Impregnation of Natural Rubber into Rubber Wood: A Green Wood Composite

Wassa Ruayruay and Sureurg Khongtong*

A green wood composite material was developed from the two environmentally friendly substrates natural rubber (*cis*-1,4-polyisoprene) and rubber wood *(Hevea brasiliensis*). Natural rubber (NR) was introduced into rubber wood by pressurization of NR latex, followed by the removal of the aqueous phase to allow only dry NR to remain inside the wood structure. Scanning electron microscopy images and the weight increase of the dry impregnated samples revealed the retention of dry NR within the rubber wood. The natural rubber enhanced the water resistance and compressive strength of the treated rubber wood.

Keywords: Wood impregnation; Wood composites; Rubber wood; Natural rubber

Contact information: Materials Science and Engineering Programs, School of Engineering and Resources, Walailak University, Thasala, Nakhon Si Thammarat 80160, Thailand;
** Corresponding author: ksureurg@wu.ac.th*

INTRODUCTION

The development of materials from sustainable sources has become a current challenge in materials science. This offers opportunities for improving material properties as well as producing more environmentally friendly materials. The focus of our research group has been the introduction of composite materials by integrating natural rubber (NR), a product from cultivated rubber trees (*Hevea brasiliensis*), and rubber wood, a by-product from aged rubber trees felled for replanting due to low latex yield.

Natural rubber and rubber wood are attractive because they are renewable raw materials and, in fact, have become the most consumed biopolymer in industry (Malaysian Rubber Board 2013). The initial form of NR is a colloidal latex, which consists of natural *cis*-1,4-polyisoprene particles in an aqueous phase collected from the bark of rubber trees (Blackley 1997) and is generally used for the manufacture of latex gloves and condoms. In addition, solidified NR latex is a major raw material for the production of vehicle tires. Rubber wood is currently an important source of timber in Southeast Asia because it is affordable as well as environmentally friendly, as it is a by-product of farm trees and normally thrown away. This wood, however, displays low durability and strength, especially when used in contact with water (Scheffer and Morrell 1998). Therefore, quality improvement is essential for wider applications.

Studies involving wood impregnation with polymeric materials to improve water resistance and strength have been reported. The inclusion of polystyrene maleic anhydride and the derivatives of polyacrylate into wood can upgrade its dimensional stability and water absorption (Uyttenhove and Tilquin 2005; Grelier *et al.* 2007). Bakraji and Salman (2003) found that the compressive strength of white poplar, cypress, and white willow can be enhanced by acrylamide and butyl methacrylate. The hardness of beech and spruce were elevated by introducing polyvinyl alcohol and polyacrylate into the wood (Solpan and

Guven 1999). Hamdan *et al.* (2010) improved the stiffness and thermal stability of wood by impregnation with dimethylacetamide. Poplar wood displayed a greater density and degree of crystallinity after impregnation with urea-formaldehyde (Wu *et al.* 2010). Additionally, rubber wood impregnation with synthetic polymers has been reported, with the result that treated rubber wood provided increased dimensional stability, water resistance, and strength (Devi *et al.* 2003; Mohamad *et al.* 2007).

Impregnation of a variety of synthetic polymers, as stated previously, is difficult and expensive for industrial-scale applications. Moreover, some monomers used for impregnation are harmful (Caldeira 2010), and thus the hazards of using polymer products have been mentioned (Teoh *et al.* 2011). Therefore, the system of NR-impregnated rubber wood proposed in this study can overcome these disadvantages. Simple pressurization of NR latex into the rubber wood prior to oven drying allows the retention of dry NR within the wood structure. Furthermore, this procedure is systematically analogous to the method of wood treatment by the impregnation of preservatives currently applied to the processing of rubber wood (Gnanaharan and Dhamodaran 1993), and may provide industrial applications for the manufacture of green wood composites with improved water resistance and strength.

