

# Relation Between Low-Frequency Dielectric Behavior of Cellulosic Materials and Their Inorganic Contaminations

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## SYNOPSIS

Results are given for dielectric constant and loss factor of some cellulosic materials of different fine structure free of and contaminated with water-soluble inorganic salts in the frequency range 0.1–100 kc/s and at temperatures 283.15–333.15 K. The analysis of the results reveals that the dielectric dispersion and loss peak found before with medicated cotton and viscose rayon, of ash contents of 0.3 and 1.4% respectively, disappear when these fibers have been “cleaned up” from inorganic impurities by acid washes. These anomalous dielectric behaviours reappear again with “cellulose-inorganic salts” admixtures. Based on these findings, it has been concluded that the secondary relaxation process observed in the low-frequency region 0.1–1 kc/s with cellulosic materials contaminated with inorganic impurities is not intrinsic but extrinsic in nature due to the displacement of ions adsorbed in the inner surfaces of such interstitial conduction system.

## INTRODUCTION

Water-absorption dielectrics and in particular those of cellulosic materials are among the most difficult systems to measure and analyse in dielectric terms. Earlier dielectric studies<sup>1–5</sup> of natural as well as manmade fibers show that in the glassy state these materials exhibit only one  $\beta$ -relaxation process in the frequency range 100–10,000 kc/s and has been attributed to the orientation of the OH groups in the disordered regions. While this is to be expected with such a system containing one polar group, a new absorption process has been reported<sup>6</sup> in the frequency range 0.1–1 kc/s and has been suggested to be local in nature and results from the polarization of the methylol groups originated in the low fractions. The same work indicates that the peak of the dielectric loss appeared only after allowance was made for conduction current loss that apparently predominates over the Debye loss. Recently,<sup>7</sup> from dielectric loss measurements, the specific conductance of some cellulosic substances of different fine

structure free of and contaminated with electrolytic impurities was determined. During the analysis of the results obtained to determine the DC loss, it was found that the conduction current loss as well as the remaining part of the measured loss are both dependent on the amount of inorganic impurities of the examined sample and that both losses decrease with the purification of cellulosic materials from such impurities by acid treatment on cold. These preliminary findings, together with the anomalous dielectric behaviour at low frequencies reported with low-humidity textile fibers containing inorganic water soluble salts,<sup>8–10</sup> as well as with aqueous colloidal suspension systems,<sup>11–19</sup> directed our attention to the important role of the ionic polarization processes that must be taken into consideration in the analysis and discussion of dielectric data of a given system contaminated even with a small amount of inorganic impurities.

Therefore, the present work has been undertaken to study the dielectric behaviour of some cellulosic materials free of and contaminated with water-soluble inorganic salts in the frequency range 0.1–100 kc/s and at temperatures 283.15–333.15 K. The results obtained will be analysed and compared with those reported before<sup>6</sup> with medicated cotton and viscose rayon.

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## EXPERIMENTAL

### Sample Preparations

#### Samples (A)

Sample A includes three cellulose samples free of inorganic impurities as judged from their undetectable ash contents and of different fine structure as followed by measurements of X-ray diffraction determined as before<sup>20</sup> and the degree of polymerization using the previous method.<sup>21</sup> These samples are: medicated cotton of crystallinity index determined by X-ray,  $Cr_x = 83$  and  $DP = 2305$ ; viscose rayon of  $Cr_x = 40$  and  $DP = 510$ ; and ground cellulose showing a diffuse X-ray pattern indicating  $Cr_x = 0$  and  $DP = 560$ .

Cotton cellulose and viscose samples were prepared by steeping the corresponding fibers of ash contents of 0.3 and 1.4%, respectively in 5N HCl at room temperature for 2 h, then washed with distilled water till free of acid. It has been reported<sup>22</sup> that this process results in the dissolution of all inorganic contaminations, which is in agreement with the results obtained here as shown by the undetectable ash contents found with the produced samples. At the same time, no change in the fine structure or dissolution of low fractions takes place as a result of acid treatment. These conclusions are based on the experimental results, which show that (1) almost the same values of  $Cr_x$  and  $DP$  were found for a given sample before and after acid treatment, and (2) when the mother liquor was tested for any dissolved sugar as previously described<sup>23</sup> a negative result was found.

The third sample of ground cellulose was prepared from the purified medicated cotton by cutting the fibers into small pieces that were then ground to pass a 20-mesh screen by using a hardened steel vial containing two steel balls. The resulting ground sample was then washed with cold 5N HCl to be sure that no inorganic contamination is present in the sample. A negative result was also found when the mother liquor was tested for any dissolved sugar. Therefore, the process of acid washes employed results only in the purification of the examined samples from inorganic impurities.

