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Characterization of plywood made from heat-treated rubberwood veneers bonded with melamine urea formaldehyde resin

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Abstract

The objective of the study is to evaluate the properties of the plywood made from rubberwood veneers thermally treated at various temperatures. Rotary-peeled rubberwood veneers were thermally treated at 160, 170, 180, and 190 °C under 1 bar pressure. 5-ply plywood panels were produced using melamine urea formaldehyde (MUF) resin as binder. Dimensional stability, i.e., water absorption (WA) and thickness swelling (TS) as well as mechanical properties of the plywood were assessed. The results demonstrated that the dimensional stability of the plywood made from thermally treated rubberwood veneers improved as indicated by lower WA and TS values. The reduction in WA and TS values increased along with treatment temperatures. Darkening in the plywood as a result of heat treatment was observed. Meanwhile, heat treatment at 170 °C and 180 °C resulted in plywood with the highest modulus of rupture (MOR), modulus of elasticity (MOE) and shear strength. However, beyond that temperature, the mechanical strength of the plywood started to decline. Based on the findings in this work it appears that heat treatment of rubberwood veneers at temperatures ranging from 160 to 180 °C could have a potential to improve overall properties of plywood panels.

Keywords Plywood, Thermally treated wood, Rubberwood, Melamine urea formaldehyde resin, Physical-mechanical properties

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Introduction

Rubberwood (Hevea brasiliensis Muell. Arg.) is a significant trading timber species in the Southeast Asia region [1]. Owing to its whitish appearance, rubberwood is widely marketed as oak wood in Southeast Asia. It is a fact that rubber tree plays a significant role on economics of Southeast Asian counties due to natural rubber production. The tree can produce rubber that can be used as a raw material for many applications, such as automobiles, cosmetics, medical equipment, and home appliances. However, rubber trees become less productive, approximately at the age of 25 years, and replantation would be required [2]. Harvested rubber trees can be served as raw material for woodbased industries. Generally, small logs having a diameter less than 4 in. will be used for the production of



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wood-based panels. Meanwhile, big logs with a diameter of more than 4 in. are used for sawmills, and any residue will be transported to bioenergy plant [3]. The veneers peeled from the rubberwood logs can be used to produce plywood, one of the engineered wood-based panels that is widely used for construction.

Plywood made of rubberwood veneers has proved to be a potential high value end use as it can serve as construction and decorative purposes [4]. However, rubberwood has poor dimensional stability and low durability against biodeterioration agents [5]. These problems have restricted rubberwood and its resultant panel products from being used exteriorly. Therefore, treatment is required to enhance the dimensional stability of rubberwood and its panel products. Thermal treatment or heat treatment, being a green treatment method without the usage of chemicals could be a feasible technique to solve the issue. Several studies investigated heat treatment of lumbers at a temperature of 130-280 °C and exposure time of 3-24 h [6-8]. It is a known fact that thermal modification of wood causes overall weight loss and enhanced dimensional stability of the member [9].

There are two types of heat treatment methods used on plywood: one applies heat treatment on the wood veneers prior to glue, and the other applies heat treatment directly on the finished plywood [10]. The latter can be called post heat treatment. In comparison to post heat treatment, studies on heat treatment on the wood veneers prior to assembling into plywood are rather limited. Thermal treatment has been shown to improve selected properties of wood veneers, allowing it to be used more efficiently in the production of plywood and laminated veneer lumber (LVL). Candan et al. [11] thermally compressed Douglas fir veneer (Pseudotsuga menziesii) sheets were at three different pressure levels of 1.0 N/ mm², 2.0 N/mm², and 2.5 N/mm² at two temperatures of 180 °C and 210 °C for 3 min. Thermally compressed veneers had a smoother surface than untreated veneers, as evidenced by a lower roughness value. The smoothness of the veneer improved as pressure levels increased. Smoother veneers are preferred in plywood production because rough veneer reduces contact between the layers, resulting in a weak glueline, low strength properties, and excessive resin consumption [12]. Bekhta et al. [13] confirmed that thermal compression can improve the surface quality of veneer. After being thermally compressed, rotary-peeled European beech (Fagus sylvatica L.) veneers showed lower surface roughness. Surprisingly, plywood made of thermally compressed veneers has good bonding strength despite using 40% less glue. Apart from that, plywood made from thermally modified wood veneers also displayed better dimensional stability and lower thickness swelling [10], lower formaldehyde emission [14] but poorer mechanical properties [15].

