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Low frequency dielectric spectroscopy characterization of microcrystalline cellulose, tablets and paper

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Cellulose fibres and particles in the form of powder, tablets and paper sheets have been investigated by very low frequency dielectric spectroscopy using a novel form of dielectric cell, in which two planar electrodes have been mounted in fixed positions at right angles. The broad pattern of response obtained from the samples is independent of the structural form of the cellulose sample, a loss peak in the 0.1–100 Hz range, and at lower frequencies a dispersion process which is dominated by an imperfect charge transport. Moisture has a significant influence on the rate of charge transport. In dry samples the dipolar loss peak was not evident but as the moisture content increased it appeared. Using a humidity normalizing technique the dielectric response for microcrystalline cellulose has been characterized over the equivalent of 14 decades in frequency. It has also been shown that there is a linear response between the capacitance and the density of microcrystalline cellulose samples. The consolidation of powder into tablets is discussed with respect of the observations of changes in capacitance, loss peak frequency and imperfect charge transport efficiency. Furthermore it was found possible to investigate differences between the dipolar relaxation rate ''in'' and ''out'' of the plane of paper in the stack. The relaxation time for dipoles ''out'' of the paper plane is 7 to 8 times longer than for dipoles ''in'' the paper plane.

1. Introduction

The technique of low frequency dielectric spectroscopy has been applied to a number of problems of pharmaceutical interest [1–4]. The present investigation has used a common tabletting excipient, microcrystalline cellulose, and used dielectric spectroscopy to investigate the properties of the material in a number of forms, dry and wet powder, tablets formed under a range of pressures and stacked paper sheets. It has been observed that the essential features of the dielectric response are maintained but that there is a change in the frequency and magnitude of the individual components of the response. In Fig. 1 we present a typical dielectric spectrum of the cellulose material. It can be seen that there are three separate mechanisms in the response. At the highest frequencies the loss component is dominated by the lead inductance of the cell, and is not of significance to us; in the frequency region between 10–100 Hz there is a loss peak dispersion which is indicative of a dipolar response and at the lowest frequencies the parallel dispersion with fractional exponent is characteristic of an imperfect charge transport process [5]. Hence we can obtain information about both charge storage, from the dipolar response, and charge transport, from the low frequency dispersion, as separate features. The measuring range covers ten decades of frequency from 10^{-4} to 10^{6} Hz and the results are presented in terms of the real, $C'(\omega)$, and imaginary, $C''(\omega)$, components of the complex capacitance $C^*(\omega)$ in the range between 0.1–10^3 pF. The complex capacitance can be expressed as

$$C^*(\omega) = C'(\omega) - iC''(\omega) \tag{1}$$

and $i = (-1)^{1/2}$ and $\omega = 2\pi f$ with f being the frequency of the applied electrical field in Hz. The complex capacitance of the sample is a product of its relative complex permittivity and the capacitance of the empty sample holder. Hence the influence of the sample holder is eliminated in the complex permittivity $\varepsilon^*(\omega)$ by dividing the measured complex capacitance $C^*(\omega)$ at frequency ω by the frequency

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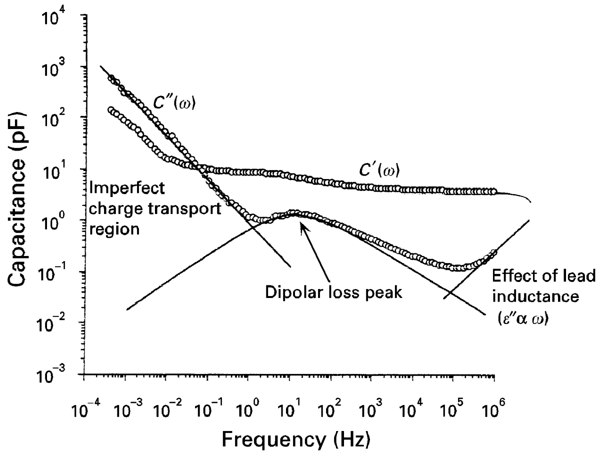

Figure 1 Dielectric spectra of microcrystalline cellulose (equilibrated over saturated K_2CO_3 salt solution). The two different dispersion mechanisms contributing to the total dielectric response are indicated, as is the inductive effect of the 0.5 m long sample leads.

capacitance of the empty sample holder C_0.

