

Effects of Adding Ammonium Dihydrogen Phosphate to a Water-soluble Extract of the Inner Part of Oil Palm Trunk on Binderless Particleboard

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This study investigated the effects of adding ammonium dihydrogen phosphate (ADP) on the physical and chemical changes of a water-soluble extract of the inner part of oil palm trunk (OPT) to clarify the bonding mechanism of the binderless particleboard. The extract's effect on ADP-added binderless particleboard was also investigated. OPT particles were treated by hot water at 60 °C for 6 h. Water-soluble extract and treated OPT particles were obtained. ADP was added to the water-soluble extract at 0, 10, and 40 wt%, and the mixtures were heated at 180 °C for 10 min. Furthermore, binderless particleboards using the treated particles were manufactured with similar condition. The 10 wt% ADP mixture changed the water-soluble extract to an insoluble substance, which was twice that of with 0 wt.% ADP addition. Infrared spectroscopy revealed peaks of furan and carbonyl in the insoluble substance. This indicated that the free sugar content in the water-soluble extract would change to furan compounds. Thermal analysis revealed that the resulting insoluble substance had good thermal stability, suggesting a high-molecular-weight substance. The insoluble substance would contribute to bonding of the binderless particleboards. In particular, a significant contribution to the water resistance was observed.

Keywords: Water-soluble extract; Oil palm trunk; Ammonium dihydrogen phosphate; Binderless particleboard; Water resistance

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INTRODUCTION

The global forest area decreases each year (FAO and UNEP 2020), resulting in limited wood resources. Accordingly, the utilization of nonwood lignocellulosic resources, especially agricultural residue, has gained attention as a raw material for wood-based materials (van Dam *et al.* 2006; Kusumah *et al.* 2016; Widyorini *et al.* 2019). Global annual production of oil palm biomass is estimated at 214.5 million tons (Tye *et al.* 2016). Once an oil palm tree reaches 25 years of age, its oil productivity generally diminishes, and the tree is replanted (Ismail and Mamat 2002). Massive numbers of trunks are felled, representing a promising nonwood lignocellulosic resource. Research into wood-based materials using oil palm trunk (OPT) can be classified into two categories: those using synthetic resin adhesives, and those in which binderless boards are prepared without usage of synthetic resin adhesives. Phenol formaldehyde resin and urea formaldehyde resin adhesives are used for various wood-

based materials such as plywood and particleboard (Hashim *et al.* 2010; Hartono *et al.* 2017; Baskaran *et al.* 2019), but the physical and mechanical properties of wood-based materials are not sufficient. One explanation is that water-soluble components, including saccharides, inhibit the curing of adhesives (Jumhuri *et al.* 2014; Bockel *et al.* 2019). To resolve this problem, an increase in resin content and the utilization of other high-performance adhesives such as polymeric diphenylmethane diisocyanate (pMDI) have been proposed (Hermanto and Massijaya 2018; Prabuningrum *et al.* 2020). However, synthetic resin adhesives are derived from fossil resources, so their use should be reduced as much as possible.

For binderless boards, chemical components in raw material are denatured by heat and moisture to develop adhesiveness (Hashim *et al.* 2011; Lamaming *et al.* 2013; Lamaming *et al.* 2014). However, the performance of binderless boards is very poor when compared with boards that use a suitable synthetic resin adhesive, especially one that is water-resistant. Moreover, another study on binderless boards (Anglès *et al.* 2001; Widyorini *et al.* 2005) was successful in improving all of the boards' properties, especially dimensional stability. But it needed some pre-treatment such as steam-treatment, which requiring high energy and cost in the manufacturing process. Therefore, a high-performance board fabricated by a method that does not use synthetic resin adhesive and a simple method without any pre-treatment are desired.

Ammonium dihydrogen phosphate (ADP) acts as a catalyst to convert sucrose into a substance that is insoluble in hot water (Umemura *et al.* 2017). Furthermore, sucrose and ADP have been used as bio-based adhesives for manufacturing particleboard (Zhao *et al.* 2018). Oil palm trunk, especially the inner part, contains many water-soluble components, including free sugars such as sucrose and glucose (Mansor and Ahmad 1990). Thus, the addition of ADP might be effective for binderless particleboard using OPT. Based on this hypothesis, our research group manufactured binderless particleboard using the inner part of OPT and ADP, resulting in good physical and mechanical properties, especially excellent water resistance (Komariah *et al.* 2019). This study used ADP contents ranging from 0 to 40 wt.%, with 10 wt.% ADP showing the optimum results. The free sugar contained in the water-soluble extract is thought to affect ADP's contribution to the qualities of binderless particleboard. However, the effect of OPT's water-soluble extract on the bonding mechanism of binderless particleboard with the added ADP has not been fully clarified. Therefore, the influence of ADP on the physical and chemical changes of a water-soluble extract of the inner part of OPT were clarified. The effect of the water-soluble extract on binderless particleboard with the addition of ADP was also investigated in this study.

EXPERIMENTAL

Preparation of Materials

Oil palm trunk with a diameter of 60 cm and an approximate age of 30 years was collected from Bogor, Indonesia. The inner part of the trunk (the inner two-thirds of the diameter) was cut and chopped into chips. The chips were dried under sunlight and then oven-dried at 70 °C for 12 h to reach a moisture content between 1 and 3%. The dried chips were ground into particles with a ring flaker (Pallman Maschinenfabrik, Zweibrücken, Germany).

The particles were screened by a sieving mill (Iida sieve shaker, Iida Seisakusho, Yokohama, Japan). The particles that remained between the aperture sizes of 2 and 0.25 mm were used as the raw material. ADP of extra-pure grade was purchased from Nacalai Tesque

(Kyoto, Japan) and used without further purification.

Hot-water Extraction Treatment

To obtain a water-soluble extract of OPT particles, the particles were immersed in hot water at 60 °C for 6 h (Lamaming *et al.* 2013), as shown in Fig. 1. The water-particles ratio use on the extraction process was 1 liter of water to 100 g of air-dried weight (moisture content ~7%) OPT particles. The solution was filtered using a Buchner funnel with medium-fine filter paper. The filtrate was freeze-dried to obtain a water-soluble extract. The treated particles were oven-dried at 105 °C for 24 h. The particles were used as raw material for manufacturing the binderless particleboard.