EXPERIMENTAL

Materials

Wood samples with dimensions of approximately 20 mm in width (radial) × 20 mm in thickness (tangential) × 60 mm in length (longitudinal) were randomly cut from lumber of rubber trees (RRIM-600 variety), grown in the area of Nakhon Si Thammarat Province, Southern Thailand, that were about 25 years of age. The cut samples were dried in an oven at 103 ± 2 °C until they reached a constant weight.

It was found that the density of these dry samples was in the range of 0.62 to 0.68 g/cm^3, which is about the same as that of the rubber wood generally used in the industry. They were then kept desiccated until use.

Concentrated NR latex (high ammonium grade, 60% dry rubber content; Rubber Estate Organization, Thailand) was used for impregnation. Before use, the NR latex was gently stirred at an ambient (26 to 28 °C) temperature for about 1 h to remove the ammonia. To vary the percentage of dry rubber content (DRC) in the NR latex (12%, 24%, 36%, 48%, and 60%), water was added and the mixture was gently mixed for 20 min. The latex viscosity was monitored by a Brookfield RTV viscometer (Massachusetts, USA), and each value reported in this study (1, 2, 3, 4, 12, and 50 centipoises) is an average of at least three measurements at temperature of 26 to 28 ºC.

Methods

Scanning electron microscopy

Scanning electron microscopy (SEM) was performed on the representatives of treated and untreated wood samples. The samples (10×10×5 mm^3) were mounted on an aluminum stub with double-sided tape, sputter-coated with gold, and examined using an FEI Quanta 400 SEM (Oregon, USA) operating at an accelerating voltage of 10 kV.

Laser particle size analysis

The particle sizes of concentrated NR latex used in this study were monitored using a Coulter LS230 (California, USA) laser light scatter particle size analyzer (LPSA). Water (10 mL) was added into 10 mL of NR latex (60% DRC), and 10 mL of a mixer was used for this analysis.

Latex pressurization

Rubber wood samples were placed in a shop-made pressurization chamber (Fig. 1) at a vacuum of 635 mmHg for 15 min to remove the air and moisture from the samples. The chamber was then filled with 0% (water) to 60% DRC of NR latex, as stated in each experiment, and pressurized to 6 bars for 2 h. These conditions were sufficient to achieve the maximum latex embedding in the pressurized samples.

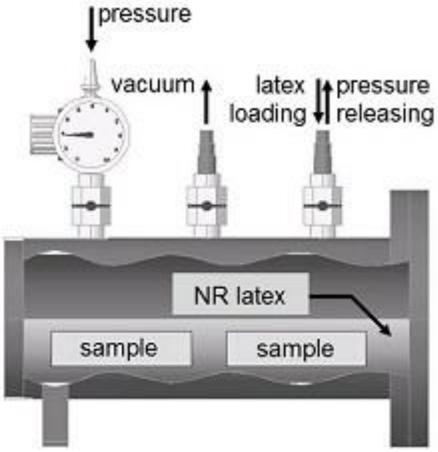

Fig. 1. An illustration of the pressurization chamber

Measurement of NR latex infiltration and dry NR inclusion

To investigate how much NR latex could infiltrate the wood samples under pressure, the excess latex on sample surfaces was removed after retrieving the samples from the pressurization chamber to ensure that the observed weight increase was due only to the embedded latex. The percentages of embedded latex (stated as the weight increase) in treated samples were calculated using Eq. 1,

$$\%\ weight\ increase = (W_2 - W_1) \times 100 / W_1 \tag{1}$$

where W_1 and W_2 are the sample weights before and after pressurization, respectively. The pressurized samples were then dried in an oven at 103 ± 2 °C until they reached a constant weight. The percentage of dry rubber inclusion in treated samples was then calculated using the same equation, where W_1 and W_2 are the sample weights before pressurization and after drying, respectively.

