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CHARACTERIZATIONS OF TWO TYPES OF TANNIN-BONDED *RHIZOPHORA SPP*. PARTICLEBOARDS AS PHANTOM MATERIALS FOR PHOTON BEAMS

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Abstract- ***Particleboards were fabricated using Rhizophora spp. wood particles as phantom material in for photon beams with a target density of 1.0 g/cm^3. Two types of tannin, namely the extracted tannin and the commercialized tannic acid were added into the particleboard to improve the physical strength of the particleboards. The strength of the particleboards was measured according to Japanese Industrial Standards (JIS A-5908:2003). The microstructure of the particleboards was investigated using a Scanning Electron Microscope (SEM) coupled with an Energy Dispersive X-ray Analysis (EDXA) to determine the elemental composition of the particleboards. The mass attenuation coefficient of Rhizophora spp. particleboards were measured by using the attenuation of x-ray fluorescent (XRF) photons from niobium, molybdenum, palladium, silver and tin metal plates that provided photon energies between 16.59 and 25.26 keV. The CT images of the particleboards were obtained to determine the CT number and mass attenuation coefficient at 80, 120 and 140 kVp x-ray energies. The results showed that tannic acid provided better improvement of physical strength of the particleboard based on the internal bond strength, thickness swelling and water absorption tests. The mass attenuation coefficient of tannic acid-Rhizophora spp. particleboards were found to be the nearest to calculated XCOM of water indicated by the χ^2 values. The CT study showed that tannic acid-Rhizophora spp. particleboards have the nearest CT numbers to water substitute at all CT energies.***

Keywords- *Rhizophora spp*. particleboards, tannin adhesive, SEM-EDXA, Computed Tomography (CT), mass attenuation coefficient.

I. INTRODUCTION

Rhizophora spp. as Phantom Materials

The suitability of *Rhizophora spp*. wood trunk as phantom materials in application of low and high energy photons had been studied in several studies (Bradley et al., 1991). This is due to its mass density and mass attenuation coefficient value close to that in water (Che Wan Sudin et al., 1991; Bauk and Tajuddin, 2008). However several limitations arise to prepare *Rhizophora spp*. trunks as phantom materials. This includes the limited size and density inhomogeneity of the *Rhizophora spp*. tree trunk and tendency of the wood to bend and crack over period of times (Marashdeh et al., 2011). Several studies had been conducted to determine the suitability of *Rhizophora spp*. particleboard as phantom material (Surani, 2008; Ngu, 2009). The particleboards has advantages over solid wood including easy to cut and shape to any size, homogenous density throughout the particleboard and no deformation over period of times without compromising its mass attenuation coefficient value (Marashdeh at al., 2012; Shakhreet et al., 2013). In order to accommodate the heavy workload as phantom materials, several studies had been carried out to improve the physical strength of *Rhizophora spp*. particleboards (Abuarra et al., 2014, Tousi et al., 2014).

Tannin Adhesives

Tannin had been commonly used in natural wood, plywood and particleboard adhesive for quite some time (Coppens et al., 1980; Pizzi and Merlin, 1981; Pizzi and Sharfetter, 1989). Tannin is suitable as adhesive due to its high reactivity towards formaldehyde indicated by its Stiasny number of 72 (Nurulhuda et al., 1990). Tannin can be obtained mainly from mangrove wood barks through an extraction process (Mohd Yusoff et al, 1988). The extraction of tannin can be made using Soxhlet extraction apparatus with hot water as solvent. A previous study showed that an extraction of tannin using 90°C of water provided as high as 21.87% yield of tannin from mangrove bark compared to other solvents such as acetone and ethanol (Tan and Kassim, 2011).