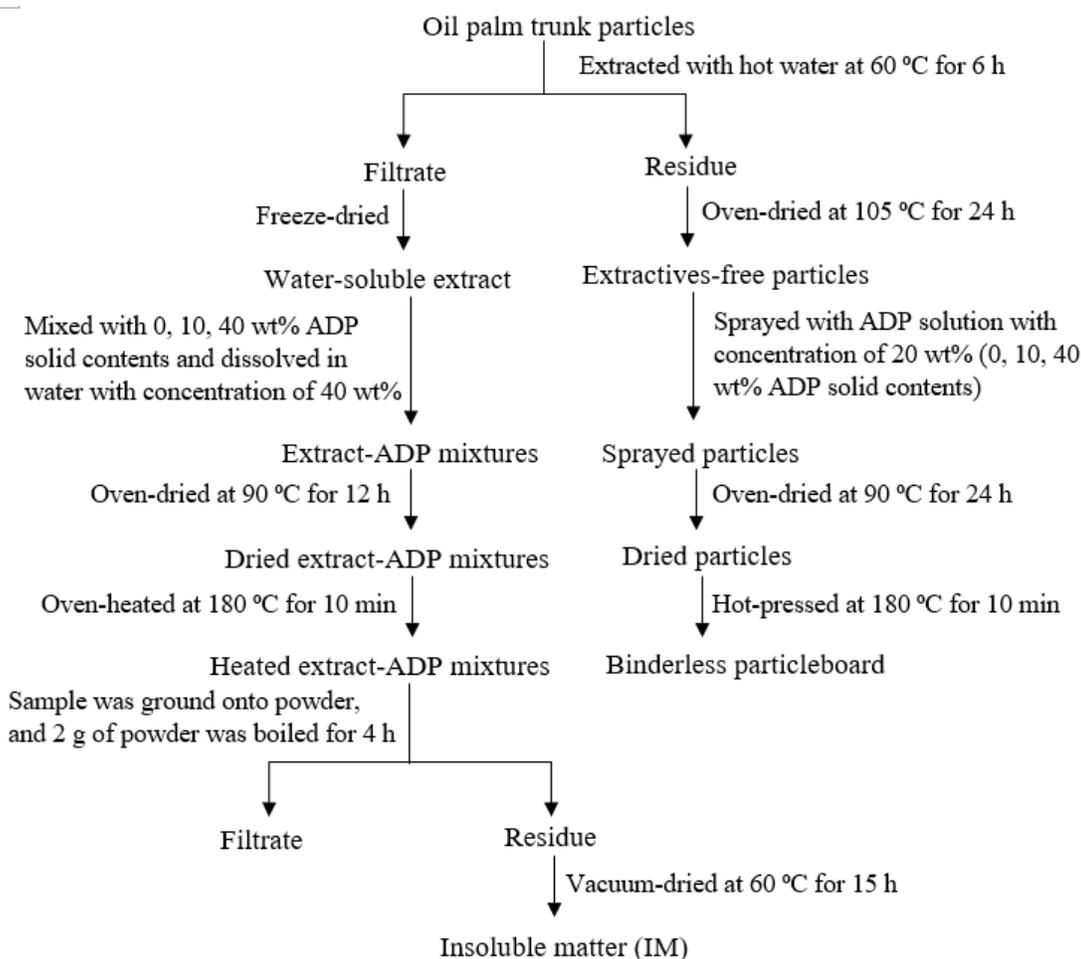


Fig. 1. Experimental flow chart to get insoluble matter and binderless board from OPT particles.

Sugar Analyses in Extract

The free sugars (glucose, sucrose, and fructose) in the water-soluble OPT extract were quantified. Briefly, the dried extract was re-solubilized in distilled water (10 mg/mL) and submitted to the assays using a glucose CII test kit (Wako Pure Chemical, Osaka, Japan), a sucrose assay kit SCA 2-1 KIT, 20 assays (Sigma-Aldrich, St. Louis, MO), and a fructose assay kit (C/F) (Bio-vision, Palo Alto, CA). Absorbance was measured with an SH-1000 Lab Microplate Reader (Corona Electric, Ibaraki, Japan). Glucose, sucrose, and fructose solutions were used as standards for calibration.

Insoluble Matter (IM) Measurement

The extract was mixed with various ADP solid contents, *i.e.*, 0, 10, and 40 wt.% based on the weight of the dried extract. After being dissolved in distilled water with a concentration of 40 wt.%, the mixtures were oven-dried at 90 °C for 12 h then heated at 180 °C for 10 min. The heat-treated mixtures were ground into powder using a mortar. Two grams of powder of each heat-treated mixture was boiled for 4 h. The results were obtained by three repetitions with the same sample. After the treatment, the solution was filtrated with a glass filter, and the insoluble matter remaining on the glass filter was vacuum-dried at 60 °C for 15 h and finally weighed. The IM was calculated based on the weight change of the mixture by the following equation,

$$IM \text{ rate } (\%) = (a / 2 \text{ g of } b) \times 100 \quad (1)$$

where *a* is weight of dried insoluble matter (g) and *b* is heat-treated mixture (g).

Fourier Transform Infrared Spectroscopy (FTIR) Measurement

The analysis was performed using OPT particles, water-soluble extract, and IM samples. Each specimen was ground into powder (passed 150 μm) using a mortar. The powder obtained was vacuum-dried at 60 °C for 15 h. Infrared (IR) spectra were obtained by using the KBr disk method with an FTIR spectrophotometer (FT/IR-4200, Jasco, Tokyo). The infrared spectra were recorded by an average of 32 scans at a resolution of 4 cm⁻¹ in the range of 400 to 4000 cm⁻¹.

Thermal Analysis Measurement

The OPT particles, OPT extract, and IM samples were pulverized (passed 150 μm). All of the samples were vacuum-dried at 60 °C for 15 h before analysis. Differential scanning calorimetry (DSC) was carried out with a DSC 25 differential scanning calorimeter (TA Instruments, Tokyo). Before the sample was prepared, tweezers were used to poke a small hole in the cover of an aluminum lid. Each sample for the DSC analysis was encapsulated in an aluminum pan and was scanned from room temperature to 400 °C at an increase rate of 10 °C/min under nitrogen purging with a flow rate of 50 mL/min. Thermogravimetric analysis (TGA) was carried out using a TGA 55 thermogravimetric analyzer (TA Instruments, Tokyo). Each sample of TGA analysis was scanned from room temperature to 400 °C under nitrogen purging with a flow rate of 50 mL/min and an increase rate of 10 °C/min.

Binderless Particleboard Manufacturing

Based on our previous study (Komariah *et al.* 2019), similar conditions on manufacturing particleboards were conducted. The ADP was dissolved in distilled water at a concentration of 20 wt% and used as an additive. The pH of 3.9 and viscosity of 3 mPa•s of the ADP solution at 21 °C was measured using a pH meter (D-51 Horiba, Tokyo) and a viscosity meter (Fungilab, Barcelona, Spain), respectively. The 20 wt% ADP solution was sprayed onto the hot-water-treated OPT particles with the various ADP solid contents, *i.e.*, 0, 10, and 40 wt% based on the weight of the dried particles. The sprayed particles were dried at 90 °C for 24 h to reduce the moisture content to around 1 to 3%. The dried particles were hand-formed into a mat using a 300 mm × 300 mm forming box. The mat was hot-pressed at 180 °C for 10 min with a 5-mm distance bar to control the board thickness. After 1 min of pressing, the pressure was reduced for 30 s to prevent blisters. The particleboard target density was 0.8 g/cm³. Particleboard with 0 wt% ADP was obtained from particles that had

not been sprayed with an ADP solution. The manufactured particleboards were then conditioned for 1 week at 20 °C and approx. 60% relative humidity.

