

Fabrication of Transparent Composites from *Pinaceae* Wood Packaging Residues

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Abstract

In the 21st century, mankind has witnessed the rapidly development of all industries with a lot of new products in a variety of types and designs. However, this development has been also causing many problems with the society and the ecological environment such as a wasteful excess of products, exhaustive exploitation of natural resources, indiscriminate deforestation, and waste pollution affecting the living environment, ecosystem, and human health. Many organizations and governments are calling for environmental protection, limit waste emissions, and find good solutions to use the recycled materials as raw materials in production plants. This study would like to provide a solution which not only utilizes waste packaging *Pinaceae* wood for recycling as raw material but also supplies to the market with a green product responding to the durability requirements in fact. *Pinaceae* wood packaging residues were chemically processed through two stages. The amount of lignin extract from *Pinaceae* was determined by the method of Tappi 222 om-02 which was significantly influenced by the extraction parameters such as temperature, time and concentration of the treatment solution. Morphological modification of wood materials was analyzed by SEM micrographs. In particular, the mechanical properties of the Epoxy/TPW2 composite green material have been significantly improved with increasing up to 206–540 % compared to the original *Pinaceae* wood. The optical properties of the wood have completely changed from opaque pine wood with the optical transmittance of 8 % into transparent composite material with the optical transmittance up to 85 % using UV-vis spectroscopy analysis.

Keywords

green composite, *Pinaceae* wood, lignin extraction, chemical treatment, transparent

1 Introduction

The solid waste pollution is currently a top concern in most countries all over the world. Indeed, the overpopulation leads to increase demand for food and consumer goods causing more waste, mainly domestic waste, agricultural by-products and industrial waste. In the past, there were many research works used the agricultural by-products such as sisal coir fiber [1], banana fiber [2], water hyacinth fiber [3, 4], cashew nut shell [5], leaf fiber [6] and bamboo fiber to reinforce in wood plastic composites based on epoxy resin [7, 8]. There are twin-screw extruders manufactured with screw length (L) at 36D with advanced technology to produce sheets from wood pulp reinforced thermoplastic composite. In which, the thermoplastics are normally high density polyethylene, polypropylene, polyvinyl chloride and even Acrylonitrile Butadiene Styrene. Meanwhile, the types of domestic waste are sorted, mixed with additives and then they are composted in many

days to produce fertilizer for the agricultural production. For the industrial solid waste, the recycling has still faced to many difficulties due to the variety of origins, containing many different components included hazards and toxic substances or heavy metals [9–14]. This is a big obstacle for the recycling and use of the industrial solid waste as raw materials for other industries.

Research by Nguyen and Nguyen [13] in 2021 treated kraft paper packaging by-products to manufacture light-weight panels with sandwich core structure, applied in the field of building materials. In addition, there are a lot of *Pinaceae* wood packaging that is discarded piled up or used as firewood with low economic value in factories. The wooden packages mainly contain machines or spare parts in industrial production. Therefore, it is necessary to have a solution for utilization this raw material as a raw material for other industries.

The wood and agricultural by-products must be treated before used as raw materials for other industries. Vaccarino et al. [15] reported that the highest amount of lignin was extracted in solution of NaOH and Na_2CO_3 at 60 °C. Silverstein et al. [16] studied the effect of sulfuric acid, sodium hydroxide, hydrogen peroxide, ozone on cotton fibers. In which, the highest value of lignin extraction at 65 % was in the solution of 2 % NaOH for 90 minutes at 120 °C. Zhu et al. [17] used sodium chlorite solution to extract lignin and the lignin content was drastically reduced with less than 3 % remaining in the samples. The use of chemical solutions to extract the lignin from *Pinaceae* was first studied by Fink [18] in 1992. According to the study, the combination of polymer and reinforcing materials have the same refraction index at 1.56 increased the light transmittance through the material. Li et al. [19] developed the above method to remove the lignin in chip wood with a thickness less than 1 cm, and then they were combined with some resins such as polystyrene and polymethylmetacrylate. The wood used in the studies had bright white color such as oak, balsa wood, ash wood and others in the form of thin sheets with a thickness of 0.5–3.0 mm.

After treated, the wood-based materials are improved on engineering properties such as strength, toughness, low density and low thermal conductivity [20, 21]. According to Li et al. [22] and Wang et al. [23], the wood-based materials can be used for insulating and cooling in buildings, producing Perovskite solar cells for saving energy [10]. According to Müller et al. [24], the absorption of light is strongly related to the chemical composition of the wood. In which, the lignin is able to absorb about 80–95 % of light transmitted through the wood. Therefore, when the light absorption of the wood or wood-based materials is rocketly reduced, they will become transparent. This can be conducted by deigninification or neutralization of pigments (chromophores) in the lignin [25, 26].

This study utilized the thin *Pinaceae* wood chips from waste wood packaging to conduct the lignin treatment and then produce green composite materials with many outstanding advantages on engineering properties compared to the original *Pinaceae* wood.

2 Materials and methodology

2.1 Raw materials

The wooden packaging was chosen in type of white *Pinaceae*, popularly grown in Canada. The packaging was often used for the packaging of machinery, industrial spare

parts or consumer goods as shown in Fig. 1. The *Pinaceae* wood was sawn into white bars, the wooden surface was naturally without being coated any material. The wood has been hulled and processed for exterior cleaning. In all experiments, the *Pinaceae* samples were cut into thin sheets with a thickness of 2.0 mm.

