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Original Article

Effect of poly (ethylene-co-vinyl acetate) grade on water resistance and mechanical properties of particleboard

Salwanee Waelaeh[1], Varaporn Tanrattanakul[1*], Kittisak Poolyarat[2], Pimpatima Hamaphat[2], and Kittitat Junnam[2]

[1] *Department of Materials Science and Technology, Faculty of Science, Prince of Songkla University, Hat Yai, Songkhla, 90110 Thailand*

[2] *Panel Plus Co., Ltd., Hat Yai, Songkhla, 90230 Thailand*

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Abstract

Three-layer particleboard was prepared in the laboratory using a similar composition and processing condition to those used in the plant production of particleboard. Poly(ethylene-co-vinyl acetate) (EVA) was mixed with natural rubber-wood particles in the core layer to reduce the water resistance of the particleboard. The properties of three EVA grades with vinyl acetate contents of 19%, 22%, and 28% were melt indexes of 530, 1.8, and 6.8 g/10 min, melting temperatures of 84, 86, and 75 °C, and molecular weights of 29000, 68000, and 61000 g/mol, respectively. The addition of 5% EVA significantly decreased the thickness swelling and water absorption properties of the particleboard. The mechanical properties of the particleboard that were determined were the modulus of rupture, modulus of elasticity, internal bond strength, and screw holding force. A statistical analysis of the data showed that the addition of 5% EVA did not deteriorate the mechanical properties. EVA22 seemed to be the optimal grade.

Keywords: particleboard, poly(ethylene-co-vinyl acetate), thickness swelling, water absorption, mechanical properties

1. Introduction

Three-layer particleboard is a well-known wood-based panel used for furniture such as cupboards, shelves, flooring and decorative interior. It is composed of top and bottom surface layers made of fine wood particles, while the core layer contains coarse wood particles (Irle, Barbu, Reh, Bergland, & Rowell, 2012). These three layers are bonded together into a sheet with thermoset adhesive under applied heat and pressure. Urea formaldehyde (UF) resin is generally used as an adhesive in the industrial production process because of low cost, good bonding properties, short press time, and relatively high water resistance (Irle & Barbu, 2010). However, the particleboard which is glued with UF has limited performance when exposed to very high humidity for long periods of time compared with other thermoset adhesives. Voids or gaps between the wood particles are normally found in the core layer more than in the surface layers because coarse wood particles were employed in the core layer. These voids or gaps plus the hydrophilic nature of wood particles affect the water resistance of the particleboard. Therefore, much research has focused on adding thermo-plastics into the wood particles. By adding 5-30 wt% low density polyethylene (LDPE) in the core layer, the dimensional stability and the core bond strength of the particleboard increased with increasing LDPE powder content (Ayrilmis, Kwon, & Han, 2012). A mixture of LDPE powder (5-30 wt%) with rice husk particles in the core layer provided a decrease in the thickness swelling and water absorption of the particleboard and an increase in the internal bond strength (Kwon, Ayrilmis, & Han, 2014). Recently our group evaluated the effects of different grades of LDPE added into

*Corresponding author
Email address: varaporn.t@psu.ac.th

the core layer on the physical and mechanical properties of the particleboard (Waelaeh, Tanrattanakul, Phunyarat, Panu pakorn, & Junnam, 2017). We found that all LDPEs increased the water resistance of the particleboard and LDPE in a powder form was more efficient than the pellet form. Recycled polyethylene applied also to the surface layer increased the water resistance and mechanical properties of the particleboard (Kargarfard & Jahan-Latibari, 2012). Recycled high density polyethylene (HDPE) from milk bottles with particle sizes of 1-3 mm was mixed with sugar cane bagasse with particle sizes of 2-15 mm to produce bagasse/plastic composite boards (Talavera, Guzmán, Richter, Dueñas, & Quirarte, 2007). It was observed that water absorption and thickness swelling of the particleboard decreased with increasing content of HDPE. A mixture of olefin polymers with an average molecular weight of 1500 to 5000 g/mol and a melting point of 45 to 80 °C was used as a waterproof agent in the surface layer of the particleboard to reduce water absorption and prevent the expansion in the thickness of the particleboard (Tsunoda, 2009). A mixture of polypropylene (PP) powder and rubber-wood fibers (60:40) increased the water resistance and internal bond strength of the wood composite (Ayrilmis & Jarusombuti, 2011). Recycled polyvinyl chloride (PVC) (10-40 wt%) added in the core layer to replace the wood particles provided better dimensional stability when the percentage of PVC increased (Thanigai, Sangeetha, Pandey, Kiran, & Prakash, 2013). A mixture of recycled polyethylene terephthalate (30-60 wt%) and sawdust decreased the moisture content and water resistance of flat-pressed wood plastic composites (Rahman *et al.*, 2013). Poly(ethylene-co-vinyl acetate) (EVA) has been used as a coupling agent in poplar wood flour/HDPE wood composite, and it was found that the water resistance and mechanical properties were enhanced with increasing EVA content (Li, Li, Hu, & Li, 2012).