Particleboard Properties

The particleboards were tested according to the Japanese Industrial Standard for Particleboard (JIS A 5908 2003). The physical and mechanical properties tested were thickness swelling (TS), bending, and internal bond strength (IB). Moreover, water absorbance (WA) was measured and cyclic aging treatment was conducted. The TS and WA after water immersion at 20 °C for 24 h were measured on a 50 mm × 50 mm × 5 mm specimen. After the TS test, the samples were subjected to a cyclic aging treatment (drying at 105 °C for 10 h, hot water immersion at 70 °C for 24 h, drying at 105 °C for 10 h, immersion in boiling water for 4 h, and drying at 105 °C for 10 h). The changes in the thickness and weight of the samples throughout the treatments were observed.

The static three-point bending test was carried out on a 200 mm × 30 mm × 5 mm sample of each type of particleboard. The effective span was 150 mm, and the cross-head speed was 10 mm/min. The modulus of rupture (MOR) and modulus of elasticity (MOE) were calculated. The IB test was performed on a 50 mm × 50 mm × 5 mm sample with a tension loading speed of approx. 2 mm/min. Each test was carried out in five replications, and the average values and standard deviation were calculated. The MOR, MOE, and IB values of the boards were corrected for each target density based on the regression lines between the obtained values and the sample densities. The means were compared using Tukey's HSD post hoc test to identify the significance of differences at the confidence level of 95%.

RESULTS AND DISCUSSION

Free Sugars in Water-soluble Extract

In this study, the amount of the water-soluble extract that obtained was 11.59%. Table 1 shows the free sugars in the extract. The glucose content was the highest, at 13.63%, followed by fructose and sucrose, with 3.93% and 2.32%, respectively. These obtained values were different from those obtained from chemical composition and free sugar analysis in the previous study (Komariah *et al.* 2019). This difference can be explained as a function of raw material size and to the difference in the methods used to obtain the extracts. In this study, the particles were extracted directly with hot water, whereas in the previous study the raw material was a powder. The solvents used to obtain free sugars also differed between studies. However, the present results are consistent with those of Lange and Simatupang (1994) and Murai and Kondo (2011), in which glucose had the highest sugar component in the hot-water extract of OPT particles.

Table 1. Free-sugar Content in the Water-soluble Extract of the Inner Part of OPT

	Glucose (%)	Sucrose (%)	Fructose (%)
Hot-water extraction	13.63 ± 0.30	2.32 ± 0.24	3.93 ± 0.80

*n=3

Insoluble Matter of Extract with ADP Added

To clarify the chemical and physical changes resulting from the addition of ADP to

the water-soluble extract, a mixture of extract and ADP was heated and treated by boiling water. Figure 2 shows the IM value after the treatment with boiling water. The IM proportion without ADP (0 wt.%) was 19.72%. This means that the extract turned into IM with just heating. When 10 wt.% of ADP was added to the extract, the IM proportion almost doubled, to 37.04%. In the case of 40 wt% ADP, the proportion was 25.80%. The reason why the value was lower than that of 10 wt% seems to be that a large amount of ADP was eluted during treatment. Meanwhile, when the practical IM was calculated based on the weight of the extract, the IM proportion of 40 wt% ADP was higher than that of 10 wt% ADP: 43.58% vs. 38.31%, respectively. It was clarified that ADP accelerates the change of water-soluble extract into an insoluble substance. This is in accordance with Umemura *et al.* (2017) and Zhao *et al.* (2018), who stated that an ADP content of 10 wt% was needed to obtain a highly insoluble substance from sucrose.

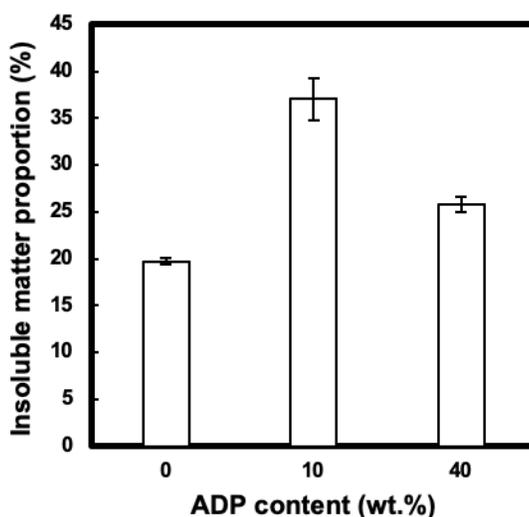


Fig. 2. Insoluble matter (IM) of OPT with added ADP

FTIR Measurement

The FTIR spectra of OPT particles, OPT extract, and each IM are shown in Fig. 3. In the OPT particles, seven characteristic absorption peaks were observed at 1630, 1515, 1405, 1361, 1241, 1077, and 1051 cm^{-1} . The absorption at 1630 cm^{-1} is probably associated with the C=O stretch of hemicelluloses (Sun *et al.* 1999). The peak at around 1515 cm^{-1} indicates C=C stretching of the aromatic ring (lignin) (Lee *et al.* 2018). The peaks at 1405, 1361, and 1241 cm^{-1} represent O-H and C-H bendings (Sun *et al.* 1999). The peaks at 1077 and 1051 cm^{-1} can be attributed to the C-OH bending from hydroxyl groups in polysaccharides (Kačuráková *et al.* 1994). In the OPT extract, all of the peaks except for 1515 cm^{-1} were observed. This means that OPT extract showed similar hemicellulose-indicative absorbances to OPT particles but did not show lignin-related absorbance.

However, the addition of ADP to OPT extract after boiling treatment (IM) showed two new peaks, at around 1515 and 792 cm^{-1} , that were not found in the OPT extract spectra. Umemura *et al.* (2017) attributed the peak around 1510 cm^{-1} to C=C of the furan ring. Also, Sun *et al.* (2019) attributed the peak at around 780 cm^{-1} to CH=CH of the furan ring, and this observation might be related to the production of 5-hydroxymethylfurfural (5-HMF) from the sugar compound on the OPT extract to form a furan polymer. Therefore, the peaks at around 1515 and 792 cm^{-1} might be attributable to the formation of the furan ring in the IM.

Also, the peak at around 1630 cm^{-1} showed increasing absorbance intensity in all IM samples irrespective of ADP content.

