

The Microwave Dielectric Constants of Water–Paper Mixtures: The Role of Sheet Structure and Composition

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Synopsis

A precise technique for determining dielectric constants at microwave frequencies from slotted line measurements is adapted to paper samples. The effects of refining, wet pressing, calendering, and TiO₂ addition on the dielectric constants vs. moisture curves are reported and discussed. The observed in-plane dielectric anisotropy is discussed. The importance of considering geometry at the fiber level in explaining the dielectric behavior of paper is stressed.

INTRODUCTION

At microwave frequencies the interaction between electromagnetic radiation and paper is very sensitive to the moisture content of the paper. Microwave moisture gages, which exploit this fact, are widely used in the paper industry. Microwave driers, which depend on the strong adsorption of microwave radiation by water, have also found applications. The fundamental parameter which determines this interaction is, of course, the dielectric constant ϵ of the paper at microwave frequencies. As will be discussed later, a number of studies of the dielectric constant of paper at various moisture levels have been published. In this paper, the studies are extended to include the effects of common paper-making process variables and additives.

Measurements are made on sheets subjected to variable levels of refining, wet pressing, and calendering. Titanium dioxide, a common paper additive which has a large dielectric constant, is found to have a small effect on the microwave dielectric constant of the sheet. This behavior is discussed. A difference in dielectric properties of sheets made from bleached and unbleached chemical pulps is observed. A small in-plane anisotropy in the microwave dielectric constant of normal machine-made papers is reported and discussed.

The experimental technique used here differs from the earlier studies. A more accurate technique is adapted for thin, flexible samples, and a cumbersome graphical analysis is performed numerically on a computer.

A large difference between the in-plane and out-of-plane dielectric constants has been noted by earlier investigators. This has been explained as a result of an anisotropy in the polarizability of the adsorped water; the water supposedly has a greater polarizability along the fiber axis. This explanation ignores the effect of geometric anisotropy at the fiber level, however, and is strongly criticized in the background section of this paper.

BACKGROUND

Below microwave frequencies water has a large real dielectric constant (~ 80) due to the motion of its permanent molecular dipole moment. At low frequencies the dipoles in an electromagnetic field can oscillate in phase with the changing direction of the electric field, thereby contributing to the real part of the dielectric constant. As microwave frequencies are approached, however, the dipoles are unable to stay in phase with the electric field, and both the real and imaginary parts of the dielectric constant are large. At still higher frequencies the permanent dipoles cannot contribute, and the dielectric constant of water decreases to about 5. The permanent dipole moment of water is the dominant factor in the interaction between paper-water mixtures and electromagnetic radiation at microwave frequencies.

To understand the dielectric behavior of paper, it is important to recognize that paper is a heterogeneous mixture of moist fibers and air. The problem of expressing the effective dielectric constant of a mixture in terms of its microscopic properties has attracted considerable attention. A book by Beran¹ and an article by Landauer² are good reviews of the subject. A fundamental concept in the study of heterogeneous mixtures is that the effective properties of the mixture cannot be determined solely from a knowledge of the properties of the constituents and their volume fractions. The geometry of the components in the mixture must also be known. With no information about the geometry, it can only be stated that the effective dielectric constant ϵ^* lies within the bounds³

$$(\overline{1/\epsilon})^{-1} \leq \epsilon^* \leq \bar{\epsilon} \quad (1)$$

Here $\bar{\epsilon}$ is the volume average of the dielectric constant and $\overline{1/\epsilon}$ is the average of $1/\epsilon$. The maximum occurs if the geometry is such that the electric field is parallel to all interfaces. The minimum occurs if the field and interfaces are always normal. The range between the two limits is large for equal mixtures of components if their dielectric constants are quite different. If the mixture geometry were stacked slabs, for example, both extremes could be measured. The maximum would exist when the applied field was parallel to the interfaces, and the minimum when it was normal. A highly anisotropic medium could be produced by an inhomogeneous mixture of isotropic components.

For paper, the geometry of the mixture is governed by the shape and orientation of the fibers. Kraft softwood fibers, for example, are ribbonlike and oriented in the plane of the sheet. For machine-made papers there is an additional orientation preference in the machine direction. When the applied electric field is in the plane of the paper, it is more nearly parallel to the air-fiber interfaces than when it is perpendicular to the sheet. Therefore, the dielectric constant is larger in the plane of the sheet than out of plane. Also, for machine-made papers the machine direction has a slightly larger ϵ than that in the cross direction. The best available model for studying the dielectric constant of paper in terms of its microscopic properties appears to be the one presented by Davies.⁴ He uses a "self-consistent approximation"¹ to estimate the dielectric constant of a medium with inclusions of infinitely long cylinders.