Water absorption and thickness swelling tests

The weight and thickness (in the tangential direction) of oven-dried specimens were taken before they were soaked in water for either 2, 24, or 48 h. After each time interval, they were retrieved from the water and the excess water on their surfaces was removed using a soft cloth. The weight and thickness of these specimens were then measured again. Water absorption and thickness swelling were calculated for these controls using the following equations,

$$\%\text{water absorption} = [(W_1\text{-}W_0) \times 100/\ W_0] \tag{2}$$

$$\%\text{thickness swelling} = [(T_1\text{-}T_0) \times 100/\ T_0] \tag{3}$$

where W_0 and W_1 are the weight before and after water soaking, respectively, and T_0 and T_1 are the thickness before and after water soaking, respectively.

Compressive strength testing

Compressive strength parallel and perpendicular to the fiber grain was evaluated using a Lloyd LR150R (Hampshire, UK) universal testing machine with a load cell of 150 kN. This test was performed at ambient temperature with a crosshead speed of 2 mm/min (ISO 3132 1975; ISO 3787 1976). The instrument software (Nexygen 4.5) was used to control the test and analyze the data. Each value reported in this study is an average of at least 10 measurements.

RESULTS AND DISCUSSION

Mechanism of Natural Rubber Impregnation into Rubber Wood

To achieve the goal of this study, *cis*-1,4-polyisoprene particles in NR latex must move through the pathways in the wood structure, as illustrated in Fig. 2, during latex pressurization. Dry rubber would then remain inside the wood structure after the pressurized samples were dried in an oven.

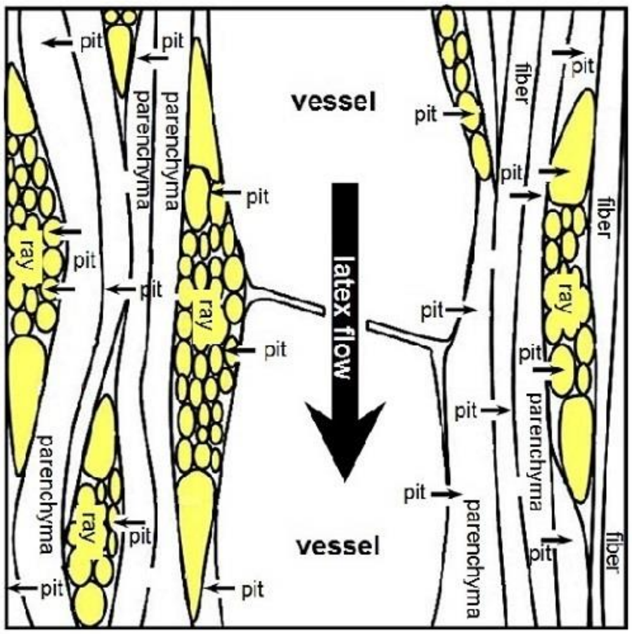

Fig. 2. Schematic illustration of the pathways for NR latex flow in rubber wood

Consistent with this hypothesis, the images obtained by a scanning electron microscope reveal the microstructure of dry rubber wood (Fig. 3). Figure 3a shows the structure of fibers and vascular tissues with diameters in the approximate range of 20 to 120 μm. Vessel pits, mainly oval in shape with the length of the minor axis about 4 to 8 μm, are seen in Fig. 3b. However, the narrowest pathway found in this system was the fiber pit, which is also oval in shape with the length of the minor axis about 2 to 4 μm (Fig. 3c). In separate experiments, NR latex was examined by a laser particle size analyzer, which found particle sizes in the range of 0.04 to 2.92 μm, with the average value of 0.80 ± 0.42 μm. The results from SEM and LPSA, therefore, suggested the possibility that NR particles should be able to penetrate the structure of rubber wood upon pressurization.