Most of the studies focused on the European wood species. Tropical wood species such as rubberwood, on the other hand, have attracted less attention. Therefore, the objective of this study was to investigate different properties of experimental plywood panels manufactured from heat-treated veneer sheets of rubberwood so that such panels can be used for interior applications with a better understanding of their behavior during their service life.

Materials and methods

Veneer sheets preparation and plywood manufacture

Twenty-five year-old rubber trees with a diameter of around 200–250 mm were harvested from forest plantation at Mueang, Surat Thani, Thailand (9°05′43.9″N 99°21′20.4″E), for the experiment. The harvested rubberwood logs were soaked in fresh water for 2 h before peeling. Rotary cutting was used to produce veneer sheets in 2 mm of thickness. Defect-free veneer sheets with the width of 600 mm and the length of 600 mm were initially dried in laboratory oven at the temperature of 70 \pm 5 °C for 3 days. The veneer surface of rubberwood rotary sheet is illustrated in Fig. 1.

Heat treatment was conducted on the veneer sheets prior to plywood manufacture. The veneer sheets were treated at the temperature of 160, 170, 180, and 190 °C, respectively, under 1 bar pressure for 15 min using a hot press machine. A total of 60 veneer sheets were treated in this study. Next, the treated veneer sheets were cut into smaller sheets with width of 500 mm and length of 500 mm. 5 plies veneers sheet were assembled perpendicularly to each other and E1 grade melamine urea formaldehyde (MUF) resin (AICA, Hatyai, Thailand) were manually applied on each layer of the veneer at a glue spreading rate of 150 g/m². The properties of MUF resin are displayed in Table 1. The assembled veneers lay-up was then cold pressed for 15 min followed by hot pressing at a temperature of 130 °C under specific pressure of 1.5 MPa for 15 min using a hot press machine. The hot-pressed plywood panels were then cooled down before they were trimmed to 300 mm × 300 mm size for properties evaluation. A set of 5-ply plywood made from untreated veneers was prepared and served as control. Three panels were made for each parameter. A total of 15 plywood panels were made in this study.

Water absorption and thickness swelling of the samples

Water absorption (WA) of the samples were evaluated by soaking in distilled water for 2 and 24 h according to ASTM D1037-12 standard. The sample size of 50 mm \times 50 mm \times 10 mm was prepared. 5 replications were used for each parameter and a total of 25



Fig. 1 The rubberwood veneer sample using rotary cutting

Table 1 Properties of melamine urea formaldehyde resin, typeE1 used in this study

Properties	Results
Viscosity at 30 °C (cps)	143
% N.V.C 3 h at 105 ℃	64.20
pH at 30 ℃	9.17
Density at 30 °C (g/cm³)	1.27
Gel time at 100 °C (s)	57

pieces were used for WA test. At the end of the 2 h and 24 h, the samples were taken out from the water and all surface water was removed by wiping before they were weighed at an accuracy of 0.01 g. The difference between before soaking and after soaking was calculated and expressed in percentage (%). The same

samples used for WA test was also used for the determination of thickness swelling (TS). The thickness of the samples before and after soaking was measured with a vernier caliper at an accuracy of 0.01 mm. The changes in thickness were determined and expressed in percentage (%).

Color changes of the samples

The color changes of the samples before and after heat treatment were determined by using a spectrophotometer, MiniScan EZ 4500, HunterLab (Virginia, USA). Three random measurements points were taken on the samples of each parameter and the value obtained was averaged. The results were reported using the color system in L^* , a^* , and b^* values. The color change (ΔE) was calculated according to the CIELab method (Hunter Lab. 1996).

Bending strength of the samples

Bending strength such as modulus of rupture (MOR) and modulus of elasticity (MOE) of the samples were determined according to the procedures specified in ASTM D1037-12 standard [16]. A total of 25 samples (5 replications for each parameter) with dimensions of 50 mm \times 300 mm \times 10 mm in width, length, and thickness, respectively, were tested. Three-point bending test was carried out using a Universal Testing Machine (UTM) (Tinius Olsen, 100KU Series, Redhill, UK). The load was applied continuously throughout the test at a speed rate of 10 m/min.

Shear strength of the samples

Shear strength of the plywood samples was determined in accordance with ISO 12466-1:2007 [17], Plywood-Bonding quality. The test samples were prepared, cut, and nicking (saw cuts) which grain direction of the layer between the glue lines under test is perpendicular to the length of the samples. Samples with dimensions of 25 mm×135 mm×10 mm in width, length, and thickness, respectively, were examined. Prior to the shear test, the samples were soaked for 24 h in water at a temperature of 25 °C. Next, the samples were evaluated using an UTM machine at a crosshead speed of 2 mm/min. The percentage of apparent cohesive wood failure of the samples was also determined by using shear strength tested samples to compare with the standard illustrations by 10% increments from 0 to 100% [17]. The determination of 5 repeated samples were calculated by consisting of an area assessment to identify wood fiber failure. The mean value was determined decimal rounding up to ten.