Another reason for the anisotropy in the dielectric constant of paper might be that moist fibers have different dielectric constants along and normal to the

fiber axis. Except in very dry sheets, water in the fibers is the major contributor to the dielectric constant at microwave frequencies. The anisotropy might arise if the polarizability of some of the water were dependent on the orientation of the electric field to the fiber axis. This would happen if the attachment between the water molecules and the polymers in the fiber was such that, on an average, water polarization was preferentially inhibited in some direction relative to the fiber. In proteins⁵ and protein fibers⁶ there is evidence that the bond between water and the protein polymers can decrease the microwave polarizability of the water. Here the "bound" water has a relaxation at about 1 GHz. For "free" water this relaxation in the motion of the permanent dipole of water does not occur until about 20 GHz. So, at microwave frequencies in paper there may be a considerable difference in the dielectric properties of "free" and "bound" water. A piece of evidence that the bond between cellulose and water can alter the microwave polarizability of water comes from Dusoiu.⁷ For paper at a given moisture constant, he shows that the dielectric constant is greater along the desorption isotherm than along the adsorption isotherm. This hysteresis is attributed to the presence of a greater ratio of "free" to "bound" water during drying. So, it is possible that the association of water with cellulose can hinder the motion of the water dipoles at microwave frequencies, decreasing the dielectric constant. We know of no direct evidence, however, that the degree of hindrance varies with the angle between the fiber and the electric field.

The fact that paper has a greater in-plane than out-of-plane microwave dielectric constant has been observed by a number of authors.⁸⁻¹² Several^{9,10} have explained this anisotropy in terms of a difference in the dielectric constant of water along and normal to the fiber. A model such as Kirkwood's,¹³ appropriate only for mixtures which are homogeneous at the molecular level, is applied to predict the anisotropy in the polarizability of water. Since the geometric anisotropy at the fiber level is ignored, this is an improper procedure. In contrast, Fleischman¹² has argued that the geometry effect at the fiber level is sufficient to explain all the anisotropy, even if the fiber properties are isotropic. This is not to say that the fiber polarizability is isotropic, but only that it is unnecessary to postulate fiber anisotropy in order to explain sheet anisotropy.

EXPERIMENTAL

Most investigators have used a shorted waveguide technique to measure the microwave dielectric constants of paper. See the article by Dusoiu et al.¹⁴ for a detailed description of this procedure. Briefly, a stack of paper is placed at the end of a shorted waveguide. The dielectric constant is calculated from the nature of the standing wave patterns resulting when continuous microwave radiation is reflected off the short with and without the paper placed at the short. A significantly different procedure has been described by Kumar and Smith,¹⁵ in which sheets of paper are placed in a resonant cavity and the dielectric constant is found from the shift in the cavity resonant frequency and quality factor.

A very sensitive apparatus is needed to make meaningful measurements on thin sheets. Therefore, we modified for use on thin specimens a slotted line technique developed some years ago by Deschamps.^{16,17} Instead of using a fixed end waveguide, a waveguide with a movable short is incorporated. A tedious graphical analysis is employed to determine the dielectric constant from mea-

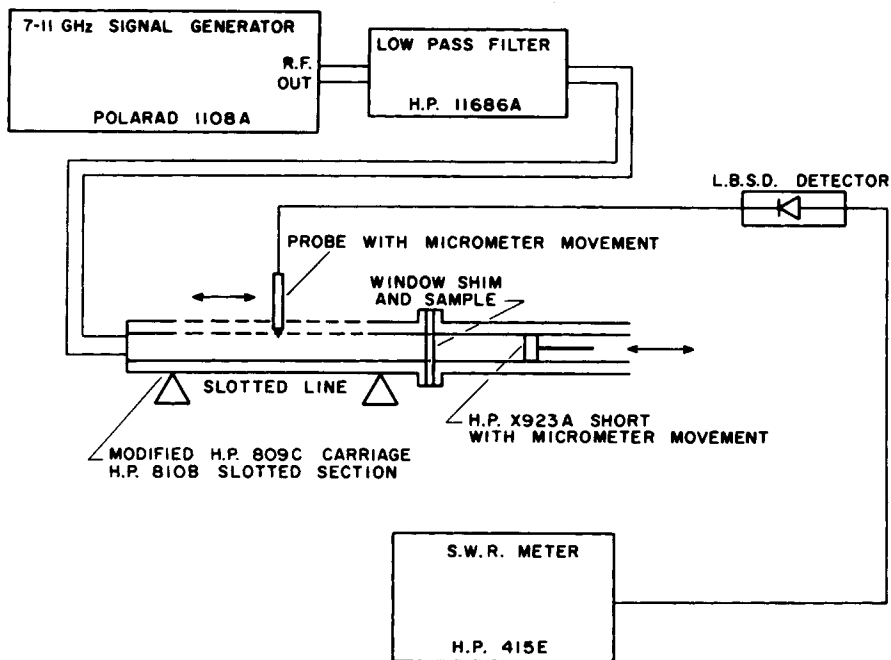


Fig. 1. Schematic of experimental apparatus.

