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EFFECTS OF WEATHERING ON MECHANICAL PROPERTIES AND SURFACE ROUGHNESS OF *ENDOSPERMUM MALACCENSE* WOOD MODIFIED WITH PROPIONIC ANHYDRIDE

This study evaluated some properties of sesenduk (Endospermum malaccense) wood treated with propionic anhydride and exposed to outdoor conditions for up to one year. Defect-free samples of sesenduk were extracted with a 3:1:1 (v/v) mixture of toluene, ethanol and acetone for 3 hours at a temperature of 100°C. The samples were then modified at the same temperature for another 3 hours using propionic anhydride and 10% sodium formate as catalyst. Modulus of elasticity (MOE), modulus of rupture (MOR) and parallel-to-grain compression strength of the samples exposed to weathering were tested. Chemical treatment reduced the MOE but slightly increased the MOR and parallel-to-grain compression strength compared with untreated samples. However, treated samples retained higher strength properties than untreated ones. The modulus of elasticity, modulus of rupture and parallel-to-grain compression strength of treated samples at radial orientation were respectively 20%, 31% and 62% higher than those of untreated samples after one year's outdoor exposure. Weathering adversely influenced the surface quality of the specimens for all exposure times.

Keywords: chemical modification, weathering; mechanical properties, surface quality, sesenduk (*Endospermum malaccense*)

Introduction

Tropical hardwoods are gaining favour around the world, particularly in European countries, owing to their larger log diameters, appealing texture and

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superior decay resistance. They have been utilized for furniture, wood composite and a wide range of outdoor applications [Kilic and Niemz 2012]. For outdoor applications, the durability of the timber against weathering is of the utmost importance. The term "weathering" describes the irreversible deterioration of appearance and properties of a material when exposed to the weather [Jankowska and Kozakiewicz 2016]. Weathering is the general term used to refer to the slow degradation of materials exposed to the weather. The degradation mechanism depends on the type of material, but the cause is a combination of factors found in nature: moisture, sunlight, heat/cold, chemicals, abrasion by windblown materials, and biological agents.

In tropical countries such as Malaysia, the interest of manufacturers has shifted to low-density hardwood, as resources of high-density wood are being depleted [Mohammad-Fitri et al. 2017]. Some lesser-known species (LKS) such as sesenduk have high potential in replacing the now depleted high-quality timber in order to sustain wood-based industry. Sesenduk (*Endospermum malaccense*) is a light-density tropical hardwood with air-dried density ranging from 305 to 655 kg/m^3. Unfortunately, due to its low density, sesenduk has been reported as a non-durable wood with an average service life of 1 year [Jackson 1965]. Therefore, treatment is always necessary to enhance the biological durability and weathering resistance of sesenduk wood.

Treatment with phenolic resin is commonly applied to improve both the biological durability and weathering resistance of sesenduk wood and other woody materials [Anwar et al. 2011]. Mohammad-Fitri et al. [2017] treated sesenduk wood with low-molecular-weight phenol formaldehyde (LMwPF) followed by hot pressing of the samples at 150 °C and exposure to accelerated ageing for 10 cycles. The results revealed that the treated wood subjected to accelerated ageing lost 22.9-38.3% of its initial modulus of elasticity, while untreated samples lost 51.3%, confirming that the phenolic resin treatment improved the weather resistance of the treated wood. Heat or thermal treatment is another effective method for protecting the wood against weathering [Candelier et al. 2016]. Yildiz et al. [2013] reported that hardwoods and softwood thermally treated using steam displayed better resistance against artificial weathering compared with untreated samples.

Apart from the aforementioned treatment methods, several other types of chemical compounds have been used to modify wood, including anhydrides, acid chlorides, carboxylic acids, isocyanates, aldehydes, alkyl chlorides, lactones, nitriles and epoxides [Militz et al. 1997]. For instance, Bhat et al. [2010] treated *Acacia mangium* and *Acacia* hybrid woods with propionic and succinic anhydride, and exposed the treated wood to weathering for a period of one year. The results revealed that the treated wood exhibited better resistance against weathering, where discoloration, weight loss and mechanical strength loss were lower than in untreated wood samples. Of the two chemicals, succinic anhydride offers better protection to the treated wood than propionic anhydride.