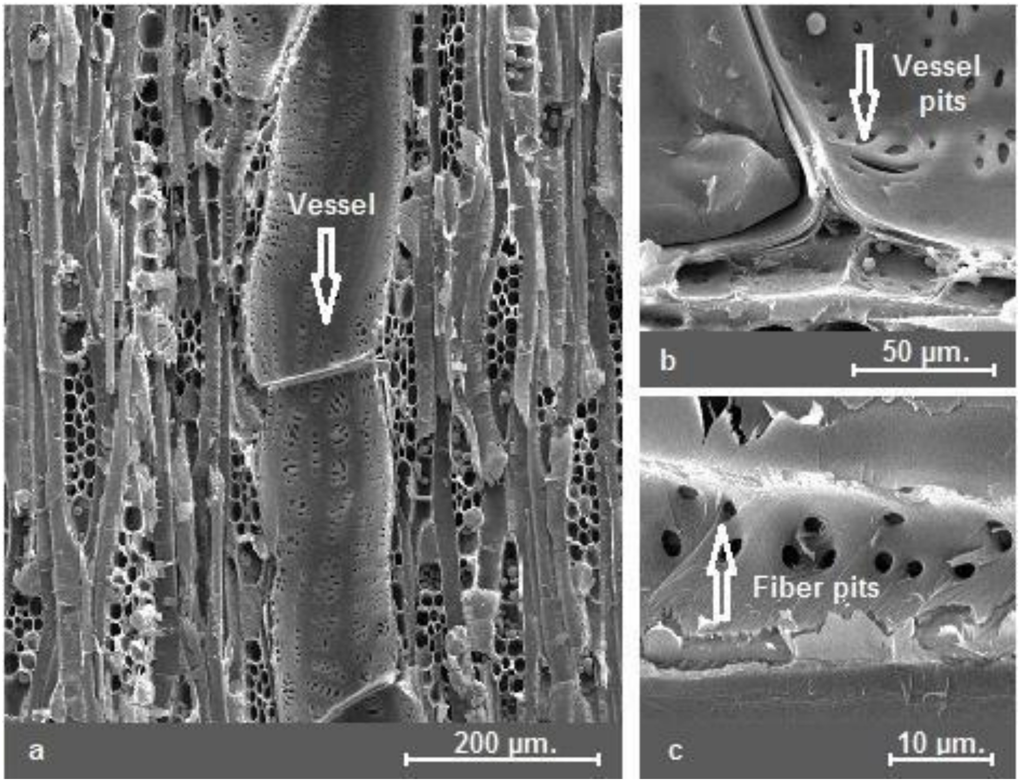

Fig. 3. SEM images of wood structure: (a) vascular tissues and fibers, (b) vessel pits, and (c) fiber pits

To test this hypothesis, wood samples were pressurized in a chamber filled with different dry rubber contents of NR latex. The weight increase after pressurization was monitored; this reflects the amounts of NR latex retained in treated samples. The result of a weight increase on latex pressurized samples (stated as embedded NR latex) is shown in Fig. 4a, suggesting that NR latex was able to penetrate into the rubber wood during pressurization. In addition, the amount of embedded latex approached the upper limit, approximately 90%, when pressurized with NR latex at 12% DRC. This limit was indicated by a comparison of the pressurized water control samples. The amounts of embedded latex, however, declined from about 90% to 50% when the DRC of latex was gradually raised from 12% to 60%. The decrease in the latex-embedded levels observed here is accounted for by an increase in the viscosity of the latex (Blackley 1997). Natural rubber latex with a lower viscosity (and values of DRC) penetrates into the wood structure more easily than NR latex at a higher viscosity.

The same set of pressurized samples was then dried in an oven at 103 ± 2 °C until a constant weight was reached, normally within 72 h. This step was performed to remove the aqueous portion of the NR latex embedded inside the pressurized wood samples. As a result, dry NR particles were retained within the samples and were monitored by the weight gain. As expected, all latex-pressurized samples displayed a weight increase after drying, while that pressurized by water did not (Fig. 4b). The additional weight found here, therefore, inferred that the fixed dry NR was inside the samples and in the range of about 4% to 8% when the DRC of latex was varied from 12% to 60%. The amount of dry NR that remained in the wood samples treated by latex with different DRC values, however, was smaller than what it should have been. For example, wood samples treated by the latex with 12% DRC displayed a value of embedded latex of about 85%, while the ideal retention of dry NR should be about 10%, rather than the 4% shown in Fig. 4b. This may be due to the coagulation of rubber particles in NR latex during pressurization into rubber wood. Basically, each pair of adjacent NR particles in latex was stabilized by the attractive and repulsive forces of the surrounding ionic molecules (Blackley 1997). During pressurization, rubber particles were forced to move through the narrow pathways, *e.g.*, vessel pits and fiber in the wood structure. This brings about the destabilization of these forces from operating between each pair of adjacent particles. Consequently, coagulation of NR particles took place and occluded the pathways inside the wood. Thus, only the aqueous phase of latex was allowed to move through the wood structure.