measurements of the standing wave patterns resulting from a series of different sample to short separation distances. Our refinements of the system resulted in two major improvements: (1) thin, flexible samples can be precisely aligned in the waveguide, and (2) the graphical technique is simulated by a computer program in order to speed the data reduction task. A brief discussion of the Deschamps method with our adaptations follows.

A schematic diagram of the experimental apparatus is shown in Figure 1. The sample is mounted in the waveguide between a movable short and a slotted waveguide. The movable short and slotted line detector probe are positioned by precision micrometers, having 0.0001-in. dial divisions. Energy is supplied by a Polarad 1108A variable frequency microwave signal generator with internal 1-kHz square-wave amplitude modulation. A Hewlett Packard 11686A Low Pass Filter is used to condition the signal before sending it to the waveguide. The signal picked up by the detector probe is rectified by an Alpha Industries DDC4561D Low Barrier Schottky Diode (LBSD) in the probe mount and sent to a Hewlett Packard 415E Standing Wave Ratio (SWR) meter having a 1-kHz bandpass filter.

The standing wave pattern in the slotted line results from interference between the incoming wave and the wave reflected by the sample and short combination. The wave pattern (phase and amplitude) can be measured by using the probe to find the location of a standing wave minimum and the standing wave ratio (SWR). All measurements reported here were taken at 9.6 GHz.

The effect of the sample in the waveguide can be characterized at some plane by a scattering matrix, S_{ij} , where $i, j = 1$ or 2 . The scattering matrix is the ratio (at the defined plane) between radiation entering the substance on the j side and

exiting from the i side, when the waveguide is terminated in its characteristic impedance on the i side. Deschamps' graphical method determines the scattering matrix at a plane a known distance from the first short location, using measurements of the wave pattern in the slotted line resulting from different short-sample separations. Four separations are needed to determine uniquely the scattering matrix. However, to average out statistical fluctuations, measurements were made at eight separations. This also enabled us to determine the scatter in S_{ij} , by using different combinations of the measured wave patterns to calculate S_{ij} .

For a homogeneous material shaped as a regular parallelepiped, the complex dielectric constant can be calculated from the scattering matrix if either the sample thickness or the distance from the sample to the reference plane is known. Unfortunately, paper is a very inhomogeneous material with a poorly defined thickness or surface plane. Nevertheless, if the scattering matrix measured for the sample is such that $|S_{11}| = |S_{22}|$, then the scattering matrix can be reproduced by a homogeneous sheet of constant thickness. Within experimental error this was the case; thus, a dielectric constant could be associated with the paper. To calculate the dielectric constant from the scattering matrix, the thickness of the homogeneous slab was set equal to the micrometer thickness of the sample. The micrometer measurements were equivalent to readings on a Schopper caliper gage.

The method for mounting paper sheets inside the waveguide deserves special mention. To obtain accurate and repeatable results, it is necessary to carefully align the paper in the waveguide without inserting extraneous materials that can alter the standing wave pattern. This was achieved in the present work by supporting the samples in metal "window shims" of the appropriate thickness. A window shim has a punched hole with a height the same as the waveguide (0.4 in.), a length slightly longer than the waveguide width (0.9 in.), and a thickness slightly less than the thickness of the specimen to be supported. The paper specimen is cut to fit tightly in the window shim. The shim is then mounted between the slotted line and the movable short. There is no first-order effect on the standing wave pattern in the waveguide due to the shim, because the currents in the waveguide are parallel to the wall gaps caused by the shim.

For thin sheets (<0.8 mm) the effect of the paper on the standing wave pattern is not large enough to allow accurate measurements of the dielectric constant. However, when measurements were made on stacks of sheets, there was no significant variation of the dielectric constant with the number of sheets in the stack. Therefore, we used measurements on stacks (usually two or three sheets) to calculate the dielectric constant.

It was possible to adjust the sample moisture content from about 4% to about 14% by varying the ambient relative humidity. Higher moisture contents were achieved by sealing wet samples in 0.013-mm-thick Saran (registered trademark, Dow Chemical Co.) wrap. Experiments conducted on samples at 50% RH showed that the thin plastic had no detectable effects on the measured dielectric constants.

