Effect of Thermo-Mechanical Modification on Mechanical Properties and Water Resistance of Plantation-Grown Vietnamese Acacia and Rubberwood

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ABSTRACT

Low density and poor mechanical performance often limit utilization of sawn wood from fast-growing plantation forests. Thermo-mechanical modification of timber (TMTM®) is one innovation for improving the utilization rate of light-weight wood species. The objective of this study was to determine the effects of thermo-mechanical modification and subsequent thermal modification on dry density, modulus of elasticity (MOE), compression strength, Brinell hardness and swelling behavior in immersion test on two fast-grown Vietnamese species, acacia (Acacia mangium) and rubberwood (Hevea brasiliensis). Test boards were modified in an industrial kiln where tangential thickness compression of 14% and 12% were aimed to acacia and rubberwood, either with or without subsequent thermal modification at 190 °C. Dry density, MOE, Brinell hardness, compression strength, and dimensional changes in water immersion of specimens were measured from the modified and reference materials, the latter ones being kiln dried at 50 °C. The results showed that changes in the mechanical properties were more evident for rubberwood than for acacia. In rubberwood, the MOE and compression strength of wood thermo-mechanically modified with or without thermal modification were higher than those of kiln-dried reference specimens throughout the thickness profile. In acacia similar differences between the modified and reference specimens were observed only in the surface layer. Density and Brinell hardness of thermo-mechanically modified rubberwood were higher than those of reference specimens, but after thermal modification they did not differ from (acacia) or were lower (rubberwood) than those of thermo-mechanically modified materials. Subsequent thermal modification increased the water resistance of thermo-mechanically modified specimens.

Keywords: acacia, drying, rubberwood, thermo-mechanical modification, thermal modification

BRIEF ORIGINAL

Vietnamese acacia (*Acacia mangium*) and rubberwood (*Hevea brasiliensis*) boards were modified using tangential thickness compression either with or without subsequent thermal modification at 190 °C. Changes in density, modulus of elasticity, compression strength, Brinell hardness and swelling behavior between untreated and modified wood were studied. The results showed that changes in the physical and mechanical properties were more evident for rubberwood than for acacia. The unfavourable effects of any single modification method can be partly compensated by applying combinations of modifications and by the use of appropriate process parameters.

1. INTRODUCTION

In general, mechanical properties of wood are highly dependent on wood density (Pelit et al. 2018). Fast-growing acacia (Acacia mangium) and rubberwood (Hevea brasiliensis) plantations have been successfully commercialized in Southeast Asia thanks to their adaptability to various conditions and short rotation period (e.g., Nguyen 2013; Hai et al. 2015). However, their applications in wood product industries are limited by their relatively low density and poor dimensional stability (Mohammed Raphy et al. 2011; Teoh et al. 2011; Nambiar et al. 2014; Shukla and Sharma 2018).

Densification or compression in transverse direction is considered as an effective and environmentally friendly method to increase the density of wood. Densification reduces the void volume of lumens and reshapes the

morphology of cells without loss of lignin, resulting in improvement of mechanical properties such as modulus of elasticity, compression strength, tensile strength, hardness, and abrasion resistance of low-density wood species (Anshari et al. 2011; Sandberg et al. 2013). However, once the atmospheric humidity and heat in compression process terminate, elastic energy stored in microfibrils of compressed wood will be released, resulting in the thickness along compression springing back towards its original shape, which is defined as "shape memory" or "set recovery" (Navi and Sandberg 2012). This phenomenon is concerned as the biggest problem regardless of the mechanical property elevation of compressed wood (e.g., Möttönen et al. 2015).

