

# Quick Determination of Dielectric Anisotropy of Paper Sheets by Means of Microwaves

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## Synopsis

This paper deals with a new type of method for determining the complex dielectric constant at microwave frequencies in about 30 s. This method determines the dielectric anisotropy and refractive index for optically opaque paper sheets. The dielectric anisotropy ascribable to the local motions of cellulose molecules is supported by electron microscopic observations and mechanical anisotropy. The angular dependence of the complex dielectric constant gives the degree of molecular orientation and the distribution of cellulose molecules in paper sheets. The direction of the average orientation of molecular chains can be inferred from the anisotropic data. It is also suggested that the cellulose molecules should be oriented on an average in the direction of long axis of pulp fibers. The accumulation of data for complex dielectric constant and refractive index at microwave frequencies gives an important chart for manufacturing the functional paper corresponding to needs.

## INTRODUCTION

Dielectric relaxations have been studied by many authors at frequencies below 10 MHz with an aim of investigating the relationship between the motions of molecular chains and their structure in polymer film.<sup>1-4</sup> However, few studies on dielectric anisotropy have been reported,<sup>5,6</sup> because the preparation of different samples such as randomly oriented and uniaxially oriented films requires a long time. Because the fine structure of molecular chains differs from sample to sample, it is essential to measure the dielectric anisotropy with the same sample. For paper sheets, it is difficult to measure the complex dielectric constant after evaporating a metal onto the quality paper sheet, because the surface of a paper sheet consisting of pulp fibers is not smooth and the evaporated metal penetrates into the paper sheet containing many voids. For such paper sheets it is necessary to measure the dielectric anisotropy without contact.

So far, few studies have been made on the dielectric properties at microwave frequencies.<sup>7,8</sup> In previous papers,<sup>9-11</sup> the author investigated the orientation of paper or polymer films at a fixed frequency by use of a Microwave Molecular Orientation Analyzer MOA-2001A developed by Kanzaki Paper Mfg. Co., Ltd. This analyzer allowed the measurement of the dielectric anisotropy of paper sheet at microwave frequencies without difficulty.

This paper describes such an experimental study made on paper sheets. It also describes an attempt at evaluating the complex dielectric constant at microwave frequencies and the dielectric anisotropy.

## EXPERIMENTAL

The samples used were tetrafluoroethylene-hexafluoroethylene copolymer films, handsheets, machine-made paper, uniaxially oriented paper, and condenser paper. The tetrafluoroethylene-hexafluoroethylene copolymer films were previously found to be random in molecular orientation by the microwave method.<sup>10,11</sup> The handsheets with a basis weight of 62 g/m<sup>2</sup> were prepared from hard wood pulps with a Canadian standard freeness (CSF)<sup>12</sup> of 400 mL beaten by a Niagara beater. A uniaxially oriented paper with a basis weight of 41 g/m<sup>2</sup> and a density of 0.48 g/cm<sup>3</sup> was prepared by using an oriented-sheet former manufactured by Kumagai Riko Co. The machine-made paper with a basis weight of 60 g/m<sup>2</sup>, a density of 0.81 g/cm<sup>3</sup>, and a 2800-mm width was also prepared at a machine speed of 650 m/min. The condenser paper with a basis weight of 28 g/m<sup>2</sup> and a CSF of 200 mL had a high density of 1.43 g/cm<sup>3</sup>.

The apparatus used was a Microwave Molecular Orientation Analyzer MOA-2001A developed by Kanzaki Paper Mfg. Co., Ltd.<sup>9-11</sup> Polarized microwaves were irradiated perpendicularly to the plane of a test sample which was inserted in a narrow gap between a pair of rectangular waveguides consisting of a cavity resonator system. The sample had been cut to a fixed size of 10 × 10 cm<sup>2</sup> and placed in a sample holder. The resonance curve of frequency vs. transmitted microwave intensity gives a resonance frequency and a *Q* value in the resonator system. Here, the resonance frequency is defined by one at a maximum of the transmitted microwave intensity in the curve, and the *Q* value by the ratio of the resonance frequency to the half-width in the curve. The electromagnetic mode used was a transverse electric wave (TE) of a type TE<sub>10L</sub>, with *L* being an odd number.<sup>10,11</sup> The resonance frequency and the half-width in the resonance curve were searched by sweeping frequency with a tuner rotated, and the dielectric constant  $\epsilon'$  and the dielectric loss  $\epsilon''$  were determined. Then, the sample was rotated to different angles around the central axis normal to the sheet plane, and measured successively to determine  $\epsilon'$  and  $\epsilon''$ . It took about 3 min to obtain the complete angular dependence of  $\epsilon'$  and  $\epsilon''$  at every 30°. The measuring frequencies were 3.4–4.0 GHz.

The mechanical breaking strength was measured for uniaxially oriented paper sheet with a basis weight of 41 g/m<sup>2</sup> and a size of 15 mm (width) × 100 mm (length) prepared at a stretching rate of 10 mm/min by the use of an Instron. The electron microphotographs were obtained using a scanning microscope JSM T300 (Japan Electron Optics Laboratory Co.).

All measurements were made at 20°C and 65% RH. The sample used in the present study contained about 6% water.