Altogether, the slotted line technique, careful specimen mounting procedures, and computer simulation of the graphical method for analyzing the slotted line data have resulted in a system capable of making dielectric constant measurements on paper with an accuracy of about 5%.

RESULTS

A main objective of this study was to investigate the effects of refining, wet pressing, and calendering on the dielectric constant of paper. To do this, bleached kraft handsheets were prepared with and without calendering, and unbleached kraft handsheets were prepared at four levels of wet pressing and two levels of refining. These sheets are described in Table I. The results of the dielectric constant measurements at all moisture contents are given in Table II. At a constant gravimetric moisture ratio, all three processes increase the dielectric constant, due largely to the increase in apparent density with refining, wet pressing, and calendering. In general, a density increase means a tighter packing of dipoles, and this gives a larger dielectric constant. At high moisture levels, water dominates the interaction between the sheet and microwave radiation. Therefore, the apparent water "density" (water mass per paper sample volume) is an important parameter in determining the dielectric constant.

Figures 2 and 3 are, respectively, plots of the real, ϵ' , and imaginary, ϵ'' , parts of the dielectric constant vs. water density for the unbleached sheets. Except at the dry end, where fiber dipoles make a significant contribution, the values of the real part of the dielectric constant lie nearly on the same curve for the different degrees of refining and wet pressing. The imaginary part of the dielectric constant depends only on water density at all moisture levels, because dry fibers have a very small imaginary dielectric constant. Similar results for the bleached samples are shown in Figures 4 and 5. The same general statements apply, although there is a significant decrease in ϵ'' with calendering at constant water density. This relative insensitivity of ϵ to process variables accounts for the success of microwave moisture gages at high moisture content.

Least square fits of the data in Figures 2 and 3 and in Figures 4 and 5 were made to Davies theory⁴ for media with cylindrical inclusions. The complex dielectric constant of the media, ϵ , is related to the complex dielectric constant of

TABLE I
Sample Characteristics

Sample no.	Description	Oven-dry basis weight (g/m ²)	Wet pressing pressure (psi)	Ash content (%)	Other
NM052	Machine-made newsprint	44.6			
BK060-00	Bleached kraft (BK)	59.2	50	0.22	
BK060-02	BK, calendered	59.2	50		Three calender passes
BK060-04	BK, ~4% TiO ₂	60.1	50	3.71	
BK060-08	BK, ~8% TiO ₂	60.4	50	8.85	
BK060-12	BK, ~12% TiO ₂	60.9	50	14.43	
UK205-1	Unbleached kraft	199.7	10		Refining time, 5 min
UK205-2	Unbleached kraft	199.8	50		Refining time, 5 min
UK205-3	Unbleached kraft	200.2	84		Refining time, 5 min
UK205-4	Unbleached kraft	200.2	400		Refining time, 5 min
UK205-5	Unbleached kraft	201.0	50		Refining time, 25 min

TABLE II
Summary of Results
(23°C, 9.6 GHz)