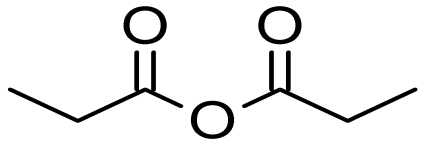
However, to the best of the authors' knowledge, no study has been reported related to the effects of weathering on the mechanical properties and surface roughness of propionic anhydride-modified sesenduk wood, especially in the tropical and ASEAN regions. Therefore, the objective of this study was to evaluate some mechanical properties and the surface quality of propionic anhydride-modified sesenduk wood following outdoor exposure.

Materials and methods

Preparation of materials

Sesenduk (*Endospermum malaccense*) trees with an average age of 19 years were harvested from a plantation located at Forest Research Institute Malaysia (FRIM), Kepong, Selangor. The harvested trees were sawn in the radial and tangential directions. Propionic anhydride and sodium formate were used as modifying chemicals in this study. The molecular structures of propionic anhydride and sodium formate are presented in Figure 1, and the molecular formulae, densities and molecular weights are given in Table 1.

Propionic anhydride



Sodium formate

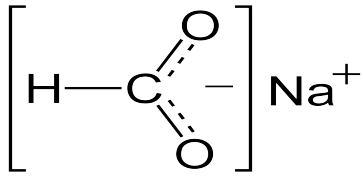


Fig. 1. Molecular structures of propionic anhydride and sodium formate

Table 1. Properties of propionic anhydride and sodium formate

Properties	Propionic anhydride	Sodium formate
Molecular formula	$C_6H_{10}O_3$	HCO_2Na
Molecular weight	130.14 $gmol^{-1}$	68.01 $gmol^{-1}$
Density	1.015 g/cm^3	1.92 g/cm^3

Preparation of propionic anhydride-modified wood

Wood samples were extracted in a Soxhlet apparatus with a 3:1:1 (v/v) mixture of toluene, ethanol and acetone at a temperature of 100 °C for 3 hours. After the extraction process was completed, the extracted samples were first air-dried and then weighed. The air-dried samples were oven-dried overnight at 103 ±2°C prior to the modification treatment. Oven-dried samples were cooled off in a desiccator.

In the next step, the samples were modified in a 2 L reaction flask. The reaction with propionic anhydride was carried out with a 10% concentration of sodium formate, used as a catalyst, at a temperature of 100°C, for 3 h. The reactor with an attached condenser was placed in an oil bath. The flask contents were stirred every 10 min. At the end of the reaction time the flask was removed from the hot oil bath and the hot reagent decanted off from the samples. The samples were then immediately soaked in ice-cold acetone for 2 h to quench the reaction.

Evaluation of mechanical properties of samples

A total of 420 modified and control samples were tested for mechanical properties. Ten samples for each outdoor exposure level, namely 1-month, 3-month, 5-month, 7-month, 9-month and 12-month exposure, were used for the tests. The samples were placed on a steel rack facing south, at an angle of 45°C, at the Forest Research Institute Malaysia (FRIM), Kepong, Selangor, Malaysia for outdoor exposure evaluation of the sample according to the procedures specified in Anwar et al. [2011]. The average temperature in FRIM, Kepong is 25.9°C and the relative humidity is more than 80% all year long. The precipitation is about 2579 mm per year.

The bending strength and compression strength parallel to the grain were tested based on BS EN 373:1957. Bending and compression test samples had dimensions of 20 × 5 × 120 mm and 20 × 20 × 60 mm, respectively. A Shidmazu AG-15 universal testing machine having two supports over a span of 100 mm with 100 kN capacity was employed for bending strength testing. All outdoor exposed samples were conditioned to an equilibrium moisture content of 12% prior to the tests.