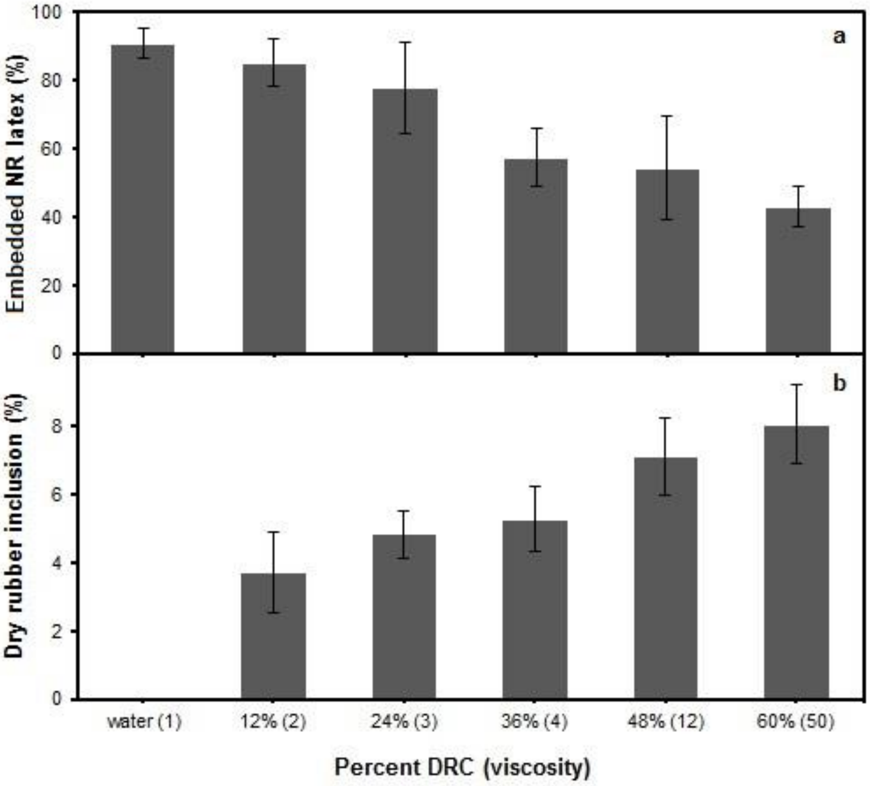

Fig. 4. Impregnation levels of treated wood samples as a function of DRC of NR latex: (a) embedded latex and (b) dry NR inclusion. The numbers the in parentheses of x axis labels refer to the latex viscosity (in centipoises) at each value of DRC.

To ensure that NR impregnated the oven-dry pressurized rubber wood, representatives of treated samples (~8% of rubber inclusion) were then compared with controls using SEM. The SEM images revealed the substrate layers and accounted for dry NR deposited on the walls of the vascular tissue of the treated samples, while the untreated controls were normal with clearly visible pits (Fig. 5).