Additionally other materials have been used in order to improve a specific property. As examples, Ayrilmis, Buyuksari, and Avci (2009) used waste tire rubber crumbs mixed with fine and coarse wood particles to increase the water resistance of particleboard. Poppy husk biomass and pine wood were used to prepare particleboard (Keskin, Kucuktuvek, & Guru, 2015). Glass and rock wools were added to enhance the fire resistance of particleboard (Ülker & Burdurlu, 2015), and date palm branches and vermiculite were employed to improve the sound absorption coefficient of particleboard (Ghofrani, Ashori, & Mehrabi, 2017). Ashori, & Nourbakhsh (2008) prepared a single-layer particleboard using low quality materials such as eucalyptus (*Eucalyptus camaldulensis*), mesquite (*Prosopis juliflora*), saltcedar (*Tamarix stricta*), and date palm (*Phoenix dactylifera*) wood.

Based on our knowledge, EVA has not been used in particleboard. EVA is an interesting polymer to enhance the water resistance of particleboard. EVA has low temperature-flexibility, toughness, and good adhesion to different materials that includes plastics and woods (Lu *et al.*, 2013). Since EVA is a copolymer of ethylene and vinyl acetate, the properties of EVA can be controlled and the properties depend on the vinyl acetate content. The objective of this study was to improve the water resistance of the particleboard by adding 5% EVA into the core layer. The water resistance was determined in terms of thickness swelling and water absorption at 2 h and 24 h. The mechanical properties of the particleboard were also investigated. Three grades of EVA were selected to determine the key parameter affecting the water resistance and mechanical properties. The parameters of interest were vinyl acetate content, melt index (MI), melting temperature (T_m), and molecular weight.

2. Materials and Methods

2.1 Materials

Fine and coarse natural rubber-wood particles and all chemicals were kindly provided by Panel Plus Co., Ltd., Thailand. The diameter range of fine wood particles was 0.23-1.70 mm. The thickness and length ranges of the coarse wood particles were 1.71-8.00 mm and 10-30 mm, respectively. Urea formaldehyde (UF) adhesive had a 1:1.3 U:F molar ratio and the solid content was 67%. Ammonium chloride (NH_4Cl) was used as a hardener. Paraffin wax emulsion containing 60% solid content was used as a sizing agent to reduce the water absorption of the wood particles. Three grades of EVA with different vinyl acetate content were used (Table 1). A T_m of <100 °C of the EVA was required in order to obtain a complete melt of EVA in the particleboard during a very short hot-pressing time.

Table 1. Physical properties of the EVAs.

Code	Manufacturer	Vinyl acetate content (%)	Melt index (g/10 min)	Melting Temperature (°C)
EVA19	Global Connections Plc.	19	530	84
EVA22	TPI Polene Co., Ltd.	22	1.8	86
EVA28	TPI Polene Co., Ltd.	28	6.8	75

EVA19, vinyl acetate content of 19%; EVA22, vinyl acetate content of 22%; EVA28, vinyl acetate content of 28%

2.2 Preparation of three-layer particleboard

The surface and core layers were compounded separately and the compositions are listed in Table 2. For the surface layer, the paraffin wax was primarily mixed with the wood particles. Then the UF adhesive, NH_4Cl hardener, and water were added successively. Paraffin wax was not applied to the core layer. In the case of 5% EVA samples, the EVA pellets were blended with wood particles after mixing with UF adhesive and NH_4Cl solution. Due to the proprietary formulation of the UF adhesive and the NH_4Cl solution, information on these materials was not available.