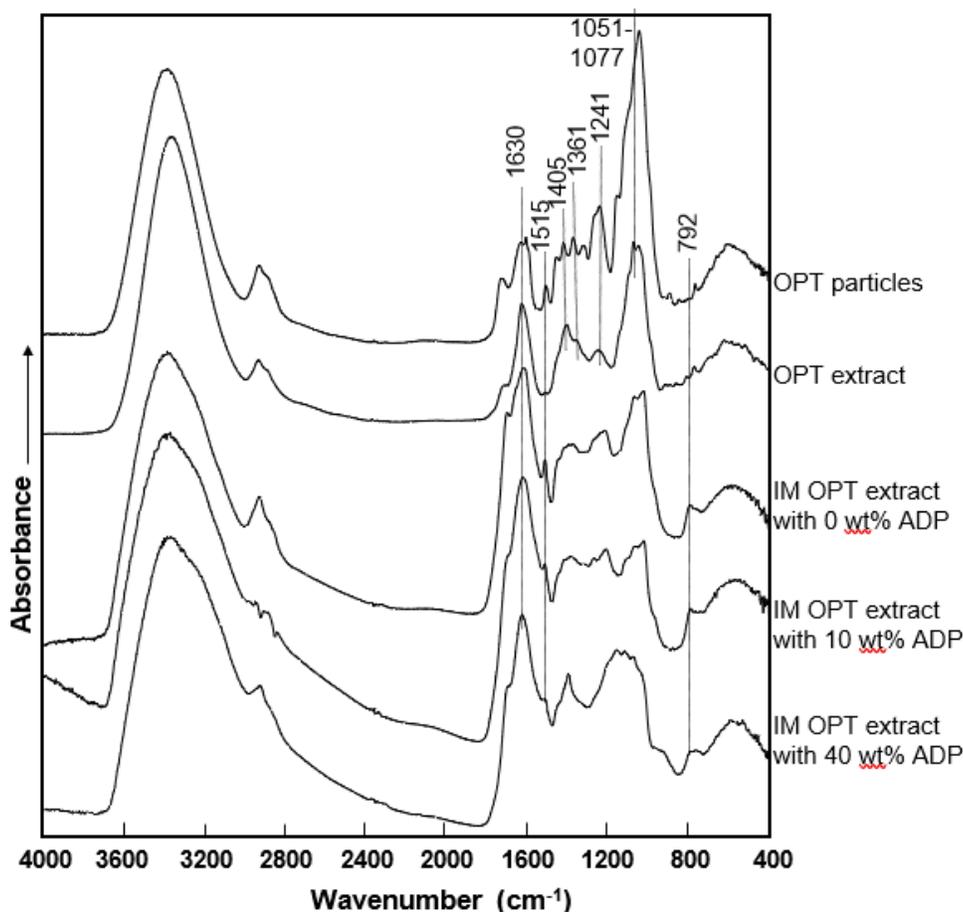


Fig. 3. FTIR spectra of OPT particles, OPT extract, and IM of OPT with added ADP (wt.%)

The FTIR results showed that the IM contained a furan ring and a carbonyl group. Therefore, it was considered that the water-soluble component was easy to change into insoluble matter containing furan and carbonyl compounds due to heating and to the addition of ADP.

Thermal Analysis Measurement

The thermal behaviors of OPT particles, OPT extract, and each IM were analyzed by DSC and TGA. The results of DSC are shown in Fig. 4. The DSC curve for the OPT particle had one endothermic peak at around $70\text{ }^{\circ}\text{C}$ and two broad exothermic peaks at approx. 270 and $330\text{ }^{\circ}\text{C}$. Meanwhile, the DSC curve for the OPT extract had two endothermic peaks at around 85 and $135\text{ }^{\circ}\text{C}$ and one broad exothermic peak at around $230\text{ }^{\circ}\text{C}$. The DSC curves of the mixtures of the various IMs were similar regardless of the ADP content; each curve showed two endothermic peaks at around 110 and $230\text{ }^{\circ}\text{C}$.

The results of thermogravimetric (TG) and derivative TG (DTG) analyses are shown in Fig. 5. And also the mass loss of the samples at specified temperature are summarised in Table 2. In the TG curves (a), the OPT particles showed reduced weight at temperatures between 200 and $350\text{ }^{\circ}\text{C}$. According to the DTG curves (b), the OPT particles lost significant

weight at ~310 °C. The onset temperature of weight reduction of the OPT extract was about 135 °C (a), and three-step weight reduction was observed in the DTG curve (b): the first step at around 179 °C, the second at around 213 °C, and the third at around 279 °C. Meanwhile, the IM results had similar weight-reduction onset temperatures at around 200 °C, and one broad weight reduction that started around 200 °C (a) was observed in the DTG curve (b) irrespective of the ADP content.

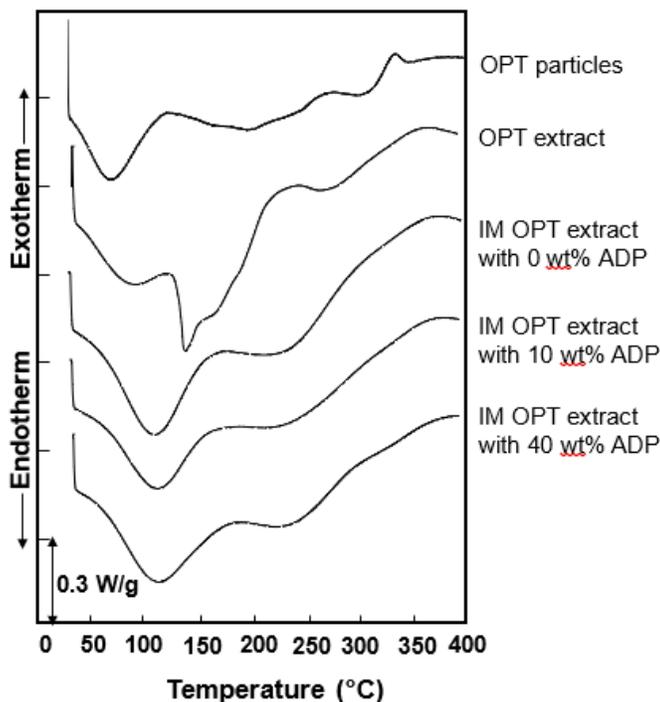


Fig. 4. DSC graph of OPT particles, OPT extract, and IM of OPT with added ADP (wt.%)

Table 2. Mass Loss (%) of the Samples by Thermogravimetric Analysis (TGA)

Samples	Mass loss observed by TGA (%) at temperature range (°C)								DTG peak temp. (°C)
	20-100	100-150	150-200	200-250	250-300	300-350	350-400	residual mass	
OPT particles	4.59	1.24	4.58	10.27	25.37	24.32	2.8	26.83	310.5
OPT extract	1.14	6.64	15.08	11.24	9.76	6.92	4.07	45.15	179.8 213.8 279.1
IM OPT extract with 0 wt% ADP	3.08	0.57	1.51	6.61	12.27	11.93	9.41	54.62	284.3
IM OPT extract with 10 wt% ADP	2.99	0.55	1.35	5.06	8.95	10.75	9.54	60.81	326.2
IM OPT extract with 40 wt% ADP	3.22	1.01	1.96	6.01	8.93	8.24	7.23	63.40	263.6

The thermal behaviors of OPT particles and OPT extract differed due to the differences in chemical compounds, as the OPT extract consisted of low-molecular-weight compounds compared to OPT particles. The endothermic peaks of DSC of around 70 to 85 °C observed in OPT particles and OPT extract indicated the weakening effect of hydrogen bonds between carbohydrates (Mehrotra *et al.* 2010); this was supported by the fact that the TG and DTG curves that did not show clear weight reduction in that temperature range. The exothermic peak at around 270 °C on OPT particles would be attributable to the decomposition of hemicellulose and the slower decomposition of lignin, according to Yang *et al.* (2007). Shebani *et al.* (2009) suggested that the exothermic peak at around 330 °C indicates the decomposition of cellulose. These phenomena were also supported by the weight reduction between 200 and 330 °C in TG of OPT particles. According to Hurtt *et al.* (2004), the melting peak temperatures of fructose and glucose were around 131 and 159 °C, respectively. This was similar to the finding of an endothermic peak at around 135 °C on the OPT extract; wherein it shows in Table 1 that the OPT extract contains glucose and fructose.