### Evaluation of Complex Dielectric Constant

The sample must be very small compared to the cavity itself so that a frequency shift is produced by the insertion of the sample, where the frequency shift is much smaller than the resonance frequency of the empty cavity. The cavity perturbation method can be used for determining the complex dielectric constant when the dielectric loss is small.<sup>13</sup>

Defining the complex angular frequency by  $\omega$ , the complex dielectric constant of a sample by  $\epsilon$ , and the electric field by  $E$ , we have

$$\frac{\omega_2 - \omega_1}{\omega_2} = \frac{(\epsilon_2 - 1) \int_{V_s} |\mathbf{E}_1| |\mathbf{E}_2| dV}{2 \int_V |\mathbf{E}_1|^2 dV} \quad (1)$$

where the subscripts 1 and 2 indicate, respectively, the values before and after the insertion of the dielectric sample,  $V$  is the total volume of the cavity,  $\mathbf{E}_1$  is the electric field in the empty cavity, and  $\mathbf{E}_2$  is the perturbed field in the sample volume  $V_s$ .

When the sample is a sheet material and its size is larger than that of the opening of each waveguide, the electromagnetic field in the cavity is substituted into eq. (1). The dielectric constant  $\epsilon'$  and dielectric loss  $\epsilon''$  for the sheet material are given by

$$\epsilon' = 1 + A \frac{c}{t} \frac{f_{1r} - f_{2r}}{f_{2r}} \quad (2)$$

$$\epsilon'' = B \frac{c}{2t} (1/Q_2 - 1/Q_1) \quad (3)$$

Here,  $c$  is a parameter related to the depth of the cavity,  $t$  the apparent thickness of the sample,  $A$  and  $B$  constants associated with the apparatus,<sup>14</sup>  $f_{1r}$  and  $f_{2r}$  the resonance frequencies, and  $Q_1$  and  $Q_2$  the values, with the subscripts 1 and 2 indicating the values before and after the insertion of the sample, as above.

According to the Maxwell equation, the refractive index  $n$  of a dielectric material with a low dielectric loss at sufficient high frequency is given by

$$n = \sqrt{\epsilon'} \quad (4)$$

### Dielectric Anisotropic Index

According to eqs. (2) and (3),  $\epsilon'$  and  $\epsilon''$  decrease with increase in  $t$ , which is an increasing function of the content of voids in the sheet if the basis weight stays unchanged.

If the fiber arrangement and the basis weight are the same for two sheets with different thicknesses  $t(1)$  and  $t(2)$ , the resonance frequency and  $Q$  value observed for them do not differ. For these sheets eqs. (2) and (3) give

$$[\epsilon'(1) - 1]/[\epsilon'(2) - 1] = t(2)/t(1) \quad (5)$$

$$\epsilon''(1)/\epsilon''(2) = t(2)/t(1) \quad (6)$$

which indicate that the ratios of  $[\epsilon'(1) - 1]/[\epsilon'(2) - 1]$  and  $\epsilon''(1)/\epsilon''(2)$  depend only  $t(2)/t(1)$ . Here,  $\epsilon'(1)$ ,  $\epsilon''(1)$  and  $\epsilon'(2)$ ,  $\epsilon''(2)$  are the values in a fixed direction for the samples with thicknesses of  $t(1)$  and  $t(2)$ , respectively.

The direction of maximum  $\epsilon'$  or  $\epsilon''$  may not differ for the two samples. Thus, eqs. (5) and (6) yield

$$[\epsilon'_{\max}(1) - 1]/[\epsilon'_{\min}(1) - 1] = [\epsilon'_{\max}(2) - 1]/[\epsilon'_{\min}(2) - 1] \quad (7)$$

$$\epsilon''_{\max}(1)/\epsilon''_{\min}(1) = \epsilon''_{\max}(2)/\epsilon''_{\min}(2) \quad (8)$$

which imply that both  $\epsilon''_{\max}/\epsilon''_{\min}$  and  $(\epsilon'_{\max} - 1)/(\epsilon'_{\min} - 1)$  are invariant for samples with different thicknesses but the same basis weight. In other words, these ratios should give an anisotropic index essentially free from the effect of voids.

More generally, the following relations can be derived for a given sample:

$$(\epsilon'_{\max} - 1)/(\epsilon'_{\min} - 1) = (f_{1r} - f_{2r(\max)})/(f_{1r} - f_{2r(\min)}) \quad (9)$$

$$\epsilon''_{\max}/\epsilon''_{\min} = (1/Q_{2(\max)} - 1/Q_1)/(1/Q_{2(\min)} - 1/Q_1) \quad (10)$$

The ratios of  $(\epsilon'_{\max} - 1)/(\epsilon'_{\min} - 1)$  and  $\epsilon''_{\max}/\epsilon''_{\min}$  may be considered to be anisotropic indices substantially free from the effect of voids.

