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Measurement of the Dielectric Properties of Paper

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Unlike dc resistivities of non-impact printing papers that are routinely measured, their dielectric properties are seldom considered. The dc resistivity alone does not predict the behavior of paper very well when paper passes through a transfer nip. If the electric field changes with time, all anisotropic frequency-dependent electrical properties of paper should be considered. We present a new method for the measurement of the electrical properties of paper in the principal directions of the sheet. In the frequency range of 20 Hz–1 MHz, the permittivity ε' and dielectric loss factor ε" or electrical conductivity σ can be measured. Measurements on coated and uncoated commercial papers showed some interesting characteristics. In spite of the low moisture content of coated paper, its dielectric constant increased with increasing relative humidity at the same rate as in copy paper. The dielectric constant of copy paper measured in the machine direction (MD) was much higher than what was measured in the thickness direction (ZD). For coated paper the difference between MD and ZD dielectric constants was small. For both types of paper the dielectric loss was much greater in machine direction than in the thickness direction. Dielectric loss increased with increasing relative humidity more in machine direction than in z-direction. Differences in the MD and ZD dielectric constant were attributed to the anisotropy of paper.

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Introduction

In the transfer step of electrophotographic printing, paper is polarized by a strong electric field and the oppositely charged toner is attracted to the paper surface.[1] The electric field is usually applied using a high voltage corona or a bias roller. With increasing printer speed, the dwell time of paper in the transfer zone decreases and dynamic properties of paper become important.

Of the electrical properties that characterize paper, (dc) surface resistivity is the one most extensively used.[2] However, dc resistivity is not well defined and it poorly represents the electrical behavior of paper. Even though several authors mention the significance of permittivity (dielectric constant) to the toner transfer efficiency,[3–7] the dielectric properties of non-impact papers have not been studied. To fully account for the toner transfer efficiency, one should consider both the permittivity and the resistivity of the paper. As an example, transfer efficiency has been found to correlate better with charge decay rate (which includes the effect of permittivity) than with resistivity alone.[3] Furthermore, the paper industry has not yet adopted any measurement method to serve this purpose.

In this article, we present a method for the measurement of the dielectric properties of paper. The effects of frequency and moisture content on the dielectric properties of commercial papers are presented.

Dielectric Properties of Paper

Permittivity ε describes the polarizability of the material in an external electric field. Because the values of permittivity are of the order of 10^{-11} F/m, it is more convenient to use the dielectric constant κ which is the ratio of the permittivity of the material and the permittivity of vacuum $\kappa = \varepsilon/\varepsilon_0$. Values of the dielectric constant range from 2 for most polymers up to 10^3 to 10^4 for ferroelectric materials such as Barium Titanate.[8] The dielectric constant of dry paper is typically 2 to 4, but it can be higher depending on density, filler content, fiber furnish, etc.

When an external electric field is applied to a dielectric, the charges in the dielectric are displaced so as to reduce the electric field inside the material. Positive charges of non-polar molecules move in the direction of the applied field and negative charges in the opposite direction. The displacement of charges depends on the forces opposing the movement and on the strength of the applied electric field. If there are polar molecules, their electric dipoles tend to align with the direction of the field. This further increases the polarization of the material. The larger the permittivity of the material, the stronger is the polarization.[9]

Permittivity is frequency dependent. As frequency increases, slow polarization mechanisms cannot keep up

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with the rapidly changing electric field and the polarization mechanism relaxes, i.e. it no longer contributes to the polarization. This reduces the dielectric constant. If there is a well-defined relaxation frequency for the polarization mechanism, it causes a drop in the dielectric constant spectrum and a corresponding peak in the dielectric loss spectrum.[10] Due to the large number of different polarization mechanisms that exist in paper, no well-defined relaxation frequencies are seen at sub-optical frequencies.