The three-layer composition was controlled by a weight ratio of wood compounds as 23:56:21 (top surface: core: bottom surface). The particleboard was prepared in a forming box with dimensions of 400x400x15 mm. The manufacturing process of the three-layer particleboard in the laboratory is shown in Figure 1. The mat was pressed at 0.03 bar for 30 sec at room temperature to control the mat height. Then the mat was hot pressed in a laboratory press under a pressure of 130 bar for 360 sec. The temperatures of the top and the bottom plates of the press were 185 °C and 180 °C, respectively.

Table 2. Compositions of wood compounds.

Sample	Surface layer		Core layer	
	Wood (%)	Wax (%)*	Wood (%)	EVA (%)
0%	44	0.5	56	-
5% EVA	44	0.5	51	5

* Based on weight of dried SL wood particles
EVA, Poly(ethylene-co-vinyl acetate).

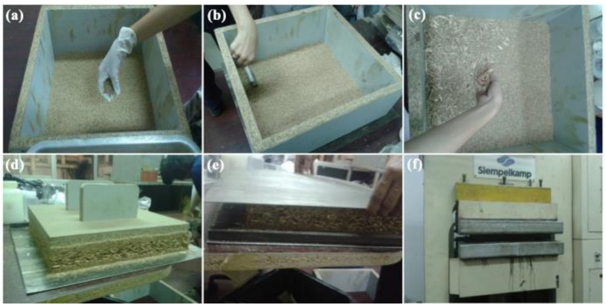

Figure 1. Manufacturing process of a three-layer particleboard: (a) placing wood compound into a forming box, (b) distributed evenly by brush, (c) placing wood compounds for the core layer and top surface layer, (d) pre-pressing the mat at room temperature, (e) placing 15-mm thick metal bars at the edges and a caul plate on top of the mat, and (f) hot-pressing under a laboratory press.

2.3 Testing of the physical properties

The particleboard was conditioned at 20 °C and 65% relative humidity for 24 h before testing the physical and mechanical properties. Specimens were cut from the particleboard sheet. The dimensions and density of the specimens were investigated using an IB600 series IMAL® Machine according to EN325 (CEN, 1993) and EN323 (CEN, 1993), respectively. The moisture content of the particleboard was examined according to EN322 (CEN, 1993). Four specimens with dimensions of 50×50×15 mm were weighed before (W_1) and after (W_2) drying in the oven at 105 °C for 4 h. The moisture content was calculated from Equation 1.

$$\textit{Moisture content}\ (\%) \quad = \quad \frac{W_1 - W_2}{W_2} \times 100 \qquad (1)$$

The thickness swelling and water absorption were measured in accordance with EN317 (CEN, 1993). Eight specimens with the dimensions of 50×50×15 mm were employed. The experiments were carried out in water at 20±2 °C for 2 h and 24 h and the results were calculated according to Equations 2 and 3.

$$\textit{Thickness swelling}\ (\%) \quad = \quad \frac{T_2 - T_1}{T_1} \times 100 \qquad (2)$$

$$\textit{Water absorption}\ (\%) \quad = \quad \frac{W_2 - W_1}{W_1} \times 100 \qquad (3)$$

where T_1 and T_2 were the thicknesses of the specimen before and after immersion, respectively, and W_1 and W_2 were the weights of the specimen before and after immersion, respectively.

2.4 Testing of the mechanical properties

All mechanical properties were tested using the IB600 series IMAL® Machine. The modulus of rupture (MOR) and modulus of elasticity (MOE) were determined according to EN310 (CEN, 1993). Six specimens with dimensions of 300×50×15 mm were employed. The testing speed was 10 mm/min. MOR and MOE were calculated from load deflection curves according to the following equations:

$$MOR\ (MPa) \quad = \frac{3PL}{2Bt^2} \qquad (4)$$

$$MOE\ (MPa) \quad = \frac{L^3 \Delta W}{4Bt^2 \Delta S} \qquad (5)$$

where P was the maximum load (N), L was the span (mm), and B and t were the specimen width (mm) and thickness (mm), respectively. ΔW was the load at the proportional limit (N) and ΔS was the deflection corresponding to ΔW.