It is important to improve the dimensional stability of densified wood in the direction of compression, because changed thickness indicates changes in properties, thereby implying the success of densification. Although the shape memory cannot be totally eliminated, it can be restricted by adjusting the heat, compressive force, and pressing time during the densification process (e.g., Navi and Girardet 2000; Kúdela et al. 2018). Depending on the combination of relative humidity and temperature used during the process, densification can be categorized into two types, thermo-hydro-mechanical modification (THM) and thermo-mechanical modification (TM). In THM process, atmospheric temperature and humidity are well-controlled and the compressive forces are subjected against the radial or tangential direction of wood (Dogu et al. 2010; Sandberg and Kutnar 2016). In TM, the compression process takes place in an open system without detailed control of relative humidity, being also widely applied in the industries (Kúdela et al. 2018). If the conditions are well-controlled in THM, higher values in density and stronger surface layers would thereby be achieved resulting in surfaces well applicable in flooring, furniture, and decorative uses (Gao et al. 2016).

Post-compression thermal treatment is known as another effective way to improve the dimensional stability of compressed wood, thereby offsetting the shape memory of THM (e.g., Gong et al. 2010; Möttönen et al. 2013; Yan and Morrell 2014; Möttönen et al. 2015; Sandberg and Kutnar 2016; Marttila et al. 2017; Sandberg et al. 2017). During the thermal modification process, wood is exposed to high temperature (≥160°C) conditions (e.g., Millet and Gerhards 1972; Hillis 1975; Kocaefe et al. 2015). In this process, the structure of wood chemical components will be altered to different extent, thereby affecting other properties of wood (e.g., hygroscopicity, equilibrium moisture content, dimensional stability, fungal and insect resistance, mechanical properties, colour, and odour) (e.g., Sandberg et al. 2017). Optimized parameters during the modification processes need to be designed for specific species, considering their different characteristics (Sandberg et al. 2013).

Fast-growing plantations of acacia (Acacia mangium) and rubberwood (Hevea brasiliensis) have been successfully commercialized in Southeast Asia thanks to their adaptability to various conditions and short rotation period (Nguyen 2013; Hai et al. 2015). However, their applications in wood product industries are limited because of their low density and poor dimensional stability of wood (Mohammed Raphy et al. 2011; Teoh et al. 2011; Nambiar et al. 2014; Shukla and Sharma 2018). The timber is mostly used by kraft pulping industries.

The objective of this article is to investigate the effect of THM and combined THM and subsequent thermal modification on selected mechanical properties, i.e., modulus of elasticity, compression strength, and Brinell hardness, as well as extreme-condition swelling behavior of acacia and rubberwood.

2. MATERIALS AND METHODS

Logs of two different tree species from Vietnam, acacia (Acacia mangium) and rubberwood (Hevea brasiliensis) were transported by sea fright in green condition to Juankoski, Finland and then sawn to boards and thermo-mechanically modified. The pilot modification kiln patented by KWS Timber Tech Ltd allows wood drying, mechanical compression, and subsequent thermal modification in one single kiln unit. Different combinations of processes can be achieved by adjusting parameters such as temperature, hydraulic pressure, relative humidity, and treatment time. The boards were divided into six groups: four groups of sawn timber treated through different ways and two reference groups dried at 50°C in the oven but without any modification (Table 1).

Before modification, the boards were placed between perforated aluminum plates where the moisture content of boards could be indirectly controlled by the air circulation rate between these plates in the kiln (Fig. 1). Drying and hydraulic compression proceeded simultaneously. The drying temperature was steadily increased up to 130°C for acacia and 120°C for rubberwood, and the nominal degrees of mechanical compression (thickness change / original thickness) for acacia and rubberwood were set to 14 and 12 per cent, respectively. After drying and mechanical compression, a three-hour thermal modification at 210°C was instantly applied to the two groups of boards. During this process, certain amount of steam was applied in the kiln to protect the boards from darkening. After the thermal modification process, the system was cooled down, and pressure was released.