2.2 The chemical treatment of *Pinaceae* wood

The samples of white *Pinaceae* wood were cut into squares of 30 ± 2 mm with a thickness of 2.0 ± 0.1 mm. The lignin in *Pinaceae* wood was treated with a mixture of NaOH and Na_2SO_3 solutions. In which, the Na_2SO_3 concentration was fixed at 0.4 M and the NaOH concentration was changed in the range of 2.0–3.0 M. The extraction temperature was carried out from 30–80 °C and the treated time was from 1–7 hours. The color changes of the lignin extract were observed and calculated the extracted lignin content. The *Pinaceae* wood samples were then washed several times with hot water and neutralized with acetic acid. The samples were further bleached with solution of 2.5 M H_2O_2 for 1–7 hours with a temperature of about 50 °C, washed again and dried to a constant volume.

The experiments were conducted in the temperature conditions of 23 °C, relative humidity of 80 % at the Material Technology Laboratory, Ho Chi Minh City University of Food Industry. The experimental processes are shown in Fig. 2.

2.3 Method of calculation for the lignin content

The method of TAPPI 222 om-02 [27] was used to calculate the lignin content in *Pinaceae* wood. The *Pinaceae* wood samples were crushed into powder and put in beaker containing 15 mL of 72 % sulfuric acid. The mixture was continuously stirred and the beaker was kept in the water bath at 20 ± 1 °C for 2 hours. Water was added from 300



Fig. 1 *Pinaceae* wood packaging for containing cargo

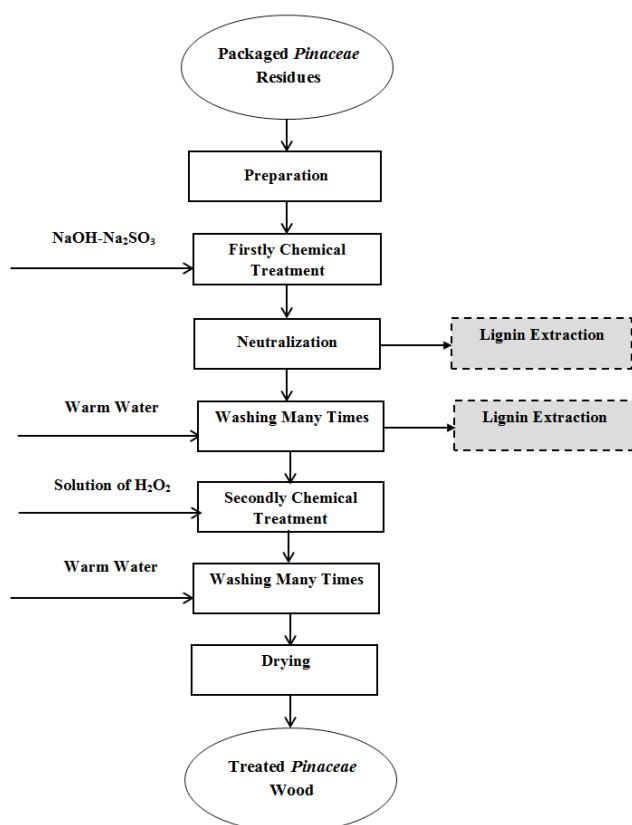


Fig. 2 The chemical treatment processes for the *Pinaceae* wood packaging residues

to 400 mL in beaker to rinse and dilute so that the concentration of sulfuric acid in solution was at 3 %. The solution was boiled for 4 hours, and then the lignin was settled to the bottom of the flask for 12 hours. The precipitate was filtered and dried at 105 ± 3 °C to constant mass.

2.4 Fabrication of green composite material based on epoxy reinforced *Pinaceae* wood residues

An epoxy resin of EPD 3120 pre-polymer was mixed with EPC 3809 hardening agent (Nan Ya Plastics Corporation, Taiwan). The reinforcement materials for the composite were samples of TPW2 sample (chemically treated *Pinaceae* wood) and UPW (untreated *Pinaceae* wood) with a thickness of 2.0 ± 0.1 mm. The *Pinaceae* wood samples were submerged in epoxy mixture for 30 minutes to have enough time for epoxy mixture to penetrate inside the wood samples. The composite samples were put in an oven with convection air and dried according to the processing parameters of 90 °C for 2 hours, 120 °C for 1 hour and continued at 140 °C for 1 hour to harden epoxy completely. And then the samples were cooled in a desiccator (containing silica gel) for 24 hours to stabilize the dimensions and durability. Finally, the samples were cut into the specimens according to mechanical measurement standards.

2.5 Microstructural characteristics of *Pinaceae* wood and the green composite

Morphology of the *Pinaceae* wood samples before and after chemical treatment was characterized using Scanning Electron Microscope (SEM) of Hitachi S-4800 (Japan) at the Institute of Nanotechnology - Vietnam National University of Ho Chi Minh City.

2.6 Analysis of optical transmission of the *Pinaceae* wood and the green composite materials

The analytical process was carried out according to "ASTM D1003-13 Standard Test Method for Haze and Luminous Transmittance of Transparent Plastics" [28] – standard measuring method for turbidity and optical transmittance of transparent resin – using UV-Vis spectrophotometer (UV-1800 Series, Shimadzu, Japan) to measure optical transmission (Transmittance). A very high intensity light source was used with the spectral region stretches from 190–1100 nm.

2.7 Evaluation on mechanical strength of the *Pinaceae* wood and the green composite materials

The compressive strength was tested according to ASTM D695-15 [29] standard using device of LLOYD LR 30K at the Mechanical Measurement Laboratory of the Material Research Center, Ho Chi Minh City University of Technology. The samples were prepared in sizes of ASTM standards D695 with length (l) \times width (b) \times height (h) at $20 \times 20 \times 12$ mm, respectively. It is noted that the compression speed was at 1.3 mm per minute.