II. METHODOLOGY

Preparation of *Rhizophora spp*. Wood Particles

The middle section of *Rhizophora spp*. trunks were chosen based on the study by Shakhreet et al. (2009). The bark was removed from the trunks and then the trunks were planed and ground to obtain the wood particles. The wood particles were screened using a horizontal sieve machine to obtain wood particles with the size of ≤ 104 μm based on available mesh size of the horizontal sieve machine. The wood particles were dried and the moisture content was kept lower than 7%. This

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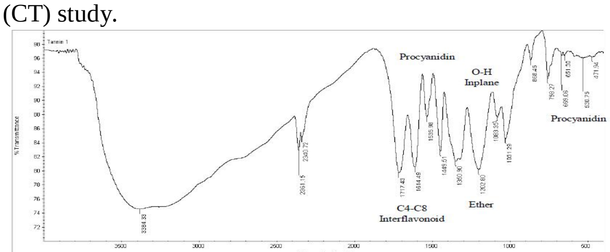
is to prevent swelling and blistering of particleboard during the hot pressing process.

Tannin Extraction

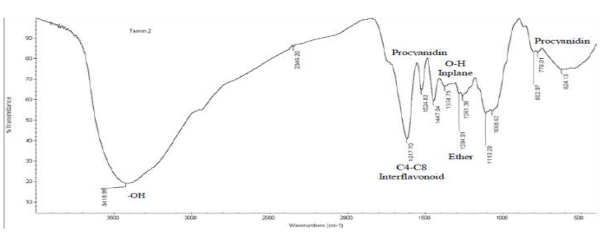
The removed bark from the tree trunks were cleaned and dried to remove soil and dirt. The bark was then ground to reduce them into smaller particle size. Tannin was extracted from the bark using Soxhlet extraction apparatus with hot water as solvent. The tannin solution obtained from the process was dried and evaporated to obtain the tannin powder.

Fabrication of Particleboards

Three types of *Rhizophora spp.* particleboards namely the binderless, tannin-*Rhizophora spp.* and tannic acid-*Rhizophora spp.* particleboards were fabricated in this study. The binderless particleboards were fabricated without any addition of adhesives. The tannin-added particleboards were fabricated by adding 10% of tannin powder obtained from the extraction into the wood particles. The tannic acid-*Rhizophora spp*. particleboards were fabricated similarly to that of the tannin-based particleboard but using commercially available tannic acid powder. In this study, a commercialized tannic acid ($C_{76}H_{52}O_{46}$) with formula weight of 1701.2 g/mol was used as adhesive in comparison to extracted tannin from *Rhizophora spp.* bark. The functional group of extracted tannin and tannic acid using Fourier Transform Infrared analysis (FTIR) is shown in Figure 1. The elemental composition of extracted tannin and tannic acid based on scanning electon microscope (SEM) and energy dispersive x-ray analysis (EDXA) is shown in Table 1. All particleboards were fabricated at a target density of 1.0 g/cm^3 to simulate water as standard phantom material. Four replications of each type of the particleboards were fabricated with three replications were made for the strength analysis and one for the mass attenuation coefficient and computed tomography (CT) study.



a



b

Fig.1 The functional group of tannin (a) and tannic acid (b) based on FTIR analysis

TABLE 1 ELEMENTAL COMPOSITION OF TANNIN AND TANNIC ACID

Sample	Elemental composition (%)				
	C	O	Cl	Na	Mg
Tannin	22.61	62.72	7.79	6.60	0.27
Tannic acid	27.29	72.71			

Strength Test Analysis

Several tests were carried out according to the Japanese Industrial Standard (JIS A-5098:2003). Test pieces of approximate size of 5.0 cm x 5.0 cm from three replications of particleboards were prepared for the internal bond (IB), thickness swelling (TS) and water absorption (WA) tests while test piece of approximate size of 20.0 cm x 5.0 cm were prepared for the modulus of rupture (MOR) test.