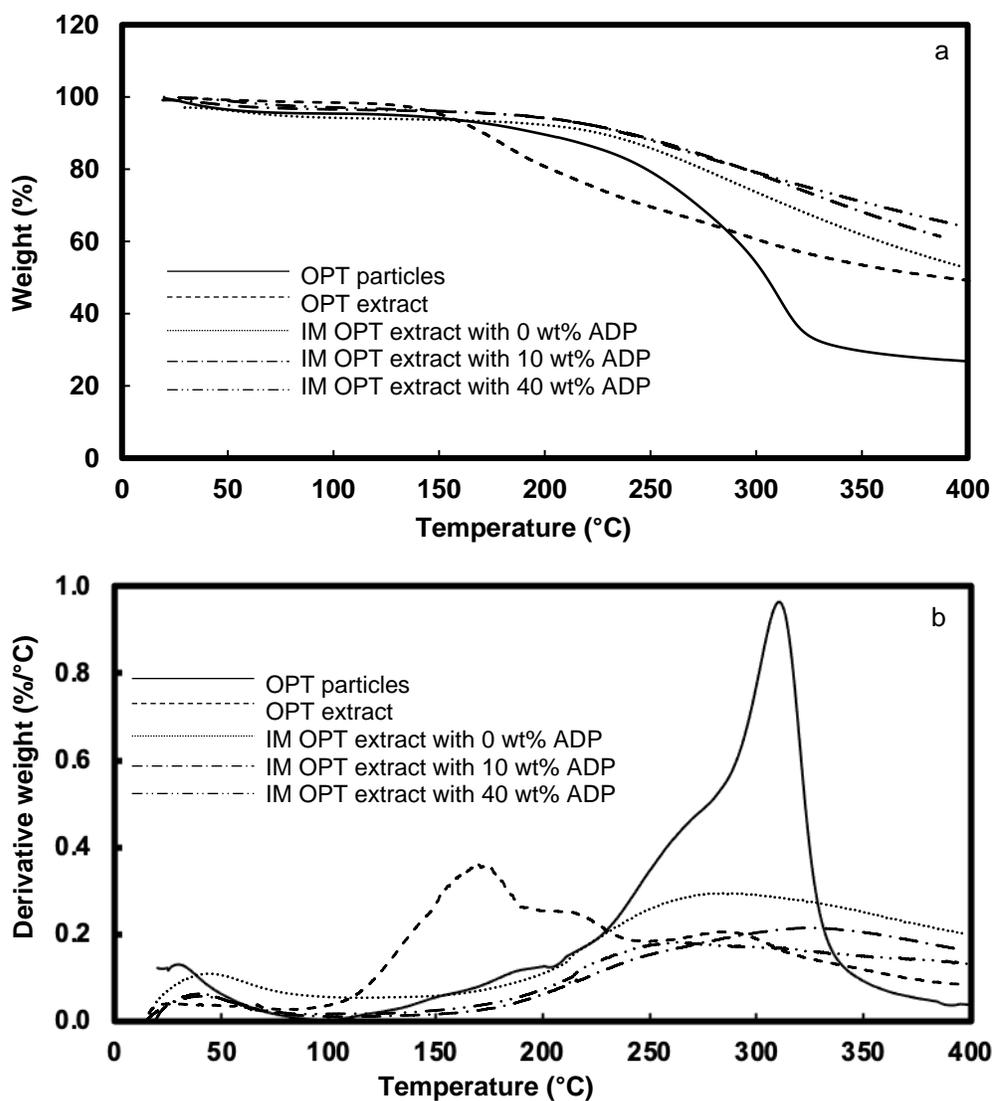


Fig. 5. Thermogravimetric (TG) (a) and Derivative TG (DTG) (b) graphs of OPT particles, OPT extract, and IM of OPT with added ADP (wt%)

The DSC and TGA of the IM mixtures showed similar behaviors regardless of ADP content. They showed the upward shifts in the onset temperature and weight reduction temperature of the IM mixtures compared to the OPT extract and the OPT particles. These results indicated that the OPT extract incurred changes in some high-molecular-weight substances due to heating and ADP addition, as indicated by the higher temperature of weight reduction. As a result, the IM mixtures had good thermal stability that contained high-molecular-weight substances.

Water Resistance Properties of Binderless Particleboard Using Hot-Water-Extracted Particles

To investigate the effects of the water-soluble extract on binderless particleboards with ADP added, boards were manufactured using hot-water-treated particles. Figure 6 shows the TS and WA of the particleboards at various ADP contents. The water resistance of the particleboards improved with the addition of ADP. The TS values of the particleboard without ADP addition (0 wt%) was 140%. The addition of ADP significantly decreased the TS values, which were 33% and 28% for 10 wt% and 40 wt% ADP additions, respectively. This was different from the authors' previous study (Komariah *et al.* 2019), in which binderless particleboard (without ADP addition) using untreated particles had a TS value of 49%. The difference was similar to that reported by Lamaming *et al.* (2014), in which an OPT binderless board with hot-water-treated particles had a higher TS value than boards made with untreated particles. The same trend observed for TS was also seen for WA values. The WA value of the particleboard without ADP addition (0 wt%) was 142%, whereas the addition of ADP 10 wt% and 40 wt% resulted in significantly decreased values, to 60.55% and 46.86%, respectively. Meanwhile Komariah *et al.* (2019) showed that binderless particleboard (without ADP addition) using untreated particles had a WA value of about 65%. Interestingly, the 10 and 40 wt% ADP did not differ significantly ($p>0.05$) in TS or WA values. This indicated that the addition of ADP played an important role in increasing water resistance regardless of the amount added. However, compared to previous study (Komariah *et al.* 2019), the TS and WA values of treated particles were higher than those of untreated particles.