## RESULTS AND DISCUSSION

### Thickness Dependence of Dielectric Constant

Figure 1 shows  $\epsilon'$  at 4.0 GHz for tetrafluoroethylene-hexafluoroethylene copolymer films with different thicknesses. It can be seen that  $\epsilon'$  is about 2.04,

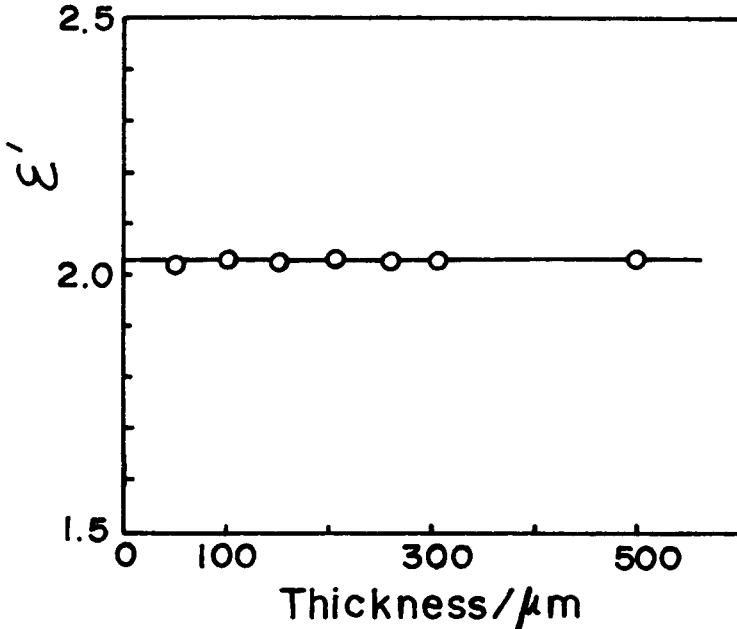


Fig. 1. Thickness dependence of dielectric constant  $\epsilon'$  at 4.0 GHz for tetrafluoroethylene-hexafluoroethylene copolymer.

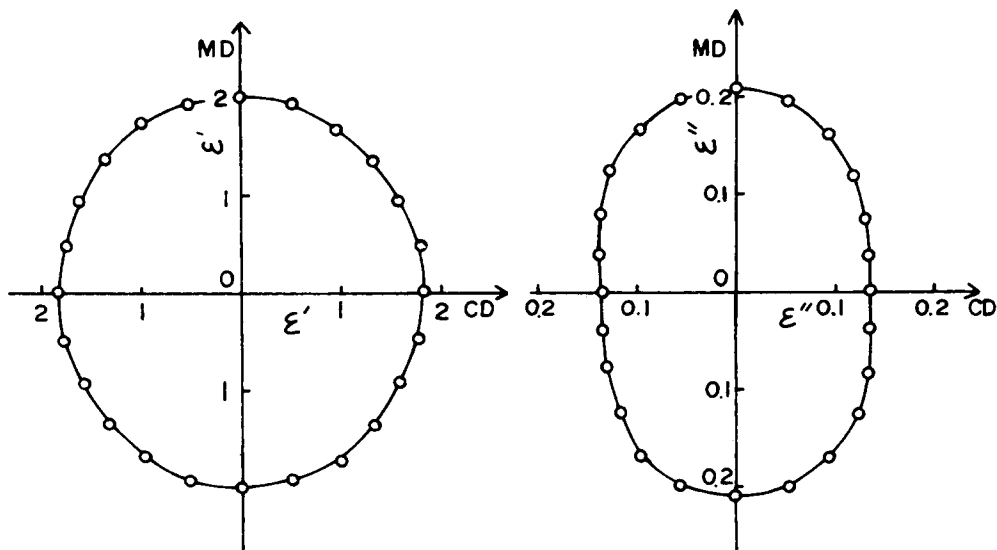


Fig. 2. Angular dependences of dielectric constant  $\epsilon'$  and dielectric loss  $\epsilon''$  at 3.4 GHz for a uniaxially oriented paper with a basis weight of 41 g/m<sup>2</sup> and a density of 0.48 g/cm<sup>3</sup>.

independent of thickness. A change in dielectric constant was less than 1.0% even when the electromagnetic mode was changed. For such a film, the value of 2.05 had been reported at 1.0 GHz,<sup>15</sup> in good agreement with our value obtained by the MOA-2001A instrument. This agreement may be taken as showing the reliability of our method for determining the dielectric constant at a microwave frequency of 4.0 GHz.

### Anisotropy of Complex Dielectric Constant for Paper Sheet

Figure 2 shows  $\epsilon'$  and  $\epsilon''$  at 3.4 GHz for a uniaxially oriented paper with a basis weight of 41 g/m<sup>2</sup> and a density of 0.48 g/cm<sup>3</sup>. Both  $\epsilon'$  and  $\epsilon''$  in the machine direction MD are larger than those in the machine cross direction CD which is perpendicular to the MD. The  $\epsilon'$  value of ca. 2.0 suggests that the paper should contain many voids. In other words, this dielectric constant is an apparent value for the bulk paper constituting pulp fibers and voids. In fact, the density was very low.

The electron microphotograph taken for the surface of the uniaxially oriented paper is shown in Figure 3. It suggests that the pulp fibers orient in a unique direction on an average. The electron microphotographs for the cross section in the MD and the CD also given in Figure 3 show that most pulp fibers orient in the MD within the plan of the paper, and support the above suggestion that the paper contains many voids.