Microstructure Study and Determination of the Effective Atomic Number, Z_{eff}

The microstructures of particleboards were investigated to observe the morphology of parenchyma cells that were related to the binding quality of each type of the particleboards. The microstructures were observed using a scanning electron microscope (SEM) on samples with approximate size of 0.5 cm x 0.5 cm. The samples were coated with gold using an ion sputterer to prevent ionization of sample during the scanning process. The elemental compositions of the samples were determined using the energy dispersive x-ray (EDXA) analysis of the SEM. The effective atomic number of the samples can be calculated using the equation

$$Z_{eff} = \left[\sum_{i=1}^{n} (\alpha_i z_i^{m})\right]^{\left(\frac{1}{m}\right)} \qquad (1)$$

where a_i and z_i are the electron fraction and the atomic number of i^{th} element in the sample while m is the experimental coefficient for biological materials and water with a value of 3.4 (Duvauchelle et al., 1999). The electron fraction of the i^{th} element can be calculated by the equation

$$\alpha_i = \frac{wi\left(\frac{z_i}{A_i}\right)}{\Sigma w_i\left(\frac{z_i}{A_i}\right)} \qquad (2)$$

where w_i and A_i are the fractional weight and the atomic mass of the i^{th} element respectively. **a**

Mass Attenuation Coefficient Study Using XRF Photons

The x-ray fluorescent (XRF) set up used to determine the mass attenuation coefficient of particleboards at low energy photons is as shown in Figure 2. An annular ^{241}Am source with a nominal activity of 100 mCi was used in conjunction with niobium, molybdenum, palladium, silver and tin pure metal plates were used to obtain $K_{\alpha1}$ x-rays of 16.59, 17.46, 21.21, 22.16, and 25.26 keV respectively. The transmitted photons through the samples were collected using a low-energy Germanium detector (LEGe) connected to a Multi-Channel Analyzer (MCA). The incident photons will be attenuated by the particleboard sample with thickness *x*. Its intensity will be attenuated and reduced according to Beer-Lambert's law calculated by

$$I = I_{\circ} e^{-\mu x} \qquad (3)$$

where $I_{\circ}$ is the initial intensity of photons, and μ is the linear attenuation coefficient of sample in cm^{-1}. The linear attenuation coefficient of sample can be calculated by modifying Equation (1) into

$$\mu = \frac{1}{x} \ln\left[\frac{I_{\circ}}{I}\right] \qquad (4)$$

The mass attenuation coefficient of particleboard samples can be calculated by dividing the value of linear attenuation coefficient with the mass density of the particleboard samples. The values of mass attenuation coefficient of samples were compared to the value of water (XCOM) (Hubbell, 1982; Berger & Hubbell, 1987). A chi-square goodness of fit calculation was used for comparison to water (XCOM) given by the equation

$$\chi^2 = \frac{(yi - xi)^2}{\sigma^2} \qquad (5)$$

where xi and yi are the values of sample and water respectively, and σ is the error of measurement.

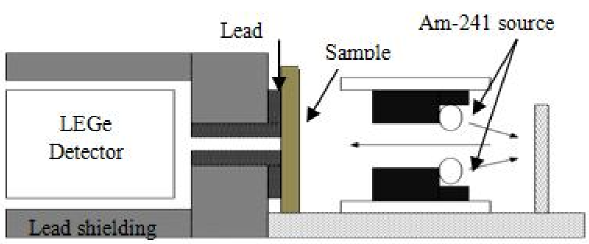


Fig. 2 The arrangement of XRF set-up

Computed Tomography Study

Density plug phantoms were constructed from the replication of each type of *Rhizophora spp.* particleboards. The particleboards were sawn circularly to obtain disk-shaped samples. The disks were then stacked and glued to obtain cylindrical-shaped density plug phantoms with dimensions of approximately 3.0 cm diameter and 5.0 cm length compatible with CT electron density phantom model CIRS 062M. The plugs were inserted in the electron density phantom along with a water-equivalent density plug phantom. The samples were scanned at 80, 120 and 140 kVp CT x-rays commonly used in clinical CT scan (Constantinou et al., 1992). The CT imaging provides the image of pixel value called CT number defined by the equation

$$CT\ number = 1000\ \frac{\mu - \mu w}{\mu w} \qquad (6)$$

where μ and μ_w is the linear attenuation coefficient of sample and the linear attenuation coefficient of water respectively. The CT Number is represented by the Hounsfield Unit (HU).