The flexural strength of samples was tested according to ASTM D790-17 [30]. This method allows the determination of the flexural properties of reinforced or non-reinforced resins including high-modulus composites. The sample was put on two supports, the load was applied in the center of the sample until the specimens were broken. The bending velocity was of 1.5 mm per minute and the distance between the two supports was at 60 mm.

3 Results and discussion

3.1 Effects of the processing parameters to changes of the extracted lignin content and the *Pinaceae* wood

3.1.1 Effects of NaOH concentration on the extracted lignin content

The mixtures of NaOH and Na_2SO_3 solution were used to extract lignin from *Pinaceae* wood samples with a fixed concentration of Na_2SO_3 solution at 0.4 M, the extracted temperature of 60 °C and the extraction time was at

5 hours. The concentration of NaOH solution was changed in the range of 2.0–3.0 M. The color of extracted lignin solutions (color bar chart) and the extracted lignin content (line graph) are shown in Fig. 3.

The changes of lignin content according to NaOH concentration which divided into three stages:

- Stage (1): concentration of NaOH solution from 2.0 to 2.3 M,
- Stage (2): concentration of NaOH solution from 2.3 to 2.5 M and
- Stage (3): concentration of NaOH solution from 2.5–3.0 M.

In the first stage (Stage (1)), the extract color had a little change, correspondingly, the lignin content increases slowly from 6.18–8.20 %. In this sodium hydroxide concentration range, the lignin content increases linearly with that of NaOH solution as shown in Fig. 3.

In Stage (2), the concentration of NaOH changed from 2.3 to 2.5 M, the lignin content increases rapidly from 8.20 % to 10.40 % according to the concentration of NaOH solution. With the concentration of NaOH 2.5 M or over, the lignin extract starts to change color clearly as shown in Fig. 3 corresponding to a lignin content of 10.40 %.

At Stage (3), with higher concentrations of NaOH from 2.5–3.0 M, the extracted lignin color changed to dark yellow corresponding to an increase of lignin content from 10.40 to 11.02 %. The change of the lignin content is linear according to the concentration of NaOH solution. However, it increases very slowly compared to the first stage.

It is easy to realize that at the NaOH concentrations less than 2.5 M, the extracted lignin capacity was not high. From Fig. 4, the fracture point appears at 2.5 M NaOH concentration with the lignin extract starts to change color and the lignin content was extracted quite a lot (over 10.40 %).

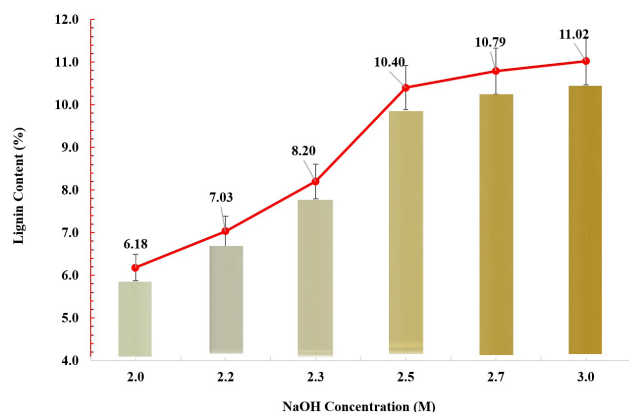


Fig. 3 The changes of lignin content and its extract color in alkaline condition with various concentration

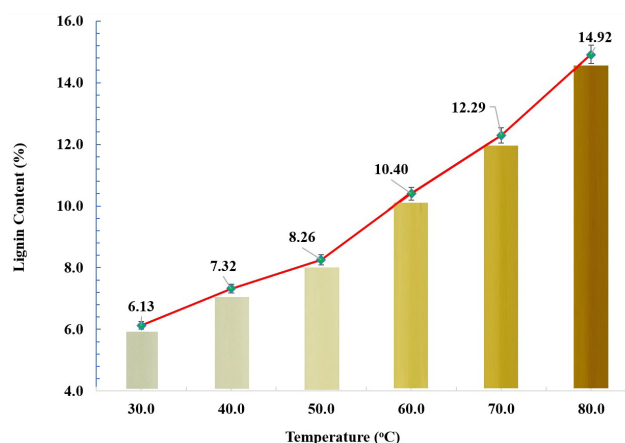


Fig. 4 Effects of the extracted temperature to the lignin content

With the NaOH concentrations from 2.5–3.0 M, the lignin extraction capacity increased slowly, specifically at the 3.0 M NaOH concentration, the lignin content was only 11.02 %. Therefore, the treatment for lignin in *Pinaceae* wood was not carried out at NaOH concentrations over 3.0 M. The concentration of NaOH at 2.5 M was suitable for pine wood treatment. The experimental results are consistent with previous studies and used for the next experiments [3, 7, 8].

3.1.2 Effects of extraction temperature on Lignin content

The extracted temperature was investigated in the range of 30–80 °C for 5 hours using a mixture of NaOH 2.5 M and 0.4 M Na_2SO_3 solution to extract lignin from *Pinaceae* wood samples. The results of the extracted lignin content (line graph) and its color changes (color bar chart) are presented in Fig. 4.

When the temperature of mixture was increased from 30 to 80 °C, the color of the lignin extraction changed from colorless to brownish yellow corresponding to the higher extracted lignin content. From 30 to 50 °C, the lignin content increases slowly; However, the extracted lignin rate increases faster in the temperature range of 50–80 °C. From 70–80 °C with higher temperature and long treatment time, the wood samples were deformed and cannot continue for the next experiments. Therefore, the extracted temperature was chosen for the experiments was at 60 °C.

3.1.3 Effects of the first extracted time using NaOH- Na_2SO_3 solution

The solution of NaOH/ Na_2SO_3 with NaOH concentration of 2.5 M was used for the experiments at temperature of 60 °C. The extraction time was carried out from 1 to 7 hours with the results as shown in Fig. 5.