Figure 4 shows the angular dependence of  $\epsilon''$  at 3.4 GHz for a condenser paper with a low CSF of about 200 mL and a high density of 1.43 g/cm<sup>3</sup>. The density is nearly equal to those of pulp fibers.<sup>16</sup> The condenser paper with less voids is also seen to show an anisotropy in  $\epsilon''$ . The direction of the maximum  $\epsilon''$  corresponds to that of the maximum in polarized infrared absorbance (see Fig. 4 or Ref. 10 for the anisotropy in polarized infrared absorbance), suggest-

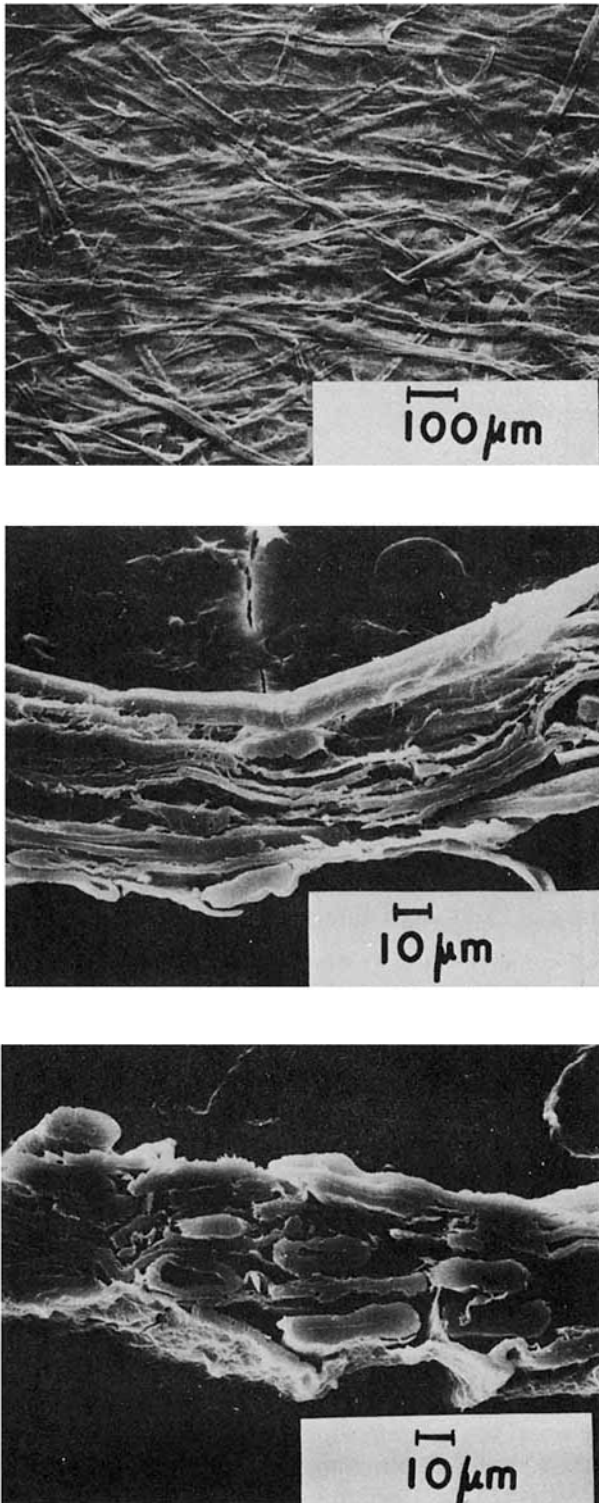


Fig. 3. Electron microphotographs of the surface and the cross sections in the MD and CD for a uniaxially oriented paper with a thickness of  $85 \mu\text{m}$  and a basis weight of  $41 \text{ g/m}^2$ .

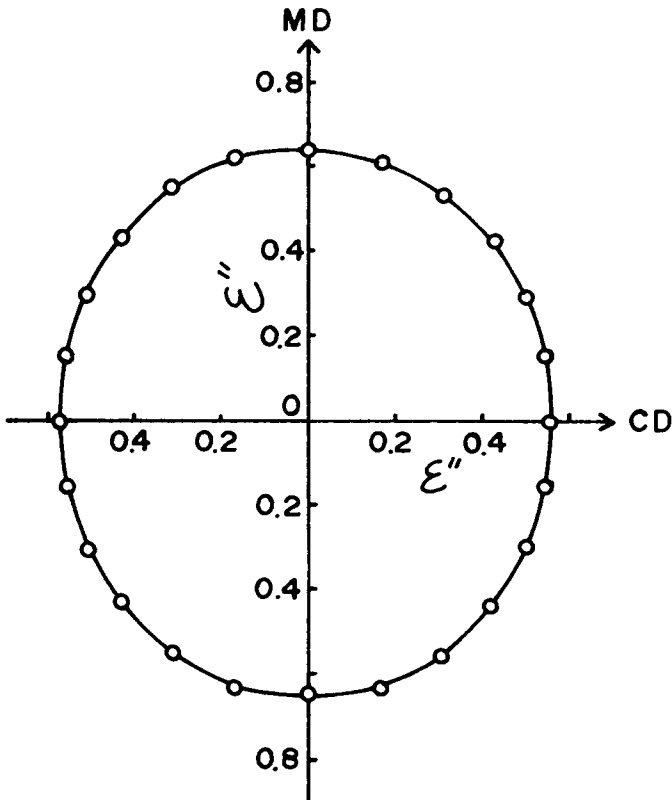


Fig. 4. Angular dependence of  $\epsilon''$  at 3.4 GHz for a condenser paper with a basis weight of 28 g/m<sup>2</sup> and a density of 1.43 g/cm<sup>3</sup>.

ing that the main chains of cellulose molecules orient predominantly in the direction of maximum  $\epsilon''$ . The dielectric constant for the condenser paper was 4.2–4.5, which indicates that this paper has a condensed texture of pulp fibers.