Fifteen slices of CT images with 1.0 mm thickness per slice were obtained from each samples. The average CT numbers and standard deviation (SD) of CT number were obtained and compared to that in water equivalent density plug.

III. RESULTS AND DISCUSSION

The average density of binderless, tannin-*Rhizophora spp.* and tannic acid-*Rhizophora spp.* particleboards and the densities of the respective plug phantom samples are shown in Table 2. From the table, it was found that all types of particleboards had achieved mass densities near to the value of water. The tannic acid-*Rhizphora spp.* particleboard was found to have the closest mass density to water and a better density uniformity shown by the SD values.

TABLE 2 AVERAGE DENSITY OF *RHIZOPHORA SPP.* PARTICLEBOARDS AND DENSITY PLUG PHANTOMS

Sample	Measured Density (g/cm^3)			SD	Density (g/cm^3)
	Average	Max.	Min.		
A	1.037	1.095	0.955	0.061	1.002
B	1.016	1.079	0.978	0.044	1.021
C.	1.004	1.040	0.980	0.023	1.022

A Binderless *Rhizophora spp.* samples
B Tannin-*Rhizophora spp.* samples
C Tannic acid-*Rhizophora spp.* samples

The average internal bond strength (IB) of binderless, tannin-*Rhizophora spp.* and tannic acid-*Rhizophora spp.* samples are shown in Figure 3. All particleboards met the reuirement of JIS A-9508 for particleboard type 18 (0.3 N/mm^2). From the figure, it was also understood that addition of tannin had improved the IB strength of *Rhizophora spp.* particleboards. Tannic acid had provided better improvement of IB strength between the two types of tannin added particleboards.

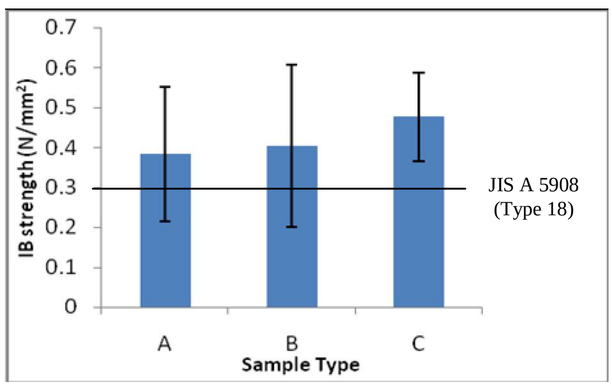


Fig. 3 The average IB strength of binderless, tannin-*Rhizophora spp.* and tannic acid-*Rhizophora spp.* particleboards according to JIS A-5908:2003.

The average modulus of rupture (MOR) of binderless, tannin-*Rhizophora spp.* and tannic acid-*Rhizophora spp.* samples are shown in Figure 4. From the figure, it is illustrated that samples met the requirement for JIS A-5908 for particleboard type 8 (8 N/mm^2). The tannic acid-*Rhizophora spp.* samples were found to have the highest

MOR value at 12.149 N/mm^2 nearing the requirement for JIS A-5908 type 13 (13 N.mm^2). The average thickness swelling (TS) and water absorption (WA) of binderless, tannin-*Rhizophora spp*.and tannic acid-*Rhizophora spp*.samples are shown in Figure 5. From the figure, it was illustrated that addition of tannin had improved the TS and WA of *Rhizophora spp*. particleboards. Both tannin-*Rhizophora spp*. and tannic acid-*Rhizophora spp*. complied with the JIS A-5908 for thickness swelling (12.0%) with tannic acid-*Rhizophora spp*. samples gives the lowest average TS compared to that by tannin- *Rhizophora spp*. samples.

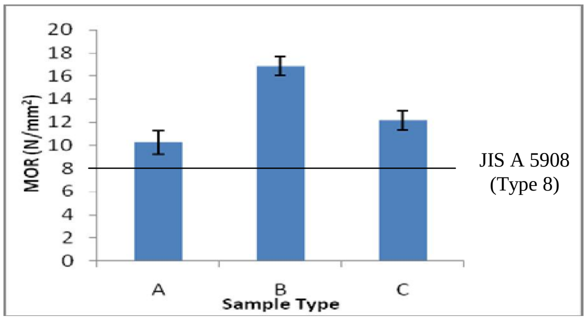


Fig.4 The average MOR of binderless, tannin-*Rhizophora spp*. and tannic acid-*Rhizophora spp*. particleboards according to JIS A-5908.