#### Samples (B)

Four samples of "cellulose-inorganic salts" admixtures of different ash contents were prepared from samples (A). These are: two viscose samples of ash contents of 0.23 and 0.68%, one cellulose sample of ash content of 0.42%, and one ground sample of ash contents of 0.53%.

These samples were prepared by steeping 10 g corresponding fibers in 100 mL aqueous solution containing different amounts of a mixture of NaCl-KCl (w/w 50%). The mixture was then left overnight at room temperature and evaporated using a water bath heated at 60°C till near dryness. The resulting "cellulose-salts" admixture samples were then used for dielectric measurements. The choice of NaCl-KCl (w/w 50%) is based on the reported values of ash contents of natural fibers that contain 90% as NaCl and KCl of almost the same weight proportion.<sup>24</sup>

### Percentage Ash Contents

The percent ash contents of the examined samples (A) and (B) was determined following the previous procedure.<sup>25</sup>

### Dielectric Measurements

The dielectric constant,  $\bar{\epsilon}$ , and loss factor,  $\bar{\epsilon}_{AC}$  were measured over the frequency band 0.1–100 kc/s and in the temperature range 283.15–333.15 K for samples (A) and (B). The apparatus used and the experimental details for such measurements, as well as the method of drying the samples, are described elsewhere.<sup>6,7</sup> Under the experimental conditions employed, the moisture contents of the examined samples was reported to be of the order of 1% and the residual water corresponding to this low moisture contents is strongly bounded by cellulose chains, particularly in the "difficulty" accessible regions.<sup>7,26–28</sup>

## RESULTS AND DISCUSSION

A primary task of the present investigation is to resolve the measured loss,  $\bar{\epsilon}_{AC}$  of the examined samples (A) and (B) into components due to different mechanisms and find out the effect of inorganic contaminations, determined as percent ash contents, on the magnitude of the resolved components in the frequency and temperature ranges studied. In the glassy state, the total measured loss of a given cellulosic sample can be considered, in general, to be the sum of two parts. The first part is due to the conduction current loss,  $\bar{\epsilon}_{DC}$  and the remaining part is due to the nonconduction loss,  $\bar{\epsilon}_{NC}$ . The latter loss may arise from several processes<sup>10</sup> that could be due to (1) molecular polarization, (2) polarization of adsorbed ions, and (3) polarization of free ions in case the resistance of the conducting paths is non-

**Table I Dielectric Loss Components and Specific Conductance of Viscose Samples Free of and Contaminated with Different Amounts of Inorganic Contaminations at 293.15 K**

Frequency (f, Hz)	Viscose Purified			Commercial Viscose <sup>a</sup> of 1.4% Ash			Viscose-Salt Admixture of 0.68% Ash			Viscose-Salt Admixture of 0.23% Ash		
	$\bar{\epsilon}_{AC}$	$\bar{\epsilon}_{DC}$	$\bar{\epsilon}_{NC}$	$\bar{\epsilon}_{AC}$	$\bar{\epsilon}_{DC}$	$\bar{\epsilon}_{NC}$	$\bar{\epsilon}_{AC}$	$\bar{\epsilon}_{DC}$	$\bar{\epsilon}_{NC}$	$\bar{\epsilon}_{AC}$	$\bar{\epsilon}_{DC}$	$\bar{\epsilon}_{NC}$
50	0.068	0.009	0.059	—	—	—	0.160	0.050	0.110	0.125	0.020	0.105
100	0.064	0.004	0.059	1.002	0.709	0.293	0.145	0.025	0.120	0.114	0.010	0.104
150	0.062	0.003	0.059	0.855	0.472	0.382	0.127	0.017	0.110	0.113	0.006	0.107
200	0.061	0.002	0.059	0.734	0.356	0.378	0.112	0.012	0.100	0.107	0.005	0.102
400	0.056	0.001	0.055	0.520	0.177	0.343	0.104	0.006	0.098	0.099	0.002	0.097
1,000	0.058	—	0.058	0.338	0.071	0.267	0.095	0.0025	0.092	0.081	0.001	0.080
2,000	0.058	—	0.058	0.251	0.035	0.215	0.099	0.001	0.098	0.070	—	0.070
4,000	0.054	—	0.054	0.225	0.107	0.207	0.102	—	0.102	0.060	—	0.060
10,000	0.056	—	0.056	0.223	0.007	0.216	0.142	—	0.142	0.056	—	0.056
Specific Conductance ( $\sigma \times 10^{13}$ )	2.5			393.8			13.8			5.5		