$$\varepsilon^*(\omega) = \frac{C^*(\omega)}{C_0} \tag{2}$$

We shall in this work make use of two simple models in the analysis of the dielectric response, the constant phase angle [5, 6] and the Debye model [7]. At the lowest frequencies the imperfect charge transport process is characterized by the constant phase angle response which is seen in Fig. 1, as the parallel capacitance components region. A general description for this process, which commonly appears in structurally complex materials without a clear defined structure, is the fractally scaling model [8–11], which is expressed in terms of the permittivity ε at frequency ω as:

$$\varepsilon\left(\mathrm{i}\frac{\omega}{\omega_c}\right) = \alpha\,\varepsilon\left(\mathrm{i}\beta\frac{\omega}{\omega_c}\right) \tag{3}$$

where α and β are the scaling of the magnitude and frequency respectively and the frequency is expressed in terms of a characteristic value, ω_c. A solution of this equation is

$$\varepsilon(\omega) \propto \left(\mathrm{i}\frac{\omega}{\omega_c}\right)^{-s} \text{ with } s = \frac{\ln\ \alpha}{\ln\ \beta} \tag{4}$$

where s is positive and fractional. From Equation 4 it can be deduced that:

$$\frac{\varepsilon''(\omega)}{\varepsilon'(\omega)} = \tan\left(\frac{s\pi}{2}\right) \tag{5}$$

where the constant phase angle, $s\pi/2$, is the characteristic of imperfect charge transport. We note that d.c. conduction occurs when $s \to 1$ since this gives a purely imaginary response which is the feature of a conductance, G, with $\varepsilon''(\omega)\varepsilon_0 = GI_f/\omega$. where I_f is the instrumental factor in units of m^{-1}. Alternatively Debye's model and variants of it, describe the rotational displacement of a spatially fixed dipole returning to equilibrium under the action of an over damped viscous force. In terms of permittivity the Debye model is characterized by the dispersion:

$$\varepsilon^*(\omega) - \varepsilon'(\infty) = \varepsilon_0\left(\frac{1}{1+\omega^2\tau^2} - \mathrm{i}\frac{\omega\tau}{1+\omega^2\tau^2}\right) \tag{6}$$

where ε_0 is the permittivity of the free space $= 8.854 \times 10^{-12}\ \mathrm{F\ m^{-1}}$ and τ is the relaxation time. This response is seldom seen except for very dilute dispersions of a dipole substance in a non-polar medium. In most cases the loss peak is broader than the Debye value but the relaxation time continues to be characterized by the frequency of maximum loss.

It is also possible to define a relaxation time for the constant phase angle response by making use of the crossover point at the upper frequency limit of the response. Taking this frequency as ω_x we have $C'(\omega_x) = C''(\omega_x) = G/\omega_x$, i.e., $\omega_x = G/C'(\omega_x) = 1/(RC'(\omega_x)) = 1/\tau_x$, where R is the resistance of the sample.

2. Experimental procedures

2.1. Dielectric technique

2.1.1. Sample holder

The sample holder was constructed with the two plane metal electrodes being at a 90° angle as is shown in Fig. 2. Thin brass electrodes were glued onto borosilicate glass microscope slides of high electrical resistance. The dimensions of the electrodes were 15 × 76.2 mm. At the bottom of the V, a narrow 1.3 mm wide gap was left between the electrodes as is shown in Fig. 2. The capacitance of the empty sample holder cell was measured to be 2.55 pF.

A model describing the sample holder was derived by assuming that a cylindrical field existed between the electrodes, as is shown in Fig. 2. Then, the electrical field E_r at a distance r from the point of electrode intersection is:

$$E_r = \frac{2U_0}{\pi r} \tag{7}$$

for an applied voltage of U_0. According to Gauss' theorem the electrical field can also be written as a function of the surface charge density on the plates ρ_r:

$$E_r = \frac{\rho_r}{\varepsilon\varepsilon_0} \tag{8}$$

so that

$$\rho_r = \frac{2\varepsilon\varepsilon_0 U_0}{\pi r} \tag{9}$$

The total plate charge Q on the electrodes is then:

$$Q = \int_0^h \int_{r_0}^{r_m} \frac{2\varepsilon\varepsilon_0 U_0}{\pi r}\,\mathrm{d}h\,\mathrm{d}r = h\,\frac{2\varepsilon\varepsilon_0 U_0}{\pi}\int_{r_0}^{r_m}\frac{\mathrm{d}r}{r} \tag{10}$$

where r_0 and r_m are the minimum and maximum radii respectively and h is the length of the electrodes. Hence the capacitance C of the sample holder is given in terms of the dimensions of the sample holder as:

$$C = \frac{\mathrm{d}Q}{\mathrm{d}U_0} = \frac{2\varepsilon\varepsilon_0 h}{\pi}\ln\frac{r_m}{r_0} \tag{11}$$