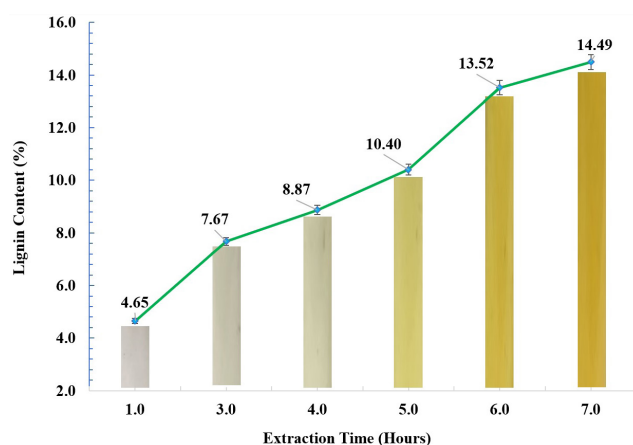


Fig. 5 The changes of lignin content and its extract color during extracted time from 1 to 7 hours using NaOH-Na₂SO₃ solution

The color of lignin extract changed from colorless to dark yellow during the extracted time from 1 to 7 hours as shown in the color bar chart. Thus, the longer the extraction time is, the more the lignin content extracted. Specifically, the lignin content increased from 4.65 % for 1 hour to 14.49 % for 7 hours. It is clear that the extracted lignin content increased more than 3 times.

From the extracted time from 1 to 5 hours, the extract color had a little change, correspondingly, the extracted lignin content increased from 4.65 % to 10.40 % which increased more than 2 times within 4 hours. However, from 5 to 6 hours, the lignin content rose from 10.40 % to 13.52 % higher than 1.3 times within 1 hour). The color of extract also changed clearly from pale yellow to dark yellow. Finally, from 6 to 7 hours, the lignin content changed slowly from 13.52 % to 14.49 % which has an increase of 1.07 times within 1 hour. Thus, the lignin content was quite low with the extracted time less than 5 hours. The lignin content tended to increase slowly with the extracted time higher than 5 hours. Therefore, the extracted time at 5 hours was selected to carry out for the experiments.

3.1.4 Effects of the second extraction time to the Lignin content using H₂O₂ solution

The first treated *Pinaceae* wood samples with NaOH 2.5 M and Na₂SO₃ solution at 60 °C for 5 hours were used to continue the second treatment with H₂O₂ solution. Concentration of H₂O₂ solution was fixed at 2.5 M during the treated time from 1 to 7 hours. The experimental results for the changes of lignin content (line graph) and the color of *Pinaceae* wood samples after treatment (color bar chart) are shown in Fig. 6.

After the second treatment with H₂O₂ solution from 1 to 7 hours, the extracted lignin content continuously changed from 17.13 to 24.34 % which has an increase of

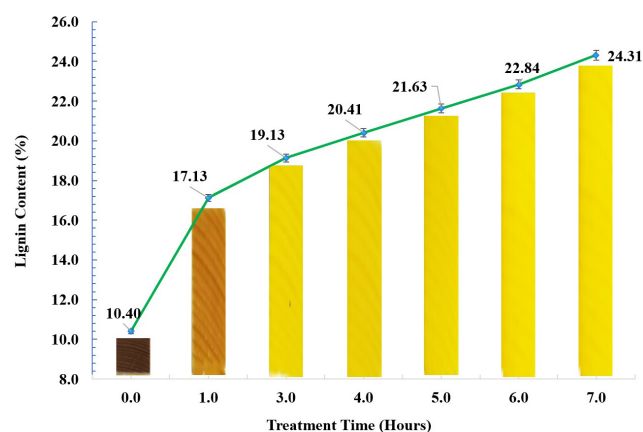


Fig. 6 The changes of the total extracted lignin content and its extract color during 1 to 7 hours using H₂O₂ solution

1.5 times as shown in Fig. 7. After treated for 1 hour with H₂O₂ solution, the color of *Pinaceae* wood changed from dark brown to lighter brown with lignin content about 17 %. After treated for 3 hours with H₂O₂ solution, the brown color of the wood sample disappeared to change yellow. With treatment for 5 hours in H₂O₂ solution, the samples had a bright yellow color corresponding to the extracted lignin content at 21.63 % calculated for the total of 2-time treatments. For the treated time over 5 hours, the extracted lignin content increased rapidly reaching to 24.31 %. Because the lignin content extracted is very high and quite close to the content of lignin in the *Pinaceae* wood. Therefore, the treatment time in H₂O₂ solution should be at 5 hours. In this treated condition, the *Pinaceae* wood samples had bright color and the extracted lignin content is not too high to affect the structure and durability of the *Pinaceae* wood.

3.1.5 The changes of *Pinaceae* wood color after secondly treatment

The changes of *Pinaceae* wood color were observed according to the first treatment time (t_1) with the mixture of NaOH/Na₂SO₃ solution for 1 to 13 hours and the second treatment time (t_2) using H₂O₂ solution with changes in the range of 1 to 5 hours. The results were simultaneously recorded with 2 parameters of t_1 and t_2 as shown in Table 1.

The longer treatment time in the solution of NaOH/Na₂SO₃ mixture is, the more the amount of extracted lignin is. This caused correspondingly the color of the *Pinaceae* wood changed from yellow brown to black after 13 hours due to the dissolution of lignin, tannins and pectin in extracted solution. Therefore, it is necessary to use H₂O₂ solution to treat wood color. The experimental results showed that the color of the *Pinaceae* wood was gradually lighter after bleaching and cleaning.

