

「超音波顕微鏡を用いたパルプ繊維壁の弾性係数測定」

走査型超音波顕微鏡(SAM)を用いてパルプ繊維壁の弾性係数を測定した。超音波材料シグニチャ(AMS)と呼ばれる、材料に特有の超音波の干渉図からこの計算が可能となる。熱処理した繊維及び未処理の繊維の横断面を調製し、この断面の表面を走るレイリー波の速度を測定すると、それぞれ $3,520 \pm 170$ m/s 及び $3,240 \pm 180$ m/s であった。これは、レイリー波の速度が速いほど弾性係数 (C_{44}) が大きくなることから考えて、熱処理した繊維の繊維壁は未処理の繊維に比べて堅くなったことを意味する。さらに、繊維は放射方向に等方体と考えられる S_2 層が主成分であると考え、レイリー波の速度は、剪断波の速度の 0.93 倍に相当し、セルロースの真密度が 1.5 g/cm^3 であるという条件を適用することができる。すると、熱処理した繊維及び未処理の繊維の繊維壁弾性係数 (C_{44}) は、それぞれ 22 ± 2 GPa 及び 18 ± 2 GPa であることがわかった。これは熱処理によって弾性率が増加することを意味するが、未叩解の熱処理した繊維から調製したシート全体の面内剪断弾性率は未処理の場合と比べて低い値を示した。ここでの結果は、繊維壁の弾性係数を増加させる熱処理の効果を表しており、一般に角質化として知られているものである。

Measurement of elastic constant of pulp fiber wall by scanning acoustic microscope

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Abstract

An acoustic micro-metrology technique of a scanning acoustic microscope, as acoustic material signature, was applied to measure the elastic constant of bleached softwood kraft pulp fiber wall. The Rayleigh wave speed measurement of cross sectional surface of heat-treated and untreated single bleached softwood kraft pulp fibers, which were $3,520 \pm 170$ m/s and $3,240 \pm 180$ m/s, respectively, was achieved by this technique. This evidently means that the wall of heat-treated fibers was stiffer than that of the untreated ones because the higher the Rayleigh wave speed, the higher the elastic constant (C_{44}). Furthermore, if the ideal component of fiber wall is supposedly composed from a majority of a transversely isotropic S_2 layer, the calculation of C_{44} of the fiber wall could possibly be demonstrated by applying a Rayleigh wave speed to shear wave speed ratio of 0.93 and a density of 1.5 g/cm^3 for a close-packed cellulosic material of the S_2 layer according to the theoretical equation. The calculated C_{44} values for the heat-treated and untreated fiber wall were 22 ± 2 GPa and 18 ± 2 GPa, respectively. However, handsheets made from unbeaten heat-treated fibers had a lower in-plane shear modulus than those from unbeaten unheated fibers. These results indicate the

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effect of heat treatment on elasticity of a fiber wall which is generally known as hornification.

Keywords: *Acoustic material signature, Elastic constant, Hornification, Scanning acoustic microscope*

Introduction

A scanning acoustic microscope (SAM) is one of the non-destructive testing instruments. Its advantage is ability to use for measuring the elastic constant of the focused microscopical area in a material, by the acoustic micro-metrology technique as acoustic material signature (AMS). The ray model¹⁻³⁾ (Fig. 1) is generally used for explaining how the SAM can be used for measuring the elastic constant of the material. When an acoustic lens scans with a defocused position normal to a surface of the material, there are two kinds of power of reflected waves from the surface of the material. The first one is power of the directly reflected wave (as is shown as A in Fig. 1) and the other is the Rayleigh wave which is bounded on the surface of the material at a critical angle and releases its power to the acoustic lens during its movement over the surface of the material (as is shown as B in Fig. 1). The reflected power from both of the waves interferes with each other and produces an oscillation pattern of relative reflected wave power ($V(z)$) called AMS, as is shown in Fig. 2, as the acoustic lens moves down during scanning for detecting the reflected wave power. This oscillation pattern of $V(z)$ can be used for computing the Rayleigh wave speed, by equation (1), which is closely related to elastic properties of the material. Thus, in the case of an isotropic elastic material, its elastic constant can be calculated if the Rayleigh wave speed is substituted in equation (2)¹⁻³⁾

$$V_R = V_0 \left\{ 1 - \left(1 - \frac{V_0}{2f\Delta zN} \right)^2 \right\}^{-1/2} \quad \dots\dots(1)$$

$$\left(\frac{C_{44}}{\rho} \right)^{1/2} = V_{shear} \approx \left(\frac{1+\nu}{0.87+1.12\nu} \right) V_R \quad \dots\dots(2)$$

where ΔzN is the periodic oscillation pattern of $V(z)$, f is the frequency of the acoustic wave, V_0 is the speed of the acoustic wave in the coupling fluid (V_0 of water = 1,500 m/s), V_R is the Rayleigh wave speed, V_{shear} is the shear wave speed, C_{44} is the elastic constant of the material, ρ is the density of the material, and ν is the Poisson's ratio of the material.

