Analysis of Microfibril Angle in Acacia Mangium Wood Using Small-Angle X-Ray Scattering

Tamer A. Tabet*[1], *Member, IAENG*, Fauziah Abdul Aziz[2] and Shahidan Radiman[3]

***Abstract*—Microfibril angle, (MFA) in hardwood is one of the key determinants of solid timber performance due to its strong influence on the stiffness, strength, shrinkage, swelling, thermal-dynamics mechanical properties and dimensional stability of wood.. A method is presented for the measurement, using small-angle X-ray scattering (SAXS), of the microfibril angle, (MFA) and the associated standard deviation for the cellulose microfibrils in the S_2 layer of the cell walls of *Acacia mangium* wood. The length and orientation of the microfibrils of the cell walls in the irradiated volume of the thin samples are measured using SAXS and scanning electron microscope, (SEM). The undetermined parameters in the analysis are the MFA, (M) and the standard deviation (σ_Φ) of the intensity distribution arising from the wandering of the fibril orientation about the mean value. Nine separate pairs of values are determined for nine different values of the angle of the incidence of the X-ray beam relative to the normal to the radial direction in the sample. The results show good agreement. The curve distribution of scattered intensity for the real cell wall structure is compared with that calculated with that assembly of rectangular cells with the same ratio of transverse to radial cell wall length. It is demonstrated that for $\beta = 45°$, the peaks in the curve intensity distribution for the real and the rectangular cells coincide. If this peak position is Φ_{45}, Then the MFA can be determined from the relation $M = \tan^{-1}(\tan \Phi_{45} / \cos 45°)$, which is precise for rectangular cells. . Here we shall present our results of the MFA in the cell wall with respect to its shape and structure as an important fast check and yet accurate towards the quality of wood, its uses and application.**

***Index Terms*—: Small-Angle X-Ray Scattering, Microfibril Angle, MFA, rectangular cell wall and real cell wall, *Acacia mangium*.**

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I. INTRODUCTION

The angle between the cellulose fibrils and the longitudinal cell axis, the microfibril angle (MFA), has been shown to be a crucial factor that influence the modulus of elasticity and the dimensional stability of wood [7]. Reference [5] stated that the microfibril angle of the S_2 layer in the fibre cell wall is known to be one of the main determinants of the thermal and dynamic-mechanical properties of the hardwood. Reference [6] reported that the microfibril angle changed as a function of the position in the tree. The mean microfibril angle of *Pinus massoniana* decreased more gradually as the distance increased from the pith and reached the same level in mature wood [6]. The varied wood resource of the future will require more definitive information about the microfibril angle to improve selection and utilization. X-ray diffraction has the potential to be a much more rapid method to determine microfibril angle than is microscopic measurement [2].

Acacia mangium wood wood is classified as a hardwood. It is the major fast growing plantation species for timber and pulp in Asia. Botanically *Acacia mangium* comes from family Legiomenosae and sub-family mimisoideae. It has a wood density ranging from 420 to 600 Kg/m^3 and specific gravity of 0.65

X-ray diffraction is a well-established method for the determination of the mean microfibril angle [3]. Additionally, Reference [2] also utilized the SAXS to investigate the spiral in the wood cell wall, but there primary assumption in the analysis is that the wood cell walls are square. It is necessary to take in to account the detailed cell structure in order to interpret diffraction and scattering data [1]. This involved using quantitative image analysis or SEM to measure the length and the orientation of the microfibrils in the irradiated volume of the thin samples. From the data, the angle of the peak scattering intensity Φ_p was calculated for a sample irradiated in a direction of 45° to the radial and transverse direction. It was thereby demonstrated that the MFA could be calculated from the formula (1).

$$M = \tan^{-1}\left(\tan \Phi_p / \cos 45^\circ\right) \quad (1)$$

where: M the MFA in S_2 layer, with an error about 1°. The relation is exact for rectangular cells [2].

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II. MATERIALS AND METHODS

The wood samples used in this study were selected from 3, 5, 7, 9, 10, 11, 13 and 15-year-old of *Acacia mangium* plantation from Sabah Forestry Development Authority (SAFODA). Two trees from each compartment were selected providing a total 16 trees for this study. Two discs of 40 mm were taken at breast height of the stem for each tree. In total, eight knot-free discs were labelled and stored in plastic bags for further sample preparation.