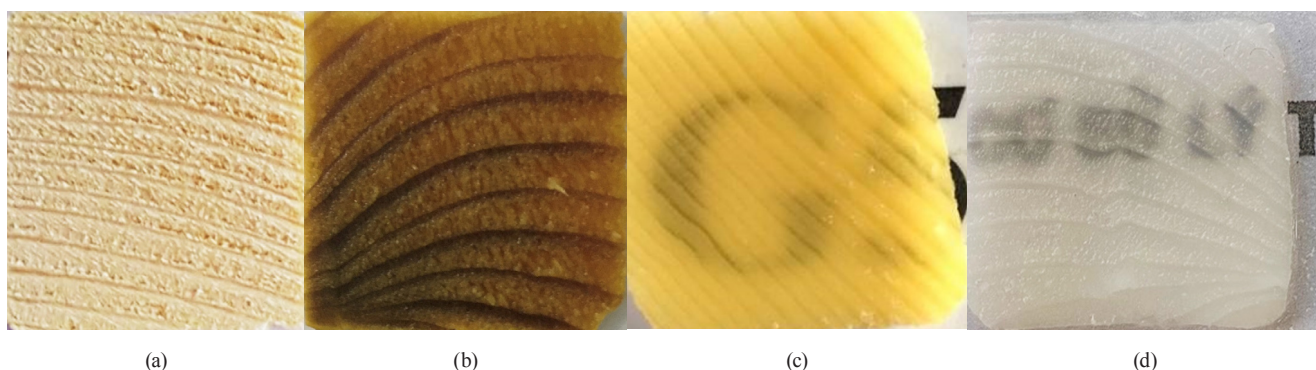


Fig. 7 The color of *Pinaceae* wood and green composite materials reinforced *Pinaceae* wood: (a) The initial white *Pinaceae* wood without treatment (UPW); (b) The *Pinaceae* wood after the first treatment using NaOH-Na₂SO₃ solution (TPW1); (c) The *Pinaceae* wood after the second treatment with H₂O₂ solution (TPW2); and (d) The green composite materials based on Epoxy/TPW2

Table 1 The changes of lignin content and the color of *Pinaceae* wood samples during the treated time from 1 to 7 hours using H₂O₂ solution

		t_1 (hours)					
		3	5	7	9	11	13
The Color of <i>Pinaceae</i> Wood after the First Treatment with Time t_1 (hours)							
The Color of <i>Pinaceae</i> Wood after the Second Treatment Time t_2 (hours)	1						
	2						
	3						
	4						
	5						
Lignin content (%)		17.13	19.13	20.41	23.12	24.15	25.01

Table 1 shows that there are 2 samples treated in conditions of $t_1 = 7$ hours; $t_2 = 5$ hours and $t_1 = 9$ hours; $t_2 = 4$ hours which are bright and uniform color, no wood grain and transparency. It is noted that t_1 is the treatment time with solution of NaOH and Na₂SO₃ and t_2 is the treatment time with H₂O₂ solution.

3.1.6 Effects of the treatment times to changes of the *Pinaceae* wood using H₂O₂ solution

The first treated *Pinaceae* wood samples with NaOH and Na₂SO₃ solution at 60 °C for 5 hours (NaOH 2.5 M solution) were used to continue for the second treatment with H₂O₂ solution. The samples were treated with 2.5 M H₂O₂ solution with the number of treatments from 1 to 5 times for the purpose of wood whitening. The experimental results are shown in Table 2.

Table 2 The color changes of *Pinaceae* wood with the number of treatments using H₂O₂ solution

Number of Treatment	1	2	3	4	5
The Color of <i>Pinaceae</i> Wood after Treatment					
Lignin Content (%)	19.13	21.24	21.32	21.53	21.92

Initially, the *Pinaceae* wood has ivory-white with several light brown streaks. The wood is completely opaque and the light cannot be transmitted it as shown in Fig. 7 (a). After treatment with the solution of NaOH and Na₂SO₃ at 60 °C, the compounds such as lignin, pectin, tannins and tree sap were separated and dissolved in the solution causing the wood color changed into dark brown as shown in Fig. 7 (b). The wood samples were continuously secondly treated with H₂O₂ solution which has light color with

fading of the wood grain and the wood color becomes homogeneous as shown in Fig. 7 (c).

Table 2 shows that the content of extracted lignin using H_2O_2 solution after the first treatment was at 19.13 %, the second treatment was at 21.24 % and others. Finally, the extracted lignin content reached to 21.92 % after treated for 5 times with the solution of H_2O_2 . It is noted that the extracted lignin content only had a little change (from 21.24 to 21.92 %) with number of treatments increased from 2 to 5 times. However, the color of *Pinaceae* wood clearly changed from dark brown with wood grain to bright and uniform color without wood grain and transparency. After combined with transparent and colorless epoxy resin, the green composite materials were fabricated using the epoxy resin reinforced with TPW2 which have bright color with the blurred wood grain and transparent. In particular, the Epoxy/TPW2 green composite sample is almost transparent allowing light to penetrate easily as presented in Fig. 7 (d).

3.1.7 The changes of *Pinaceae* wood weight after chemical treatment

According to Pettersen [31], the content of lignin in white *Pinaceae* wood is about 27 % in weight of the wood. The experimental results in Table 1 show that after the first treatment for 13 hours with solution of NaOH and Na_2SO_3 and the second treatment for 2 hours, the extracted lignin content was at 25.01 %. Or after the first treatment for 11 hours with solution of NaOH and Na_2SO_3 and the second treatment for 3 hours with H_2O_2 solution, the extracted lignin content was at 24.15 %. The samples of *Pinaceae* wood were extract a part of lignin and some un-cellulosic compounds such as pectin, tannins and sap. Because the lignin is a biopolymer with a role in binding the hemicelluloses and cellulose walls in the wood. If the lignin is extracted too much, it will affect the properties of composite products [32]. The content of lignin remaining in *Pinaceae* wood after treatment is about 2 % which are consistent with the study of Li et al. [20, 22]. In these studies, the residual lignin content after treatment was less than 3 %. Therefore, after 2 times of chemical treatment, the lignin content was extracted about 25 %. This proves that the volume of *Pinaceae* wood has been significantly reduced to 25 % less than that of initial wood (UPW) after treatment.