The internal bond (IB) strength was examined in accordance with EN319 (CEN, 1993) using four specimens with dimensions of 50×50×15 mm. The testing speed was 2 mm/min. The force at break (P_b) was recorded and IB was calculated using Equation 6, where *l* was the length of the specimen.

$$IB\ (N/mm^2) \quad = \frac{P_b}{Bl} \qquad (6)$$

The surface and edge screw holding forces were determined according to EN320 (CEN, 1993). Four specimens with dimensions of 50×50×15 mm were tested at the testing speed of 2 mm/min. The applied force was recorded.

2.5 Statistical analysis of data

The analysis of variance (ANOVA) was performed using SPSS software (version 11.5) at 95% confidence levels ($P<0.05$). The significant differences among the samples were analyzed using the least significant difference (LSD) method. Different letters (e.g., A, B, C, AB, and BC) were used to identify the samples when the average values were significantly different at $P<0.05$.

2.6 Micrograph characterization

The molten EVA in the particleboard was observed from the fracture surfaces of the IB tested samples. A stereo microscope (Olympus® D617) was employed and photographs were recorded at magnifications of 7x and 20x.

3. Results and Discussion

3.1 Physical properties

Moisture content, thickness swelling, and water absorption of all particleboards are shown in Figures 2-4, respectively. Table 3 shows the results of the statistical analysis which were used to determine the significant effects of the EVA type on the properties. The data indicated any group which was significantly different from others at 95% confidence level. Based on the LSD, the highest value was set as "A" and the values are ranked in the following order: A>AB>B>BC>C. A, B, and C differed significantly from each other (P<0.05). Between A and AB (B and AB), they were insignificantly different, which was similar to B-BC and C-BC.

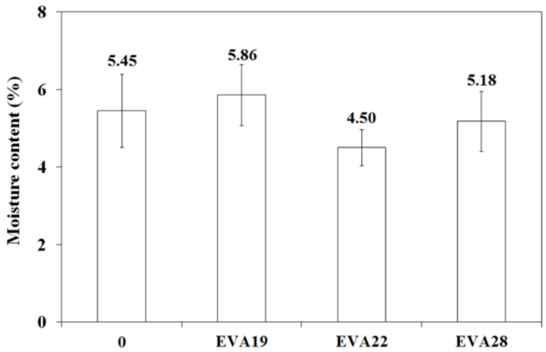

Figure 2. Effect of EVA on the moisture content of particleboards.

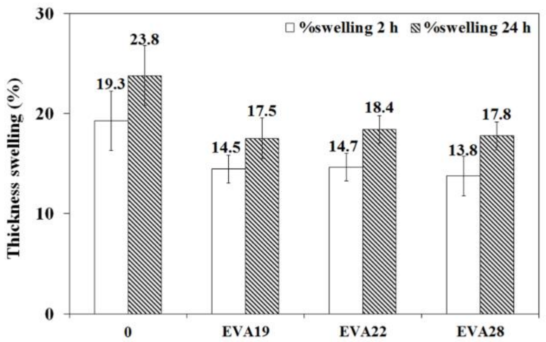

Figure 3. Effect of EVA on the thickness swelling at 2 h and 24 h of particleboards.

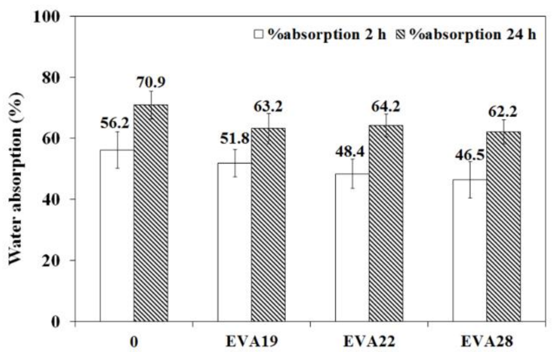

Figure 4. Effect of EVA on the water absorption at 2 h and 24 h of particleboards.