<sup>a</sup> These results are reproduced from an earlier work.<sup>6</sup>

uniform. In absence of these processes, a low loss magnitude may arise that is referred to as background loss. The contribution of each of these components to the measured AC loss depends mainly on the structure of the sample examined, its purity, the experimental conditions of temperature, and the frequency ranges employed. As representative examples and for the sake of comparison, Tables I and II give typical data of analysis of the total measured

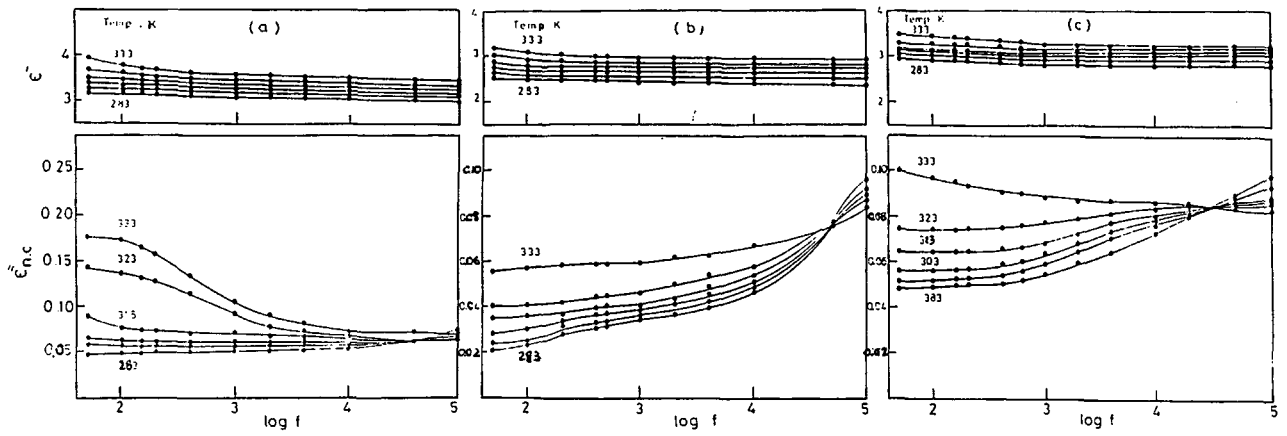
AC loss,  $\bar{\epsilon}_{AC}$ , into  $\bar{\epsilon}_{DC}$  and  $\bar{\epsilon}_{NC}$  loss components at two temperatures, namely 293.15 and 313.15 K, and at different frequencies for four viscose samples free of and contaminated with water-soluble inorganic salts as well as commercial inorganic impurities of different ash contents. The same analysis was made for the different samples examined here in the temperature and frequency ranges studied.