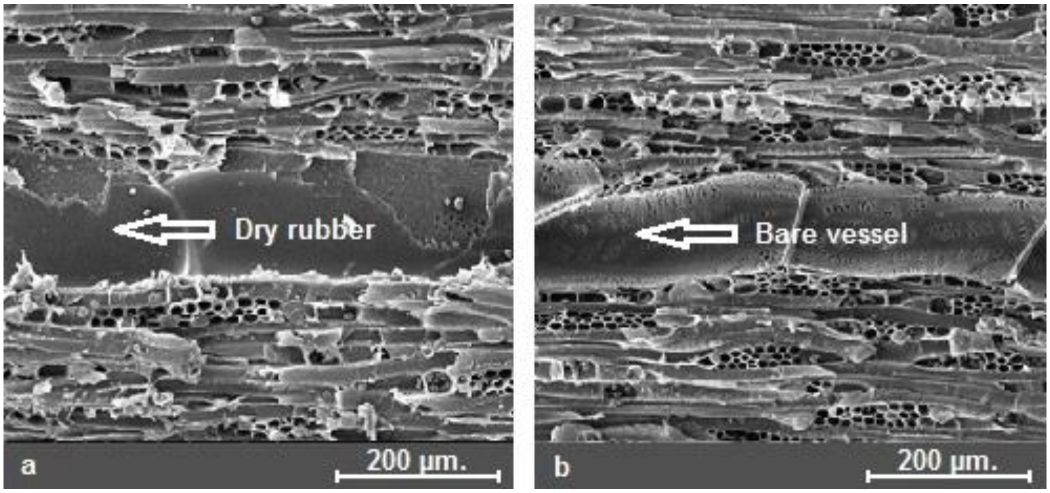

Fig. 5. SEM images of (a) oven dry pressurized rubber wood and (b) untreated rubber wood

Water Resistance and Compressive Strength of Treated Rubber Wood

As depicted in Fig. 5, the structure of wood was impregnated with dry NR, which may enhance water resistance to rubber wood, as this hydrophobic natural polymer acts as a barrier against water. To test this, water absorption and thickness swelling experiments were conducted on treated samples, pressurized by NR latex with 12%, 24%, 36%, 48%, and 60% DRC, compared with the controls; therefore, the levels of dry rubber inclusion were in the range of 0% to about 8%, as the result of dry rubber inclusion previously shown in Fig.4b. As expected, treated samples (about 4% to 8% inclusion) exhibited lower levels of water absorption than did the untreated samples (0% inclusion), especially when the soaking interval reached 48 h (Fig. 6a). Differences in the level of water absorption between treated and untreated samples likely reflect the role of the rubber portion in the wood structure. Additionally, at all soaking periods, samples with a greater level of rubber inclusion displayed the tendency to better resist water uptake. The similar result was also observed when thickness swelling was evaluated from the same set of samples (Fig. 6b). This result, however, became apparent in the samples with higher degree of rubber inclusion, *e.g.*, about 6% to 8% inclusion.

It was also expected that dry NR impregnated in the wood structure could strengthen the treated rubber wood, since it partly filled the void inside the wood structure. The compressive strength of treated samples, conducted parallel to the fiber grain, was therefore evaluated and found to increase as a function of rubber inclusion (Fig. 7a). The compressive strength of rubber wood was enhanced up to about 62% as rubber inclusion increased from 0% to ~8%. The same tendency was also observed when the experiment was conducted perpendicularly to the grain of fibers (Fig. 7b), with the magnitude of increasing strength about 66%. The result shown in Fig. 7b, however, revealed greater values of standard deviation (as shown by the error bars), especially on the treated samples, than did the parallel test. This might reflect the non-uniformity of dry NR impregnated in the samples of each treatment, as only the middle part of the specimens was compressed by the test instrument in the later experiment (ISO 3132 1975).

