DSC profile of pure MO (100:0) has a sharper first and second endothermic peak at 200 and 225 °C, respectively (Fig. 4). This DSC profile was quite similar to the MO adhesive used in other studies (Sutiawan *et al.* 2023). However, these endothermic peaks were lower than those of the sucrose adhesive used by Kusumah *et al.* (2017a). In addition, the endothermic profile of the MOCA adhesives with 75:25 and 50:50 mixture ratios showed a shift of their first endothermic peak to a much lower temperature, particularly at 140 and 110 °C, respectively. Furthermore, the endothermic peak widened significantly after adding CA. This phenomenon indicates that the addition of CA within the adhesive formulation alters the melting point of MO adhesives due to the carboxyl group of CA reacting with the hydroxyl group of MO. A similar phenomenon also occurred

in the CA-sucrose formulation, which reduced the endothermic peak compared with CA and sucrose alone (Kusumah *et al.* 2017a).

Physical Properties of OPF Particleboard Manufactured Using Bio-adhesive Based Molasses and Citric Acid (MOCA)

The properties of the OPF particleboard bonded using pure MO and MOCA adhesives at different mixture ratios are presented in Table 2. The physical properties observed, including moisture content (MC), density, water absorption (WA), and thickness swelling (TS), were relatively higher in the OPF particleboard bonded with pure MO (100:0). Moreover, the addition of CA to the MO adhesive decreased the physical properties of the OPF particleboard bonded with the MOCA formulation (Table 2).

Table 2. Physical Properties of OPF Particleboard Manufactured Using MOCA Adhesive at Different Mixture Ratios

Parameters	Composition MO:CA (w/w%)			ANOVA (p<0.05)
	100:0	75:25	50:50	
Moisture content (%)	7.19 ± 0.18^{b}	7.06 ± 0.06^{b}	6.54 ± 0.07^{a}	**
Density (g.cm^{-3})	0.74 ± 0.06^{a}	0.70 ± 0.05^{a}	0.70 ± 0.08^{a}	ns
Water absorption (%)	50.05 ± 4.70^{b}	46.26 ± 2.66^{ab}	35.51 ± 8.58^{a}	ns
Thickness swelling (cm)	12.26 ± 1.22^{c}	8.26 ± 1.48^{b}	5.99 ± 0.28^{a}	**

Note: The subscript alphabet above describes the interaction between each factor using Duncan's multiple distance test; ns describes not significant, while ** describes significant at p<0.05.

As can be seen in Table 2, the MC and density of OPF particleboard ranged from 6.54 to 7.19% and 0.70 to 0.74 g.cm^{-3}, respectively. Similar results for MC values were also found in the plywood products bonded using the same adhesive at different mixture ratios. However, the density is much lower (0.46 to 0.53 g.cm^{-3}) than this obtained result (Sutiawan *et al.* 2024). Previous research reported that particleboard density bonded with CA and sucrose (27:75 of mixture ratio) is 0.4 to 0.8 g.cm^{-3} (Umemura *et al.* 2015). In addition, the MC and density values slightly decreased as the CA ratio within the MO adhesive increased. This phenomenon follows the ANOVA results (Table 2), which show that the MC values of the OPF particleboard were significantly different at different mixture ratios. In contrast, the density values showed the opposite phenomenon. The MC and density of the OPF particleboard bonded with the MOCA adhesive were reduced by 1.81 to 9.04% and 5.40%, respectively, compared to those of the OPF particleboard bonded with the pure MO adhesive. However, all these results were still comparable to the requirements of JIS A-5908 (JIS 2003).

Furthermore, the WA and TS of OPF particleboard ranged from 35.51 to 50.05% and 5.99 to 12.26%, respectively. Those values were much lower than that of particleboard bonded (board density of 0.6 g.cm^{-3}) with CA-sucrose (25:75 mixture ratio) studied by Umemura *et al.* (2015), as well as sorghum-bagasse particleboard bonded with CA-sucrose at the same mixture ratios reported by Kusumah *et al.* (2017a). Owing to the different mixture ratios of MOCA adhesive, the WA and TS values reduced along with the increase in CA ratios. This phenomenon is following the ANOVA results (Table 2). The WA and TS values of the OPF particleboard bonded with the MOCA adhesive were reduced by 7.57 to 29.05% and 32.62 to 51.14%, respectively, compared to those of the OPF particleboard bonded with the pure MO adhesive. Lower values of WA and TS can give a good sign of

the dimensional stabilization of OPF particleboard, particularly at a 50:50 mixture ratio, since those two parameters are related. Umemura *et al.* (2015) described the possible interaction between CA and sucrose during particleboard manufacturing. Citric acid may interact with sucrose, which produces hydroxyl groups and ester linkages (Kusumah *et al.* 2017a; Umemura *et al.* 2015). The formation of these linkages can significantly affect the physical properties of OPF particleboards bonded with MOCA adhesive by inhibiting their WA uptake, thus directly affecting their final TS value. In addition, Sutiawan *et al.* (2024) also reported a possible mechanism between MO and CA, which contains similar interactions with previous studies using sucrose compounds.