Dielectric properties are affected by the moisture in paper. The dielectric constant of water is 78.4, over one order of magnitude larger than that of paper.[8] This is not surprising considering that water molecules have a permanent dipole moment. However, the water molecules in paper are neither free to move around nor free to choose their orientation. This means that their effective dielectric constant is much smaller than that of free water and depends on the orientation of the electric field.[11] A water molecule attached to a cellulose chain is relatively free to orient itself parallel to the field only if the field is parallel to the cellulose chain. If the field is perpendicular to the chain, the orientation is inhibited by the presence of the chain. Because cellulose chains in paper point in different directions, only a small fraction of the water molecules can align perfectly with the electric field. Subsequently, the contribution of water to the dielectric constant of paper is smaller than could be expected if the water molecules were free.[12] The dielectric constant depends also slightly on temperature. In general, a temperature rise will decrease the restraints of the dipoles causing an increase the dielectric constant. However, in paper the effect is very small in the temperature range of interest (only 5% in the range 20 to 120°C) and it can usually be ignored.[13]

The dielectric constant of paper is greater along the sheet plane than normal to the sheet plane (in the z-direction). In addition, in sheets with fiber orientation the dielectric constant has a maximum at the direction of the fiber orientation angle. Earlier work shows that dielectric anisotropy in paper can be attributed to fiber orientation without having to assume that the fiber itself is anisotropic.[14] This fact can be used in the measurement of fiber orientation angle and strength.

The dielectric constant of paper increases with increasing density. The density dependence of the dielectric constant is given with reasonable accuracy by the Clausius-Mossotti relation[15]

$$\frac{\kappa' - 1}{\kappa' + 2} \propto \rho_d \qquad (1)$$

where κ' is the real part of the complex dielectric constant and ρ_d is the density of paper. Fillers affect the dielectric properties of paper in two ways: First, they increase the density of paper, and second, dielectric constants of filler materials are higher than the dielectric constant of wood fibers, e.g., the dielectric constant of a typical filler material, $CaCO_3$, is 8.3 to 8.7 at 94 GHz depending on the orientation of the field.[8] Fillers should therefore increase the dielectric constant of paper.

Measurement of Dielectric Properties

The sensor head is shown in Fig. 1. It employs two sets of interdigital electrodes that are carefully aligned so that they are located exactly opposite to each other. Various interdigital electrodes have been used earlier to study not only the dielectric properties of materials but also for monitoring thickness, mechanical properties, moisture dynamics, etc.[16–21] Our device employs the same principle of measuring the impedance between interdigital electrodes although it was not designed on the basis of a particular existing device.

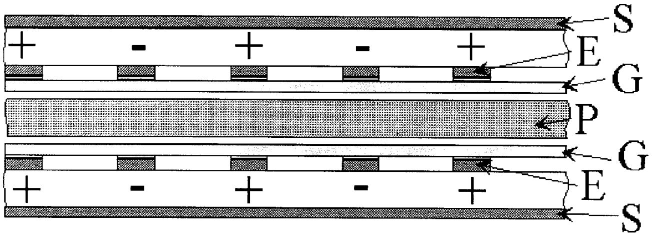

Figure 1. Partial cross-sectional view of the sensor head of the dielectric analyzer DIANA. The copper Electrodes (E) are machined on printed circuit boards whose other side's copper layers (S) act as guards. The paper (P) is inserted between two glass plates (G). The width of the electrodes is 200 μm and their center-to-center distance is 600 μm. The thickness of the copper electrodes and the glass plates are 40 and 30 μm, respectively.

The paper is placed between two printed circuit boards with a set of interdigital electrodes on each. The length of the electrodes and their lateral extent are such that the sensor measures the electric properties averaged over an area of 41 × 36 mm^2. Interdigital electrodes on the boards can be connected with relays in two ways: 1) The electrodes of one board are connected in parallel and the impedance between the electrodes of the boards is measured. In this configuration the electric field points through the paper and thus parameters in z-direction (perpendicular to the sheet plane) are measured. 2) The electrode of one board is connected to the facing electrode of the other board. In the impedance measurement, the electrodes from two sets are connected to opposite potentials. This connection measures the in-plane electric properties of the paper.