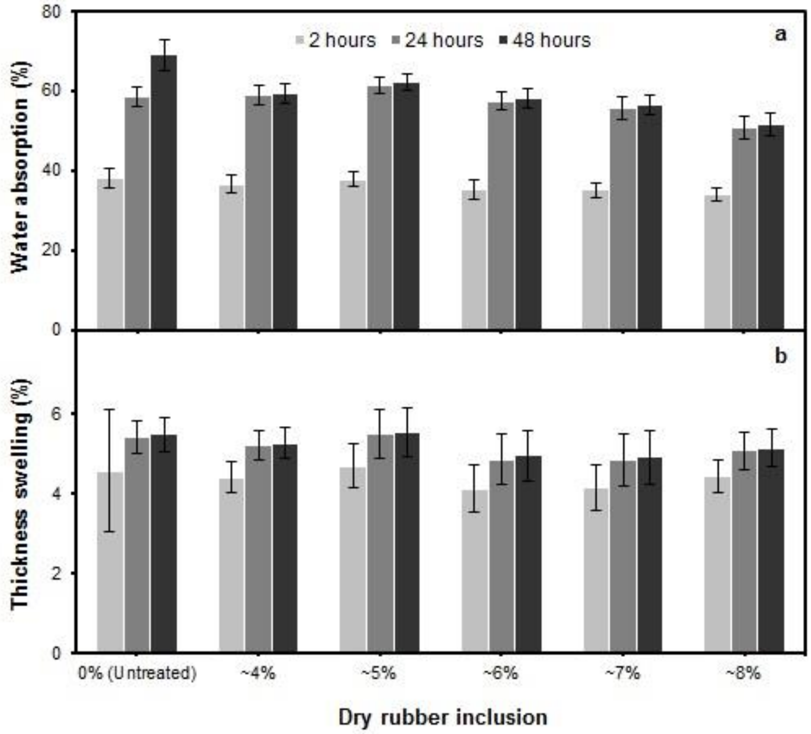

Fig. 6. Water resistance test of rubber wood at soaking periods of 2, 24, and 48 h as a function of dry rubber inclusion: (a) water absorption and (b) thickness swelling.

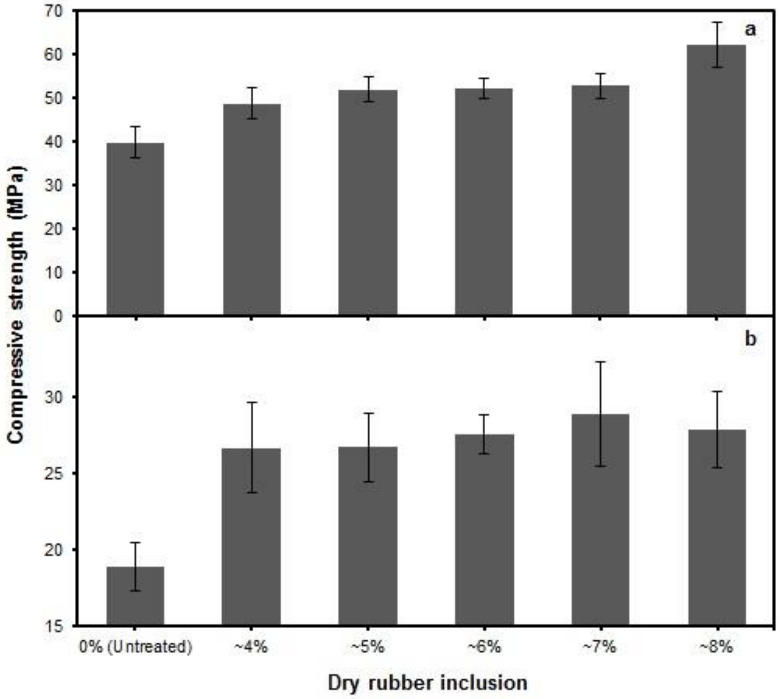

Fig. 7. Compressive strength of rubber wood as a function of dry rubber inclusion: (a) parallel to the fiber grain and (b) perpendicular to the fiber grain

CONCLUSIONS

1. Natural rubber was impregnated into rubber wood by the pressurization of NR latex and the pressurization of rubber wood specimens with greater DRC values of latex provided a higher degree of impregnation.
2. Scanning electron microscope and LPSA data suggest that rubber particles in NR latex were able to penetrate into the wood structures.
3. The evidence of NR inclusion in dry, pressurized samples was revealed by both weight gain and by SEM images.
4. The water resistance results indicate that impregnation of NR into rubber wood enhanced water absorption and thickness swelling resistance to the treated samples.
5. Compressive strength tests, conducted parallel and perpendicular to the fiber grain, displayed the improvement of this mechanical property up to about 62% and 66%, respectively, after applying the impregnation treatment to the specimens.