Because the SAM can yield values of local elastic properties of a material to a diameter as small as 10 μ m or to an area as small as 15-20 μ m² (2, 4-6), therefore, the advantage of this technique is expectedly to achieve evaluation of actual elastic constants of pulp fiber walls in this study. Though there are many methods⁷⁻¹⁰⁾ for evaluating the elastic constants of a single fiber wall, but their interpretation procedures are quite limited to practical cases due to complicated modeling assumptions and data analysis methods.

Materials and Methods

Both heat-treated (soaked in water overnight and then heated at 105 °C for 24 hours) and untreated bleached softwood kraft pulp (BSKP) fibers were used as the samples in this study. A straight single fiber was aligned on a glass slide and one end was cut off for a smooth cross sectional surface to adjust it at the same height with an edge plane of the glass slide, followed by coating with a waterproof substance (SUPER-FIXA, Sakura Color Products Corp., Japan). An image of this cross sectional surface was taken and reflected acoustic wave power ($V(z)$) was detected by scanning a spherical acoustic lens (100 MHz) of SAM (Scanning Acoustic Microscope HSAM 210, Hitachi, Japan) in the direction normal to the XY plane like a schematic diagram in Fig. 3. During this whole process, the sample and the lens were kept in

water. Furthermore, handsheets were also made from both heat-treated and untreated BSKP fibers, without beating, for measuring their elastic constants or shear moduli (G) as such by sonic velocity measurement of a sonic sheet tester (SST-110, Nomura Shoji Co., Ltd., Japan). The principle of this method has been introduced and clearly ascertained by Taylor and Craver¹¹⁾. In this study, the sample of single pulp fibers and handsheets were conditioned following the TAPPI test method for the subsequent measurements.

Results and Discussions

Acoustic images of a cross sectional surface of a single fiber were taken when the focal plane of the spherical acoustic lens was exactly on it (at $z=0$), as is shown in Fig. 4. A two-dimensional image of a $V(z)$ pattern (Fig. 5) was obtained when the spherical acoustic lens was moving down in the Z direction gradually, from $z=0$ to $z=-400\ \mu\text{m}$, during scanning over the cross sectional surface of the single fiber along a line scan in the middle of Y -axis of Fig. 4. The oscillation pattern of $V(z)$ along the dotted line from A' to B' in Fig. 5, which expectedly matched $V(z)$ at the fiber wall position of the cross sectional surface, indicates the AMS of the fiber wall substance as is shown in Fig.6. The wave velocity, which is also presented in the Fig. 6 ($=3,862\ \text{m/s}$), was completely calculated from the oscillation pattern of $V(z)$ by the specific computer software of the SAM. This wave velocity is the speed of the Rayleigh wave (V_R) that was bounded on the cross sectional surface of the single pulp fiber and could not be determined by any other methods. The accuracy and precision of V_R measurement were also determined by the V_R of a surface of waterproof substance coated over the sample. The V_R of the waterproof substance coated on a glass slide surface was higher than that of an uncoated surface by about 7 % (Fig. 7). That is, the V_R of all samples, which have been coated with the waterproof substance in this study, was also assumed to be identically higher than their actual V_R