3.2 Microstructure and morphologies of the materials

There were 3 materials used to observe the microstructure and morphologies included UPW, TPW2, and composite based Epoxy/TPW2. Fig. 8 (a) and (b) shows that

the untreated *Pinaceae* wood (UPW) has smooth fiber surfaces because the fibers were covered with initial insoluble mainly compounds such as lignin, pectin, tannins, insoluble wax compounds. In addition, the compounds of lignin and binders made closely bonds with cellulose and hemicellulose in the microstructure of the *Pinaceae* wood. Therefore, it is difficult to observe the fiber surface as well as the arrangement of structure among the fibers in the original *Pinaceae* wood sample.

For the secondly treated *Pinaceae* wood (TPW2), Fig. 8 (c) shows that the surface of TPW2 fibers became rougher than UPW, no longer as smooth as the original UPW sample. Because a part of lignin and the above uncellulose components had been extracted and separated from the *Pinaceae* wood fibers in high alkaline condition of NaOH and Na_2SO_3 solution. These experimental results are consistent with the previous research of Nguyen and Nguyen [3] when treatment for hyacinth fibers in high alkaline solution.

In addition, the arrangement of the fibers in *Pinaceae* wood was also changed due to the loss of lignin not only on the fiber surfaces but also among the wood fibers. This leads to the expansion of micropores among the *Pinaceae* wood fibers due to the lignin loss in the alkaline solution as shown in Fig. 8 (d). These proves that the *Pinaceae* wood treatment had changed the microstructure and morphologies of the samples.

During the fabrication of modified *Pinaceae* wood reinforcement composite (Epoxy/TPW2), the *Pinaceae* wood was impregnated with epoxy resin. Therefore, the epoxy resin not only completely covered the surface of *Pinaceae* wood fibers but also were permeable the inner structures of fibers. Therefore, it is difficult to clearly observe the surface structure of the wood fibers as shown in Fig. 8 (e). The micropores among the *Pinaceae* wood fibers were also filled with the epoxy resin blocks causing difficult to observe the structural arrangement of the fibers inside the composite as shown in Fig. 8 (f). The experimental results are consistent with previous studies of Nguyen and Nguyen [3] when treatment of water hyacinth and bamboo fibers in high alkaline condition to reinforce composite materials.

3.3 Optical transmission of the materials using UV-Vis spectroscopy with transparent characteristic of the composite based Epoxy/TPW2

The samples of UPW 2 mm, Epoxy/TPW2 2 mm, Epoxy/TPW2 5 mm, and Pure Epoxy 2 mm were used to test for optical transmission. Fig. 7 (a) shows that the original untreated *Pinaceae* wood (UPW sample) has a light brown