ACKNOWLEDGMENTS

We gratefully acknowledge support of this study from the Office of the Higher Education Commission (OHEC), Thailand. We also thank the Thailand Research Fund (TRF) and Nakhon Si Thammarat Rajabhat University for providing partial support for this study.

REFERENCES CITED

Bakraji, E. H., and Salman, N. (2003). “Properties of wood–plastic composites: Effect of inorganic additives,” *Radiation Physics and Chemistry* 66(1), 49-53.

Blackley, D. C. (1997). *Polymer Latices Science and Technology,* Chapman and Hall, London.

Caldeira, F. (2010). “Boron in wood preservation a review in its physico-chemical aspects,” *Silva Lusitana* 18(2), 179-196.

Devi, R. R., Ali, I., and Maji, T. K. (2003). “Chemical modification of rubber wood with styrene in combination with a crosslinker: Effect on dimensional stability and strength property,” *Bioresource Technology* 88(3), 185-188.

Gnanaharan, R., and Dhamodaran, T. K. (1993). “A pilot plant investigation of boron treatment of rubber wood: arriving at an economical treatment schedule,” *Holz als Roh- und Werkstoff* 51(4), 279-282.

Grelier, S., Castellan, A., and Podgorski, L. (2007). “Use of low molecular weight modified polystyrene to prevent photo degradation of clear softwoods for outdoor use,” *Polymer Degradation and Stability* 92(8), 1520-1527.

Hamdan, S., Rahman, M. R., Ahmed, A. S., Talib, Z. A., and Islam, M. S. (2010). “Influence of N,N-dimethylacetamid on the thermal and mechanical properties of polymer-filled wood,” *BioResources* 5(4), 2611-2624.

ISO 3132 (1975).*Wood-Testing in Compression Perpendicular to Grain*. International

Standards Organization, Geneva, Switzerland.
ISO 3787 (1976). *Wood-Test method-Determination of Ultimate Compression Stress Parallel to Grain*. International Standards Organization, Geneva, Switzerland.
Malaysian Rubber Board (2013). *Natural Rubber Statistics 2013*, (http://www.lgm.gov.my/nrstat/nrstats.pdf), accessed December 2013.
Mohamad, J. S., Rozman, H. D., and Rahim, S. (2007). "Rubberwood-polymer composites: The effect of chemical impregnation on the mechanical and physical properties," *Malaysian Polymer Journal* 2(2), 1-11.
Scheffer, T. C., and Morrell, J. J. (1998). "Natural durability of wood: A worldwide checklist of species," *Forest Research Laboratory, Oregon State University*, Research Contribution 22, pg. 24.
Solpan, D., and Guven, O. (1999). "Preservation of beech and spruce wood by allyl alcohol-based copolymers,"*Radiation Physics and Chemistry* 54(6), 583-591.
Teoh, Y. P., Don, M. M., and Ujang, S. (2011). "Assessment of the properties, utilization, and preservation of rubberwood (*Hevea brasiliensis*): A case study in Malaysia," *Journal of Wood Science* 57(4), 255-266.
Uyttenhove, A., and Tilquin, B. (2005). "Wood xylowall: New process to reduce water exchange by an intra-graft of polymer," *Nuclear Instruments and Methods in Physics Research* 236(1-4), 137-140.
Wu, G. F., Lang, Q., Qu, P., Jiang, Y. F., and Pu, J. (2010). "Effect of chemical modification and hot-press drying on poplar wood,"*BioResources* 5(4), 2581-2590.

Article submitted: March 19, 2014; Peer review completed: June 26, 2014; Revised version received and accepted: July 11, 2014; Published: July 21, 2014.