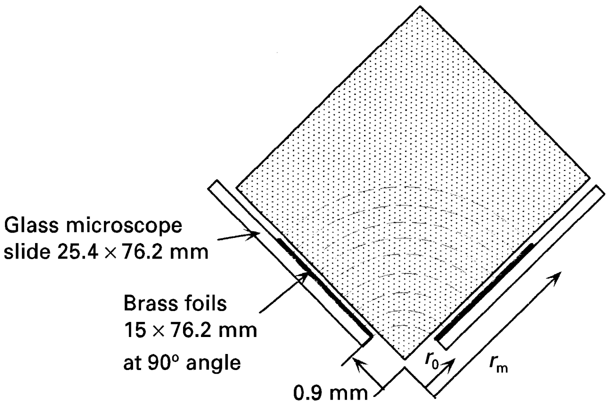

Figure 2 A sketch of the sample holder with the plane electrodes mounted at a right angle forming a V in which the sample was placed. The thin brass foil electrodes were glued to two highly insulating borosilicate glass slides. The empty cell capacitance was measured to be 2.55 pF. The dashed radial curves indicate the electrical field lines used in the deduction of Equation 10 which predicts an empty cell capacitance of 1.23 pF.

with the instrumental function $I_f = 2h/\pi \ln r_m/r_0$. By entering the dimensions of the sample holder; $r_m = 15.9 \times 10^{-3}$ m, $r_0 = 0.9 \times 10^{-3}$ m, $h = 76.2 \times 10^{-3}$ m, $\varepsilon_0 = 8.854 \times 10^{-12}$ F m^{-1}, $\varepsilon = 1$; we obtain a theoretical sampleholder capacitance of 1.23 pF, approximately half of the measured value.

2.1.2. Equipment and measurements

The equipment, a PC V Opus computer, a 1255 HF frequency-response analyser (Solatron Instruments) and a Chelsea interface and MP 3200 *xy* plotter (Graphtech Corp.) were supplied by Dielectric Instrumentation Ltd. UK. The measurements were performed using a reference technique (reference capacitance = 10.104 pF and a reference conductance = 1.200×10^{-11} mho). Four to twelve measurements were made per decade of frequency. A 1 $V_{r.m.s.}$ signal was applied to the sample, measurements with a 0.1 $V_{r.m.s.}$ signal were also taken and the results were equivalent to that obtained with the 1 $V_{r.m.s.}$ signal but with a higher noise level. The total measuring time for the spectra containing twelve measuring points per frequency decade varied between hours (frequencies down to 10^{-2} Hz) to several days (frequencies down to 10^{-4} Hz).

2.2. Materials

Microcrystalline cellulose powder (Avicel PH 101, FMC, Ireland, lot 6240) was used either "as delivered" or after being pressed in a squared die of size 20 × 20 mm, with a hydraulic press, to give tablets with densities of 497, 600, 794, 976 and 1228 kg m^{-3}; the density of a single cellulose particle is 1570 kg m^{-3} and this is the maximum value possible. During compression the upper punch moved downwards towards the static plane. Lactose, a different tablet excipient, (Pharmatose, DMV, Netherlands) was used "as delivered". The volume between the electrodes ≈ 16 cm^3, was completely filled with the sample of loose powder, tablets or paper.

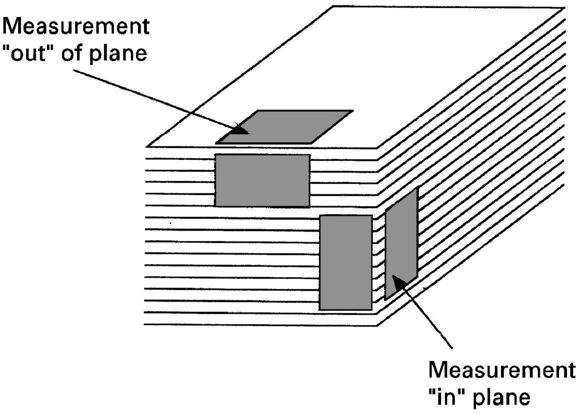

Figure 3 The application of the electrodes on a stack of paper to obtain "in" and "out" of plane measurements. On the stack of paper there are 4 edges giving "in" plane measurements and 8 edges giving "out" of plane measurements.

The paper, arranged into a stack 8 cm high of 8 × 8 cm paper sheets, (80 g m^{-2}, Datacopy sheets, Mo och Domsjš, Sweden) was placed in all of the twelve possible orientations in the holder. This gave 4 different edges "in" the paper plane and 8 edges "out" of the plane, Fig. 3.