For comparison, the angular dependence of mechanical breaking strength for the uniaxially oriented paper with a density of 0.48 g/cm<sup>3</sup> is shown in Figure 5. The breaking strength in the MD is larger than that in the CD, indicating that the pulp fibers orient mainly in the MD, as found above from electron microphotographs.

The relation between the dielectric constant  $x$  and the mechanical breaking strength  $y$  for the uniaxially oriented paper was found to be expressed, in a first approximation, by the following empirical equation:

$$y = -24.1 + 13.6x \quad (11)$$

with a correlation coefficient between  $x$  and  $y$  of 0.93 and a standard deviation of 0.37. In more strict treatment,  $y$  may be expressed in terms of a quadratic equation of  $x$ , it was found that

$$y = 282.1 - 30.4x + 824.0x^2 \quad (12)$$

with a standard deviation of 0.31 (see Figure 6).

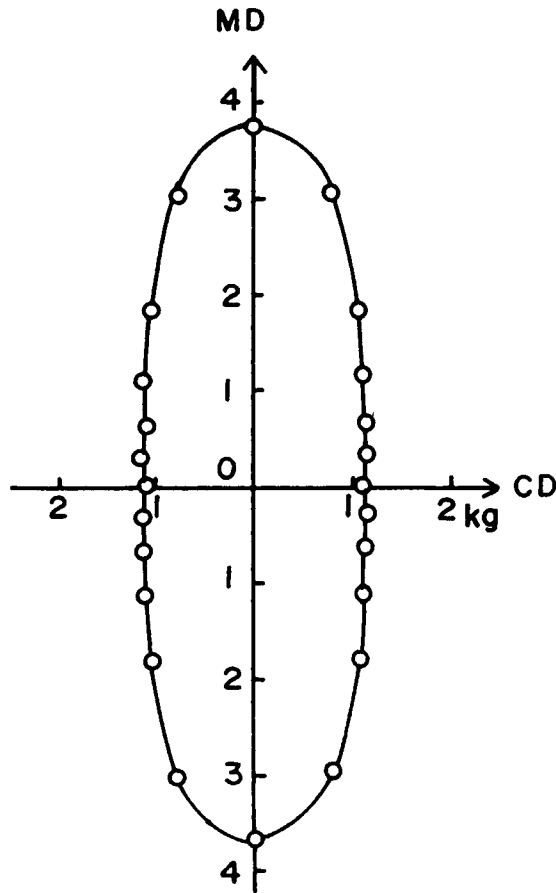


Fig. 5. Angular dependence of mechanical breaking strength for the uniaxially oriented paper with a thickness of  $85 \mu\text{m}$ , a sample width of 10 mm, a basis weight of  $41 \text{ g/m}^2$ , and stretching rate of 5 mm/min.

The relation between  $y$  and the dielectric loss  $z$  for the uniaxially oriented paper could be expressed by

$$y = -3.6 + 32.2z \quad (13)$$

with a correlation coefficient of 0.92 and a standard deviation of 0.40 or by

$$y = 15.3 - 189.4z + 636.5z^2 \quad (14)$$

with a standard deviation of 0.222 (see Fig. 6). Thus, we may conclude that good correlations exist between  $x$  and  $y$  and between  $z$  and  $y$ .

As discussed above, the dielectric data reflect the anisotropy of the cellulose molecules rather than the macroscopic pulp fibers while the mechanical anisotropy is considered to originate from the arrangement of the pulp fibers. It is well established by X-ray diffraction<sup>17,18</sup> that cellulose molecules orient on an average along the long axis of pulp fibers and that the actual fibers are anisotropic in molecular orientation.



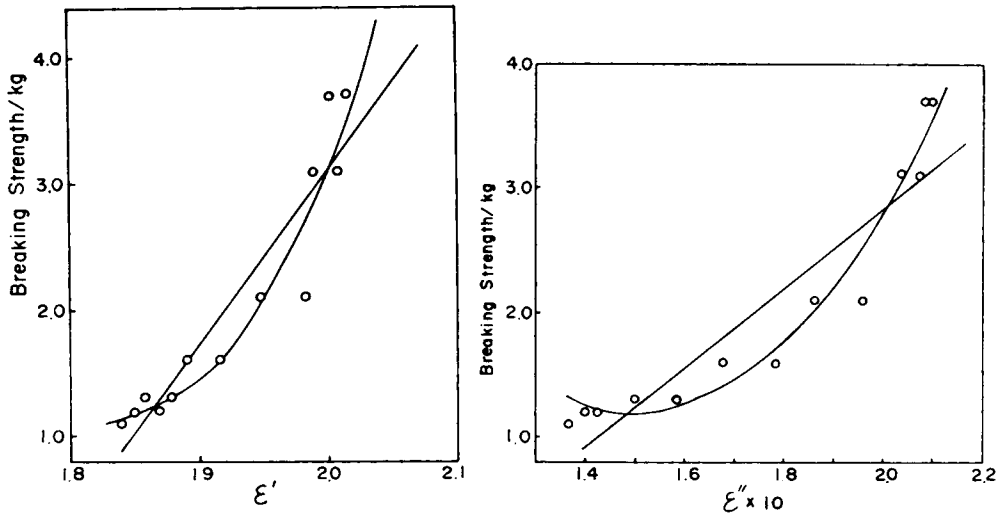


Fig. 6. Relationships between the mechanical breaking strength  $y$  and the dielectric constant  $x$  and between  $y$  and the dielectric loss  $z$  and 3.4 GHz for a uniaxially oriented paper with a basis weight of 41 g/m<sup>2</sup>.

