DENSITY DEPENDENCE OF COMPLEX DIELECTRIC CONSTANT OF PAPER SHEET AT MICROWAVE FREQUENCIES

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ABSTRACT: Dielectric constant $\varepsilon'$ and dielectric loss $\varepsilon''$ at microwave frequencies were determined for handsheets of various densities prepared by changing freeness, mesh size for fractionation and pressure for wet pressing. Both $\varepsilon'$ and $\varepsilon''$ at 4.0 GHz increased linearly with increasing density. Empirical formulae $\varepsilon' = 0.531 + 3.07 \rho$ and $\varepsilon'' = -0.098 + 0.619 \rho$ for handsheets of hardwood pulps were obtained for densities between 0.5 and 1.0 g/cm$^3$. The correlation coefficient between $\varepsilon'$ and $\rho$ and between $\varepsilon''$ and $\rho$ were 0.994 and 0.984, respectively. These formulae were interpreted on the basis of proportionality of density to the number of dipoles per unit volume in the paper sheet.

1. INTRODUCTION

It is important to study the effect of density on physical properties of paper sheet consisting of pulp fibers and voids for the production of high quality papers. While the paper density affects dielectric properties of paper sheet, dielectric measurements at audio frequencies suffer from difficulties arising from incomplete deposition of metal on rough surfaces of paper sheet and penetration of the electrode metal into paper sheet. Therefore, the dielectric measurement at microwave frequencies, which eliminates the needs for contact between sample and electrodes, would give useful information on the fine structure of pulp fibers in paper sheets.

A new type of instrument using microwaves has been developed for determining the fiber orientation in paper sheets, blood vessels, and nonwoven fabrics and molecular orientation in polymer films. The new microwave method provides accurate and quick non-contact measurement of dielectric properties and should be useful for studying the density dependence of dielectric constant and dielectric loss, i.e., the relationship between the complex dielectric constant and the density of paper sheet.

The present paper describes such an experimental study on the density dependence of dielectric properties of paper sheets by using the microwave method.

2. EXPERIMENTAL

Most of the samples were handsheets prepared from hardwood pulps beaten by a Niagara beater. Canadian standard freeness (C.S.F.) for the hardwood pulps ranged from 100 ml to 700 ml. The pulp was fractionated by a classifier according to fiber length, and handsheets were prepared from each fraction. The screen meshes of classifier were 14, 24, 42 and 80 wires per inch. The density of handsheet was higher for smaller mesh and lower freeness. Handsheets with different densities were also prepared by changing the pressure of wet pressing for a pulp with a freeness of 580 ml C.S.F. The handsheets prepared from softwood pulps were also used.

All handsheets were preconditioned at 25°C and 65% RH for 10 days. Since the dielectric behavior is significantly dependent on moisture content, the behavior under this standard condition was studied by focusing on the influence of sheet density.

Measurements of complex dielectric constant were carried out by means of microwave molecular orientation analyzer model No. MOA-2001A. Polarized microwaves were irradiated perpendicularly to the plane of a sample sheet which was inserted in a narrow gap of a cavity resonator system. The sample of 100 mm × 100 mm was placed in the sample holder. The frequency dependence of transmitted microwave intensity gives the resonance frequency and the Q-value, defined as the ratio of the resonance frequency to the half width of the resonance curve. The dielectric constant $\varepsilon'$ and the dielectric loss $\varepsilon''$ were calculated from these values. It took about...
30 s for determining the complex dielectric constant for each sample. The measuring frequency was between 3.6 and 4.2 GHz.

The density of each paper sheet was determined by measuring its weight and thickness. In the present study, the average values of $e'$ and $e''$ at angles of 0° and 90° were used for eliminating the effect of anisotropy of handsheets.

### 3. COMPLEX DIELECTRIC CONSTANT

A perturbation theory is applicable for the determination of the complex dielectric constant when the dielectric loss is very small and the frequency shift is much smaller compared with the resonance frequency without the sample. The cavity perturbation method can be used for determining complex dielectric constant when the dielectric loss is small.

When the sample sheet is larger than the opening of each waveguide, $e'$ and $e''$ of the sheet at microwave frequencies are given by

$$
e' = 1 + \frac{c}{t} \left( \frac{1}{f_1} - \frac{1}{f_2} \right)$$

$$
e'' = B \frac{c}{2t} \left( \frac{1}{Q_2} - \frac{1}{Q_1} \right)$$

Here, $c$ is a parameter related to the depth of the cavity, $t$ is the apparent thickness of the sample, $A$ and $B$ are the instrumental factors, $f_1$ and $f_2$ are the resonance frequencies, and $Q_1$ and $Q_2$ are the $Q$-values, with the subscripts 1 and 2 indicating the values before and after insertion of the sample respectively.