2.2.1. Conditioning the samples

The samples were stored for at least a week in a desiccator at 20–22 °C. A range of air moistures, and hence water pickup by the sample were obtained by covering the bottom of the desiccator with P_2O_5 powder or saturated salt solutions of K_2CO_3 or KCl. Cellulose in contact with air equilibrated over P_2O_5 powder is referred to as the dry sample, while microcrystalline cellulose powder in equilibrium with a saturated salt solution of K_2CO_3 contained 8.1 wt% water, and with KCl it contained 14.6% water.

Equilibrium was checked by ensuring that the measurements were stable over time. The measurements were performed on samples held in the desiccator by using 0.5 m long leads that passed through the sealed top of the desiccator and then went to the measuring instrument.

3. Results

3.1. Block sample holder

The empty cell capacitance was measured to be 2.55 pF. To check the suitability and effectivity of the sample holder and to obtain a calibration, the permittivities of paraffin oil and water were obtained by immersing the holder in these liquids. The results, Table 1, using $C_0 = 2.55$ pF, show a good agreement between the measured values and those reported in the literature. A second check of the response of the sample holder was to change the density of the tablets and plot the high frequency capacitance ($> 10^5$ Hz) as a function of the density of the cellulose sample, as in Fig. 4.

The best fit line to the data was determined to be

$$C' = 2.39 + 5.55 \times 10^{-3} \text{ (density) pF} \qquad (12)$$

TABLE I Calibration of sample holders with paraffin oil, microcrystalline cellulose and water as references. Measured relative permittivity versus literature data

Sample	Measured[a]			Literature [12]
	C' (pF)	C_0 (pF)	ε	ε
Paraffin oil	5.68	2.55[b]	2.23	2.2
Microcrystalline cellulose	11.1[c]	2.55[b]	4.35	4.5
Water	231	2.85	81.0	80

[a] Capacitance C' at 10^5 Hz.

[b] Capacitance of empty sample holder used in this work.

[c] Calculated from Equation 12.

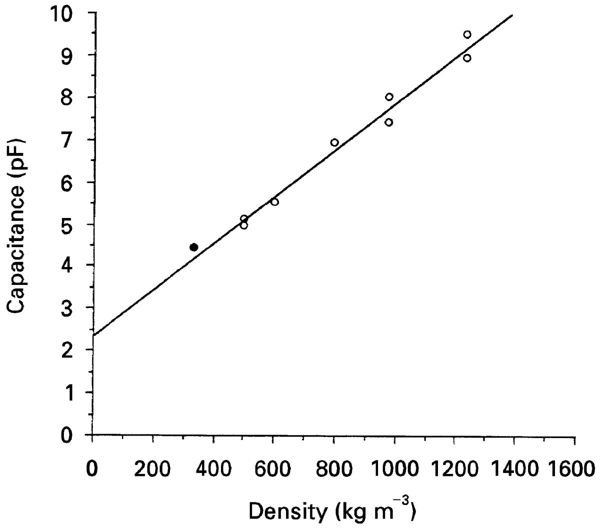

Figure 4 Capacitance $C'(\omega)$ at 10^5 Hz versus density for powder and compressed tablets of microcrystalline cellulose (equilibriated over saturated K_2CO_3 salt solution). The best fit line, Equation 12, with a linear correlation coefficient of 0.982 line indicates an empty cell capacitance of 2.39 pF which can be compared to the empty cell capacitance of 2.55 pF. Powder sample (●) and tablet sample (○).

with a linear correlation coefficient $R^2 = 0.98$. A zero density results in an empty cell capacitance of 2.39 pF. Using the density of a cellulose particle, 1570 kg m^{-3}, gives a permittivity of 4.35 for cellulose, Table I.

3.2. Powder

All cellulose samples whether in the form of microcrystalline cellulose powder, tablets or papers, showed the same form of response to that in Fig. 1. In Fig. 5. the dielectric responses of microcrystalline cellulose and lactose in a humid environment are presented. There are several similarities. At high frequencies the samples have an almost identical capacitance magnitude and the lead inductance which is constant for all samples tends to equalize the loss component as expected. Furthermore the crossover point, $C'(\omega) = C''(\omega)$ is located at the same frequency (≈ 0.1 Hz) and magnitude (≈ 10 pF) for both samples. The major difference is the presence of a loss peak (≈ 10 Hz) in the response of the microcrystalline cellulose, which is not apparent in the response from the lactose powder. Also the imperfect charge transport is close to d.c. conduction (for which $s = 1$) for lactose ($s = 0.97$) but substantially lower for cellulose ($s = 0.85$).