The parallel relationship between the dielectric and mechanical anisotropies may be understood reasonably from such a structural character of the paper sheet. The complex dielectric constant for handsheet was essentially independent of the direction, and consistent with the result for the sheet from the mechanical breaking strength which showed no anisotropic behavior. Thus, the present microwave method is found to be a promising tool for studying the orientation of molecular chains quickly.

Figure 7 shows the angular dependence of  $n$  at 3.4 GHz for the same uniaxially oriented paper as in Figure 2. A remarkable anisotropy can be observed, as is the case for the dielectric constant; from this figure, the birefringence, which can be defined by the difference in  $n$  between the MD and the CD, is determined to be 0.075. Since  $n$  in different directions can be obtained, birefringences in different directions can also be determined. In this way, the present microwave method allows the refractive index and birefringence even of the opaque materials to be determined.

### APPLICATIONS

Figure 8 shows the angular dependence of  $\epsilon''$  at three different positions (front, center, and back) in the CD for the machine-made paper with a basis weight of 60 g/m<sup>2</sup>, a density of 0.81 g/cm<sup>3</sup>, and a 2800-mm width; samples were prepared by cutting a 2800-mm wide sheet to 28 different portions. The patterns differ at different positions. The orientation angles defined as the direction of maximum  $\epsilon''$  relative to the MD are plotted against the different positions in the CD in Figure 9. The orientation angle changes linearly with changing position in the CD. However, the orientation angle did not change for different samples in the same direction of the MD. Such angular dependence of complex dielectric constant reflects the distribution of pulp fibers within the paper plane.

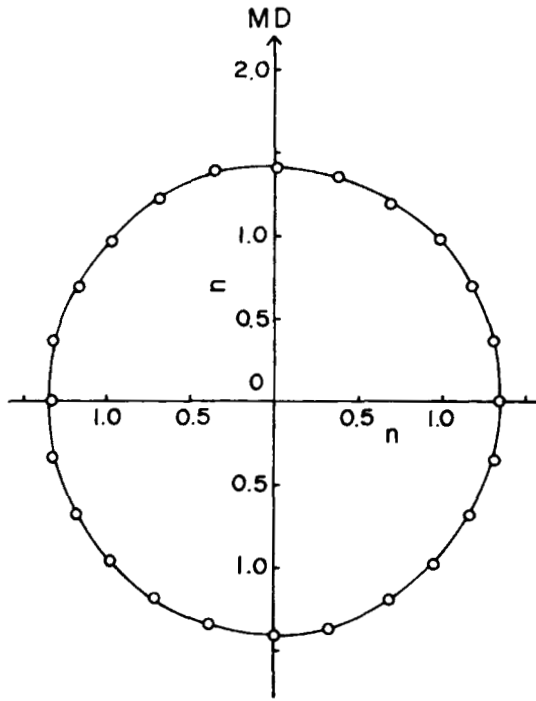


Fig. 7. Angular dependence of refractive index  $n$  at 3.4 GHz for a uniaxially oriented paper with a basis weight of  $41 \text{ g/m}^2$  and a density of  $0.81 \text{ g/cm}^3$ .

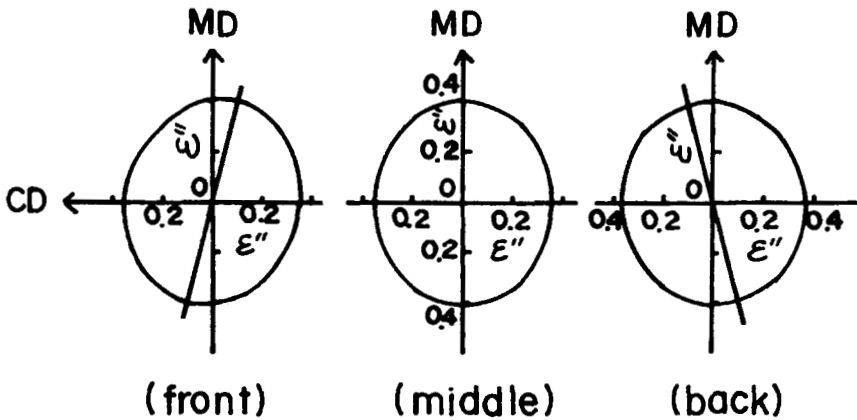


Fig. 8. Angular dependence of  $\epsilon''$  at 4.0 GHz at three different positions (front, center, and back) in the machine cross-direction CD for machine-made paper with a width of 2800 mm, a basis weight of  $60 \text{ g/m}^2$ , and a density of  $0.81 \text{ g/cm}^3$ .

The orientation angle was larger in the front and the back, but smaller in the center. This finding suggests that the axis of molecular chains deviates from the MD on the edge of the sheet, while most of the molecular chains are parallel to the MD in the center.