Equipotential lines for both electrode settings are illustrated in Fig. 2. They were calculated with a two-dimensional Finite Element Method (FEM) simulation program (Maxwell 2D Parameter Extractor, Ansoft Corp.) to verify that the direction of the electric field inside paper is correct. The direction and strength of the electric field is defined by the equipotential lines—the direction of the field at a given point is perpendicular to the equipotential line and the field strength is given by the line density. From Fig. 2 we see that the portion of the electric field penetrating to paper is indeed pointing to the right direction, especially in those areas where the field in the paper is the strongest. Even though the simulated electric field inside paper looks almost unidirectional in both x- and z-direction, the measurements are not completely independent. This is mainly due to the finite conductivity and dielectric anisotropy of paper both of which cause deviation from the calculated field in Fig. 2. In fact, in papers with strong fiber orientation the z-directional dielectric constant depends to a small extent on the sheet orientation.

The impedance between the electrode sets is measured with a Hewlett-Packard Precision LCR Meter 4284A in the C_p–R_p mode. The measurement is controlled with a PC microcomputer. The PC sets the relays in z-direction (ZD) and machine direction (MD) configuration and the impedance at predetermined frequencies is measured. The bias voltage is 1 V.

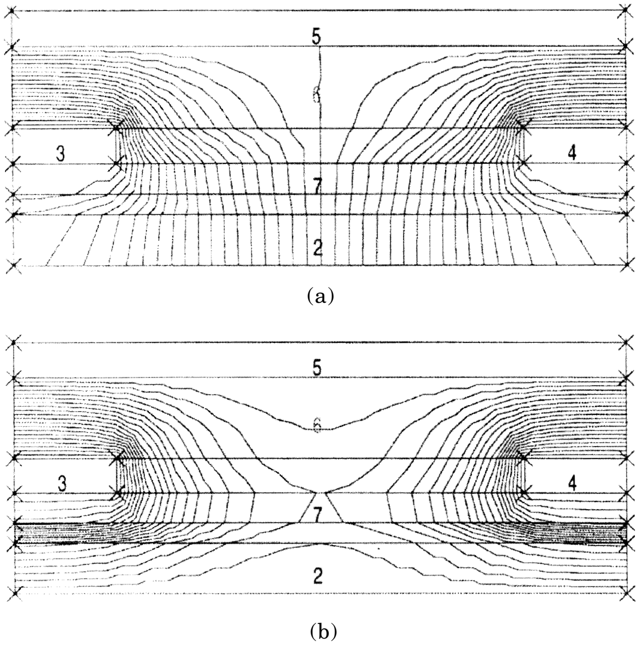

Figure 2. Equipotential lines calculated with FEM for two cases: (a) neighboring electrodes at opposite potential and (b) neighboring electrodes at the same potential. For symmetry reasons the lower half has been omitted. The labels are: 2 = top half of the paper (κ' = 3), 3–5 = copper, 6 = insulation, 7 = glass (κ' = 4).

TABLE I. Standard Errors ($\sigma^2/\sqrt{N}$) of Dielectric Constant κ_x' and Dielectric Loss Term κ_x" for Two Paper Samples at Different Frequency and Relative Humidity.

Sample/Humidity	Standard error (%)			
	κ_x'		κ_x"	
	100 Hz	1 MHz	100 Hz	1 MHz
Coated paper				
20%	2.9	2.6	3.6	2.9
50%	5.3	4.1	6.4	4.7
Copy paper				
20%	4.6	3.2	7.6	4.1
50%	9.6	5.2	39.4	6.6

Measurements were performed for six different sheets of each sample, i.e., the sheet-to-sheet variability of the samples is included in the error estimate

TABLE II. Dielectric Constants of Known Polymers Measured in the Thickness Direction and the Corresponding Literature Values.

Polymer	Specimen Thickness (μm)	Dielectric Constant Measured	Dielectric Constant Literature	Frequency (Hz)
PE-HD	104	2.19	2.3	10^3
PE-LD	210	2.34	2.3	10^3
PP	30	2.76	2.3	10^3
		2.72	2.3	10^6
Mylar®	193	3.65	3.25	10^3
		3.37	3.0	10^6
Kapton®	127	3.79	3.9	10^3

The comparison frequency is indicated. All values are from Ref. 8, except Kapton®, which is from Ref. 22.