A. Sample Preparation

Please A 4cm × 6cm rectangular block was obtained from each disc at different angles of grain. The samples subsequently stored under controlled temperature and relative humidity (23 ˚C ± ˚C 1and 55% ±3%) to achieve equilibrium moisture content about 9%. Thin strips of uniform thickness about 50 microns were cut along the radius rotary microtome. The sample dimensions were 20mm length × 10mm width.

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B. Measuring MFA Using Small-Angle X-Ray Scattering Technique

Samples of 9 different orientations were cut. The angle β shown is between the wider face of the sample and the radial direction. The samples were irradiated with the X-ray beam has a circular cross-section of 0.6mm in diameter directed normal to the face, β is the angle between the normal to the radial plane and the direction of the X-ray beam. The various cell-wall orientations were obtained by cutting specimens with different angles, so that the scattered radiation would pass through the same length of specimen material in all cases. Without any further treatment, it is encapsulated in plastic foil to keep them from drying and shrinking in the vacuum chamber of the x-ray equipment. A SAXS device (HMBG-SWAX, SAXS PW 3830 X-ray generator) was used to determine the MFA in each of the eight investigated trees. The measurements were carried out in point focus geometry using Cu Kα radiation of wave length 1.54 nm. The beam width at the sample position was 200 µm. A position detector was used to record the scattering patterns. The distance of the sample to the detector was 5.14 mm. The experimental set-up consisted of 40 Kv and 20 mA. The incoming x-ray beam had a circular cross-section of 0.6 mm in diameter.

C. Theory

The equations used to calculate the scattered intensity and MFA were derived by Reference [2] and using a peak fit method developed by Reference [7]. Recently, the MFA was calculated based on the value of parameter *T* using X-ray diffraction technique [5]. In Figure 1, *z* is the direction of the cell axis, y is the radial direction and χ is the transverse direction. A cell wall will shown with two sets of S_2 microfibrils f_1 and f_2 lying at the microfibril angle *M* to the cell axis direction *z*. The incident X-ray beam is directed along χ axis. The normal to the cell wall lies at an angle α to the direction of the X-ray beam. The azimuth angle Φ_1 for the scattered intensity from the fibril f_1 is given by:

$$\tan \Phi_1 = -\cos \alpha \tan M \quad (2)$$

The corresponding azimuth angle Φ_2 for scattering from f_2 is given by:

$$\tan \Phi_2 = -\cos(\alpha + \pi) \tan M = \cos \alpha \tan M \quad (3)$$

and so $\Phi_1 = -\Phi_2$ and the scattered intensity is symmetrical about $\Phi = 0°$.

A sample is cut so that the radial direction is at an angle β to the front face of the wood section. The X-rays are directed normal to the front face. For a cell wall lying at an angle θ to the radial direction, the value of α is:

$$\alpha = (\beta + \theta) \quad (4)$$

and thus the azimuthal angle for scattering from f_1 fibrils in the cell wall is given from equation (5) by:

$$\tan \Phi_1 = -\cos(\beta + \theta) \tan M \quad (5)$$

and for scattering from the f_2 microfibrils, Φ_2 is given from equation (6) by

$$\tan \Phi_2 = \cos(\beta + \theta) \tan M \quad (6)$$

Measurement on 9 samples will be used for the angle:

β = 0, 10, 20, 30, 35, 40, 45, 50 and 90°.

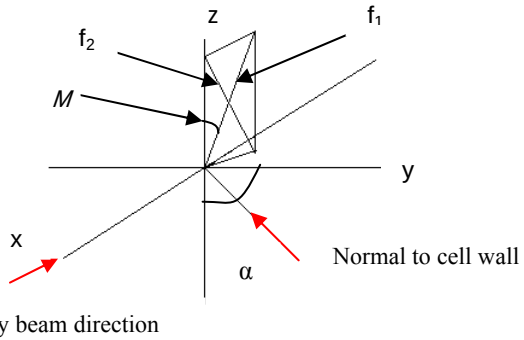

Fig. 1: Diagram showing the relationship between a cell wall, the two S_2 microfibrils and the direction of the X-ray beam [2].