The moisture content (MC, %) and dry density (ρb, kg/m3) of the specimens from all boards were determined by the gravimetric method. The size of specimens which were taken from the other end of boards was 50 × 80 × 50 mm and 60 × 80 × 50 mm (thickness × width × length) for modified and unmodified boards, respectively. Modulus of elasticity (MOE) was determined by static three-point bending test according to the standard ISO13061-4, where the applied forces caused the deflection in the mid-span of each board. Matertest model FMT-MEC 100kN material testing device was used to carry out MOE test and record the MOE value of each board. Prior to MOE test, the boards of groups of A and K were dried in an oven with a temperature of 50°C for roughly 2 weeks, while those of AC, ACT, KC, KCT were conditioned indoors for two weeks in order to achieve the adequate moisture level (12±5%) required by the tests.

Compression strength parallel to the grain of each test specimen was determined according to ISO 13061-12:2017. All samples were stored in the conditioning chamber at 20 °C, 65% relative humidity until their mass was stable. The ultimate stress (σ, MPa) was determined using gradually increasing load in parallel to the grain direction:

$$\sigma = F_{max}/(a*b) \quad (1)$$

where Fmax is the maximum load force (N), a and b are the cross-sectional dimensions of the test specimen (mm).

The moisture contents of the mechanical test specimens from groups A and K were computationally adjusted to 12% by using the following formulae, which are valid for moisture contents of 12±5%:

$$E_{12} = E_w/(1-\alpha*(W-12)) \quad (2)$$

$$\sigma_{12} = \sigma*(1+ \alpha*(W-12)) \quad (3)$$

where E12 is the calibrated MOE value at the moisture content of 12%, σ12 is the calibrated compression strength at the moisture content of 12%, α is the correction factor (0.02) for the moisture content, W is the moisture content of wood during the test, determined according to ISO 13061-1.

Before immersion test, all test specimens were stabilized in the normal climate chamber at 20 °C, 65% relative humidity until their equilibrium moisture content (EMC) was reached, i.e., the mass did not change anymore. After that, the initial length, width and thickness of specimens were measured by caliper. Next, the specimens were soaked in buckets filled with water and stored in room temperature for 14 days. The dimensions were measured from the same positions once again after the 14 days of soaking. The dimensional changes were determined according to the formula:

$$\alpha = ((L_1-L_0)/L_0) *100\% \quad (4)$$

Where α is the swelling rate in length, width, and thickness (%), L0 is the initial dimension (length, width, and thickness) (mm), L1 is the dimension (length, width, and thickness) after the 14-day immersion (mm).

Brinell hardness (HB) (kg/mm2) of the test specimens was determined according to the standard EN 1534 (2010):

$$HB = 2F/(g*\pi*D*(D-(D2-d2)1/2) \quad (5)$$

where F is the maximum load applied (1,000 N), g is the acceleration of gravity (9.81 m/s2), π is the "pi" factor (3.14), D is the diameters of the indenter (10 mm), d is the average value of the diameter of the two residual indentations [(d1+d2)/2 mm] on specimen surface, d1 and d2 being the diameters of the residual indentation along the grain and across the grain, measured by caliper.

3. Results and Discussion

3.1. Dry Density

Mean values and standard deviations (STD) of moisture content (MC), dry density, modulus of elasticity (MOE), Brinell hardness (HB), compression strength from core part (CS-C), compression strength from surface part (CS-S) of acacia (A), thermo-mechanically modified acacia (AC), thermo-mechanically modified acacia with subsequent thermal modification (ACT), rubberwood (K), thermo-mechanically modified rubberwood (KC), and thermo-mechanically modified rubberwood with subsequent thermal modification (KCT) are shown in Table 2. Mean values, STDs, and significance levels (* the mean difference is significant at 0.05 level) of dimensional stability in different directions (T=thickness, L=length, W=width; thickness is along the direction of compression, length is along the longitudinal direction) of experimental groups of A, AC, ACT, K, KC, and KCT are given in Table 3. ANOVA

results of dry density, MOE, HB, CS-C, CS-S, and swelling T (thickness direction) of experimental gropus of A, AC, ACT, K, KC, and KCT are listed in Table 4.