Water basis, wt/sheet (g/m ²)	Grav moist (%)	$\rho_{\text{H}_2\text{O}}$ (g/cm ³)	No. sheets	Caliper per sheet (in.)	ϵ'	ϵ''
<i>Sample: BK060-00</i>						
<i>ODBW: 58.57 g/m² per sheet</i>						
78.00	57.11	0.610	3	0.0050	30.3	10.3
59.27	50.30	0.470	3	0.0050	18.8	6.8
46.69	44.36	0.380	3	0.0048	13.9	4.6
32.29	35.54	0.265	3	0.0048	10.5	3.7
16.22	21.68	0.139	3	0.0046	5.1	1.7
5.45	8.52	0.058	3	0.0037	2.8	0.3
8.87	13.02	0.093	6	0.0038	3.5	1.0
4.50	7.06	0.049	6	0.0036	2.7	0.3
2.28	3.71	0.025	6	0.0036	2.3	0.1
<i>Sample: BK060-02</i>						
<i>ODBW: 58.57 g/m² per sheet</i>						
82.62	58.51	0.707	3	0.0046	33.5	10.6
73.81	55.76	0.646	3	0.0045	29.0	9.7
65.14	52.66	0.583	3	0.0044	25.5	7.1
54.10	48.01	0.488	3	0.0044	23.2	6.8
41.80	41.64	0.380	3	0.0043	14.5	4.2
27.68	32.09	0.253	3	0.0043	10.4	3.5
14.40	19.73	0.153	3	0.0037	5.9	1.7
5.73	8.91	0.071	3	0.0032	3.1	0.2
8.36	12.37	0.106	6	0.0031	3.6	0.9
4.50	7.06	0.064	6	0.0028	3.0	0.3
2.41	3.91	0.034	6	0.0028	2.6	0.1
<i>Sample: UK205-1</i>						
<i>ODBW: 198.36 g/m² per sheet</i>						
381.92	65.87	0.569	1	0.0265	21.4	9.0
340.09	63.22	0.508	1	0.0264	19.0	7.6
308.30	60.91	0.468	1	0.0260	16.7	6.4
262.28	57.00	0.400	1	0.0259	15.0	5.8
207.48	51.19	0.324	1	0.0253	10.7	4.1
171.51	46.43	0.271	1	0.0250	8.6	3.3
125.08	38.73	0.203	1	0.0243	7.0	2.6
84.08	29.83	0.143	1	0.0232	5.0	1.7
47.27	19.28	0.092	1	0.0203	3.1	0.8
23.43	10.59	0.048	1	0.0194	2.1	0.2
30.78	13.36	0.062	2	0.0196	2.5	0.5
17.48	8.05	0.037	2	0.0188	2.0	0.2
9.50	4.54	0.020	2	0.0182	1.8	0.1
<i>Sample: UK205-2</i>						
<i>ODBW: 199.20 g/m² per sheet</i>						
492.35	71.25	0.793	1	0.0245	33.8	15.6
449.69	69.35	0.740	1	0.0240	30.9	13.3
408.27	67.26	0.683	1	0.0236	26.5	10.0
340.92	63.18	0.573	1	0.0235	19.9	7.5
280.27	58.52	0.477	1	0.0232	16.7	6.2
235.09	54.19	0.403	1	0.0230	13.4	4.9
192.01	49.14	0.337	1	0.0225	10.5	4.4
156.87	44.12	0.281	1	0.0220	8.9	3.2
109.18	35.46	0.202	1	0.0213	6.1	2.4
73.62	27.04	0.148	1	0.0197	5.0	1.8

TABLE II. (Continued from previous page.)

Water basis, wt/sheet (g/m ²)	Grav moist (%)	ρ_{H_2O} (g/cm ³)	No. sheets	Caliper per sheet (in.)	ϵ'	ϵ''
23.43	10.55	0.054	1	0.0172	2.3	0.2
30.59	13.27	0.070	2	0.0173	2.6	0.6
17.48	8.04	0.042	2	0.0166	2.2	0.2
9.31	4.45	0.023	2	0.0161	1.9	0.1
<i>Sample: UK205-3</i>						
<i>ODBW: 199.62 g/m² per sheet</i>						
355.49	62.75	0.607	1	0.0218	26.2	9.8
287.38	59.07	0.525	1	0.0216	20.6	7.5
240.53	54.71	0.444	1	0.0214	16.5	6.2
197.02	49.74	0.370	1	0.0210	12.8	5.4
143.90	41.95	0.276	1	0.0206	9.1	3.8
107.09	34.97	0.209	1	0.0202	7.1	2.9
61.91	23.72	0.130	1	0.0188	4.9	1.7
23.43	10.53	0.057	1	0.0163	2.5	0.3
30.21	13.11	0.076	2	0.0157	2.9	0.7
16.91	7.79	0.044	2	0.0150	2.2	0.2
8.74	4.18	0.024	2	0.0145	2.0	0.1
<i>Sample: UK205-4</i>						
<i>ODBW: 200.04 g/m² per sheet</i>						
407.85	67.15	0.732	1	0.0220	31.9	13.0
357.66	64.19	0.648	1	0.0218	28.7	9.5
314.99	61.22	0.578	1	0.0215	22.8	7.8
266.05	57.14	0.500	1	0.0210	19.5	6.9
221.71	52.63	0.427	1	0.0205	15.8	5.7
185.73	48.21	0.368	1	0.0199	12.8	5.0
144.74	42.04	0.298	1	0.0192	10.3	4.2
106.67	34.84	0.226	1	0.0186	8.4	3.1
68.18	25.47	0.151	1	0.0178	5.6	3.8
20.50	9.32	0.055	1	0.0147	2.5	0.2
30.78	13.32	0.092	2	0.0132	3.5	0.9
17.29	7.95	0.054	2	0.0126	2.6	0.3
9.12	4.36	0.029	2	0.0122	2.2	0.1
<i>Sample: UK205-5</i>						
<i>ODBW: 201.30 g/m² per sheet</i>						
381.08	65.49	0.671	1	0.0224	26.6	12.6
342.18	63.02	0.606	1	0.0223	25.6	9.4
299.09	59.83	0.537	1	0.0220	22.2	8.3
262.70	56.68	0.480	1	0.0216	19.1	7.1
209.57	51.07	0.398	1	0.0208	16.1	5.9
156.45	43.79	0.306	1	0.0202	11.3	4.1
113.78	36.17	0.230	1	0.0195	8.2	3.1
78.22	28.04	0.166	1	0.0186	6.5	2.5
46.85	18.92	0.112	1	0.0165	4.5	1.3
20.50	9.26	0.053	1	0.0154	2.4	0.2
30.40	13.14	0.084	2	0.0143	3.2	0.8
16.72	7.68	0.049	2	0.0135	2.4	0.3
8.55	4.08	0.026	2	0.0131	2.1	0.1