Evaluation of surface roughness properties

The effects of weathering on the surface quality of the samples were also evaluated, using stylus-type equipment. Two roughness measurements were taken from the surface of each sample across the grain orientation prior to outdoor exposure. A T-500 Hommel profilometer was used for the roughness measurements. The device consists of a main unit and a pick-up with a skid-type diamond tip stylus having a radius of 5 μm. Three roughness parameters – average roughness (R_a), mean peak-to-valley height (R_z) and maximum

roughness (R_{max}) – were used to evaluate the surface quality of the control and outdoor exposed samples. The tracing span and cut-off length for the roughness measurements were 5 mm and 0.25 mm respectively.

Statistical analysis

The data were analysed statistically using analysis of variance (ANOVA) with mean separation by the least significant difference (LSD) method. All statistical analysis was performed at $p \leq 0.05$.

Results and discussion

Bending strength

The weight percent gain of the wood samples after treatment with propionic anhydride was around 22%. The results of the mechanical and surface roughness tests are given in Table 2. The control specimens, in both grain orientations, had the highest modulus of elasticity (MOE) value among the samples. Average MOE values of 9369 N/mm^2 and 9665 N/mm^2 were recorded for the control samples in the radial and tangential orientation respectively. However, following treatment with propionic anhydride with sodium formate used as a catalyst, their stiffness was reduced, as indicated by the lower MOE values. The MOE values for propionic anhydride-modified samples were 8899 N/mm^2 in the radial direction and 8953 N/mm^2 in the tangential direction. On the other hand, the modulus of rupture (MOR) in radial samples was higher after modification. The radial MOR value was 78.0 N/mm^2 for control samples and 83.5 N/mm^2 for treated samples. Bhat et al. [2010] attributed the improvement in MOR to the reaction of hydroxyl groups of cell wall polymers with the anhydride.

After a 1-month exposure, the control radial samples retained 87.87% of their initial MOE, while modified radial samples retained 92.48% of their initial MOE. In other words, MOE loss for modified radial samples was less than that of the control radial samples after a 1-month exposure. Similar findings were also recorded for the tangential samples, where control and modified samples retained respectively 88.97% and 96.65% of initial MOE after a 1-month exposure. The findings suggest that modification with propionic anhydride provides better protection against weathering. The difference between control and modified samples became more and more significant with an increase in the exposure period (3 to 12 months). After 12 months' exposure, the control radial and tangential samples retained only 63.75% and 67.81% respectively of their initial MOE. Meanwhile, modified radial and tangential samples respectively retained 83.64% and 79.05% of their initial MOE.

The trend for MOR mirrored that of MOE. Control radial samples retained 84.23% of their initial MOR after a 1-month exposure. The strength decreased as