In Figure 10, the maximum to minimum ratio of  $\epsilon''$  is plotted against different positions in the CD for the machine-made paper with a basis weight

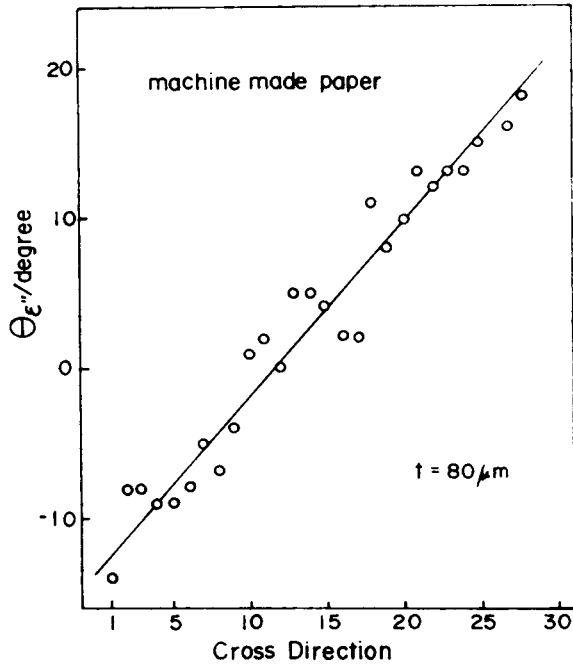


Fig. 9. Orientation angles determined from the angular dependence of  $\epsilon''$  at 4.0 GHz for the samples prepared at 28 different positions in the CD of machine-made paper with a width of 2800 mm, a basis weight of 60 g/m<sup>2</sup>, and a density of 0.81 g/cm<sup>3</sup>.

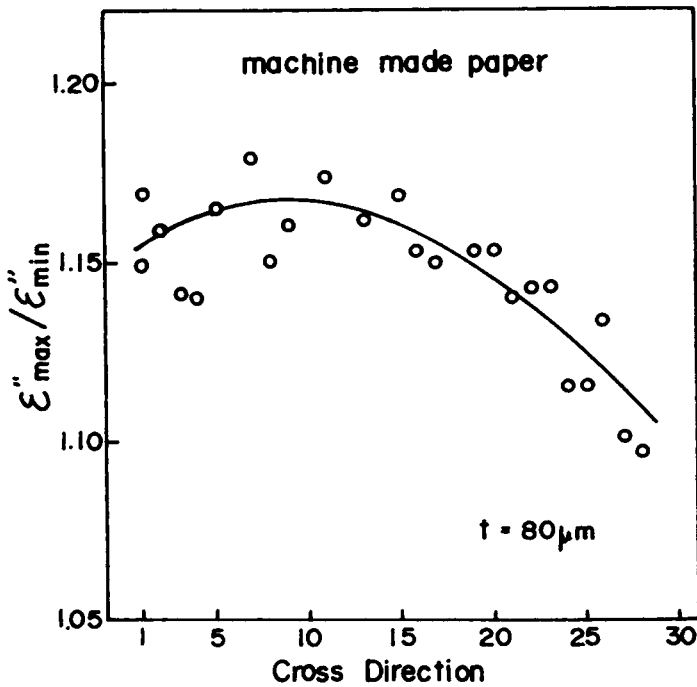


Fig. 10. Maximum to minimum ratio of  $\epsilon''$  at 4.0 GHz at many different positions in the CD of machine-made paper with a width of 2800 mm, a basis weight of 60 g/m<sup>2</sup>, and a density of 0.81 g/cm<sup>3</sup>.

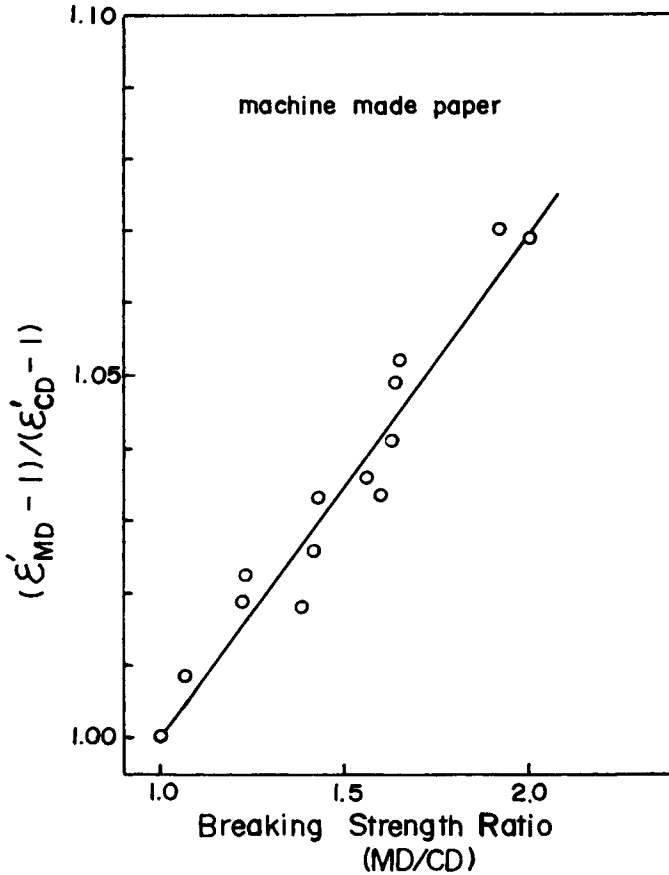


Fig. 11. Relationship between ratios of  $(\epsilon' - 1)$  at 4.0 GHz and of mechanical breaking strength in the MD to CD for machine-made paper with various basis weights.