by about 7 %. The measured V_R for the cross sectional surface of heat-treated and untreated single BSKP fibers were $3,520 \pm 170$ m/s and $3,240 \pm 180$ m/s, respectively, as is also shown in Fig. 7. This result means that the heat-treated fiber had a stiffer wall than the untreated fiber, namely, the higher the V_R , the higher the V_{shear} as well as the higher the C_{44} , according to equation (2). However, in this research the C_{44} values of the fiber wall of those single fibers examined cannot be evaluated directly by substituting their measured V_R values in equation (2) because the fiber wall is not an isotropic material. Methods for determining the elastic constant of an anisotropic material by the scanning acoustic microscope has been presented clearly by Lee et al.¹²⁾. They used the line-focus-beam acoustical microscope to measure the normalized difference of V_R between in the [100] direction and in the [110] direction () of the cubic symmetrical bare solid, and also illustrated its relationship to anisotropic factor () and ratio of V_R to V_{shear} . If the material is isotropic, the value will be equal to zero. That is, of the material and the ratio of V_R to V_{shear} are typically about 1 and 0.93, respectively. Therefore, if the fiber wall has a majority of a transversely isotropic S_2 layer, the value of the fiber wall is ideally about zero, the calculation of C_{44} of the fiber wall could possibly be demonstrated by applying the 0.93 of V_R/V_{shear} ratio and a 1.5 g/cm^3 density of a close-packed cellulosic material of the S_2 layer whose structure supposedly did not change appreciably by the heat treatment to equation (2). The calculated C_{44} values for the heat-treated and untreated fiber walls were 22 ± 2 GPa and 18 ± 2 GPa, respectively, in this case. This evidently meant that the heat treatment increased the elastic constant of the pulp fiber wall as is known as hornification.^{13, 14)} However, this increase in C_{44} was not consistent with the common knowledge that the in-plane shear modulus (G) of handsheets per se made of unbeaten-heated-fibers decreased probably because stiff heat-treated fibers had their inferior conformability in forming a handsheet (Fig. 8). Namely, the elastic modulus of the handsheet was reduced because of the resultant less area of inter-fiber bonding.^{15, 16)}

Conclusions

Heat-treated BSKP fibers were found to have a stiffer wall than untreated BSKP fibers, as is known generally as hornification, by applying scanning acoustic microscopy. The C_{44} of the fiber wall was 22 ± 2 GPa and 18 ± 2 GPa for the heat-treated and untreated fibers, respectively. The heat-treated fibers provided the lower in-plane shear modulus of the handsheet per se made of themselves because of loss in fiber conformability during forming the handsheet, thus reduced the elastic modulus of the handsheet.

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Figure legends

Fig. 1. Ray model for determining the oscillation pattern of $V(z)$.¹⁻³⁾

Fig. 2. Oscillation pattern of $V(z)$ for determining a Rayleigh wave speed.

Fig. 3. Positioned sample for the SAM operation.

Fig. 4. Acoustic image of cross sectional surface of single pulp fiber (arrowhead).

Fig. 5. Two-dimensional image of $V(z)$ pattern in depth profile.

Fig. 6. Oscillation pattern of $V(z)$ along a dotted line from A' to B' in Fig. 5.

Fig. 7. Accuracy and precision of measured Rayleigh wave speed values.

Fig. 8. Increase in C_{44} of a heat-treated fiber wall that is not consistent with decrease in in-plane shear modulus (G) of a handsheet made of the fibers.

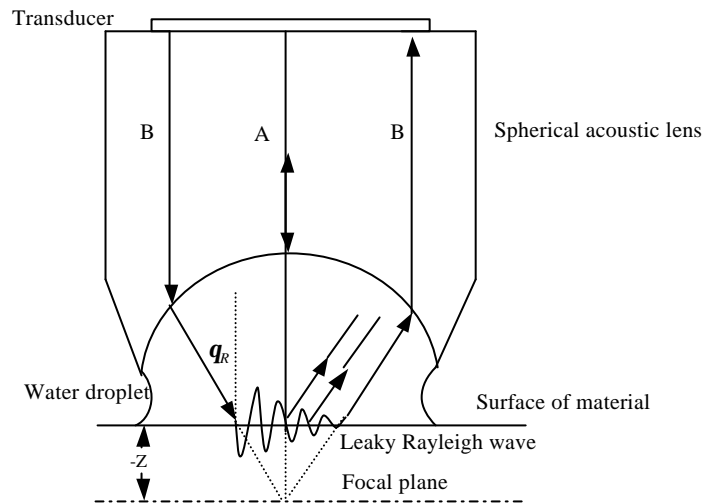


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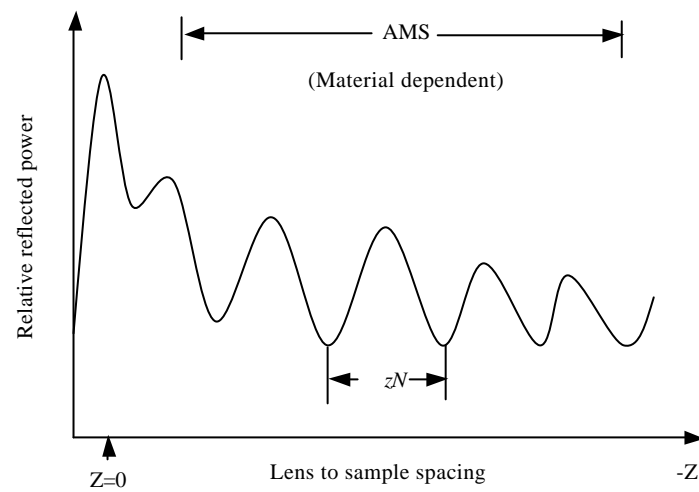