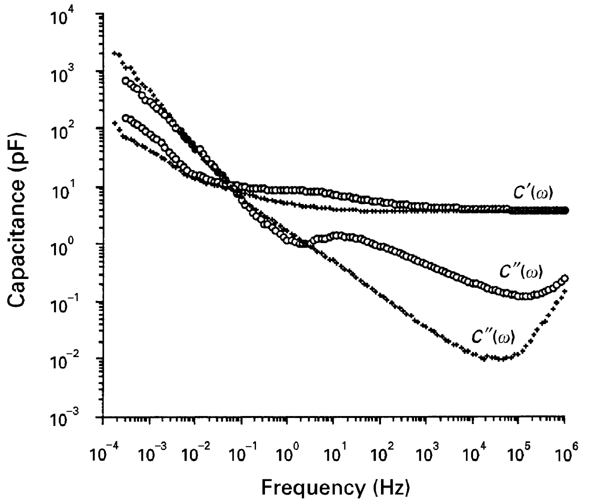

Figure 5 Comparison of dielectric spectra of powder, lactose (+) and microcrystalline cellulose, (○), (both equilibrated over saturated K_2CO_3 salt solution).

Water has a significant influence on the dielectric response of microcrystalline cellulose, Fig. 6. There is no dispersion in the response for dry microcrystalline cellulose powder for $f > 10^{-3}$ Hz, and consequently the loss capacitance C'' is low. With a moisture content of 8.1 wt% in the cellulose sample both imperfect charge transport and a dipole loss peak were present. At still higher water contents, 14.6 wt% moisture in the cellulose sample, the imperfect charge transport increases in magnitude and dominates the response.

An extended dielectric spectra for microcrystalline cellulose, Fig. 7, has been obtained by normalization [13] over the crossover point for the two dielectric spectra of microcrystalline cellulose of Fig. 6. The two datum points in the bottom left of Fig. 7, indicate the relative frequency shift between the two spectra i.e., the distance the spectra have been translated in order to enable normalization. It can be seen that, in this case, the normalization only required translation in the frequency and not in the capacitance. Hence the increased water content does not change the magnitude of the response, only the rate of the relaxation process.

3.3. Tablets

The quantified influence of the sample density on the dielectric properties of tablets made by pressing powders under different pressures are presented in Table II. In order to get a better view of how the sample density affects the magnitude and rate of the different charge transport processes, the dielectric spectra of microcrystalline cellulose were normalized over the crossover point as shown in Fig. 8a, and separately for the loss peak, Fig. 8b.

The initial treatment of the particles during consolidation into a solid body increases the capacitance and decreases the frequency, Fig. 8 (a and b). At higher densities the results differ depending on which feature

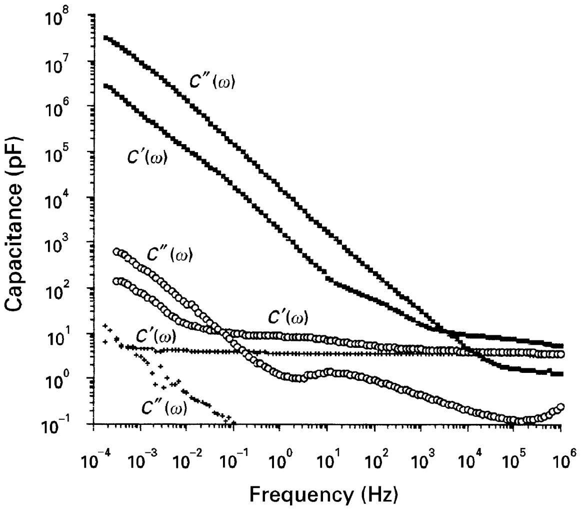

Figure 6 Influence of water on the dielectric response of microcrystalline cellulose powder. The different water contents obtained by conditioning of the powder in air over P_2O_5, powder referred to as the dry sample (+), saturated salt solution of $K_2CO_3 = 8.1$ wt % water (○) and saturated salt solution of KCl = 14.6 wt % water (■).