Table 3. Physical properties of the particleboards.

Sample	Moisture content (%)	Thickness swelling (%)		Water absorption (%)	
		2 h	24 h	2 h	24 h
0%	5.45^{AB} (0.95)	19.3^{A} (3.0)	23.8^{A} (3.0)	56.2^{A} (5.9)	70.9^{A} (4.6)
5% EVA19	5.86^{A} (0.79)	14.5^{B} (1.4)	17.5^{B} (2.0)	51.8^{B} (4.5)	63.2^{B} (5.0)
5% EVA22	4.50^{C} (0.47)	14.7^{B} (1.3)	18.4^{B} (1.4)	48.4^{C} (4.8)	64.2^{B} (3.7)
5% EVA28	5.18^{B} (0.78)	13.8^{B} (2.0)	17.8^{B} (1.4)	46.5^{C} (6.0)	62.2^{B} (3.9)

Average values with different letters in the same column are significantly different at P<0.05. Numbers in parenthesis are standard deviations.
h, hour; EVA19, vinyl acetate content of 19%; EVA22, vinyl acetate content of 22%; EVA28, vinyl acetate content of 28%.

The moisture content of the control particleboard (0%) and the particleboards containing 5% EVA fulfilled the prescribed standard of EN312:2010 (CEN, 2010) for 15-mm thick particleboard which has a limit in the range of 5-13%. A significant decrease was observed in the sample containing 5% EVA22. The lowest MI and the highest T_m of EVA22 may be attributed to the lowest moisture content. A very high melt index of EVA19 may cause slightly higher moisture content. The EVA19 could flow very well and penetrate into the wood particles so that it could not cover the gaps between wood particles as well as EVA22 and EVA28 performed. Therefore, lots of voids were visually observed in the 5% EVA19 sample, which was similar to the control (0%). In contrast, the samples that contained EVA22 and EVA28 did not show clear voids.

Water resistance of the particleboard was described in terms of thickness swelling and water absorption at 2 h and 24 h. The replacement of 5% EVA into the wood particles in the core layer showed significant improvement in the water resistance in accordance with the statistical analysis. We believe that the molten EVA covered the surface of the wood particles and filled the gaps between the wood particles, which attributed to a decrease in hydrophilicity of the particleboard. A similar result was reported by Ayrilmis and Jarusombuti (2011). They concluded that the addition of polypropylene powder in the wood plastic composite reduced the thickness swelling and water absorption because the plastic powder filled the micropores of the wood and decreased the number of polar groups (i.e. hydroxyl groups) in the wood composite. As reported by Li *et al.* (2012), the addition of EVA as a coupling agent in the wood composite (HDPE/wood flour) decreased the stretching vibration band of the OH groups of the wood composite. This resulted in the reduction of water uptake and thickness swelling of the wood composite. They also mentioned that wood flour was coated by EVA and the OH groups of the wood flour could react with the carbonyl group of EVA. In the present study only 5% EVA was added which made it difficult to investigate any change in the Fourier transform infrared spectrum of the particleboard. The total number of OH groups based on wood particles in the particleboard may decrease due to the substitution by EVA. No significant effect of EVA type on the thickness swelling was observed, and a similar behavior was also found in the water absorption at 24 h. In contrast, the EVA type affected the water absorption at 2 h. EVA22 and EVA28 provided lower water absorption than EVA19, and EVA28 showed the lowest values of water absorption among all samples. This should be due to more voids in the 5% EVA19 sample. However, after testing for 24 h the samples reached the maximum water uptake and showed a similar value.