Mechanical Properties of OPF Particleboard Manufactured Using Bio-adhesive Based Molasses and Citric Acid (MOCA)

The mechanical properties of the OPF particleboard bonded using the pure MO and MOCA adhesives at different mixture ratios are presented in Table 3. The bending properties, including modulus of elasticity (MOE) and modulus of rupture (MOR), as well as internal bonding (IB) and screw holding strength (SHS), were relatively lower in the OPF particleboard bonded with pure MO (100:0). Moreover, the addition of CA to the MO adhesive tended to enhance the mechanical properties of the OPF particleboard bonded with the MOCA formulation (Table 3).

Table 3. Mechanical Properties of OPF Particleboard Manufactured Using MOCA Adhesive at Different Mixture Ratios

Parameters	Composition MO:CA (w/w%)			ANOVA (p<0.05)
	100:0	75:25	50:50	
MOE (GPa)	1.85 ± 0.11^{a}	2.03 ± 0.27^{a}	3.26 ± 0.08^{b}	**
MOR (MPa)	6.32 ± 0.20^{a}	6.53 ± 1.46^{a}	12.40 ± 0.76^{b}	**
Internal bonding (N.mm^{-2})	1.09 ± 0.11^{a}	0.96 ± 0.20^{a}	2.10 ± 0.19^{b}	**
Screw holding strength (N)	73.18 ± 0.90^{a}	80.21 ± 4.30^{a}	127.08 ± 18.92^{b}	**

Note: The subscription alphabet above describes the interaction between each factor using Duncan's multiple distance test; ns is not significant, while ** is significant at p<0.05.

As can be seen in Table 3, the MOE and MOR of OPF particleboard ranged from 1.85 to 3.26 GPa and 6.32 to 12.40 MPa, respectively. These values were much lower than those of sorghum-bagasse particleboard bonded with CA-sucrose at the same mixture ratios (Kusumah *et al.* 2017a), as well as particleboard bonded with CA-sucrose at 25:75 mixture ratios (Umemura *et al.* 2015). However, those values were quite similar with the OPF particleboard bonded with CA-starch at different mixture ratios ranging from 2.3 to 2.5 GPa and 7.32 to 10.24 MPa, respectively (Zakaria *et al.* 2021). In addition, the MOE and MOR values increased significantly as the CA ratio within the MO adhesive increased. This phenomenon is following ANOVA results (Table 3). The MOE and MOR of the OPF particleboard bonded with the MOCA adhesive increased by 8.87 to 43.25% and 3.22 to 49.0%, respectively, compared to those of the OPF particleboard bonded with the pure MO adhesive. Even though the OPF particleboard bonded with MOCA adhesive significantly increased the bending properties, none of the samples met the JIS A-5908 (<18 MPa) of the type 18 standard (JIS 2003), except the MOE values of the OPF particleboard with a 50:50 mixture ratio (>3 GPa).

On the other hand, the IB and SHS of OPF particleboard ranged from 0.96 to 2.10 $N.mm^{-2}$ and 73.2 to 127.1 N, respectively. These IB values were much better than those of sorghum-bagasse particleboard and recycled wood-based particleboard bonded with CA-sucrose at the same mixture ratios (Kusumah *et al.* 2017a; Umemura *et al.* 2013), as well as much higher than those of OPF particleboard bonded with CA-starch at different mixture ratios, which ranged from 0.34 to 0.52 $N.mm^{-2}$ (Zakaria *et al.* 2021). These SHS values were much lower than those of sorghum-bagasse particleboard bonded with CA and CA-sucrose at different mixture ratios ranging from 348 to 525 N (Kusumah *et al.* 2017a, 2017b). Owing to the different mixture ratios, the IB value of the OPF particleboard bonded with the MOCA adhesive showed a slight decrease of 11.9% at a 75:25 mixture ratio. Then, it increased significantly by 48.1% at a 50:50 mixture ratio compared to pure MO. Meanwhile, the SHS values of OPF particleboard bonded with MOCA adhesive showed a higher increase by 8.8% and 42.4% at 75:25 and 50:50 mixture ratios, respectively, than that of pure MO only. This phenomenon follows the ANOVA results, which show that these values were significantly different at different mixture ratios (Table 3).

However, these phenomena were opposite to previous research, which found that CA-sucrose with a 25:75 ratio resulted in higher IB than that of CA-sucrose with a 50:50 mixture ratio (Kusumah *et al.* 2017a). The OPF particleboard bonded with MOCA adhesive was able to significantly increase the IB and SHS values; therefore, all of the samples met the JIS A-5908 (<18 Mpa) of the type 18 standard (JIS 2003) in terms of IB values (>0.3 N/mm^2), but not in terms of SHS values (<500 N).