of 60 g/m<sup>2</sup> and a density of 0.81 g/cm<sup>3</sup>. The ratio  $\epsilon''_{\max}/\epsilon''_{\min}$ , which gives a measure for the degree of orientation of pulp fibers, depends on the position in the CD. The ratio decreased with changing position from the center to the back, indicating that the anisotropy is not uniform in the CD.

Figure 11 shows a relationship between the ratios of  $(\epsilon' - 1)$  and of breaking strength in the MD to CD. The dielectric ratio increases almost linearly with increasing mechanical ratio which is a measure of anisotropy. This suggests that the microwave method is useful for estimating the anisotropy without destruction, instead of mechanical breaking strength.

### CONCLUSIONS

In this study, it was found that only apparent values of  $\epsilon'$  and  $\epsilon''$  were obtained for the machine-made paper sheet containing many voids. These values should approach the values intrinsic to pulp fibers when the content of voids is increased. An increase in voids increases with paper thickness  $t$ . Thus, according to eqs. (2) and (3),  $\epsilon'$  and  $\epsilon''$  increase as the density of paper increases. This is because the number of dipoles per unit volume increases.

The anisotropy of  $\epsilon'$  or  $\epsilon''$  depends not only the degree of the average orientation of pulp fibers but also on the density. If  $\epsilon'$  changes by wet pressing, the change should be due entirely to that of  $t$ . However, if the fiber orientation does not appreciably change within the plane of the sheet on wet pressing, i.e., with an increase in density, the molecular anisotropy should be invariant. Our point is that although  $\epsilon'$  and  $\epsilon''$  increase on wet pressing, the ratios of  $\epsilon''_{\max}/\epsilon''_{\min}$  and  $(\epsilon'_{\max} - 1)/(\epsilon'_{\min} - 1)$  are not significantly affected by the content of voids and are a measure of the dielectric anisotropy of the paper sheet. The effects of voids and thickness are eliminated in these ratios.

We obtained  $\epsilon''_{\max}/\epsilon''_{\min} = 1.0$  for a randomly oriented paper and  $\epsilon''_{\max}/\epsilon''_{\min} = 1.49$  for a uniaxially oriented paper. The difference between these ratios may be used for estimating the degree of molecular orientation. Because of the good correlations are obtained among the complex dielectric constant, mechanical anisotropy, and electron microscopic observations, we expect that the average orientation of molecular chains can be inferred from our anisotropic dielectric data.

The remarkable anisotropy of  $\epsilon'$  and  $\epsilon''$  in the poly(ethylene terephthalate) film<sup>19</sup> containing no voids suggests that the dielectric anisotropy is ascribed not to the voids but to the orientation of macromolecular chains. The angular dependence of  $\epsilon''$  for the condenser paper in Figure 4 shows the same thing. Since the angular dependence of polarized infrared absorbance<sup>10</sup> and mechanical breaking strength also shows a maximum in the MD, the cellulose molecules must be oriented on an average in the MD where the long axis of pulp fibers orients mainly. Hence, the dielectric anisotropy may be explained as the result of local motions which leads to a change in dipole moment in the direction of main axis of cellulose molecules.

An effect of water on dielectric behavior was observed. For example,  $\epsilon'$  and  $\epsilon''$  increased with increasing the content of water. However, for the paper containing 5–7% water,  $\epsilon'$  did not increase appreciably. Therefore, water contributes to the increase in  $\epsilon'$  and  $\epsilon''$ , but does not significantly change the anisotropy of pulp fibers. However, the orientation of water adsorbed on cellulose molecules could not be observed by FT-IR measurements.<sup>20</sup> Therefore, the changes in  $\epsilon'$  and  $\epsilon''$  because of the presence of water should contribute to a circular pattern in the angular dependence of  $\epsilon'$  and  $\epsilon''$ . In short, although the observed  $\epsilon'$  and  $\epsilon''$  are the superposition of contributions from water and the pulp fibers, the anisotropy of  $\epsilon'$  and  $\epsilon''$  comes not from the water but from the pulp fibers.

The accumulation of data for  $\epsilon'$ ,  $\epsilon''$ ,  $n$ , orientation angle, and the maximum to minimum ratio of  $(\epsilon' - 1)$  and  $\epsilon''$  will provide an important data base for manufacturing paper corresponding to needs. Actually these dielectric data are important for studying effects of additives, water content, compactness of pulp fibers, species of pulp in paper, etc., and also for producing the high quality paper with respect to the dimensional stability, such as thermal shrinkage.<sup>11</sup>

Drying conditions for paper sheets are undoubtedly important factors affecting fiber orientation. In this work, however, we did not go into this problem; we just focused on developing a quick determination of the complex dielectric constant and the anisotropy of paper sheets by our MOA-2001A instrument. Studies of effects of drying conditions on the anisotropy of sheets are now in progress in our laboratory.

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