Table 2. Mechanical properties of sesenduk wood after weathering

Exposure time (months)	Mechanical properties (N/mm^2)									
	modulus of elasticity				modulus of rupture				compression strength	
	radial		tangential		radial		tangential			
	modified	control	modified	control	modified	control	modified	control	modified	control
0	8899.1^a	9369.1^a	8952.7^a	9665.0^a	83.5^a	78.0^a	80.6^a	84.6^a	39.28^a	32.14^a
	(1140.46)	(1026.5)	(1297.6)	(1425.9)	(10.5)	(7.39)	(11.64)	(10.96)	(2.93)	(3)
1	8230.1^a	8232.4^a	8652.4^a	8598.7^a	71.0^a	65.7^b	74.9^a	71.4^a	37.08^a	28.69^a
	(1107.2)	(1185.4)	(1167.36)	(1014.8)	(6.29)	(9.1)	(10.8)	(8.51)	(8.7)	(4.03)
3	8030.7^a	7604.7^a	8691.9^a	8542.1^a	70.8^a	62.6^a	76.8^a	70.9^a	33.71^a	20.97^a
	(934.76)	(1198.7)	(1276.97)	(1020.7)	(8.19)	(9.79)	(11.86)	(7.53)	(5.66)	(8.3)
5	7873.3^a	7569^a	8521.7^a	7511.6^a	64.7^a	58.5^a	63.7^a	58.4^a	31.27^b	17.45^a
	(1147.08)	(1265.7)	(987.44)	(1171.4)	(7.53)	(9.19)	(9.32)	(9.45)	(7.16)	(6.24)
7	7694.6^a	7377.7^a	7771.7^a	7443.5^a	64.6^a	53.5^b	64.6^a	57.0^a	29.41^b	9.60^a
	(476.93)	(1221.46)	(1001.4)	(1249.8)	(11.31)	(7.4)	(6.29)	(11.64)	(7.1)	(2.27)
9	7546.6^a	6935.2^a	7751.3^a	7409.4^a	62.4^a	52.4^a	66.4^a	50.3^b	11.08^b	5.63^a
	(861.2)	(1246.18)	(833.09)	(901.86)	(12.46)	(12.67)	(7.6)	(8.45)	(1.16)	(9.71)
12	7443.5^a	5972.7^a	7077.0^a	6553.4^a	61.5^a	42.4^b	52.3^a	48.2^a	10.1^a	3.77^a
	(1249.82)	(666.5)	(1499)	(974.65)	(5.72)	(7.53)	(5.73)	(8.5)	(7.51)	(4.4)

Numbers in parentheses are standard deviations. Means followed by the same letter are not significantly different at $p \leq 0.05$.

the exposure period increased. After a 12-month exposure, control radial samples retained only 54.36% of MOR. In comparison, modified radial samples exhibited higher resistance against weathering, retaining 73.65% of the initial MOR value after a 12-month exposure. For tangential samples, the control samples retained 56.97% of initial MOR after a 12-month exposure, while modified samples retained 64.89%. The findings were in line with Homkhiew et al. [2014] and Zhu et al. [2015], who reported gradual decrements in the strength properties of samples as a function of exposure time. Sandberg [1999] reported that more cracks were formed on the tangential surfaces of wood after outdoor exposure due to the fact that the magnitude of shrinkage and swelling in the tangential direction is twice that in the radial direction. Therefore, in this study, modified tangential samples retained lower MOR and MOE values than their radial counterparts.

Cellulose depolymerization is the main factor contributing to strength loss due to weathering [Sander et al. 2003]. Anhydride molecules are able to self-condense and able to provide protection to cellulose from photodegradation, resulting in better resistance in treated wood. The dimensional stability of the treated wood is greatly enhanced. Therefore, the shrinkage and swelling of the treated cells are much lower than those of untreated cells. This phenomenon leads to lower distortion in the wall layers of treated wood, resulting in lower strength loss after weathering [Imamura 1993]. According to Rowell and Winandy [2005], lignin acts as a matrix material that binds carbohydrate molecules together within the cell wall and contributes little to the wood's stiffness. Upon exposure to outdoor conditions, lignin is the most sensitive component and is rapidly degraded photochemically by UV radiation. Cellulose becomes exposed to the weather elements as lignin degrades and is eventually washed off the wood surface during rain. Next, new lignin-rich surfaces are exposed and the degradation cycle starts over, contributing to the continuous loss of strength of weathered wood [Rowell and Winandy 2005].

Compression strength

The initial compression strengths for control and modified samples were 32.14 and 39.28 N/mm^2 respectively. Modified samples had higher compression strength than the control samples. After exposure for 1 month, the control samples retained 89.27% of their initial compression strength, while modified samples retained 94.40%. The difference is even greater and more prominent after a 12-month exposure. The compression strength of the control samples was recorded as 3.77 N/mm^2, which means that 88.27% of their initial strength was lost after exposure for 12 months. A similar trend, though to a lesser extent, was observed for the modified samples. After a 12-month exposure, the modified samples retained 25.71% of their initial compression strength. Although this is also a substantial loss of strength, it is much better than in the control samples.