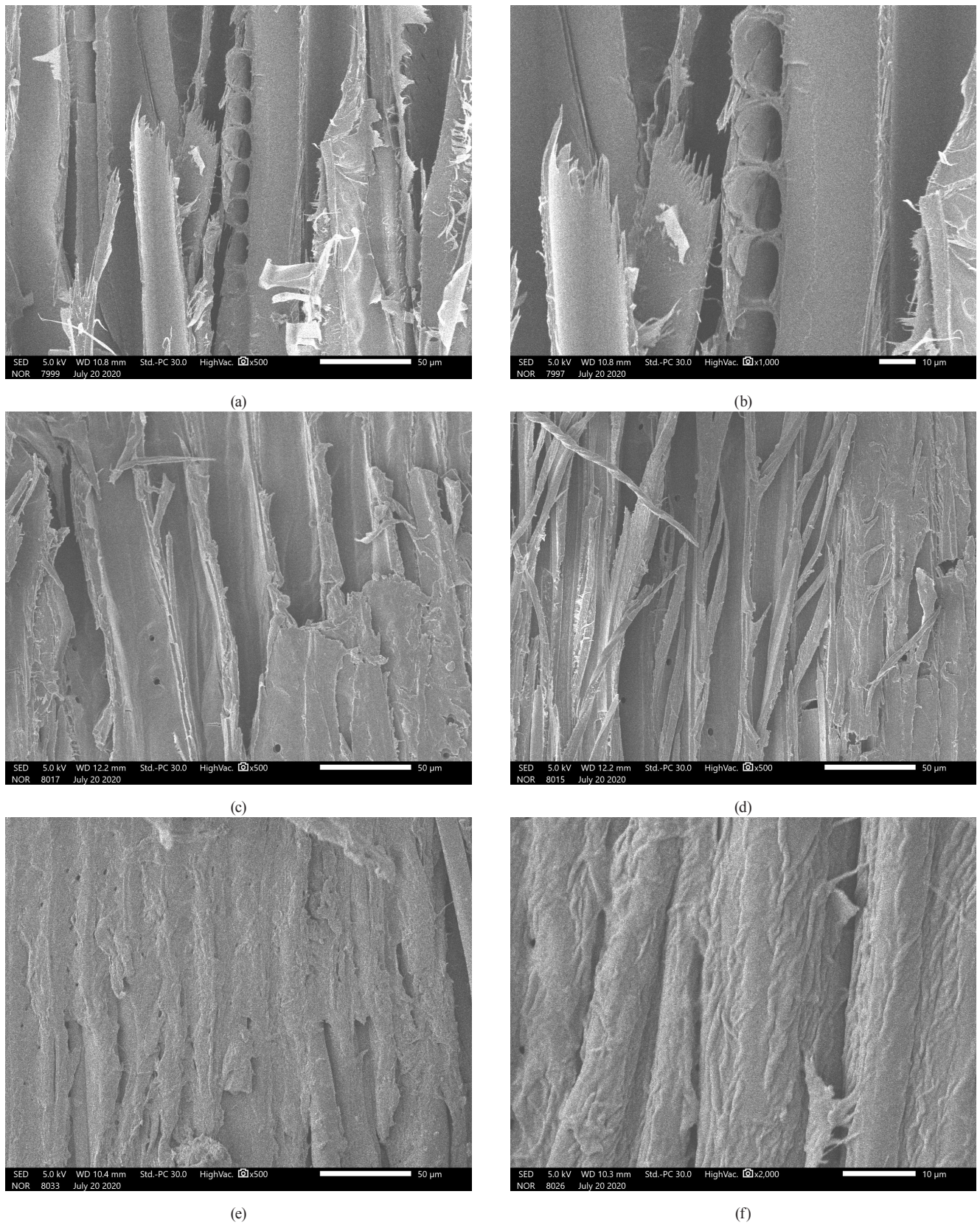


Fig. 8 The microstructure and morphologies of the *Pinaceae* wood fiber surfaces with samples of UPW, TPW2, and Epoxy/TPW2: (a) Initial UPW at magnification of 500×; (b) Initial UPW at magnification of 1000×; (c) TPW2 at magnification of 500×; (d) TPW2 at magnification of 1000×; (e) Epoxy/TPW2 at magnification of 500×; and (f) Epoxy/TPW2 at magnification of 1000×

color due to the presence of some light-absorbing components such as lignin, chlorophyll and tannins. In addition, the UPW sample is completely opaque and the light cannot be transmitted. The analysis results by UV-Vis spectrophotometer showed that the optical transmittance of UPW samples is very low only at 8 % as shown in Fig. 9 which is consistent with research by Müller et al. [24].

Fig. 7 (d) shows that the color of the wood lightens due to the chemical treatment, most of the lignin, chlorophyll and tannins have been dissolved and extracted. In combination with a transparent epoxy resin substrate, the green composite of Epoxy/TPW2 has very high transparency compared to the untreated *Pinaceae* wood sample. The UV-vis spectroscopy shows that the optical transmittance of the Epoxy/TPW2 sample increased rocketly up to 85 %, completely consistent with the loss of lignin content in raw material. It is noted that the transparency of the pure epoxy resin is at 95 % (transparent form) and consistent with previous studies [12, 33]. It is noted that the sample of Epoxy/TPW2 2 mm with 2 mm thickness had transparency at 85 % and it is higher than that of sample Epoxy/TPW2 5 mm with 5 mm thickness with transparency at 42 %.

3.4 Effects of the treatment processes to mechanical strength of the green composite based epoxy reinforced the *Pinaceae* wood

3.4.1 Evaluation on flexural strength of the materials

There are 4 materials tested the mechanical strength including UPW (untreated *Pinaceae* wood), TPW2 (treated *Pinaceae* wood after 2 treated stages), Epoxy/

UPW (the composite based epoxy reinforced untreated *Pinaceae* wood), and Epoxy/TPW2 (the composite based epoxy reinforced treated *Pinaceae* wood after 2 treated stages). The samples were prepared with the thickness at 5.0 ± 0.1 mm according to ASTM D790-17 [30]. The values of flexural strength and flexural modulus are shown in Fig. 10 and Table 3.

Fig. 10 show that the sample of TPW has a flexural strength of 12.11 MPa, which is lower than that of the original UPW sample at 14.63 MPa. Thus, after treatment, TPW sample was reduced 17 % values of flexural strength compared with UPW sample. This shows that the extraction of lignin from the *Pinaceae* wood decreased the flexural strength of the material. Because the lignin is a cross-linked biopolymer roling as a binder among cellulose, hemicelluloses and other components in the natural wood structures. The chemical solutions broke the bonding partials of the above compounds leading to a decrease in the mechanical properties of the *Pinaceae* wood samples.

The epoxy-based composite materials reinforced UPW and TPW2 have high values of flexural strength at 30.15 MPa and 65.20 MPa, respectively. Specifically, the composite sample of Epoxy/UPW increased 206 % compared with the flexural strength of UPW. The flexural strength of Epoxy/TPW2 composite sample reached to 65.20 MPa with an increase of 540 % compared to the flexural strength of the TPW sample at 12.11 MPa. The experimental results show a marked improvement in flexural strength when the samples of UPW and TPW2 were used as raw materials to fabricate epoxy-based composites.