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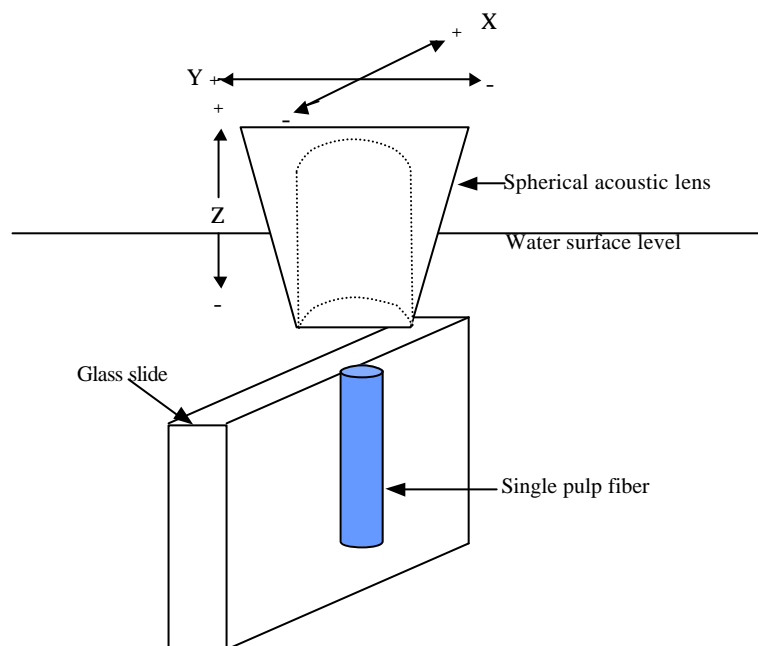


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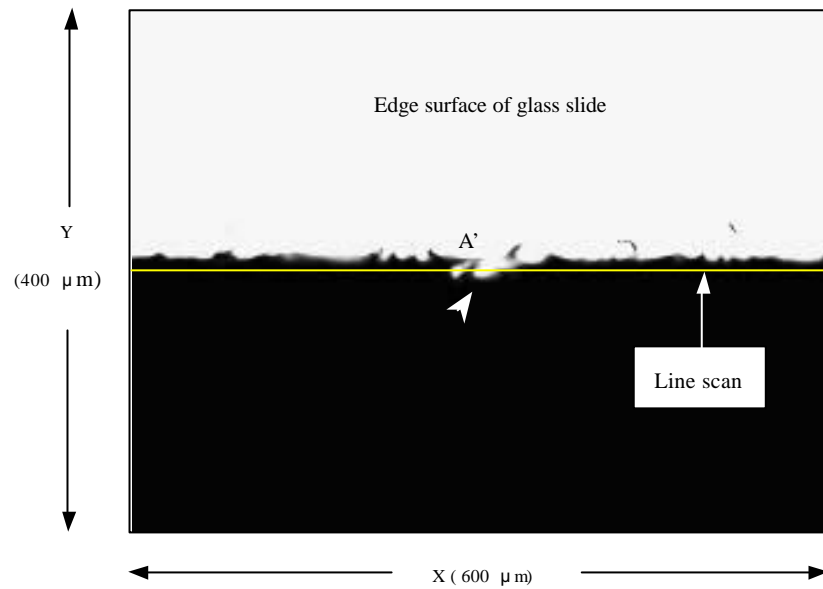


Fig. 4. Acoustic image of cross sectional surface of single pulp fiber (arrowhead).