Statistically significant difference was detected between K&KC, KC&KCT, suggesting that thermo-mechanical modification had a positive effect on dry density of rubberwood (increase by 7.3%) while subsequent thermal modification decreased the dry density from KC to KCT by 7.5%. This might indicate that the thermo-mechanical modification changed the structure of rubberwood tissue to contain less void space and more fibres per unit volume, thereby increasing the dry density (Kutnar et al. 2008; Fang et al. 2012). After the subsequent thermal modification, degradation of holocellulose, evaporation of certain extractives or volatile organic compounds (VOCs) could lead to the mass loss of cell walls (Sandberg and Kutnar 2016; Severo et al. 2016; Shukla and Sharma 2014).

On the other hand, no difference was found between the experimental groups of acacia. High initial moisture content or inappropriate compression temperature, pressure, or time might lead to the failure of compression where the whole thickness profile of acacia samples was not totally penetrated by the compressive pressure (Kúdela et al. 2018). Another reason could be that the strong spring-back effect in AC specimens counterbalanced the positive effect of compression on the density of acacia (Pelit et al. 2018). The long lasting sea fright of logs at green state may also have had adverse effects on physical and mechanical properties of wood.

3.2. Modulus of elasticity (MOE)

Regarding MOE values, thermo-mechanical modification and thermo-mechanical modification with subsequent thermal modification lead to an improvement of 39.5% and 35.9% in rubberwood, whereas there was no increase observed in acacia.

Several studies show that densification is an effective way to improve the mechanical properties of wood (e.g., Kutnar et al. 2008; Anshari et al. 2011; Fang et al. 2012; Möttönen et al. 2015; Gao et al. 2016). Generally, the bending stiffness of wood improves proportionally to density increase as a result of compression (Kutnar 2012). Increasing temperature up to 150 °C during compression also has a positive effect on MOE value of wood as it reduces the spring-back effect of compressed wood, although the increase of MOE caused by high temperature is not as obvious as that caused by high compression ratio (Tabarsa and Chui 1997; Lamason and Gong 2007).

The effect of thermal modification is dependent on the wood species, temperature, initial moisture content, surrounding atmosphere, and reaction time (Mitchell 1988). Therefore, the effects of thermal modification on MOE can be either positive (Fang et al. 2012; Shi et al. 2007) or negative (Gong et al. 2010; Johansson and Morén 2006), depending on the parameters applied during the process. High temperature and long processing time may decrease the MOE due to the deterioration of wood components, hemicellulose in the first hand (Yan and Morrell 2014; Korkut and Aytin 2015). In addition, since the proportion of amorphous cellulose decreases with increasing temperature (e.g., Sivonen et al. 2002; Yildiz and Gümüşkaya 2007), an increase in crystallinity of cellulose may cause an increase of the MOE. However, elevation in the MOE is associated with the reduced moisture content in the modified wood (Xie et al. 2013). Usually moisture content is negatively correlated with MOE below the FSP (Kretschmann 2010).

3.3. Compression Strength

Both thermo-mechanical modification and thermo-mechanical modification with subsequent thermal modification increased the compression strength of surface specimens of acacia by 23%, while the change in compression strength of core specimens was not significant. In case of rubberwood, the increments in compression strength of surface and core specimens were 19.9% and 25.0%, respectively.

There are several explanations for increment of compression strength achieved by thermal treatment. Firstly, crystallization and degradation of cellulose in amorphous region increases the proportion of crystalline cellulose, which increases the stiffness of wood in its longitudinal direction (Anderson et al. 2005; Yildiz et al. 2006); secondly, increased cross-linking of lignin polymer network can better connect and stiffen cellulose fibrils and prevent them from bending or crashing when they are subjected to compressive forces, thereby increasing the longitudinal compression strength of wood (Boonstra et al. 2007). It could also be explained by the fact that the lower moisture content detected in all modified specimens, i.e., decreased amount of bound water, resulted in increased compression strength of wood.

3.4. Swelling behavior in immersion test

The results show that compared to untreated specimens, both modification treatments and both species swell more in the direction of compression, i.e., 385%, 218%, 172%, and 114% higher dimensional changes in AC, ACT, KC, and

KCT, respectively, compared to their references. It seems that the irreversible set recovery phenomenon explained the swelling in the modified samples, in addition to the reversible hygroscopicity of wood itself (Fang et al. 2012).