Functional Group Analysis

The functional group alteration of OPF particleboard bonded with pure MO only and MOCA adhesive at different mixture ratios can be observed using FTIR spectra absorbance within the wavenumber range of 400 to 4000 cm^{-1} (Fig. 5). The absorbance spectra were interpreted by referring to other related research and comparing them to the observations made during the experiment. Some apparent changes in the presented absorbance can be noticed, which indicated the difference in the chemical composition for each observed treatment. Several band shifts and higher absorbance intensities were discernible in the OPF particleboard bonded with pure MO only and MOCA adhesive, particularly in the fingerprint wavenumber, compared to the OPF particle absorbance (Fig. 5). This could be associated with the addition of each adhesive prepared in this study, which may interact with OPF particles during particleboard manufacturing (Kusumah *et al.* 2017a; Sutiawan *et al.* 2024; Umemura *et al.* 2015).

As shown in Fig. 5, the changes in absorbance spectra, particularly the shifting and stretching, were attributed to the formation of hydrogen bonds between the OH group in the sucrose structure within the molasses compound and lignocellulose materials. It can be seen that absorbance changes in OPF particleboard bonded with MOCA adhesives were evident at specific wavenumbers, notably 700, 1250, and 1603 cm^{-1}. According to Syahfitri *et al.* (2022), the fingerprint absorbance for MO adhesives is detected at 700, 1240, and 1603 cm^{-1}. The specific wavenumber at 700 cm^{-1} was assigned to the unsubstituted CH=CH of the furan ring. Moreover, the 1240 and 1603 cm^{-1} wavenumbers can be attributed to the C=O stretching and C-O stretching of the dimethylene ether bridge generated from 5-HMF. This compound is obtained from the hydrolysis of the sucrose structure in molasses, particularly when a high-temperature process is applied. In addition, the presence of CA can be detected at specific wavenumbers, notably 1245 and 1727 cm^{-1}, which were

assigned to C=O stretching due to the vibration band of the carboxyl group or ester group, respectively (Kusumah *et al.* 2017a).

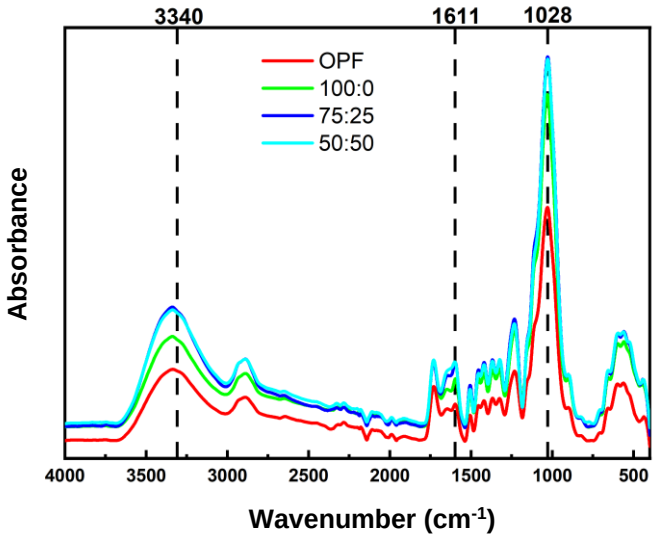

Fig. 5. FTIR observation of OPF particleboard bonded with pure MO and MOCA adhesive at different mixture ratios

CONCLUSIONS

A bio-based citric acid and molasses (MOCA) adhesive was successfully synthesized and applied in the oil palm frond (OPF) particleboard manufacturing at different mixture ratios. The basic properties and thermal behaviour of the MO adhesive changed with the addition of CA. An increase in the CA ratio was found to significantly alter these values. The MOCA adhesives were found to have a lower gelation time, viscosity, pH and comparable solid content than pure MO adhesives. The thermal behaviour of the MOCA adhesive showed an alteration in the melting point, evidenced by the prolonged temperature applied (190 to 210 °C) with slightly lower weight loss during thermal degradation. Applying this adhesive in OPF particleboards manufacture significantly improved their physical properties, including dimensional stabilization and mechanical properties. The OPF particleboard bonded with MOCA adhesive at a 50:50 mixture ratio generated a product with higher dimensional stabilization and the best mechanical properties. The latter product fulfilled the JIS A 5908:2003 standard except for MOR and SHS parameters. The FTIR observation revealed discernible alteration due to the interaction of MO and CA adhesive, attributed to the formation of hydroxyl groups and ester linkages. In addition, MO and MOCA adhesives for wood panel products could become a viable alternative to traditional, petrochemical-based bonding agents, supporting cost savings and environmental goals.

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