the cylinders, ϵ_c , the complex dielectric constant of the matrix, ϵ_0 , and the volume fraction of the cylinders, ρ by

$$\epsilon = [(3\rho - 2)(\epsilon_c - \epsilon_0) + [(3\rho - 2)^2(\epsilon_c - \epsilon_0)^2 - 8(\rho - 2)\epsilon_0\epsilon_c + 8\rho\epsilon_c^2]^{1/2}]/4 \quad (2)$$

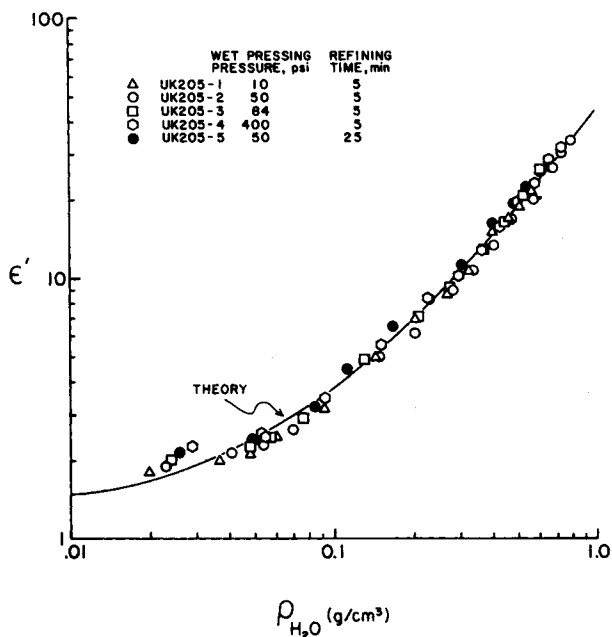


Fig. 2. The real part of the dielectric constant as a function of ρ_{H_2O} for unbleached kraft samples differing in wet pressure and refining. ρ_{H_2O} is the weight of water divided by the sample volume. Measurements are at 9.6 GHz. The theoretical curve is from eq. (2) with $\epsilon_c = 45.5 - 18.6i$ and $\epsilon_0 = 1.2 + 0.1i$.

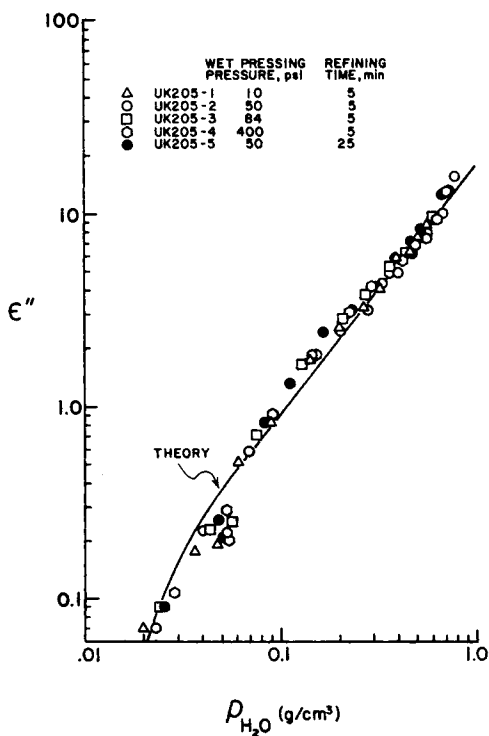


Fig. 3. The imaginary part of the dielectric constant as a function of ρ_{H_2O} for the unbleached kraft samples. Measurements are at 9.6 GHz. The theoretical curve is from eq. (2) with $\epsilon_c = 45.5 - 18.6i$ and $\epsilon_0 = 1.2 + 0.1i$.