As much as 21% less thickness swelling was detected in KCT specimens compared with KC specimens, which indicates that the set-recovery of rubberwood was partially eliminated. This might be explained by the following mechanisms: Firstly, thermal modification above 200°C can increases the cross-linking network in lignin, which increases the hydrophobicity of wood and reduces its swelling (Santos 2000). Secondly, the stored elastic stresses are released because of the degradation of hemicellulose in high temperature (Navi and Sandberg 2012). Elimination of some hygroscopic hydroxyl groups due to the depolymerization of carbohydrates explains the improved hydrophobicity in thermally modified wood (Xie et al. 2013). In addition, in cellulose components, amorphous regions in cellulose is degraded due to acid catalysis in high temperature, which increases the crystallinity and thereby decrease the accessibility of water molecules in hydroxyl groups (e.g., Cai et al. 2018; Kúdela et al. 2018).

3.5. BRINELL HARDNESS

Elevation of Brinell hardness of 58% was detected in KC samples, whereas no difference was found between A and AC. Flattening of fibre lumens and vessels in thermo-mechanical modification may increase the Brinell hardness of wood (Fang et al. 2012). However, slight loss of Brinell hardness takes place due to increased temperature during the thermo-mechanical modification (Fang et al. 2012). Typically, higher degree of compression leads to higher Brinell hardness value, although Rautkari et al. (2013) argued that Brinell hardness is mainly influenced by the density and hardness of the surface layer. In addition to varying densities among wood species, other factors such as modification temperature, final density, moisture content, techniques of measurement, and measuring conditions (e.g., load level, loading time) also have influence on the results (Holmberg 2000; Gašparík et al. 2016).

On the other hand, compared to AC and KC, subsequent thermal modification caused the reduction of Brinell hardness in ACT and KCT by 33% and 41%, respectively, which was probably due to the deterioration of cell wall structures (Pelit et al. 2015). High temperature and long processing time typically result in greater reduction in Brinell hardness because of the degradation of hemicellulose and lignin in high temperatures (Fang et al. 2012; Salca and Hiziroglu 2014). In addition to that, reduction of hardness due to the heat treatment was found to be highest for wood species with high density (Kesik et al. 2014; Salca and Hiziroglu 2014).

3.6. ECONOMIC ASSESSMENT OF THE PROCESS

A comprehensive assessment of the economy of thermo-hydro-mechanical modification would require an analysis of the entire value chain, from raw materials to end products (c.f., Sandberg et al. 2017). Compared to conventional processes the advantages gained by the THM modification include shorter production lead time, lower raw material and energy costs per cubic meter and improved properties and quality of end products. High temperature drying and modification of THM modification reduces the process time to minimum of 2 days compared to 10-20 days with conventional methods. As a result of reduced process time, labour, energy and product related capital costs are also lower. Low-priced domestic small diameter timber can be utilised in THM process enabling the replacement of more expensive and imported large diameter timber. The lower quality of small diameter timber can be compensated by densification which, to some extent, increases the hardness and strength of wood. The smaller production capacity of the THM modification kiln compared to conventional kiln of comparable size or price range is the main disadvantage from the economic point of view.

4. CONCLUSIONS

Thermo-mechanical modification is a potential technique to improve some properties of rubberwood (e.g., density, MOE, compression strength, Brinell hardness) and acacia (surface compression strength), but modified wood swells more than non-modified under extreme conditions. Post-compression thermal modification improves the dimensional stability under such conditions, but reduces the hardness of the surface.

The unfavourable effects of any single modification method can be partly compensated by applying combinations of modifications and by the use of appropriate process parameters. Therefore, further experiments are needed to investigate the influence of treatment time, compressive force, and modification temperature on acacia and rubberwood properties.

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Table 1. Treatments and number of boards in the six treatment groups.