The sensor is calibrated with the help of a PTFE (Teflon) sheet of known thickness. First the impedance of the empty sensor is measured. Then the PTFE sheet is inserted and the impedance is measured again. The calibration measurement is done separately in the sheet plane and normal to the sheet plane, i.e., both relay settings have their own calibration parameters. In the frequency range used in this study, the dielectric constants of air and PTFE are 2.10 and 1.00, respectively. Both are also practically perfect insulators. These values are then used by an interpolation algorithm to calculate the dielectric constant and dielectric loss of the samples by using the measured impedance and sample thickness values.

Any air gaps in a dielectric measurement cause serious deterioration of its reliability. This is handled by the measurement program by taking the air gap into account in the calculation. For this reason the thickness of the sample has to be inserted into the calculation. Basis weight and thickness do not have intrinsic effects on the results. This was verified by measuring papers in a basis weight range of 70 to 250 g/m^2 having the same mineral content and density. No basis weight (or thickness) dependency was found. Paper roughness and formation could pose a problem but the area of the electrodes is large enough to average the result to be independent of local variations. In the experiments we measured six sheets of each sample. We found that for commercial samples the sheet to sheet variation did not exceed the variation found in a single sheet.

The available measurement frequency range is limited by the LCR meter to 20 Hz–1 MHz. At high relative humidity the standard deviation at low frequency was greater than at high frequency (cf. Table I). More importantly, measurements at low frequency at a lower relative humidity than measurements at a higher frequency. This can be attributed to the calibration procedure which assumes that the dielectric constant is much larger than the dielectric loss term. If the dielectric loss term becomes comparable to the dielectric constant the electric field does not penetrate into paper as much as it would in the case of an insulator and the relation of the capacitance to the dielectric constant is not the same as during calibration. This poses a limit to the highest relative humidity at which the measurement can be performed reliably. In practice the limit is 50% RH for copy papers. Low frequency means high dielectric loss and consequently 1 MHz was chosen as the measurement frequency in studies of moisture content effects.

A comparison of measurement results to literature values of known polymer materials is given in Table II. The measured values coincide fairly well with literature, except for PP (polypropylene) which was by far the thinnest sample. The greater the thickness of the air gap between the specimen and the electrodes, the poorer the accuracy of the calibration procedure. The frequency dependence of the dielectric constant of PP seems to be approximately correct (practically no change from 1 kHz to 1 MHz), indicating that the difference is probably due to calibration, not erroneous functioning of the measurement device.

A commercial device (JCI 155 Charge Decay Test Unit) was employed in the charging capacity and charge decay time measurements of Figs. 7 and 8. In this method, the paper is charged for a predetermined time with a high voltage corona. After charging, the corona wires

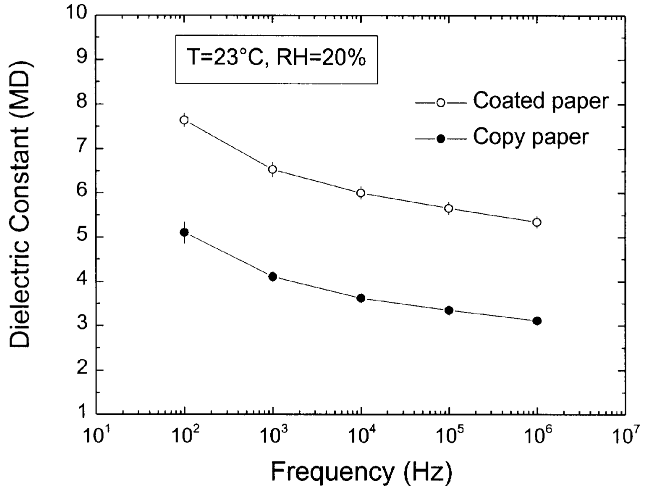

Figure 3. MD dielectric constant vs. frequency for a 130 g/m^2 coated paper and an 80 g/m^2 copy paper at 20% RH.