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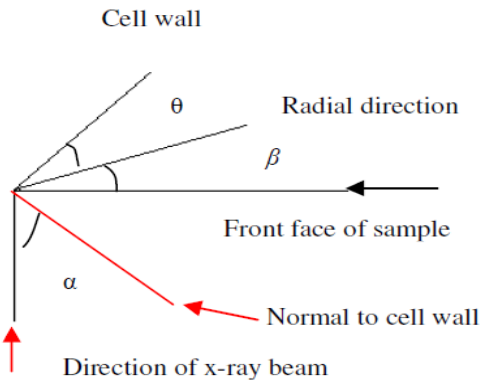

Fig. 2: Diagram showing the relation between front face of the thin sample, the radial direction (β), the cell-wall direction (θ) and the direction of the X-ray beam normal to the face of sample [2].

III. RESULTS

Figure 5 presents Figure 5 presents the determination of MFA by plotting intensity I against the azimuthal variation of scattered intensity. The azimuthal angle Φ ranging from -90° to 90°; outside this range the scattered intensity is very small. Table 1 reports the estimated values for MFA and the orientation angle of the grain, β. As can be seen there is reasonable consistency between the values derived from the nine samples. It's clear that the lower values of β give lower values of MFA and that the higher flanks of the intensity distributions are critical values of the β. That mean, for values of β less than 45°, the values of MFA are increased as the grain angle increased. For the β greater than 45°, the data of Table 1 indicate that the value of MFA is slightly less than that estimated as less than 45°. In Figure 4, a typical intensity distribution for β = 45° versus the azimuthal angle Φ. The higher peak arises from the S_2 layer and the lower peak is generated by S_1 and S_3 microfibrils. Table 1 indicate that possibly the value of estimated MFA using SAXS technique is slightly less than that for the grain orientation, β. In Figure 4, σ_Φ is the half-width at inflection point. The width T has been shown to be correlated to the MFA [7]. The "T" parameter was developed for wide-angle diffraction data but here no reason in principle why it should not be used for SAXS intensity distribution. Here the MFA values were estimated from the reference [7] equation as following:

$$MFA = 0.6\,T \quad (7)$$

$$T = MFA + 2\,\sigma_\Phi \quad (8)$$

The Comparison between Measured Value of MFA for the Real Cell Wall and the Rectangular Cell Wall.

It was found that the MFA of intensity distribution for the real cell of *Acacia mangium* used and the rectangular structure cell wall in a direction at β = 45° were close together.

The intensity for β = 45° is plotted in Figure 5. The peak intensity at Φ_{45} = 24°. The relation between this azimuthal angle and MFA for the perfect rectangular cells is given by [4].

$$MFA = \tan^{-1}(\tan\phi_{45} / \cos 45) \quad (9)$$

$$MFA = \tan^{-1}(0.445 / 0.707)$$

$$MFA = 32.16^{o}$$

The measured value of the cell wall of *Acacia mangium* MFA= 29.4°, so the use of the relation for rectangular cells to interpret the measured data gives a good estimate of MFA. Figure 3 shows a SEM micrograph for the real cell wall used for the MFA estimation while Figure 4, shows the rectangular cell wall *Norway spruce.* [9].

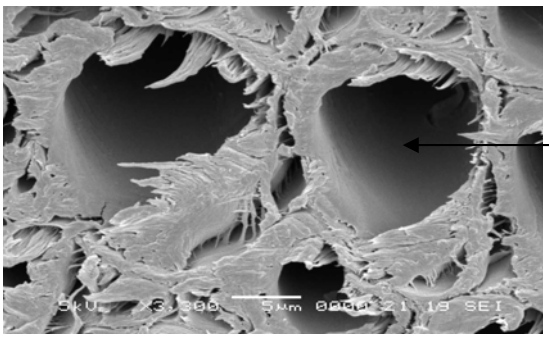

Fig. 3: SEM micrograph at magnification X 3,300, showing the shape of the real cell wall of *Acacia mangium* used for MFA estimation, sample taken from the pith region of tree 10 year-old.

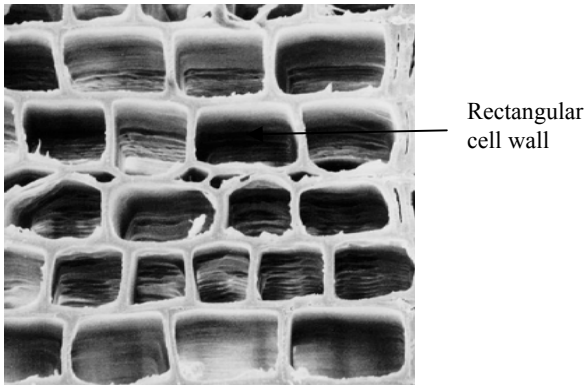

Fig. 4: SEM micrograph at magnification showing the shape of the rectangular cell wall of *Norway spruce.* [9].