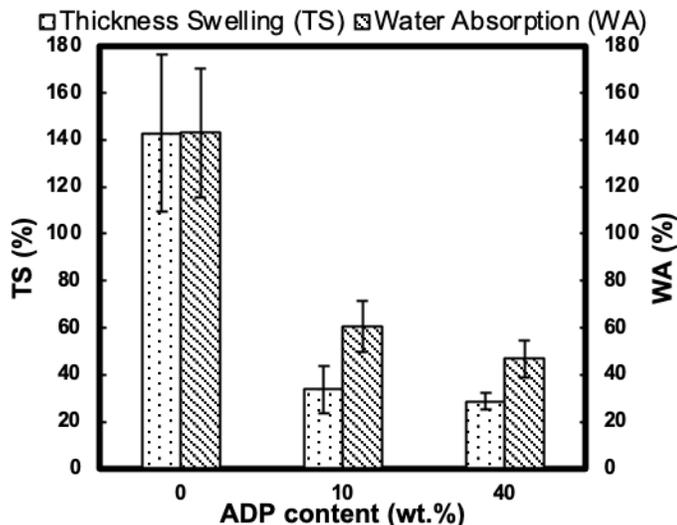


Fig. 6. Thickness swelling and water absorption of the binderless particleboards

The changes in thickness and weight by the cyclic aging treatment were observed further to investigate the particleboard's water resistance under severe conditions. The results are shown in Fig. 7. The extent of the thickness change at each stage of the cyclic aging treatment decreased with the addition of ADP. The specimens with ADP maintained their shapes throughout the stages of the cyclic aging treatment. However, the specimen without ADP (0 wt%) decomposed while being boiled in water for 4 h. The thickness change values of particleboards with 10 and 40 wt% ADP addition in the final stage of cyclic aging treatment were 32.7 and 43.0%, respectively. These values were higher than those of Komariah *et al.* (2019), which were about 0.2 and 8.63%, respectively, when using untreated particles. The weight changes of the particleboards also decreased with the addition of ADP, regardless of ADP content, compared to that without ADP addition.

The results showed the importance of extract consisting of components that prevent water absorption. In this case, when the extract was removed, ADP would decompose the chemical components of the OPT particles to some degree, which subsequently turned into an insoluble component that prevented water absorption, as shown in the insoluble matter rate value (Fig. 2). In other words, the addition of ADP and the existence of the extract are important in the manufacturing of water-resistant particleboard.

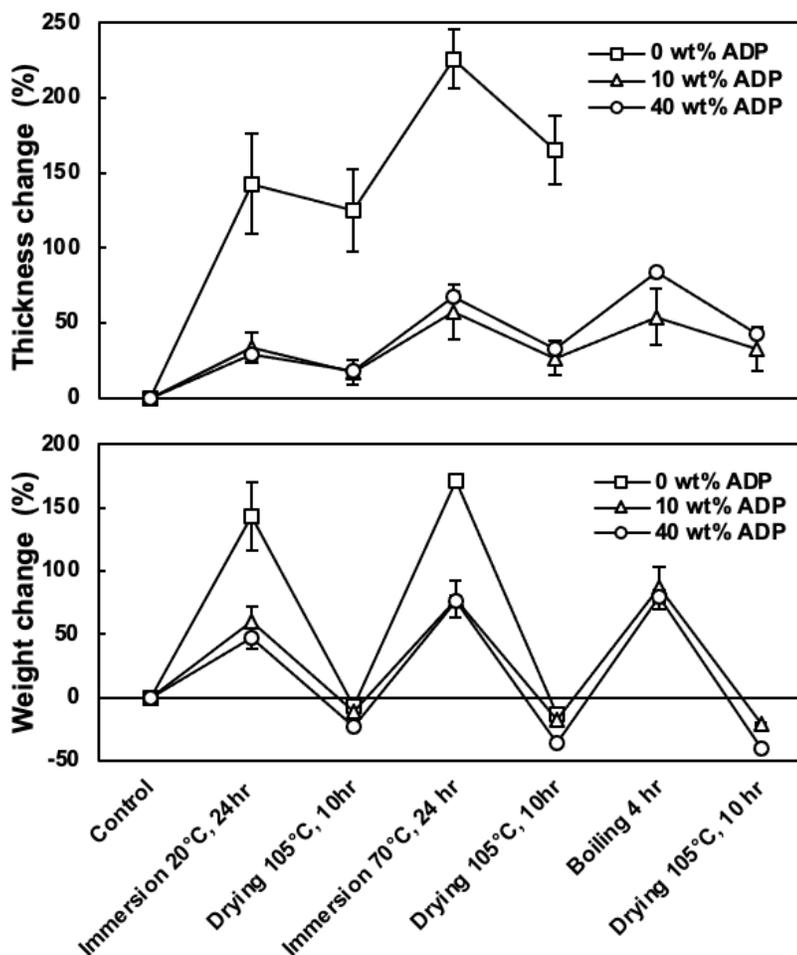


Fig. 7. Thickness changes and weight changes of the binderless particleboards in the cyclic aging treatment

Mechanical Properties of Binderless Particleboards

Figure 8 shows the bending properties of the particleboards with ADP addition. The MOR value of the particleboard without ADP (0 wt%) was 2.80 MPa. The MOR values approximately tripled when 10 wt% ADP content was added compared to the particleboard without ADP. The addition of 40 wt% ADP showed a slightly lower MOR value. This was due to the excess of ADP that caused a decrease in particle proportion and thus did not contribute to the bondability of the particleboards.

The performance of the particleboard without ADP was similar to the result of Lamaming *et al.* (2014) using treated particles; the MOR value was 3.92 MPa. However, this value was lower than those in Komariah *et al.* (2019) and Lamaming *et al.* (2014), which were 5.23 and 5.25 MPa, respectively, with untreated particles. Furthermore, the hot-water treatment of the particles did not affect the MOR values of the particleboards with ADP addition, as the present results were similar to those in Komariah *et al.* (2019) with untreated particles. The trend with MOR values was also seen in the MOE values of the particleboards, as shown in Fig. 7. The MOE value of the particleboard without ADP (0 wt%) was 1.15 GPa. After the addition of ADP, the MOE values doubled. This shows that adding ADP enhanced the strength characteristics of the particleboards using treated OPT particles.