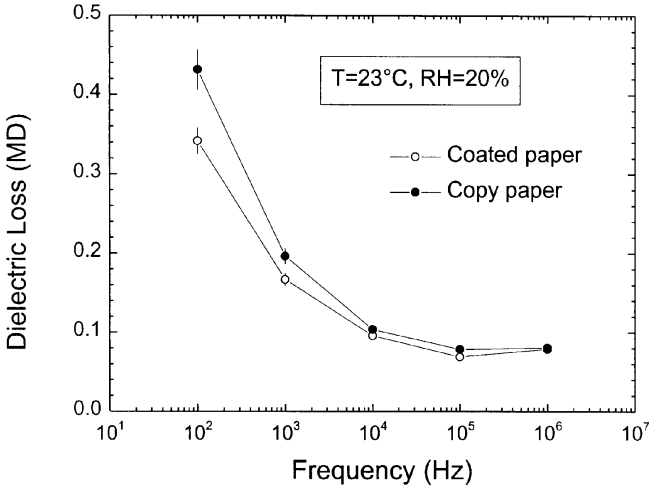

Figure 4. MD dielectric loss versus frequency for the coated paper and copy paper of Fig. 3 at 20% RH.

are rapidly moved aside revealing a high speed electrostatic field meter and the measurement starts. Charging capacity is the maximum value of the measured electrostatic potential versus time curve and charge decay time is the time elapsed between the peak voltage and its decay to half of the peak value.[23,24] The charging voltage used was 6.0 kV with 7.0 s charging time.

All measurements were performed in a humidity chamber where the relative humidity of air could be controlled within ±0.5%. The temperature of the chamber was held to 23.0 ± 0.5°C at all times. Each sample was conditioned in the measurement humidity for at least 12 h. In order to avoid hysteresis in the moisture content the samples were conditioned for 24 h at 10% RH before the beginning of the experiments.

Results and Discussion

We studied three triple coated papers with basis weight of 130 g/m^2, one coated paper of 80 g/m^2 with a single coating layer, and five uncoated woodfree copy papers with different basis weights ranging from 80 to 100 g/m^2. The filler content of the triple coated 130 g/m^2 samples was 40 wt.%. The filler content of the 80 g/m^2 coated paper was 30 wt.% while the filler contents of the copy papers varied between 18 and 22 wt.%. In the coated grades approximately half of the filler material was in the coating layer. $CaCO_3$ was the primary filler material or coating mineral in all samples. Only small traces of other filler materials were found in the samples so that we can safely assume that for all practical purposes the filler affecting the dielectric properties is $CaCO_3$.

The dielectric constant and dielectric loss factor of each sample was measured as a function of frequency in the machine direction (MD) and thickness direction (ZD). Figure 3 shows the MD dielectric constant of a coated paper and a copy paper against frequency at 20% RH. The dielectric constants of both samples decrease as the frequency increases. This is an indication of a relaxation process or several relaxation processes taking place in this frequency range. The coated paper has a higher dielectric constant than the copy paper. This is probably due to the higher $CaCO_3$ content (40.2% versus 21.3%) and higher density (1320 kg/m^3 versus 750 kg/m^3) of the coated grade. No direct relation between dielectric constant and filler content or density was found for our set of samples although the dielectric constant of samples with high filler content was generally high. This suggests that filler content increases the dielectric constant but does not alone determine it.

Figure 4 shows that dielectric loss decreases with increasing frequency for both the copy paper and the coated paper. For any material with finite electrical conductivity the apparent complex permittivity is given by[26]

$$\tilde{\varepsilon}(\omega) = \varepsilon'(\omega) - i\left[\varepsilon''(\omega) + \frac{\sigma_0}{\omega}\right] \qquad (2)$$

where ω is angular frequency and σ_0 is the dc conductivity. The dielectric loss given by the measurement is the imaginary part of Eq. 2 and thus depends on both the electrical conductivity and the polarization losses of the dielectric. For low conductivity materials like paper, electrical conduction is significant only if the frequency is low, i.e., the conductivity term of the imaginary part is proportional to the dielectric loss. The curves in Fig. 4 are not directly proportional to the reciprocal of the frequency (cf. Eq. 2) indicating that conduction is not a significant mechanism in the whole frequency range of 100 Hz–1 MHz.