For the dielectric relaxation ($\beta$-relaxation) due to local motion of macromolecules, the dielectric strength $\epsilon'_o - \epsilon''_o$ can be written as

$$
\epsilon'_o - \epsilon''_o = \frac{4\pi}{3} N \mu^2 \gamma \left( \frac{E_r}{E} \right)$$

where $\epsilon'_o$ is the dielectric constant at low frequencies, $\epsilon''_o$ is the dielectric constant at high frequencies, $N$ is the number of dipoles per unit volume, $\mu$ is the dipole moment, $\gamma$ is the restoring force, $E_r$ is the local electric field, and $E$ is the applied electric field.

From equation (3) we express the dielectric constant $\epsilon' (\omega)$ and dielectric loss $\epsilon'' (\omega)$ for the dielectric $\beta$-relaxation as

$$
\epsilon' (\omega) = \epsilon'_o - \epsilon''_o = \frac{4\pi}{3} N \mu^2 \gamma \sum \frac{g(\tau_i)}{1 + (\omega \tau_i)^2} \left( \frac{E_r}{E} \right) \propto N \propto \rho$$

$$
\epsilon'' (\omega) = \frac{4\pi}{3} N \mu^2 \gamma \sum \frac{g(\tau_i)}{1 + (\omega \tau_i)^2} \left( \frac{E_r}{E} \right) \propto N \propto \rho$$

which indicate that $\epsilon' (\omega) - \epsilon''_o$ and $\epsilon'' (\omega)$ at an angular frequency of $\omega$ are linearly related to $N$. Here, $g$ is the relaxation spectrum and $\tau$ is the relaxation time. Since $N$ is proportional to the density $\rho$, both $\epsilon' (\omega) - \epsilon''_o$ and $\epsilon'' (\omega)$ at a fixed microwave frequency should vary in proportion to the density $\rho$ of a paper sheet.

### 4. RESULTS

Figures 1 and 2 show the relations between $\rho$ and $\epsilon'$ at 4.0 GHz and between $\rho$ and $\epsilon''$ at 4.0 GHz, respectively, for various handsheets consisting of hardwood pulps. Both $\epsilon'$ and $\epsilon''$ increased linearly with increasing $\rho$. The indicated straight lines represent

$$
\epsilon' = 0.531 + 3.07 \rho$$

![Fig. 1](image1.png)

Fig. 1 Density dependence of $\epsilon'$ at 4.0 GHz for handsheets consisting of hardwood pulps. Handsheets were prepared by three different methods: _D_, freeness; _A_, mesh; _O_, pressure of wet pressing.

![Fig. 2](image2.png)

Fig. 2 Density dependence of $\epsilon''$ at 4.0 GHz for handsheets consisting of hardwood pulps. Handsheets were prepared by three different methods: _D_, freeness; _A_, mesh; _O_, pressure of wet pressing.
4.1 Effect of Freeness

The density of handsheets increased gradually with a decrease in freeness. This increase in density was large especially at freenesses below 200 ml. A lower freeness obtained by beating pulp fibers may be ascribed to more pronounced entanglement induced by cutting and fibrillation of pulp fibers. The value of $\varepsilon'$ for handsheets increased especially below a freeness of 200 ml.

Figures 1 and 2 include data for handsheets consisting of hardwood pulps with different freenesses of 100 ml to 700 ml.

Figure 4 shows $\varepsilon'$ at 4.0 GHz for mixtures of two kinds of pulps with freenesses of 100 ml and 600 ml C.S.F. It can be seen that $\varepsilon'$ decreases almost linearly with increasing the content of the pulp of freeness of 600 ml. This increase in $\varepsilon'$ may be caused by the increase in density due to an increase in the content of pulp with a lower freeness. We have also found that $\varepsilon''$ decreases linearly with increasing content of pulps with freeness of 600 ml.

4.2 Effect of Fiber Length

The density of handsheets increased with increasing mesh number (wires per inch). The length of pulp fibers decreased with increasing mesh number. Thus, an increase in mesh number should result in the increase in entanglement of pulp fibers, leading to an increase in density and have the increase in $\varepsilon'$.

Figure 5 shows the density dependence of $\varepsilon'$ at 4.0 GHz for the handsheets prepared by fractionating pulp fibers with screens of 14, 24, 42, and 80 mesh. As expected, $\varepsilon'$ increased with increasing mesh number; note that the density dependence of $\varepsilon'$ for the handsheets fractionated with different meshes has been shown in
Fig. 1.