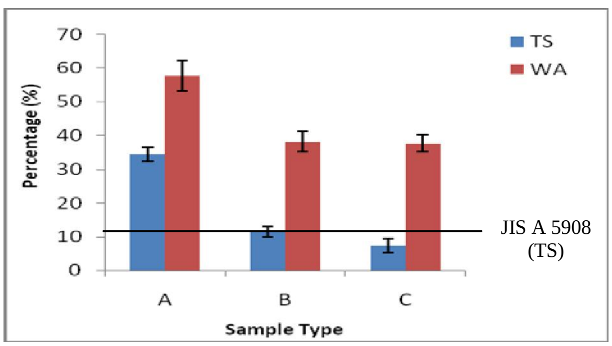


Fig. 5 The average TS and WA of binderless, tannin-*Rhizophora spp*. and tannic acid-*Rhizophora spp*. particleboards according to JIS A-5908.

The micrographs of binderless, tannin-based and tannic acid-based *Rhizophora spp*. samples are shown in Figure 6. Micrograph images were taken at 1000x magnification to observe the morphology and binding quality of the particleboards. The binderless *Rhizophora spp*. sample shows the presence of void spaces between the wood particles and the lumen of parenchyma cells that reduced the compactness of the particleboard. The micrograph of tannin-*Rhizophora spp*. sample shows reduced void spaces between the wood particles by tannin particles. This has improved the compactness as well as the physical strength of the *Rhizophora spp*. particleboard. The micrograph of tannic acid-*Rhizophora spp*. samples, on the other hand, shows successful filling of void spaces and lumen of wood particles by tannic acid particles. This had further improved the compactness and the physical strength of the particleboard.

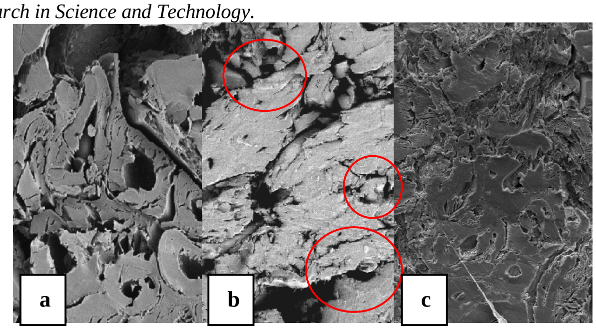


Fig. 6 Cross-sectional micrographs of (a) binderless, (b) tannin-*Rhizophora spp*. and (c) tannic acid-*Rhizophora spp*. samples at 1000x magnification using SEM.

The elemental composition measured using EDXA and the calculated effective atomic number, Z_{eff} of samples are presented in Table 3. The EDXA results had indicated the similarity of all samples to resemble the human soft tissue that basically consists of carbon and oxygen. The value of Z_{eff} can be used to determine the energy absorption and attenuation properties of a medium (Tursucu et al., 2013). Therefore materials with similar or close Z_{eff} value can be postulated to have similar energy absorption and attenuation.

TABLE 3 ELEMENTAL COMPOSITIONS BASED ON EDXA

Sample	Elemental composition (%)					Z_{eff}
	H	C	O	N	F	
A	-	32.93	38.98	28.08	-	7.185
B	-	51.25	43.11	-	5.64	7.221
C	-	24.80	66.07	-	9.12	7.723
Water[a]	11.11	-	88.89	-	-	7.500

[a]AAPM (1983)

The mass attenuation coefficients of all samples at photon energies between 16.59 and 25.26 keV using XRF are shown in Figure 6. From Figure 6, it is illustrated that tannic acid-*Rhizophora spp*. samples having the closest values of mass attenuation coefficient to the value of water calculated using XCOM.