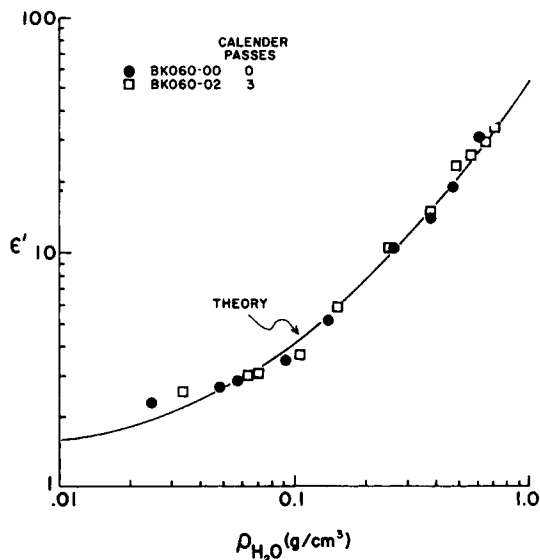


Fig. 4. The real part of the dielectric constant vs. $\rho_{\text{H}_2\text{O}}$ for the bleached kraft samples. The theoretical curve is from eq. (2) with $\epsilon_c = 50.7 - 17.0i$ and $\epsilon_0 = 1.4 + 0.1i$.

The values of ϵ_c and ϵ_0 were numerically optimized to obtain the curves giving the least variance on the log-log plots. The values of ρ were taken as numerically equal to the measured water density in g/cm^3 . The results of this exercise are shown in the figures. For UK samples 98.2% of the total variance in ϵ was accounted for by the best fit curve, while 97.0% was the corresponding number for the BK samples. Least square fits to straight lines were also performed; however, the error was approximately twice that of the eq. (2) curves and results are not shown.

The three major factors that determine the magnitude of the apparent dielectric constant of wet paper sheets are (1) the apparent density of the moisture, (2) the degree to which molecular bonding between the water and the fiber inhibits the ability of the water to interact with the microwave radiation, and (3) the geometric distribution of the water at the fiber level. The insensitivity of the dielectric constant vs. water density curves to wet pressing, refining, and calendering indicates that these processes have little effect on factors 2 and 3. The role of the second factor at high moistures can be estimated by extrapolating the theoretical curves to a water density of 1 and comparing this with the dielectric constant of pure water. (The third factor is probably not important in this case because the geometry of the water at the fiber level must become highly connected as water density approaches 1.) The result of this extrapolation for the unbleached kraft handsheets is $\epsilon \sim 45.5 - 18.6i$, while the dielectric constant of pure water is $55 - 30i$ at this frequency and temperature. For the bleached kraft samples the same extrapolation gives $\epsilon \sim 50.7 - 17.0i$. This is significantly different from the value for the unbleached kraft samples. In fact, the ϵ' curve for the bleached kraft samples is slightly above the unbleached curve over the range of water densities studied. We take these observations as evidence that the water molecules in the unbleached kraft samples are more strongly bound to the fiber than in the bleached kraft; i.e., they are not as free to oscillate and

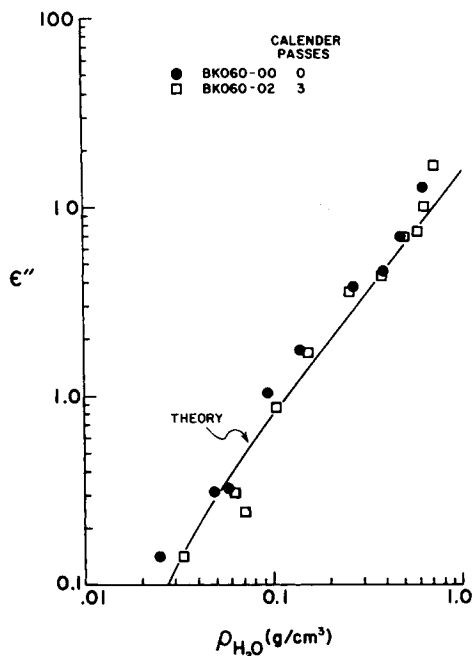


Fig. 5. The imaginary part of the dielectric constant vs. ρ_{H_2O} for the bleached kraft samples. The theoretical curve is from eq. (2) with $\epsilon_c = 50.7 - 17.0i$ and $\epsilon_0 = 1.4 + 0.1i$.

contribute to ϵ' . In addition, at the same relative humidity the bleached kraft contains less moisture than the unbleached kraft ($\sim 7\%$ vs. 8% at 50% RH), suggesting that some water in the unbleached sheet is associated with the lignin and hemicellulose material, which may be bound more tightly than in the case of the cellulose-water interaction.