4.3 Effect of Pressure under Wet Press

Figures 1 and 2 also include data for the handsheets prepared by wet pressing with different pressures. The density of the handsheets increased with increasing pressure. This is related to the increase in $\varepsilon'$ and $\varepsilon''$ with increasing density observed in the figures.

5. DISCUSSION

The density dependence of $\varepsilon'$ and that of $\varepsilon''$ for our handsheets are expressed by empirical formulae (6) and (7). The dependence can be interpreted by equations (4) and (5) for the relation between the density and the number of dipoles per unit volume. We note that formulae (6), (7) and (8) hold for $\rho$ between 0.5 and 1.0 g/cm$^3$ but not for lower densities. The increase in $\varepsilon'$ caused by increase in density must result from an increase in the number of dipoles per unit volume. In this work, the density was increased by decreasing freeness, increasing mesh number, and increasing pressure.

At a given $\rho$, $\varepsilon'$ is higher for handsheets with softwood pulps than for those with hardwood pulps, as is shown in Fig. 5. This fact may be ascribed to difference in the magnitude of the effective dipole moment or the number of dipoles per unit volume between these pulps.

It is known that $\varepsilon'$ and $\varepsilon''$ at 4.0 GHz for handsheets may be ascribed to the local motion of cellulose molecules including absorbed water molecules in the amorphous region. In order to uniformize the effect of water upon $\varepsilon'$ and $\varepsilon''$ for handsheets and to focus on the influence of sheet density, we preconditioned the handsheets at 25°C and 65% RH for 10 days.

If the $\varepsilon''$ comes from polarization due to reorientation of molecules in the amorphous region, the crystallinity will effect the value of $\varepsilon''$. It is unnecessary to consider the effect of crystallinity upon $\varepsilon'$ and $\varepsilon''$ for the handsheets prepared by wet pressing with different pressures, because the handsheets consist of hardwood pulps with the same freeness of 580 ml C.S.F. On the other hand, it may be necessary to consider the effect of crystallinity upon $\varepsilon'$ and $\varepsilon''$ for the handsheets prepared from the pulps with different freenesses and different mesh numbers because of difference in the crystallinity due to fibrillation of pulps. Actually, however, all data obtained from the samples with different freenesses and different mesh numbers approximately followed the same empirical formula to that for the samples prepared by wet pressing with different pressures. From this fact, the effect of crystallinity may be negligible for the samples used in the present study. In the future, it is expected to obtain the generalized formulae in a wide range of density by strictly examining the effect of factors such as crystallinity, water content and species of pulp fibers upon $\varepsilon'$ and $\varepsilon''$ for the handsheets.

The characteristics of a paper sheet can be evaluated from the empirical formulae (6) and (7) if $\rho$ is measured. From the complex dielectric constant estimated from $\rho$, we can obtain information on the species of pulp fibers, additives and impurities in the unknown paper sheets and then compare them with the standard sample. Such data will also reveal the difference in papermaking method. Thus, the measurement of complex dielectric constant at microwave frequencies is useful for evaluation of the compactness of pulp fibers in paper sheet, difference in species of pulps, and the structure and physicochemical properties. Such dielectric properties at microwave frequencies will be useful for solving various problems in quality occurring in paper sheets and then for developing new functional papers.

ACKNOWLEDGEMENT

The author is grateful to Y. Miura of Kanzaki Paper Mfg. Co., Ltd. for preparing the samples.

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マイクロ波域における紙シートの複素誘電率の密度依存性

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マイクロ波域において、種々の密度を持つ手抄紙の誘電率 \( \varepsilon' \) および誘電損失率 \( \varepsilon'' \) を測定した。密度の異なる手抄紙は、フリーネス、メッシュサイズを変えての分別、あるいはウエットプレス圧などを変えることによって調製した。\( \varepsilon' \) および \( \varepsilon'' \) は、密度の増加とともに増大した。広葉樹バルブから成る手抄紙の密度 \( \rho \) が0.5から1.0g/cm\(^3\)の範囲で、

\[
\varepsilon' = 0.531 + 3.07 \rho, \quad \varepsilon'' = -0.098 + 0.619 \rho
\]

なる実験式が得られた。ここで、\( \varepsilon' \) と \( \rho \), \( \varepsilon'' \) と \( \rho \) の間の相関係数は、それぞれ0.994, 0.984と良好であった。これらの密度依存性は、密度が紙シート中の単位体積当たりの双極子の数に比例するという事実から説明することができる。