Figure 5 illustrates the stereo micrographs of the fractured surfaces of IB tested specimens. It was observed that all EVAs in the core layer of particleboard were completely melted under the processing conditions. The behavior of melted plastic depended on the T_m, MI, and the individual characteristics of the EVA grade. Although the T_ms of the EVAs were similar (84, 86, and 75 °C for EVA19, EVA22, and EVA28, respectively), the MIs of the EVAs were different which could be ranked in the following order: EVA19 (530 g/10 min)>EVA28 (6.80 g/10 min)>EVA22 (1.80 g/10 min). EVA22 and EVA28 showed a similar melt behavior in the particleboard because of a small difference in the MI and T_m between both EVAs. Both EVAs have a relatively low MI and these molten EVAs became a plastic binder of wood particles and covered the voids or gaps between the wood particles. This phenomenon caused a decrease in the water absorption of the particleboard. In contrast, EVA19 had a very high MI. Thus, during the hot pressing the molten EVA19 could flow very well and penetrate into the wood particles in the core layer of the particleboard. Consequently it could not fill the voids or cover the gaps between the wood particles (Figure 5d). It appeared that EVA19 did not act as an effective binder. This contributed to a decrease in the physical and mechanical properties of the particleboard. Furthermore, the adhesion between the wood particles of the particleboard containing EVA19 was lower than the particleboards containing EVA22 and EVA28.

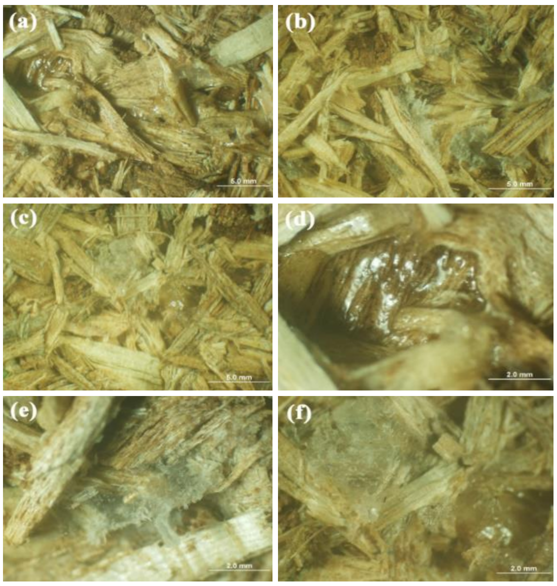

Figure 5. Stereo micrographs of the fracture surfaces of the IB tested samples: (a, d) EVA19, (b, e) EVA22 and (c, f) EVA28. The magnification of the upper and lower figures was 7x and 20x, respectively.

3.2 Mechanical properties

The mechanical properties of the particleboards are shown in Figures 6-8 and the results of the statistical analysis are given in Table 4. Basically, the mechanical properties of the particleboard are dependent on sample density. Jaru sombuti, Hiziroglu, Bauchongkol, and Fueangvivat (2009) reported that sample density affected the bending properties of wood composite. Rofii, Yumigeta, Kojima, and Suzuki (2014) also stated that the IB strength of particleboard was related to the particleboard density. Therefore, the density of specimens was determined. No significant change in density was found after adding 5% EVA (Figures 6-7). This confirmed that the mechanical properties of the particleboards in the present study were not influenced by the sample density, and it clearly

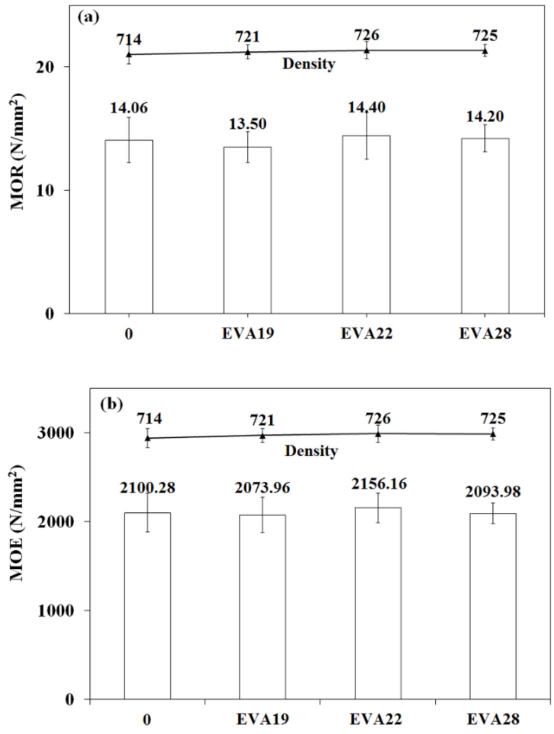

Figure 6. Effect of EVA on the modulus of the particleboards: (a) MOR and (b) MOE.