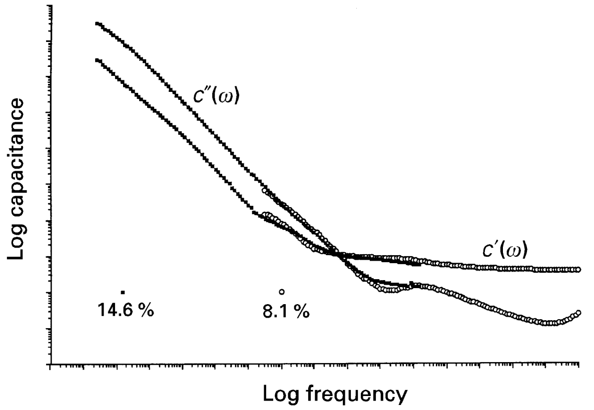

Figure 7 Normalized dielectric spectra for the two sets of microcrystalline cellulose powder data. Normalization has been carried out over the crossover point where $C'(\omega) = C''(\omega)$. The symbols in the bottom of the plot indicate the shift the curves have been moved in order to enable normalization. On both the original data plots the reference point was at $f = 10^{-3}$ Hz and $C = 1$ pF. 8.1 wt % (○), and 14.6 wt % (■).

has been used for the normalization. Normalization over the cross over point as in Fig. 8a, gives a slight increase in frequency but no change in capacitance as the compression proceeds. However, normalization over the Debye-like loss peak, Fig. 8b, indicates a return towards higher frequencies and lower capacitance under equivalent conditions. A further observation is that the shape of the high frequency wing of the Debye-like loss peak decreases when powder is compressed into a solid body and then remains constant as the tablet density increases.

In addition the exponent *s*, characterizing the imperfect charge transport, Equation 4, shows an increase during the initial phases of compact consolidation. The value of *s* increases from 0.85 in the powder to 0.92–94 at tablet densities above 600 $\text{kg}\,\text{m}^{-3}$, Table II.

3.4. Paper

The dielectric spectra obtained from the paper stack are presented in Fig. 9. The magnitude of the charge transport process at low frequencies is unaffected by the paper orientation. The imperfect charge transport exponent *s*, is only slightly higher for measurements made "in" ($s = 0.89$) compared to "out" ($s = 0.84$) of the paper plane. The difference due to paper orientation decreases as the frequency increases, but in the mid-range, around 100 Hz, a loss peak is clearly visible for the "in" paper plane measurements. For the "out" of plane measurements the loss peak is less visible since it is shifted down in frequency. The loss component reaches a capacitance of 2 pF for "in" the paper plane at 8.72 kHz (std. dev. = 34%) and "out" of the paper plane at 1.11 kHz (std. dev. = 38%).

4. Discussion

4.1. Sample holder

The new design of a sample holder with the electrodes at a right angle has enabled characterization of the different structural forms of cellulose-based products namely; powder, tablets and paper sheets using a single cell. The fixed position of the electrodes gives the opportunity of measuring the empty sample holder capacitance and thereby enabling a calculation of the empty cell capacitance and hence the relative permittivity of the samples. All the results reported here in terms of capacitance can be transformed into permittivity by dividing by the empty sample holder capacitance, 2.55 pF.

TABLE II Dielectric characterization data of microcrystalline cellulose in powder and tablet form

Sample	Density ($\text{kg}\,\text{m}^{-3}$)	Dipolar relaxation			Conduction exponent
		Frequency (Hz)	Time (ms)	Amplitude (pF)	*s*
MCC-powder	330	10	16	1.4	0.85
MCC-compacts	497	0.32	498	23	–[a]
”	600	1.0	159	14	0.82
”	794	1.8	88	18	0.94
”	976	3.2	50	13	0.94
”	1228	3.2	50	12	0.92

[a] The constant phase response was not developed in the range above 10^{-4} Hz.