We see from Fig. 4 that dielectric loss is higher in the copy paper than in the coated paper. One of the possible explanations could be the higher moisture content of copy paper (due to lower filler content). On the other hand, the dc conductivity of copy paper is also higher indicating that there are more impurities such as metal ions in the copy paper. Ionic and other impurities tend to increase the dielectric loss.[11] The values of the dc surface resistivity (average of both sides) measured with ASTM method D257-93 (100 V, 30 s charging) at 20% RH show that the copy paper is more conductive than the coated paper. The resistivity values were 1.7×10^{12} and 2.8×10^{13} Ω for the copy and the coated paper, respectively.

Figure 5 shows the effect of relative humidity of air on the dielectric constant of paper. The dielectric constant increases slowly as a function of relative humidity. The behavior of the samples resembles data reported previously for electrical papers.[25] The dielectric constant of the coated paper is higher than that of copy paper over the whole relative humidity range. The

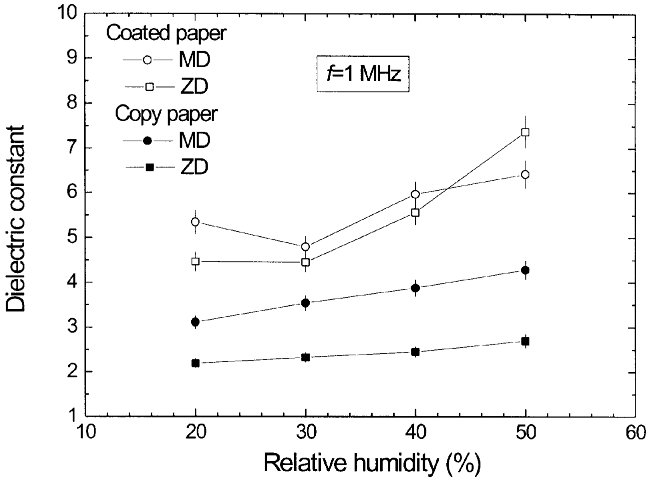

Figure 5. MD and ZD dielectric constants of the coated paper and copy paper of Fig. 3 as a function of moisture content (f = 1 MHz).

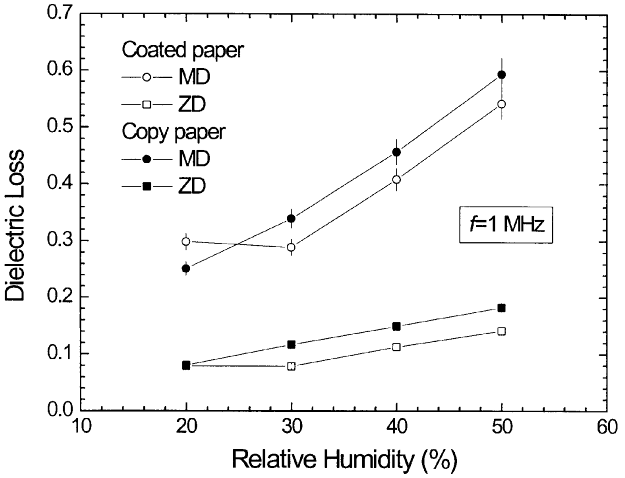

Figure 6. MD and ZD dielectric loss of the coated paper and copy paper of Fig. 3 as a function of moisture content (f = 1 MHz).

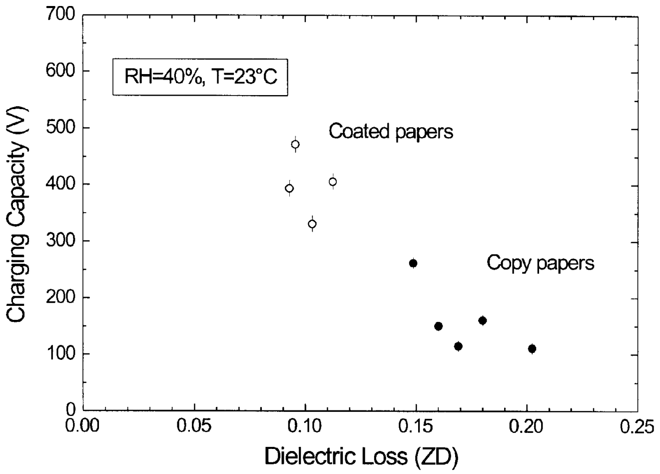

Figure 7. Charging capacity (maximum voltage) of all measured paper samples as a function of ZD dielectric loss (f = 1 MHz).