Data analysis

Analysis of variance (ANOVA) (SPSS Statistics version 22) was used to determine whether there were significant differences between the five types of plywood samples with a *p*-value of 0.05. Duncan's multiple range tests were also employed for additional analysis.

Results and discussion

Water absorption and thickness swelling

Table 2 displays density, thickness, WA, and TS of the plywood samples produced in this study. The density of the rubberwood plywood reduced from 1071 kg/m³ for the untreated samples to $983-1000 \text{ kg/m}^3$ in the plywood treated at temperature range of 160-190 °C. The extent of density reduction increased with increasing treatment temperatures. A density loss of 6.63% was recorded in the plywood thermally treated at 160 °C and the density loss was 8.22% when being treated at 190 °C. This study showed that the surface of the thermally treated veneers had smooth, and less lathe check or partial disappearance of lathe check could be observed as shown in Fig. 2. Overall, WA and TS value of the plywood samples decreased as a result of heat treatment, indicating that the heat treatment of veneers bestow better dimensional stability to the resultant plywood panels. WA value after 2- and 24-h soaking showed a decreasing trend with the increasing treatment temperatures. The lowest WA2h and WA24h was observed in the plywood type T190. On the other hand, for TS2h, the lowest TS of 1.11% was obtained from T180 sample. T190 sample showed higher TS2h than T180 sample, but the difference is not significant. TS values of the plywood samples exhibited a similar trend with WA after 24-h soaking, with the lowest TS24h of 2.22% found in the T190 samples. The findings suggested that the thermal treatment reduced the swelling of plywood effectively. A reduction of 5.35 percentage

Type of panel	Density (kg/m ³)	Density loss (%)	WA (%)		TS (%)	
			2 h*	24 h*	2 h	24 h*
Control	1071 ^b (61.17)		17.21 ^d (1.10)	38.26 ^c (2.27)	1.41 ^b (0.23)	7.57 ^d (0.37)
T160	1000 ^a (15.68)	6.63	15.97 ^{cd} (1.33)	37.07 ^c (3.04)	1.17 ^a (0.14)	3.69 ^c (0.33)
T170	997 ^a (30.51)	6.91	14.56 ^{bc} (1.44)	33.49 ^b (1.34)	1.15 ^a (0.13)	3.39 ^{bc} (0.19)
T180	996 ^a (10.41)	7.00	13.64 ^{ab} (1.05)	32.92 ^b (1.06)	1.11 ^a (0.04)	3.21 ^b (0.41)
T190	983 ^a (7.40)	8.22	12.30 ^a (0.64)	28.86 ^a (1.11)	1.17 ^a (0.13)	2.22 ^a (0.16)

Table 2 Physical properties of plywood made from rubberwood veneers thermally treated at different temperatures

The numbers in parentheses are the standard deviation values

*P-value < 0.001 and means with the different letter are significantly different



Fig. 2 Investigation of 3 points discoloration of each sample. a Non-treatment, b 160 °C, c 170 °C, d 180 °C, and e 190 °C of heat treatment

points in TS24h was recorded for plywood thermally treated at 190 °C.

Previous studies also showed the thermal modification of woods at the range of temperature 130–220 °C improved WA and TS [3, 18, 19]. A proportionate correlation was found between density and WA and TS of the plywood, where the WA and TS reduced along with decreasing density of plywood. However, the improvement in the dimensional stability as indicated by lower WA and TS in this study could be more likely due to the reduced hygroscopicity of the rubberwood veneers because of reduction of free hydroxyl groups in the wood [20]. Therefore, thermally treated wood had lower equilibrium moisture content (EMC) and the moisture absorption ability [20]. Consequently, lower WA and TS were observed in the rubberwood plywood.