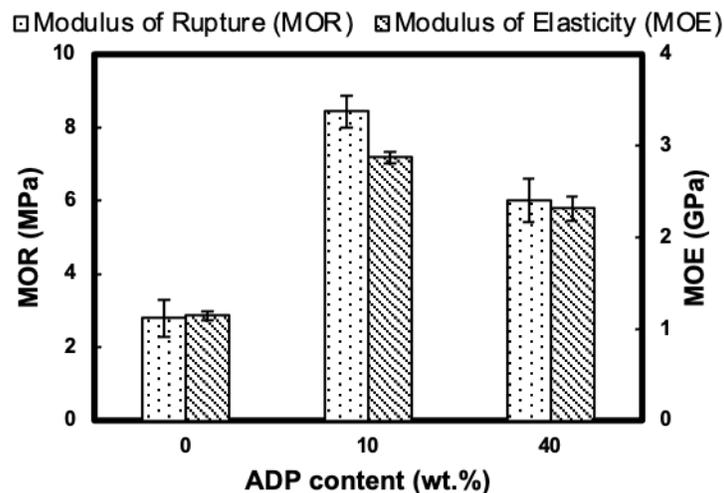


Fig. 8. Bending properties of the binderless particleboards

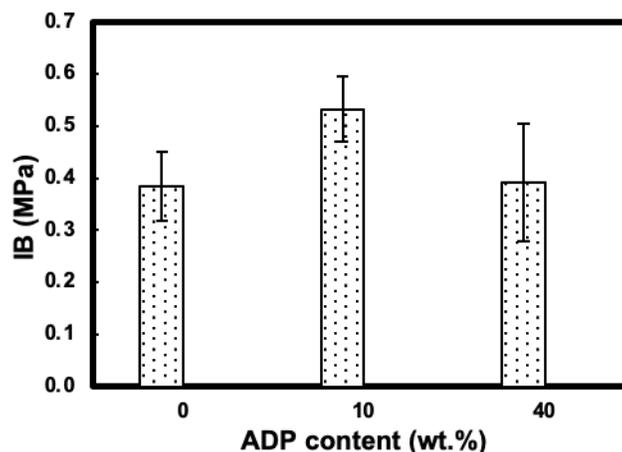


Fig. 9. Internal bond (IB) of the binderless particleboards

Figure 9 shows the IB strength of the binderless particleboards. It can be seen that the IB strength was increased with the addition of 10 wt% ADP. The IB of the particleboard without ADP (0 wt%) was 0.38 MPa, similar to the result of Komariah *et al.* (2019) with untreated particles. This result showed that the hot-water treatment hardly affected the IB strength of binderless particleboard. In addition, the IB strength of binderless particleboard with the addition of 10 wt% ADP was increased by 70%, to 0.53 MPa. Judging from the results obtained, the addition of 10 wt% ADP could improve the mechanical properties of particleboards using treated particles.

CONCLUSIONS

1. The oil palm trunk (OPT) extract changed into an insoluble substance by heating, and the addition of 10 wt% ammonium dihydrogen phosphate (ADP) doubled the insoluble substance amount. Based on the FTIR results, the formation of furan compounds derived from sugars was inferred. And the thermal analysis showed that the formed insoluble substance had good thermal stability, suggesting a high-molecular-weight substance.
2. The boards' physical and mechanical properties irrespective of ADP addition tended to be inferior to those of previously reported particleboards without extracted treatment. In particular, the water-soluble extract was essential for the water resistance of the particleboard. This means that the water-soluble extract contributed to bonding of the particleboard.
3. The particleboard with 10 wt% ADP showed the best values for modulus of rupture (MOR) (8.4 MPa), internal bond (IB) (0.53 MPa), and thickness swelling (TS) (33.8%). In addition, the particleboard had good water resistance in cyclic aging treatment. Therefore, ADP addition was judged to be effective for improving particleboard properties even in the absence of water-soluble extract.

ACKNOWLEDGMENTS

The authors are grateful for the support of the Science and Technology Research Partnership for Sustainable Development (SATREPS) and the Indonesian Institute of Sciences (LIPI) for supplying raw materials. The first author also gratefully acknowledges financial support from the Ministry of Finance of the Republic of Indonesia through an Indonesia Endowment Fund for Education Scholarship (BUDI LN-LPDP).

REFERENCES CITED

- Anglès, M. N., Ferrando, F., Farriol, X., and Salvadó, J. (2001). "Suitability of steam exploded residual softwood for the production of binderless panels. Effect of the pre-treatment severity and lignin addition," *Biomass Bioenergy* 21(3), 211-224. DOI: 10.1016/S0961-9534(01)00031-9
- Baskaran, M., Hashim, R., Sulaiman, O., Awalludin, M. F., Sudesh, K., Arai, T., and Kosugi, A. (2019). "Properties of particleboard manufactured from oil palm trunk waste using polylactic acid as a natural binder," *Waste Biomass Valor.* 10(1), 179-186.

- DOI: 10.1007/s12649-017-0026-7
- Bockel, S., Mayer, I., Konnerth, J., Harling, S., Niemz, P., Swaboda, C., Beyer, M., Bieri, N., Weiland, G., and Pichelin, F. (2019). "The role of wood extractives in structural hardwood bonding and their influence on different adhesive systems," *Int. J. Adhes. Adhes* 91, 43-53. DOI: 10.1016/j.ijadhadh.2019.03.001
- Food and Agriculture Organization of the United Nations (FAO) and United Nations Environment Programme (UNEP) (2020). *The State of the World's Forests 2020. Forest, Biodiversity and People*, Rome, Italy. DOI: 10.4060/ca8642en
- Hartono, R., Iswanto, A. H., Sucipto, T., and Lubis, K. M. (2017). "Effect of particle pre-treatment on physical and mechanical properties of particleboard made from oil palm trunk," in: *7th International Symposium for a Sustainable Humanosphere*, Bogor, Indonesia. DOI: 10.1088/1755-1315/166/1/012006
- Hashim, R., Saari, N., Sulaiman, O., Sugimoto, T., Hiziroglu, S., Sato, M., and Tanaka, R. (2010). "Effect of particle geometry on the properties of binderless particleboard manufactured from oil palm trunk," *Mater. Design* 31(9), 4251-4257. DOI: 10.1016/j.matdes.2010.04.012
- Hashim, R., Said, N., Lamaming, J., Baskaran, M., Sulaiman, O., Sato, M., Hiziroglu, S., and Sugimoto, T. (2011). "Influence of press temperature on the properties of binderless particleboard made from oil palm trunk," *Mater. Design* 32(5), 2520-2525 DOI: 10.1016/j.matdes.2011.01.053
- Hermanto, I., and Massijaya, M. Y. (2018). "Performance of composite boards from long strand oil palm trunk bonded by isocyanate and urea formaldehyde adhesives," in: *2nd International Conference on Biomass*, Bogor, Indonesia. DOI: 10.1088/1755-1315/141/1/012012
- Hurttä, M., Pitkänen, I., and Knuutinen, J. (2004). "Melting behaviour of D-sucrose, D-glucose and D-fructose," *Carbohydr. Res.* 339(13), 2267-2273. DOI: 10.1016/j.carres.2004.06.022
- Ismail, A., and Mamat, M. N. (2002). "The optimal age of oil palm replanting," *Oil Palm Industry Economic Journal* (Malaysian Palm Oil Board) 2, 11-18, (<http://palmoilis.mpob.gov.my/publications/OPIEJ/opiejv2n1-2.pdf>), Accessed 21 September 2018.
- JIS A 5908 (2003). "Particleboards," Japanese Standards Association, Tokyo, Japan.
- Jumhuri, N., Hashim, R., Sulaiman, O., Noor, W., Wan, A., Salleh, K. M., Khalid, I., Saharudin, N. I., and Razali, M. Z. (2014). "Effect of treated particles on the properties of particleboard made from oil palm trunk," *Mater. Design* 64, 769-774. DOI: 10.1016/j.matdes.2014.08.053
- Kačuráková, M., Ebringerová, A., Hirsch, J., and Hromádková, Z. (1994). "Infrared study of arabinoxylans," *J Sci Food Agric* 66(3), 423-427. DOI: 10.1002/jsfa.2740660323
- Komariah, R. N., Miyamoto, T., Tanaka, S., Prasetyo, K. W., Syamani, F. A., Subyakto, Umezawa, T., Kanayama, K., and Umemura, K. (2019). "High-performance binderless particleboard from the inner part of oil palm trunk by addition of ammonium dihydrogen phosphate," *Ind. Crops Prod.* 141, 111761. DOI: 10.1016/j.indcrop.2019.111761
- Kusumah, S. S., Umemura, K., Yoshioka, K., Miyafuji, H., and Kanayama, K. (2016). "Utilization of sweet sorghum bagasse and citric acid for manufacturing of particleboard. I: Effects of pre-drying treatment and citric acid content on the board properties," *Ind. Crops Prod.* 84, 34-42. DOI: 10.1016/j.indcrop.2016.01.042
- Lamaming, J., Sulaiman, O., Sugimoto, T., Hashim, R., Said, N., and Sato, M. (2013).