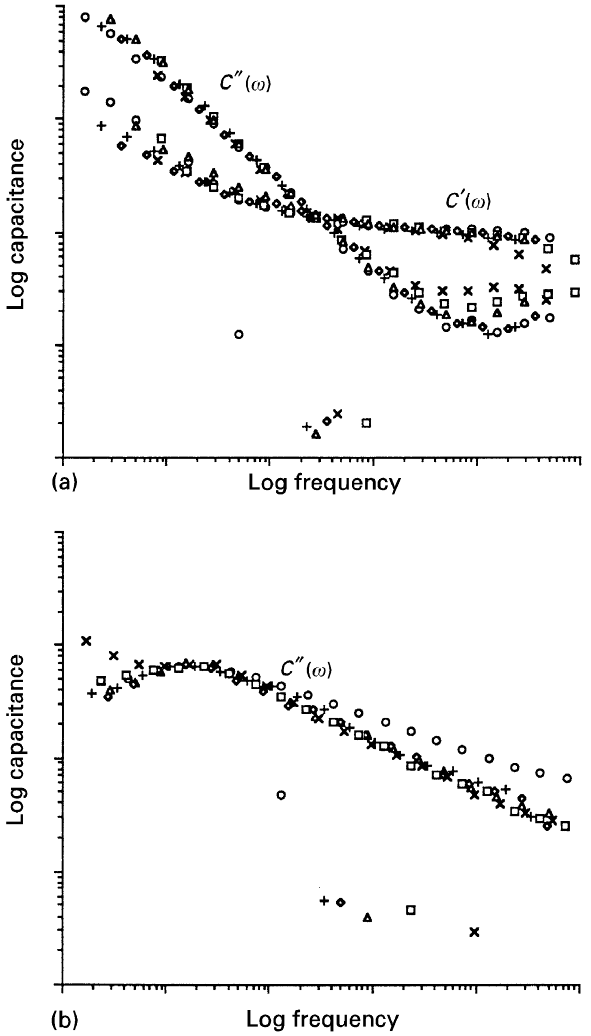

Figure 8 Normalised dielectric spectra of microcrystalline cellulose compressed to different densities. The symbols in the bottom of the plot indicate the shift the curves have been moved in order to enable normalization. On the original data plots the reference points; powder (○), tablets with densities (×) 497 kg m^{-3}, (□) 600 kg m^{-3}, (△) 794 kg m^{-3}, (◇) 976 kg m^{-3}, (+) 1228 kg m^{-3}; are located at $f = 10^{-2}$ Hz and $C = 1$ pF. (a) Normalization over the crossover point where $C'(\omega) = C''(\omega)$ and (b) normalization over the dipole loss peak.

The difference between the measured empty cell capacitance, 2.55 pF, and the value calculated from the best fit to the line in Fig. 4, 2.39 pF, is likely to be due to density variations in the tablets [14]. A larger difference has been found between the measured and the predicted value. However, the theoretical field, Equation 10, assumes no fringe effects even in the region of the confined corner which is an unreasonable assumption and considered to be the reason for the observed deviation.

When the sample holder was designed, a balance had to be made in the spacing between the electrodes. On the one hand the size of the particles to be studied required a narrow gap, whilst on the other hand the spacing must be finite to avoid electrical breakdown. The chosen configuration is a compromise and works well in practice. The good agreement between literature data and the measured permittivities for paraffin oil, cellulose and water proves the validity of this novel type of sample holder.

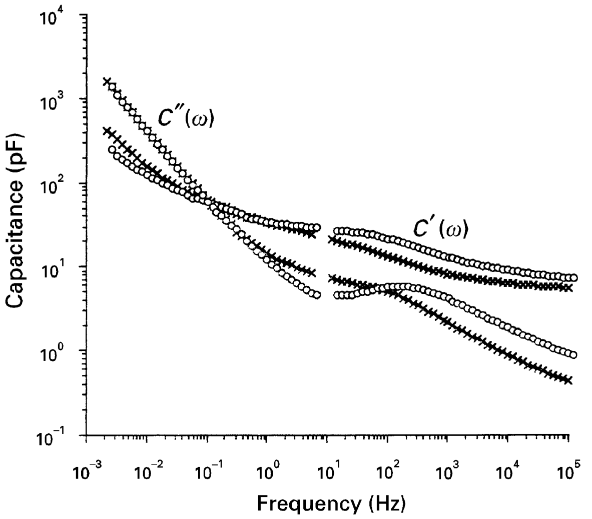

Figure 9 Dielectric spectra of paper, normalized about the crossover point $C'(\omega) = C''(\omega)$. The data missing from the middle of the spectra is an instrumental effect, due to a change in measuring technique. The symbols used are; "in" the paper plane (○), and "out" of the paper plane (×).

4.2. Characterization of powder

4.2.1. Influence of structure

Lactose has a similar chemical composition to microcrystalline cellulose, i.e., carbohydrates with OH groups as the polarizable part of the unit molecule. We suggest the difference in dielectric response to be due to the known different water solubilities of the materials. While cellulose is practically insoluble, 1 part lactose can be dissolved into 4.63 parts of water at 25 °C [15]. The high *s* value of the imperfect charge transport for lactose indicates a near perfect conduction. It therefore seems likely that the response of lactose comes from solvated molecules that are almost free to move around in the lactose particles. This almost free diffusion of the molecules explains why no loss peak has been observed in the dielectric spectra. The cellulose molecules are bonded into a solid insoluble structure making diffusion within the structure impossible.