Color changes

The color changes of the plywood samples as a function of treatment temperature are shown in Table 3. The control plywood samples without heat treatment have a lightness (L^*) value of 70.12, a^* value of 5.55 and b^* of 20.76. Positive a^* and b^* values indicate that the rubberwood plywood has a reddish and yellowish appearance. After heat treatment, the color becomes darker as indicated by the decreasing L^* values. The extent of decrement increased along with increasing treatment temperature. At temperature of 190 °C, the L^* value is 52.10, which is significantly lower than that of control samples. Meanwhile, both a^* and b^* values also increased as a result of heat treatment. As the treatment temperature

Table 3	Color	properties	of	plywood	l samples	made	from
rubberw	vood ve	neers therm	ally	treated at	t different te	emperat	tures

Type of panel	Color parameters					
	L*	a*	<i>b</i> *	ΔΕ		
Control	70.12 ^e (0.94)	5.55 ^a (0.24)	20.76 ^a (0.28)	0.00		
T160	65.75 ^d	6.22 ^b	22.27 ^b	4.67 ^a		
	(1.40)	(0.27)	(0.59)	(1.30)		
T170	62.19 ^c	7.65 ^c	23.69 ^c	8.71 ^b		
	(1.87)	(0.85)	(0.39)	(1.49)		
T180	56.07 ^b	8.64 ^d	25.23 ^d	15.06 ^c		
	(1.24)	(0.25)	(0.48)	(1.03)		
T190	52.10 ^a	9.23 ^d	26.31 ^e	19.21 ^d		
	(0.96)	(0.41)	(0.40)	(1.06)		

The numbers in parentheses are the standard deviation values.

*P-value < 0.001 and means with the different letter are significantly different

increased, the rubberwood plywood samples became more reddish and yellowish as indicated by the increasing a^* and b^* value. The total color change (ΔE) of the samples compared to that of control ranged from 4.67 to 19.21. According to the classification of the overall color change by Cividini et al. [21], ΔE value between 3 to 6 can be visible by medium quality filter. Meanwhile, ΔE values between 6 to 12 indicate distinct color changes while ΔE of more than 12 indicates a different color. Based on the findings of the current study, the rubberwood samples treated at temperature of 170 °C displayed a distinct color changes as shown by ΔE value of 8.71. Meanwhile, rubberwood samples treated at temperature of 180 °C and

Table 4	Mechanical	properties	of	plywood	made	from
rubberw	ood veneers	thermally trea	ated	at differen	t tempera	itures

Type of panel	MOR* (MPa)	MOE* (MPa)	Shear strength* (MPa)	Wood fiber* failure (%)
Control	99.1 ^a (8.54)	11300 ^a (1017)	1.5 ^a (0.09)	60 ^a (4.47)
T160	116.5 ^a (8.37)	17462 ^b (1015)	1.6 ^{ab} (0.08)	70 ^b (4.47)
T170	143.6 ^b (14.64)	24220 ^c (2029)	1.9 ^c (0.09)	80 ^{bc} (5.48)
T180	150.2 ^b (19.77)	22680 ^c (2324)	1.9 ^c (0.19)	80 ^c (7.07)
T190	102.8 ^a (6.50)	11320 ^a (907)	1.7 ^b (0.13)	70 ^b (7.07)

The numbers in parentheses are the standard deviation values

*P-value < 0.001 and means with the different letter are significantly different

190 °C exhibited a completely different color (ΔE value of 15.06 and 19.21, respectively) compared to that of control samples. However, the color change of this experiment is not uniform, but rather darker and lighter colored areas are distributed along the fiber. The visual appearance of the control samples and samples treated at different temperatures is shown in Fig. 2. An obvious color change is visible on the samples being treated beyond 170 °C.

Chromophores that exist in lignin and extractives decide the color of the wood. Degradation of lignin after thermal treatment changes the wood color [22]. During thermal treatment, additional chromophores produced as a result hemicellulose degradation could change the color of the wood [23]. Apart from that, migration of quinones, extractives, low molecular sugars and amino acids to the surface of wood veneers as a result of thermal modification could also be a probable reason that caused the darkening of wood veneers [24]. Also, the reduction in lightness of wood veneers could be contributed by the degradation of pentosan [24]. Furthermore, the chemical characteristics of the rubberwood veneers thermally treated at different temperatures in this study will be reported in the near future.

Bending and shear strength

The bending strength, shear strength and wood failure of the plywood samples are listed in Table 4. It can be noted that the plywood made from heat-treated rubberwood veneers had higher MOR values than the control sample. The same observation was also made for MOE values. Plywood made from rubberwood veneers heat treated at 180 $^{\circ}$ C (type T180) had the highest MOR



Fig. 3 Modulus of rupture (MOR) of plywood made from rubberwood veneers thermally treated at different temperatures



Fig. 4 Shear strength of plywood made from rubberwood veneers thermally treated at different temperatures

value of 150.20 MPa, a 51.5% increment compared to control samples. Meanwhile, the highest MOE value of 24,220 MPa was recorded in the plywood samples made from rubberwood veneers heat treated at 170 °C (type T170), an increment of 114.3% than the control samples. Nevertheless, when being treated at 190 °C, both MOR and MOE values of the plywood samples dropped drastically to 102.8 MPa and 11,320 MPa, respectively.