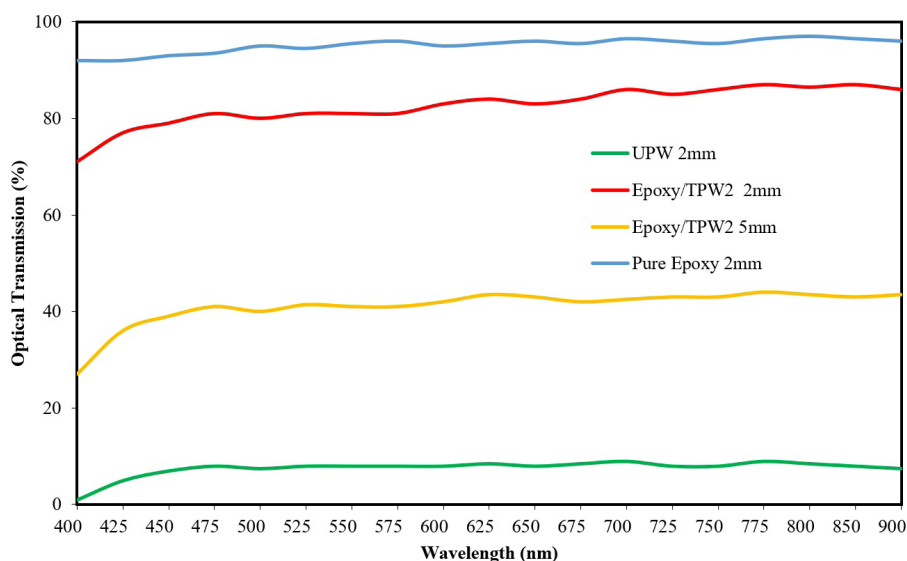


Fig. 9 The optical transmission of samples with various thickness: UPW 2 mm, Epoxy/TPW2 5 mm, Epoxy/TPW2 2 mm, and Pure Epoxy 2 mm

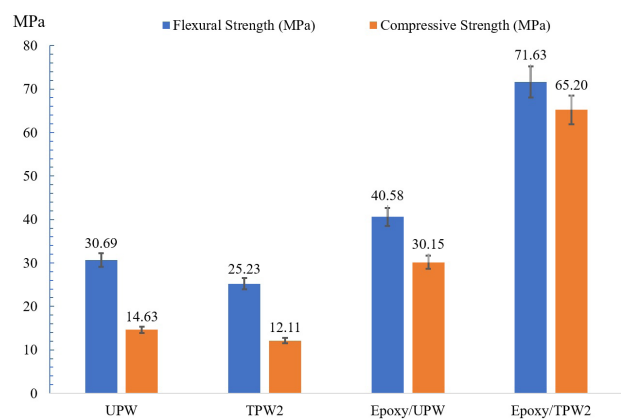


Fig. 10 Improvement on flexural strength of the composite based on epoxy reinforced the treated *Pinaceae* wood

Table 3 Flexural modulus and compressive modulus of the materials

Modulus	Materials			
	UPW	TPW2	Epoxy/UPW	Epoxy/TPW2
Flexural Modulus (MPa)	150.20	112.34	458.22	1673.85
Compressive Modulus (MPa)	320.90	276.60	382.70	424.73

Moreover, the flexural strength of the Epoxy/TPW2 composite reaches to 65.20 MPa, which is 216 % higher than that of the Epoxy/UPW composite at 30.15 MPa. This result shows a role of the wood chemical treatment in improving flexural strength of the composite materials reinforced the wood.

The results are suitable to the studies of Faruk et al. [34] and Zhang et al. [35] on the strength improvement of composite reinforced with the treated bamboo fibers or the study of Nguyen and Nguyen [3] on the composite reinforced the treated water hyacinth and the treated bamboo [7, 8]. This confirms the important role of *Pinaceae* wood treatment to improve the flexural strength of the composite.

3.4.2 Evaluation on compressive strength of the materials

The compressive strength and compressive modulus of the materials of UPW, TPW2, Epoxy/UPW, and Epoxy/TPW2 were tested according to ASTM D695-15 [29] with the results as shown in Table 3 and Fig. 10.

Fig. 10 shows that the compressive strength of the sample of UPW is at 30.69 MPa, which is 17.79 % higher than that of the sample of TPW2 at 25.23 MPa. Thus, after the chemical treatment, the *Pinaceae* wood was reduced the compressive strength due to partial extraction of lignin as similarly to the flexural strength. The composites of Epoxy/UPW and Epoxy/TPW2 increased dramatically

the compressive strength compared to that of the initial samples of UPW and TPW2. In which, the compressive strength of Epoxy/UPW composite sample increased to 40.58 MPa. Similarly, the compressive strength of Epoxy/TPW2 composite sample reached to 71.62 MPa. Thus, there were rocketly changes up to 234 % in the compressive strength of composite materials from 30.60 MPa (Epoxy/UPW) to 71.62 MPa (Epoxy/TPW2). The increase in compressive strength indicates that the successful treatment of *Pinaceae* wood has rocketly increased the compressive strength of the composite-based materials.

The values of compressive strength in this study are consistent with the previous studies for improvement on mechanical strength of the composites reinforced by the recycling materials with chemical treatment. The investigations with the same results included the studies of Akash et al. [1] on the composite using sisal and coir fiber to reinforce; Nguyen and Nguyen [3] on using bamboo to reinforce the composites; Abdul Motaleb et al. [6], used banana and fiberglass blends; and Naveen Reddy et al. [2] fabricated the composite based on epoxy reinforced pineapple fiber; as well as Do et al. [36], Maisuriya et al. [37], Antov et al. [38, 39] studied on glass fiber, banana, wood fiber, agricultural waste for reinforcing the composite-based materials.

4 Conclusions

The *Pinaceae* wood had a significantly changes on the properties of physical, chemical, morphologies, microstructure, and UV-vis, and SEM after the chemical treatment. The low alkaline solutions of NaOH and Na₂SO₃ and H₂O₂ mainly extracted and separated lignin and pectin and tannin compounds in the *Pinaceae* wood. The experimental results show that the chemical type, concentration and extraction time had significantly affected on the extracted lignin content. The secondly treated *Pinaceae* wood (TPW2) was used to reinforce for composite materials based on epoxy resin which improved dramatically on its mechanical properties. In particular, the optical properties of the wood have changed from a completely opaque to an almost transparent after combined with an epoxy resin substrate. The successful recycling of *Pinaceae* wood packaging waste contributes to the manufacture of green composite materials that are environmentally friendly products as well as reducing input costs due to reusing the wood waste. Further research should be conducted testing different application properties of composite materials such as thermal conductivity, sound insulation, chemical resistance, and others.

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