As mentioned earlier, it is well known that the out-of-plane dielectric constant is smaller than that measured in plane. Dusoiu et al.¹¹ have also shown that there is a difference between the two principal in-plane directions in highly oriented sheets. We investigated the effect of changing the alignment of the electric field to the machine direction for machine-made papers and found that there was a small, but measurable, decrease in dielectric constant in going from the machine to cross-machine direction. Figure 6 gives a plot of dielectric constant vs. field orientation for a commercial newsprint sample. The values of ϵ' and ϵ'' are both larger in the machine direction, apparently due to the preferential alignment of fibers in that direction.

It has been reported that fillers and coatings^{18,19} have little effect on the total moisture content reading of commercial microwave moisture gages. One common filler, TiO_2 , has a real dielectric constant that is very large, $\epsilon \approx 117$. It might be expected, therefore, that TiO_2 would have a large influence on microwave moisture gages, especially those whose sensitivity depends on the real part of the dielectric constant of the sheet. We investigated the role of TiO_2 on the dielectric constant by making measurements on bleached kraft handsheets with varying levels of TiO_2 . Figure 7 shows the results of those measurements, all at 50% RH. The increase in ϵ' due to the TiO_2 filler is small when compared with

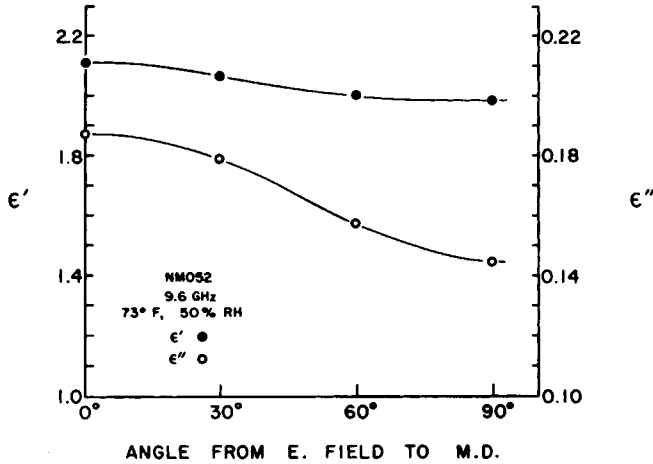


Fig. 6. Dielectric constant vs. orientation of electric field for a machine-made newsprint sample.

the increase caused by the addition of an equal weight of water. The higher density of TiO_2 (4.26 g/cm^3) compared with water explains some of the lack of sensitivity. In addition, the more highly doped sheets have a lower water density, and this compensates for the addition of TiO_2 . This lower water content explains the small decrease in ϵ'' . The volume addition of TiO_2 is about 3.5 times the volume loss of water and TiO_2 has a dielectric constant twice that of water at this frequency. Thus, a significant increase in the dielectric constant of the sheet is expected due to the addition of the TiO_2 . This increase is not observed.

The contribution of any component to the dielectric constant of a mixture depends upon its dielectric constant, volume fraction, and geometry. The in-

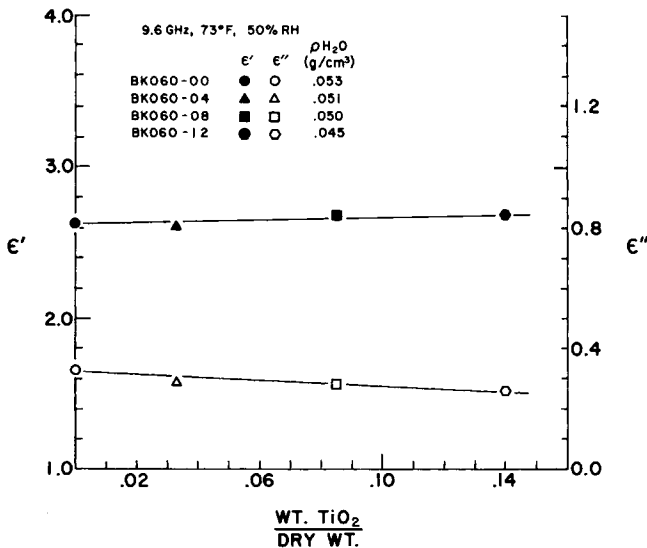


Fig. 7. Dielectric constant vs. TiO_2 content.

sensitivity of the dielectric constant to TiO_2 addition is probably due to the geometry of the TiO_2 , which exists in isolated clumps and particles on the fiber surface. This is a disconnected topology that results in a low effective dielectric constant [near the minimum of eq. (1)]. Water, however, is distributed throughout the fiber, giving it a more connected topology and making it much more effective in increasing the in-plane dielectric constant.

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