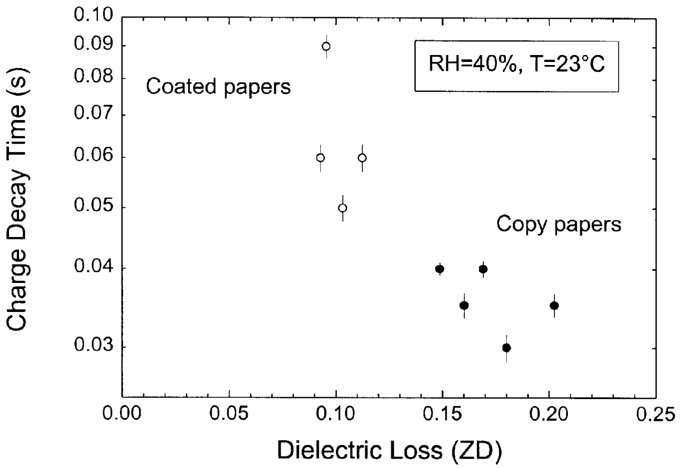

Figure 8. Charge decay time of all measured paper samples as a function of ZD dielectric loss (f = 1 MHz).

relative humidity dependence of the ZD dielectric constant is stronger in coated paper than in copy paper. Because the moisture content of the fibers changes in the same way in both papers the difference should come from the layered structure of the coated paper. It could be that the coating layers enhance polarization in the z-direction more at high relative humidity. Support to this theory is given by the fact that the MD dielectric constant increases at the same rate for both samples. However, the difference is too small to be conclusive.

Dielectric loss is plotted versus relative humidity in Fig. 6. Dielectric loss is higher in the copy paper but the relative humidity dependence is the same for both papers. This indicates that dielectric loss is related to water in the fibers. It should be noted however that free water would not contribute to dielectric loss at as low a frequency as 1 MHz, but the dielectric relaxation behavior of water molecules change when they are at the fiber surface.[10]

Contrary to theoretical considerations, charging capacity and charge decay time were found to be independent of the dielectric constant.[24] This can attributed to the fact that charge decay time depends on resistivity as well as on the dielectric constant. Because resistivity differences between samples are much greater than differences in the dielectric constant its effect is overshadowed by the effect of resistivity. However, there is a relationship between charging capacity, charge decay time and ZD dielectric loss as can be seen in Figs. 7 and 8.

Charging capacity measurements cannot be translated directly into transfer efficiency values because the dynamics of practical printers are very different from the measurement. Whether the value of the charging capacity has any reflection on transfer efficiency or not depends on a number of parameters, such as printer design, paper velocity, relative humidity of air, etc.

Summary

In this article, we presented a new device for measuring the dielectric properties of paper. We also presented the first published comparison of the dielectric properties of coated and uncoated digital printing papers.

Our measurement device employs a set of interdigital electrodes on each side of the paper and can measure the dielectric constant and dielectric loss in both the sheet plane and normal to the sheet plane in the frequency range of 20 Hz-1 MHz. Dielectric constants measured for known polymers showed that the measurement yielded realistic values except for very thin sheets. The standard error in the measurement varied with frequency and moisture content from 3 to 10%. Statistical variation increased with decreasing frequency and increasing relative humidity.

The dielectric constant of coated paper was higher than that of copy paper is due to the higher filler content and greater density of the coated paper. However, no distinct trend could be established between the dielectric properties and the filler content and density of paper. The dependence of the dielectric constant and dielectric loss on relative humidity was found to be same for copy paper and coated paper. This indicates that moisture related polarization mechanisms are due to water molecules in fibers only.

There is a difference in dielectric constant and dielectric loss whether it is measured in the machine (MD) or thickness (ZD) direction of paper. In all samples the MD dielectric constant was greater than the ZD dielectric constant. Even bigger differences were found for the dielectric loss values. This difference can be attributed to the structural anisotropy of paper. However, in the case of coated paper the difference between MD and ZD dielectric constants was small indicating that a layered structure might enhance polarization.

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