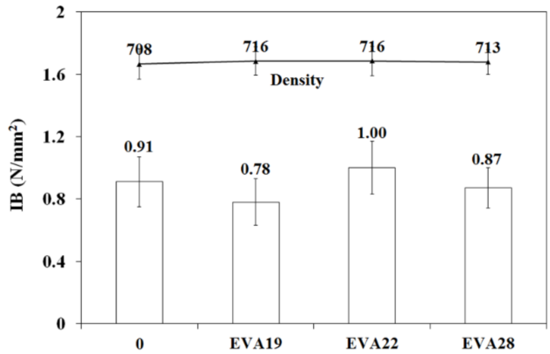

Figure 7. Effect of EVA on the internal bond strength of particle-boards.

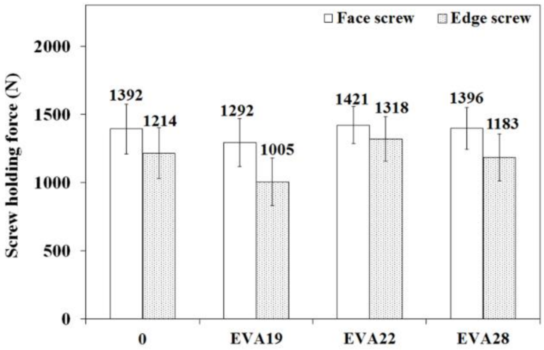

Figure 8. Effect of EVA on the face and edge screw holding force of particleboards.

Table 4. Mechanical properties of particleboards.

Sample	MOR (N/mm^2)	MOE (N/mm^2)	IB strength (N/mm^2)	Screw holding force (N)	
				Face	Edge
0%	14.06^A (1.84)	2100.28^A (218.29)	0.91^{AB} (0.16)	1392^A (182)	1214^A (186)
5% EVA19	13.50^A (1.26)	2073.96^A (197.51)	0.78^C (0.15)	1292^A (176)	1005^B (173)
5% EVA22	14.40^A (1.88)	2156.16^A (166.40)	1.00^A (0.17)	1421^A (134)	1318^A (164)
5% EVA28	14.20^A (1.10)	2093.98^A (117.18)	0.87^{BC} (0.13)	1396^A (152)	1183^A (171)

Average values with different letters in the same column are significantly different at P<0.05. Numbers in parenthesis are standard deviations.
MOR, modulus of rupture; MOE, modulus of elasticity; IB, internal bond; N, newtons; EVA19, vinyl acetate content of 19%; EVA22, vinyl acetate content of 22%; EVA28, vinyl acetate content of 28%.