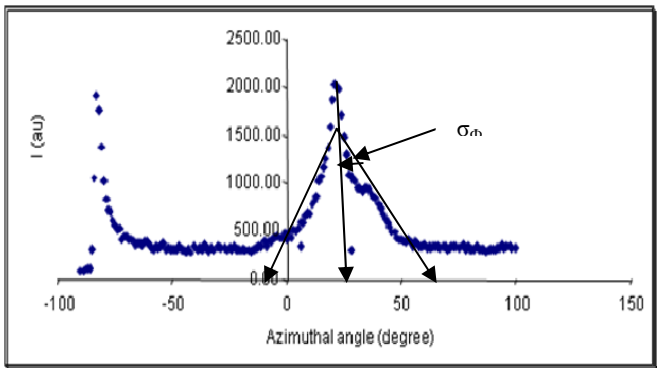

Fig. 5: The intensity distribution for β = 45° is plotted against azimuthal angle. MFA=29.4° and σ_Φ = 9.8°.

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The strong relationship between MFA and the distance from pith to bark which has previously been shown using XRD [8] was confirmed in this study using SAXS when the regression analysis showed that a straight line fit the data very well. It was found that 95.97% of the variation of MFA can be attributed to the distance from pith to bark of wood model 10-year-old. Based on the results obtained from the *Acacia mangium* wood model of 10-year-old, this study can deduce the results for other samples of different age. This observation support those of Bonham and Barnett (2004) who found that MFA in *Betula pendula Roth* varied from 10˚ to 18˚ with the distance from pith towards bark.

Table 1: Estimated MFA and the standard deviation σ_Φ for each value of β.

Sample No.	Distance from pith (mm)	β (°) ± 0.5°	MFA (°) Mean MFA=24.2°	σ_Φ
1	10	0	18.0	6.0
2	20	10.0	19.8	6.6
3	30	20.0	19.2	6.4
4	40	30.0	22.2	7.4
5	50	35.0	24.0	8.0
6	60	40.0	25.8	8.6
7	70	45.0	29.4	9.8
8	80	50.0	28.8	9.6
9	90	90.0	30.6	10.2

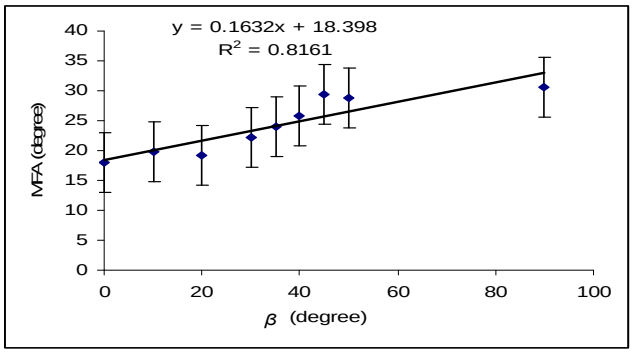

Fig. 6: Variation of the measured MFA and the angle β in *Acacia mangium* wood of 10-year-old.

IV. DISCUSSION

The results show that MFA varied from 18.0° at distance 10.0 mm from the pith center to 30.6° at 90.0 mm from the pith. The highest MFA has been found near the outer bark at the breast height of the tree. This result supports the statement made by [3] who found that MFA in hardwoods were lower than in softwoods. This was shown in *Norway spruce* where MFA after the 7th year ring varies between 6° to 10° [1]. *Acacia mangium* wood as a hardwood contains vessel elements in which the MFA can be higher. On this basis, The MFA values measured in this wood model is reasonable. The presence of vessels, especially in high abundance might be expected to increase x-ray diffractometric estimates MFA for hardwoods in comparison to softwoods (Bonham and Barnett, 2004). Removal of the contribution of vessels from the average values found in hardwoods might therefore result in an even lower value of MFA for the fibres [2].

V. CONCLUSION

MFA can be used a good indicator to estimate of hardwoods and softwood based on its value [8]. Wood containing fibres with an MFA of 30° or below have been termed hardwoods wood, while wood containing fibres with MFA of 30° or more have been termed softwoods (Bonham and Barnett, 2004). This can be proved again in *Acacia mangium* wood from Sabah where the mean value of MFA from pith to bark was found to be 24.2° in the wood disc of 10-year-old.

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