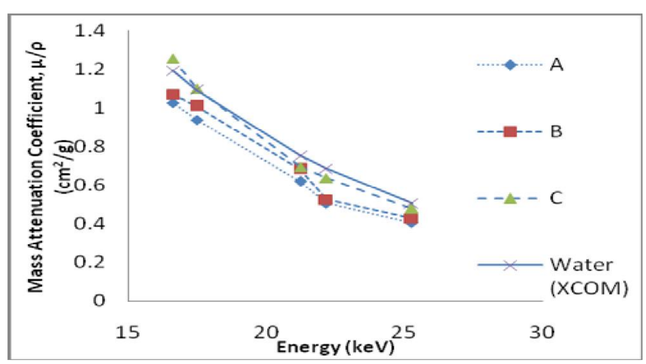


Fig. 6 Comparison of the mass attenuation coefficients against photon energy for *Rhizophora spp*. samples to water (XCOM).

The values of χ^2 for all samples to the values of water (XCOM) are presented in Table 4. From Table 4, it was found that tannic acid-*Rhizophora spp.* particleboards having the nearest value of mass attenuation coefficient to the calculated value of water (XCOM). This is due to the improved compactness of the sample as shown in Figure 5(c) that gave better attenuation to the XRF photon at all nominal energies.

TABLE 4 THE χ^2 VALUE OF MASS ATTANUATION COEFFICIENT TO WATER

Energy (keV)	χ^2 value with water		
	A	B	C
16.59	1.813	0.039	0.294
17.46	3.949	1.414	2.770
21.21	6.114	11.010	0.385
22.16	1.852	1.320	0.404
25.26	12.129	0.624	0.306
Total χ^2	**25.859**	**13.008**	**4.159**

The average CT numbers for the samples at 80, 120 and 140 kVp CT x-ray energies are shown in Table 5. From the table, it is shown that tannic acid-*Rhizophora spp.* sample gave the nearest CT number to water equivalent density plug phantoms. This is also supported by the value of χ^2 for tannic acid-*Rhizophora spp.* sample as shown in Table 6.

Table 5. The average and the standard deviation of CT number for binderless, tannin-*Rhizophora spp.* and tannic acid-*Rhizophora spp.* density plug phantoms.

Samples	Average CT Number ± SD		
	80 kVp	120 kVp	140 kVp
A	-32.814 ± 46.65	-28.539 ± 39.08	-18.713 ± 19.23
B	-32.570 ± 47.96	-27.057 ± 39.78	-25.969 ± 19.01
C	-29.385 ± 48.43	-18.429 ± 40.37	-13.688 ± 20.01
Water	-13.577 ± 47.14	-11.601 ± 37.19	-9.716 ± 16.28

Table 6. The total χ^2 values for CT number of *Rhizophora spp.* samples to water.

CT Energy (kVp)	χ^2 to water substitute		
	A	B	C
80	0.012	0.015	0.011
120	0.017	0.014	0.014
140	0.023	0.028	0.004
χ^2	**0.052**	**0.057**	**0.029**

IV. CONCLUSION

The results indicated the suitability of binderless, tannin-*Rhizophora spp.* and tannic acid-*Rhizophora spp.* particleboards as phantom material in applications of low energy x-ray and CT imaging. The addition of tannin and tannic acid had increased the strength of *Rhizophora spp.* particleboards as shown by the internal bond strength, modulus of rupture, thickness swelling and water absorption tests based on JIS A-5908. The microscopic study conducted using SEM-EDXA showed the function of tannic acid in reducing the void spaces and cell lumens that contributed to the increased strength of *Rhizophora spp.* particleboards. The XRF study showed that the tannin-*Rhizophora spp.* and tannic acid-*Rhizophora spp.* particleboards having the value of mass attenuation close to water (XCOM) at photon energies between 16.59 and 25.26 keV. The CT study showed the close attenuation properties and CT numbers of tannic acid-*Rhizophora spp.* particleboards to the value of water at all CT x-ray energies indicating its near water-equivalence property as phantom materials.

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