As for shear strength, a similar trend was also observed as the plywood made from heat-treated veneers exhibited higher shear strength compared to the control samples. Plywood type T180 had the highest shear strength of 1.9 MPa, 30% higher compared to 1.5 MPa in control. The wood fiber failure of the sheared samples was evaluated, and it ranged between 60 and 80%. The percentage of wood fiber failure increased as the treatment temperature increased. The highest wood fiber failure percentage of 80% was observed in T180 samples.

MOE of some thermally treated wood tends to increase as proven by some researchers [24-27]. When being thermally treated in hot air at temperatures ranging from 115 to 175 °C, the MOE of several wood species increased initially and then started to drop when the treatment condition becomes more severe [27]. A study

by Lee et al. [28] on thermally treated rubberwood also reported the same observation. The MOE of the rubberwood particleboard increased when being treated at milder temperatures. However, the MOE value started to decline when being treated at 160 °C. Wang et al. [29] attributed the increment in MOE to the high crystallization of cellulose and lignin condensation. At treatment temperature of 160 °C and 180 °C, the crystallization of cellulose increased. Also, hemicellulose was degraded and produced furfural that can lead to lignin condensation through cross-linking reaction.

Surprisingly, in this study, the MOR and shear strength of the rubberwood plywood also showed increasing trend as the treatment temperature increased and it only showed decrement when the temperature reached 190 °C (Figs. 3, 4). One possible explanation for the increased mechanical strength is that the thermally treated plywood had a smoother surface and the partial disappearance of lathe checks, as shown in Fig. 2. Low surface roughness may help to improve layer contact and result in a strong glueline, even at low resin spreading levels [12]. Furthermore, the presence of lathe checks reduces continuous bond lines and deepens adhesive penetration, lowering bonding strength of plywood [30]. The thermal treatment reduced the number of lathe checks on the veneer's surface and ensured that the bond lines were continuous, resulting in a stronger plywood. Moreover, according to Bekhta et al. [31], the findings could also be due to the formation of acetic acid and increase the acidity of the wood veneers and subsequently facilitated the curing of the acid-catalyzed MUF resin which led to improved MOR and shear strength of the plywood samples.

However, at extremely high temperatures, drying defects such as cracks tend to be formed and cause strength loss [31]. Moreover, low wettability of the wood veneers surface will lead to poor wetting of resin and hence lead to inferior mechanical strength [32]. It is believed that the density decrement caused the observation in plywood thermally treated at 190 °C. As shown in Table 2, plywood made from thermally treated veneers at 190 °C had the greatest density loss of 8.22% when compared to the other treatment temperatures. Because the mechanical properties of plywood increased with density [33], a significant decrease in density is expected to have a negative impact on the mechanical strength of plywood.

Conclusions

Plywood panels have been fabricated using rubberwood veneers thermally treated at various temperatures in this study. The results of the study revealed that the dimensional stability of the plywood has been improved as indicated by lower TS and WA compared to that of untreated rubberwood plywood. The improvement in dimensional stability increased along with increasing treatment temperatures. Darkening of plywood surface has been observed and the distinct color changes can be observed at 170 °C onwards. As for mechanical properties, the highest MOR and shear strength were observed at the samples treated at 180 °C and started to decline when being treated at 190 °C. Meanwhile, the highest MOE value was recorded in the plywood samples being treated at 170 °C. Therefore, it can be concluded that, using the treatment conditions of 160-180 °C under 1 bar pressure for 15 min could be beneficial to enhance the properties of rubberwood plywood.

Abbreviations

ANOVA	Analysis of variance
ASTM	American Society of Testing and Material
EMC	Equilibrium moisture content
ISO	International Organization for Standardization
LVL	Laminated veneer lumber
MOE	Modulus of elasticity
MOR	Modulus of rupture
MUF	Melamine urea formaldehyde
TS	Thickness swelling
UTM	Universal testing machine
WA	Water absorption

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Author contributions

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Availability of data and materials

The data sets used or analyzed during the current study are available from the corresponding author upon reasonable request.

Declarations

Competing interests

The authors declare that they have no competing interests regarding the publication of this paper.

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