- “Influence of chemical components of oil palm on properties of binderless particleboard,” *BioResources* 8(3), 3358-3371. DOI: 10.15376/biores.8.3.3358-3371
- Lamaming, J., Hashim, R., Sulaiman, O., Sugimoto, T., Sato, M., and Hiziroglu, S. (2014). “Measurement of some properties of binderless particleboards made from young and old oil palm trunks,” *Meas. J. Int. Meas. Confed.* 47, 813-819. DOI: 10.1016/j.measurement.2013.10.007
- Lange, W., and Simatupang, M. H. (1994). “Chemical composition of oil palm trunk extractives,” in: *3rd Tropical Pulp and Paper Conference*, Kuala Lumpur, Malaysia.
- Lee, S. H., Ashaari, Z., Ang, A. F., Abdul Halip, J., Lum, W. C., Dahali, R., and Halis, R. (2018). “Effects of two-step post heat-treatment in palm oil on the properties of oil palm trunk particleboard,” *Ind. Crops Prod.* 116, 249-258. DOI: 10.1016/j.indcrop.2018.02.050
- Mansor, H., and Ahmad, A. R. (1990). “Carbohydrates in the oil palm stem and their potential use,” *J. Tropical For. Sci.* 2(3), 220-226.
- Mehrotra, R., Singh, P., and Kandpal, H. (2010). “Near infrared spectroscopic investigation of the thermal degradation of wood,” *Thermochim. Acta* 507-508, 60-65. DOI: 10.1016/j.tca.2010.05.001
- Murai, K., and Kondo, R. (2011). “Extractable sugar contents of trunks from fruiting and nonfruiting oil palms of different ages,” *J. Wood Sci.* 57, 140-148. DOI: 10.1007/s10086-010-1144-z
- Prabuningrum, D. S., Massijaya, M. Y., Hadi, Y. S., Abdillah, I. B. (2020). “Physical-mechanical properties of laminated board made from oil palm trunk (*Elaeis guineensis* jacq.) waste with various lamina compositions and densifications,” *J. Korean Wood Sci. Technol.* 48(2), 196-205. DOI: 10.5658/WOOD.2020.48.2.196
- Shebani, A. N., van Reenen, A. J., and Meincken, M. (2009). “The effect of wood extractives on the thermal stability of different wood-LLDPE composite,” *Thermochim. Acta* 481(1-2), 52-56. DOI: 10.1016/j.tca.2008.10.008
- Sun, R. C., Fang, J. M., Mott, L., and Bolton, J. (1999). “Fractional isolation and characterization of polysaccharides from oil palm trunk and empty fruit bunch fibres,” *Holzforschung* 53(3), 253-260. DOI: 10.1515/HF.1999.043
- Sun, S., Zhang, M., Umemura, K., and Zhao, Z. (2019). “Investigation and characterization of synthesis conditions on sucrose-ammonium dihydrogen phosphate (SADP) adhesive: Bond performance and chemical transformation,” *Materials* 12(24), 4078. DOI: 10.3390/ma1224078
- Tye, Y. Y., Lee, K. T., Wan Abdullah, W. N., and Leh, C. P. (2016). “The world availability of non-wood lignocellulosic biomass for the production of cellulosic ethanol and potential pretreatments for the enhancement of enzymatic saccharification,” *Renew. Sust. Energ. Rev.* 60, 155-172. DOI: 10.1016/j.rser.2016.01.072
- Umemura, K., Hayashi, S., Tanaka, S., and Kanayama, K. (2017). “Changes in physical and chemical properties of sucrose by the addition of ammonium dihydrogen phosphate,” *J. Adhes. Soc. Japan* 53(4), 112-117. DOI: 10.11618/adhesion.53.112
- van Dam, J. E. G., van den Oever, M. J. A., Keijsers, E. R. P., van der Putten, J. C., Anayron, C., Josol, F., and Peralta, A. (2006). “Process for production of high density/high performance binderless boards from whole coconut husk. Part 2: Coconut husk morphology, composition and properties,” *Ind. Crops Prod.* 24(2), 96-104. DOI: 10.1016/j.indcrop.2005.03.003
- Widyorini, R., Umemura, K., Soraya, D. K., Dewi, G. K., and Nugroho, W. D. (2005). “Manufacture and properties of binderless particleboard from bagasse. I: Effects of raw

material type, storage methods, and manufacturing process,” *J. Wood Sci.* 51(6), 26-32. DOI: 10.1007/s10086-005-0713-z

- Widyorini, R., Umemura, K., Soraya, D. K., Dewi, G. K., and Nugroho, W. D. (2019). “Effect of citric acid content and extractives treatment on the manufacturing process and properties of citric acid-bonded *Salacca frond* particleboard,” *BioResources* 14(2), 4171-4180. DOI: 10.15376/biores.14.2.4171-4180
- Yang, H., Yan, R., Chen, H., Lee, D. H., and Zheng, C. (2007). “Characteristics of hemicellulose, cellulose and lignin pyrolysis,” *Fuel* 86(12-13), 1781-1788. DOI: 10.1016/j.fuel.2006.12.013
- Zhao, Z., Hayashi, S., Xu, W., Wu, Z., Tanaka, S., Sun, S., Zhang, M., Kanayama, K., and Umemura, K. (2018). “A novel eco-friendly wood adhesive composed by sucrose and ammonium dihydrogen phosphate,” *Polymers* 10(11), 1251. DOI: 10.3390/polym10111251

Article submitted: April 1, 2021; Peer review completed: June 20, 2021; Revised version received and accepted: July 12, 2021; Published: July 15, 2021.

DOI: 10.15376/biores.16.3.6015-6030