depended on the grade of EVA. The MOR and MOE of all particleboards were higher than the particleboard prescribed standard of EN312:2010 for use in dry conditions, which is 11 N/mm^2 and 1600 N/mm^2, respectively. The IB strength and screw holding force of all samples were also higher than the standard values of 0.35 N/mm^2 for IB strength and 500 N for face screw holding force. The addition of EVA did not affect the MOR, MOE, or face screw holding force and little change was derived based on the statistical analysis. Remarkably, the 5% EVA19 sample showed the lowest mechanical properties and lower than the control sample. Based on the statistical analysis, the inferior property from the addition of EVA19 was noticeable in the edge screw holding force, which was a significant decrease (1005 N), whereas the edge screw holding forces of the control and the 5% EVA22 sample were 1214 and 1318 N, respectively. As mentioned in the stereo micrographs of the EVA19 sample (Figures 5a and 5d), the voids and gaps observed in the core layer were not filled with plastic around the screw threads. Thus, poor load transfer occurred between the matrix of the particleboard and the screw threads which resulted in the lowest edge screw holding force of the particleboard containing EVA19. Ayrilmis and Jarusombuti (2011) reported that the highest screw holding force was observed in particleboard containing the highest PP powder because the PP powder could conform around the threads of the screw and this phenomenon affected the continuous load transferring along the thread. A comparison of the EVA grade among these mechanical properties showed that EVA22 provided the highest values which were higher than the control sample as well. The IB strength evidently showed the effect of EVA grade and substantiated that EVA22 was the best one. Consequently, the effect of the EVA grade on the mechanical properties of the particleboard could be ranked in the following order: EVA22>EVA28>EVA19. The molecular weight of the EVA should play a major role in these mechanical properties. The relative molecular weight of EVA was in the following order: EVA22>EVA28>EVA19. Their gel permeation chromatography (GPC) traces are shown in Figure 9. The apparent number-average molecular weight (M_n) was determined from GPC traces using tetrahydrofuran as the solvent. Without a correction factor, the M_ns of EVA19, EVA22, and EVA28 were 29000, 68000, and 61000 g/mol, respectively. All EVA grades melted completely during particleboard fabrication because the T_ms were low (<100 °C). The highest molecular weight and lowest melt index of EVA22 seemed to be the key parameters in the present study which attributed to the highest mechanical properties. Similar results by Waelaeh *et al.* (2017) reported that the mechanical properties of particleboard were affected by the MI and M_n of the plastic. The MOR, MOE, IB strength, and screw holding force of the particleboard containing low density polyethylene (LDPE) pellet was higher than particleboard containing ultra-low density polyethylene (ULDPE). ULDPE has a very low MI (0.48 g/10 min) although the M_n of ULDPE was lower than the LDPE. Due to limited flow ability, the molten ULDPE could not flow well and could not fill the voids and gaps between the coarse wood particles. Therefore, the interfacial adhesion and stress transfer between the wood particles were lower than the particleboard containing LDPE.

As mentioned earlier, the molten EVA22 could flow relatively well and filled the voids and gaps between the wood particles which acted as a binder between the wood particles. From the GPC results, EVA22 had the highest molecular weight which might promote strong interfacial adhesion between the wood particles for good stress transfer between the wood particles within the particleboard. For this reason, the mechanical properties and water resistance of the particleboard containing EVA22 were better than the control particleboard.

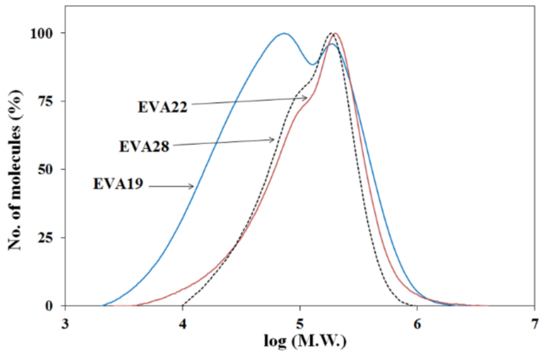

Figure 9. Molecular weight distribution curves of EVA from a gel permeation chromatography analysis.

4. Conclusions

This study investigated the effects of EVA on water resistance and mechanical properties of three-layer particleboard made from natural rubber-wood particles and UF adhesive. Three grades of EVA (EVA19, EVA22, and EVA28) were used and 5% of EVA was added into the core layer fraction. All EVA grades increased water resistance of the particleboard as demonstrated by the thickness swelling and water absorption tests. The EVA28 provided the lowest water absorption and much lower than the control sample (0% EVA). There were no significant differences in the MOR, MOE, and screw holding force between the control sample (0% EVA) and the 5% EVA samples. No significant effects of the EVA grade on those properties were observed, but the sample containing EVA22 showed the highest values for all mechanical properties. The results of the IB strength test could differentiate the effects of EVA grades which could be rated as EVA22>EVA28>EVA19. Likewise, this ranking could be noticed in all mechanical properties. The lowest melt index and highest molecular weight of EVA22 contributed to the high water resistance and mechanical properties because the molten EVA22 could fill the gaps or voids between the wood particles which became a binder for the wood particles. In contrast, the very high melt index of EVA19 made it flow easily into the wood particles so that it could not act as a binder or fill the gaps or voids between the wood particles.

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