4.2.2. Influence of water on cellulose

Using the normalization technique it has been possible to extend the investigations over a larger effective frequency range by changing the moisture in the sample. This is because the humidity affects the rate and the magnitude of the processes involved but not their basic nature, as can be seen from the experimental data.

The humidity normalized dielectric spectra of microcrystalline cellulose, Fig. 7, covers 15 decades in frequency and is a combination of data from two different moisture content samples translated by almost five decades in frequency. We observe that the normalization is performed only in frequency, not in magnitude. This is surprising because an introduction of 6 wt% of free water, should increase the magnitude of the capacitance by a factor of the order of 2. As this has not been observed, Fig. 7, we have to assume that the water is bonded into the cellulose structure in such a way that it does not contribute to the dipolar dielectric response.

4.3. Consolidation of tablets

It is known that the dielectric response of high polymers is affected by pressure [16]. The finding here is that when the cellulose particles are compressed into a solid body (tablet or compact) there is a clear increase in both the capacitance and the relaxation time, Fig. 8 (a and b), indicating that dipoles on the average can move over larger distances but that they require longer times in order to relax. The cellulose chains appear to have become more flexible.

From crystallinity measurements it has been suggested that drying stresses in microcrystalline cellulose are released during the initial stages of compacting [17]. The present work confirms this hypothesis. Eliminating drying stresses, stressing and stretching the molecule chains in the particles would increase the flexibility of the dipoles and thereby result in an increased capacitance. As the physical density of the tablets increases, the cellulose chains return to a more stressed and "locked up" state, Fig. 8b, and the dipolar flexibility decreases.

There are two indicators that the surface area taking part in the charge transport between particles does not increase with further increase in the tablet density. First, the imperfect charge transport shows that, after the initial stage of compaction there is only a small increase in the crossover frequency, Fig. 8a, with no increase in magnitude. Secondly, the s value increases only for low densities, after the initial increase the s-value is constant, Table II. A better charge contact between particles should result in an improved conduction [18] and s should increase towards its limiting value of unity.

4.4. Characterization of paper

The dielectric properties of paper and cellulose fibres has been previously reported in the literature but the measurements have been made in a range of high frequencies [19–22] or equilibrated in very high humidities [23]. At high humidities we have seen that the imperfect charge transport dominates the Debye process and at high frequencies neither of the processes contributes to the response.

Here we have examined the dielectric properties of paper, equilibrated with air at 44% relative humidity, in the mid frequency range 10^{-1} to 10^{4} Hz and observed that the dipolar relaxation is faster "in" than "out" of the paper plane, Fig. 9. Since the magnitude of the capacitive dispersion is the same for both orientations it is reasonable to assume that the dipolar contribution to the two processes are the same. We only observe the loss peak more clearly for measurements "in" than "out" of the plane, because, the loss peak for "in" plane measurements appears at a higher frequency and is not subsumed into the high frequency tail of the lower relaxation frequency. Hence it is the rate of relaxation that is affected by the orientation and not the magnitude, i.e., relaxation "in" the paper plane is easier.

5. Conclusions

Cellulose in various forms, (powder, tablets or sheets), has been characterized using dielectric spectroscopy in conjunction with a novel sample holder in which the electrodes are mounted at right angles to each other. The broad pattern of response is independent of the structural form of the samples. At low frequencies, below 10^{-2} Hz, the response is dominated by an imperfect charge transport process and in the region of 0.1–100 Hz a Debye-like loss peak was observed.

Humidity in the sample showed a significant influence on the charge transport process. A decreasing moisture content moves the low frequency dispersion down in frequency while increasing the moisture content gives the reversed response. In dry samples the dipolar loss peak was not evident but as the moisture content increased it appeared. In samples with a high moisture content the imperfect charge transport dominated the charge transport process. Moisture has a significant influence in increasing the rate of transport of charge.

The dielectric response for microcrystalline cellulose has been characterized over the equivalent of 15 decades in frequency. There is a linear correlation between the bulk density of the microcrystalline cellulose sample and the high frequency constant capacitance, indicating that the latter is a bulk property of the material.

The consolidation of microcrystalline cellulose particles into tablets proceeds by an initial increase in dipolar flexibility probably due to the release of drying stresses. As the tablet sample densities increases further the dipolar flexibility decreases since now the hydrocarbon chains are "locked up" in the compressed structure.

It has been shown that it is possible to characterize both long range transport of charge and the dipole orientation in the paper sample. This information is of interest in homogeneity studies of paper for the constant phase angle response process at low frequencies indicates a homogeneity at long range, while the dipolar loss process suggests a significant heterogeneity at the short range of the fixed dipoles in the material.

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