Code	Group	Treatment	Number of boards	Compression degree	Time and temperature in thermal treatment
A	Unmodified acacia	Reference, 50°C oven-drying	14	-	-
AC	Thermo-mechanically modified acacia	Compression, kiln drying	10	14%	-
ACT	Thermo-mechanically modified acacia with subsequent thermal modification	Compression & thermal treatment	9	14%	210 °C, 3h
K	Unmodified rubberwood	Reference, 50°C oven-drying	12	-	-
KC	Thermo-mechanically modified rubberwood	Compression, kiln drying	11	12%	-
KCT	Thermo-mechanically modified rubberwood with subsequent thermal modification	Compression & thermal treatment	10	12%	210 °C, 3h

Table 2. Mean value ($\bar{x}$) and standard deviation (s) of physical and mechanical properties

Treatment	MC (%)		Dry density (kg/m³)		MOE (MPa)		HB (kg/mm²)		CS-C (MPa)		CS-S (MPa)	
	$\bar{x}$	s	$\bar{x}$	s	$\bar{x}$	s	$\bar{x}$	s	$\bar{x}$	s	$\bar{x}$	s
Untreated (A)	65.8	27.3	571	65	10.6	2.3	2.6	0.9	49.6	2.0	49.6	2.0
Compression (AC)	7.1	2.2	590	79	9.8	1.2	2.8	1.1	56.5	2.8	61.2	1.9
Compression & thermal (ACT)	3.1	0.4	553	70	10.6	1.0	1.9	0.6	58.8	1.9	61.0	3.1
Untreated (K)	47.9	7.2	620	40	8.1	0.8	2.5	0.5	49.7	1.4	49.7	1.4
Compression (KC)	6.0	0.3	665	40	11.3	1.4	3.9	0.9	59.4	1.9	59.8	2.4
Compression & thermal (KCT)	4.2	0.2	615	38	11.1	0.8	2.3	0.4	60.8	1.7	63.5	1.2

Table 3. Mean value ($\bar{x}$), standard deviation (s), and significance level (p) of dimensional stability in tangential (T), longitudinal (L) and radial (W) directions

Treatment		Swelling T	Swelling L	Swelling W
A	$\bar{x}$	1.1%	0.2%	0.7%
	s	0.7%	0.1%	0.5%
	p	0.001*	0.666	0.202

AC	$\bar{x}$	5.4%	0.1%	0.9%
	s	1.5%	0.3%	0.6%
	p	0.000*	0.978	0.616
ACT	$\bar{x}$	3.5%	0.1%	1.3%
	s	2.9%	0.1%	0.7%
	p	0.045*	0.996	0.022*
K	$\bar{x}$	2.7%	0.1%	2.3%
	s	0.8%	0.3%	1.1%
	p	0.000*	0.958	0.001*
KC	$\bar{x}$	7.4%	-0.1%	1.3%
	s	1.1%	0.1%	0.4%
	p	0.000*	0.987	0.231
KCT	$\bar{x}$	5.8%	-0.0%	1.8%
	s	0.9%	0.1%	0.4%
	p	0.000*	1.000	0.001*

Table 4. Significance level of the difference in dry density, MOE, Brinell hardness (HB), compression strength in the core (CS-C) and surface layer (CS-S), and swelling (T) between different treatments (significant at $p \leq 0.05$, marked with*)

Factor	Dry density	MOE	HB	CS-C	CS-S	Swelling T
A-AC	0.786	0.478	0.538	0.176	0.004*	0.000*
A-ACT	0.826	0.998	0.008*	0.124	0.021*	0.012*
AC-ACT	0.493	0.584	0.001*	0.974	1.000	0.085
K-KC	0.027*	0.000*	0.000*	0.007*	0.005*	0.000*
K-KCT	0.959	0.000*	0.578	0.002*	0.000*	0.000*
KC-KCT	0.019*	0.806	0.000*	0.981	0.597	0.002*

Fig. 1. A set of acacia boards (60×80×1200 mm) after the compression and thermal modification process.