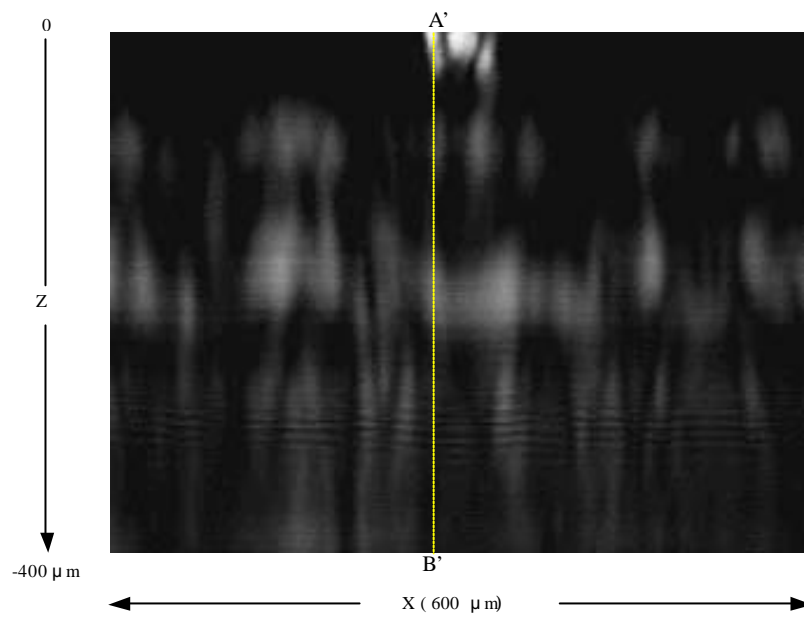


Fig. 5. Two-dimensional image of $V(z)$ pattern in depth profile.

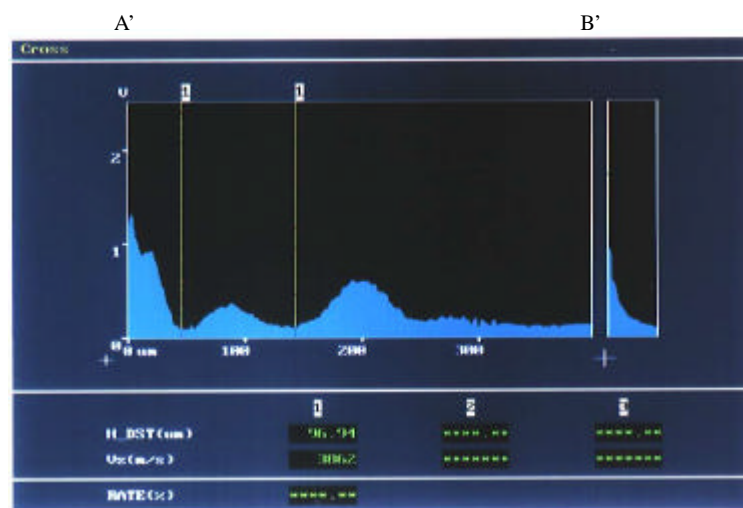


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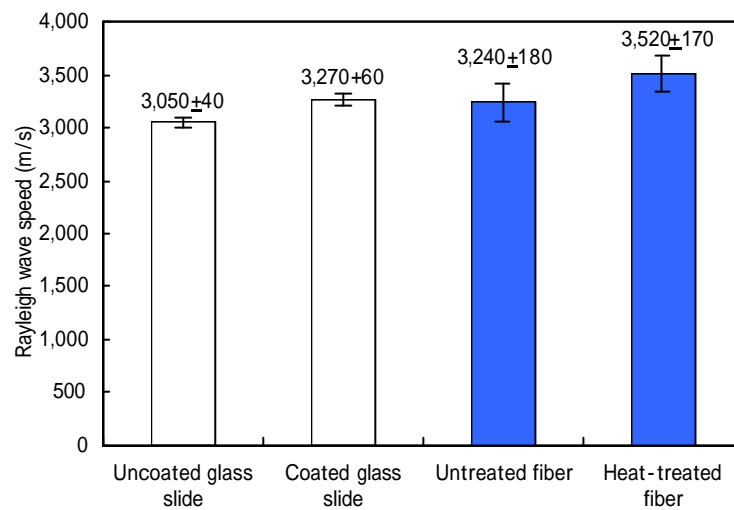


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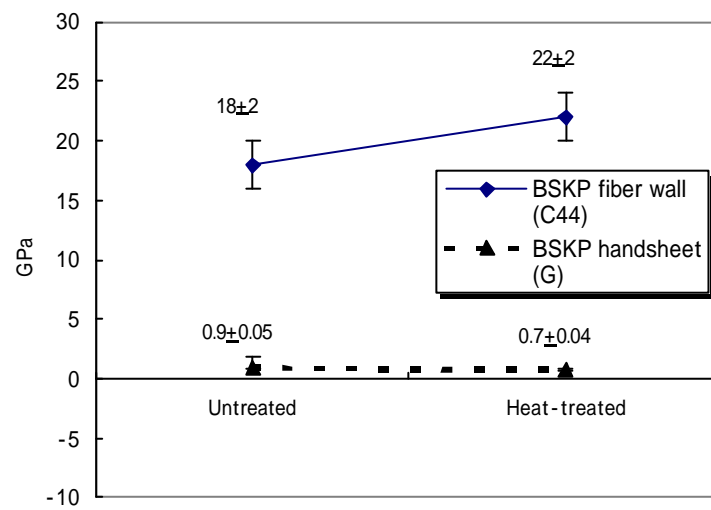


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