Jankowska and Kozakiewicz [2016] found that the loss in compression strength of wood is due to the cyclical changes in humidity and temperature during weathering. The rapid changes in both humidity and temperature caused cracking of wood as strong sorption stress exceeded the internal cohesion of wood. Consequently, the wood lost its initial strength after weathering. However, after modification with propionic anhydride, the wood's sorption of water vapour was greatly reduced, as reported by Papadopoulos [2006]. As a result, the modified wood was subject to lower stress, and therefore the loss in strength was also reduced.

Surface quality

The surface roughness of control and modified wood after one year's outdoor exposure is shown in Table 3. Average Ra values of control and modified samples before exposure were 2.87 μm and 3.15 μm respectively. As they were exposed to weathering, their surface quality was adversely affected. For

Table 3. Surface roughness of modified and unmodified wood samples after 1-year outdoor exposure

Exposure time (months)	Modified (μm)			Control (μm)		
	R_a	R_z	R_{max}	R_a	R_z	R_{max}
0	3.15	31.78	49.18	2.87	23.25	26.74
	(0.57)	(5.39)	(10.92)	(0.34)	(4.24)	(14.75)
1	10.23	48.91	82.69	7.14	41.32	59.76
	(3.35)	(51.14)	(26.55)	(2.08)	(9.68)	(22.02)
3	11.44	56.58	83.39	9.18	49.87	67.80
	(1.93)	(6.52)	(14.10)	(2.05)	(10.49)	(16.56)
5	5.01	43.94	58.17	6.17	57.62	67.04
	(1.11)	(6.33)	(2.5)	(0.98)	(5.63)	(7.54)
7	5.67	53.96	64.53	5.8	56.68	66.85
	(1.15)	(7.69)	(9.49)	(0.16)	(5.62)	(9.02)
9	5.23	55.21	65.61	6.46	59.24	71.24
	(0.74)	(1.58)	(7.01)	(0.71)	(3.96)	(10.77)
12	7.97	62.37	71.33	6.83	59.45	74.91
	(1.17)	(2.81)	(4.3)	(1.22)	(4.33)	(11.55)

Numbers in parentheses are standard deviations.

example, modified samples had an Ra value of 5.01 μm, compared with 6.17 μm for control samples, as a result of a 5-month exposure, indicating that the overall surface quality of both types of samples deteriorated. The results were in agreement with Yildiz et al. [2013], who reported that the surface of thermally treated wood samples became rougher with increasing time of weathering. Hiziroglu et al. [2008] reported a similar observation, as the surface quality of tropical hardwoods started to degrade after a 4-week exposure period. Degradation of wood polymers and leaching of degraded wood materials are the main factors causing weathered wood to have a rougher surface [Behbood and Saei 2015]. However, although not significant, it seems that the chemical treatment had some positive impact on the surface quality of the modified samples. After a 9-month exposure, modified wood samples exhibited smoother surfaces than the untreated samples. However, as the weathering period progressed, the surface of the modified wood became rougher than the untreated samples by the end of the exposure period of 12 months.

Conclusions

This study examined the mechanical properties and surface properties of propionic anhydride-modified sesenduk wood after exposure to weathering. The results revealed that the treatment using propionic anhydride enhanced the weathering resistance of sesenduk wood. The modified wood lost 7.52% of its initial MOE, while unmodified wood lost 12.13%, after a 1-month exposure. A similar trend was also observed for MOR, where the modified wood retained higher strength after weathering. The compression strength of the control samples decreased by as much as 88.27% after a 12-month exposure. Meanwhile, modified samples lost 74.29% of their initial compression strength. As for surface quality, all samples became rougher after weathering. The results obtained in this study indicate that propionic anhydride provides a certain degree of protection to sesenduk wood against weathering.

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List of standards

BS 373:1957 Methods of testing small clear specimens of timber

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