Figure 1 shows the variation with frequency of

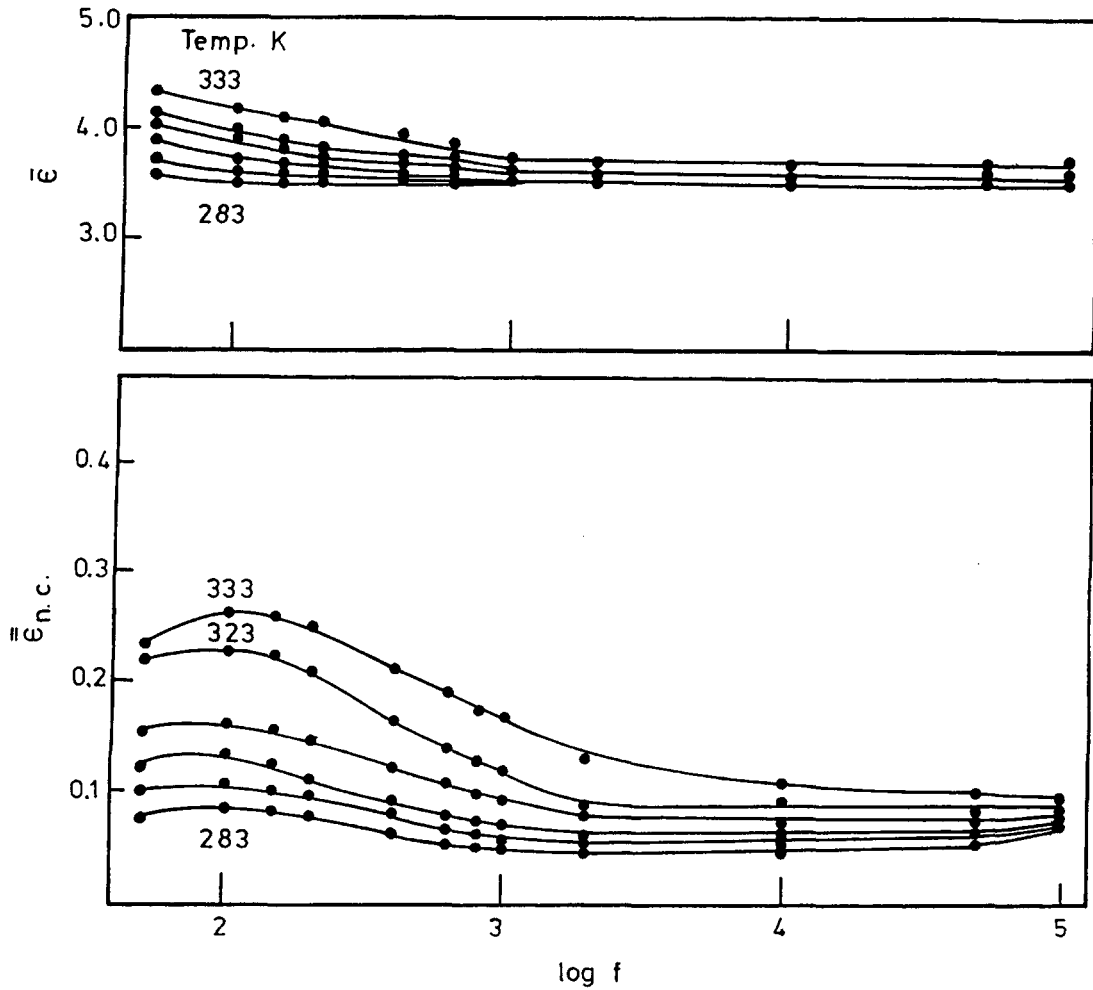
**Table II Dielectric Loss Components and Specific Conductance of Viscose Samples Free of and Contaminated with Different Amounts of Inorganic Contaminations at 313.15 K**

Frequency (f, Hz)	Viscose Purified			Commercial Viscose <sup>a</sup> of 1.4% Ash			Viscose-Salt Admixture of 0.68% Ash			Viscose-Salt Admixture of 0.23% Ash		
	$\bar{\epsilon}_{AC}$	$\bar{\epsilon}_{DC}$	$\bar{\epsilon}_{NC}$	$\bar{\epsilon}_{AC}$	$\bar{\epsilon}_{DC}$	$\bar{\epsilon}_{NC}$	$\bar{\epsilon}_{AC}$	$\bar{\epsilon}_{DC}$	$\bar{\epsilon}_{NC}$	$\bar{\epsilon}_{AC}$	$\bar{\epsilon}_{DC}$	$\bar{\epsilon}_{NC}$
50	0.167	0.08	0.087	—	—	—	0.771	0.50	0.271	0.301	0.15	0.151
100	0.114	0.04	0.074	2.763	2.218	0.545	0.572	0.250	0.322	0.234	0.075	0.159
150	0.101	0.027	0.074	2.099	1.473	0.621	0.498	0.166	0.332	0.203	0.050	0.153
200	0.090	0.020	0.070	1.783	1.109	0.674	0.353	0.125	0.228	0.182	0.0375	0.145
400	0.083	0.01	0.073	1.236	0.555	0.681	0.283	0.063	0.220	0.143	0.0187	0.125
1,000	0.074	0.004	0.070	0.822	0.222	0.600	0.226	0.025	0.201	0.098	0.0075	0.091
2,000	0.066	—	0.066	0.552	0.111	0.441	0.187	0.0125	0.175	0.076	0.0037	0.073
4,000	0.066	—	0.066	0.425	0.055	0.370	0.156	0.006	0.150	0.073	—	0.073
10,000	0.066	—	0.066	0.315	0.022	0.293	0.140	0.003	0.137	0.073	—	0.073
Specific Conductance ( $\sigma \times 10^{13}$ )	22			1232			138			41.66		

<sup>a</sup> These results are reproduced from an earlier work.<sup>6</sup>



**Figure 1** Variation with  $\log f$  of the dielectric constant  $\bar{\epsilon}$  and the nonconduction loss  $\bar{\epsilon}_{NC}$  for samples (A) free of inorganic contaminations. a, viscose rayon; b, cotton cellulose; c, ground cellulose.



**Figure 2** Variation with  $\log f$  of  $\bar{\epsilon}$  and  $\bar{\epsilon}_{NC}$  for viscose salt admixture of 0.23% ash contents.

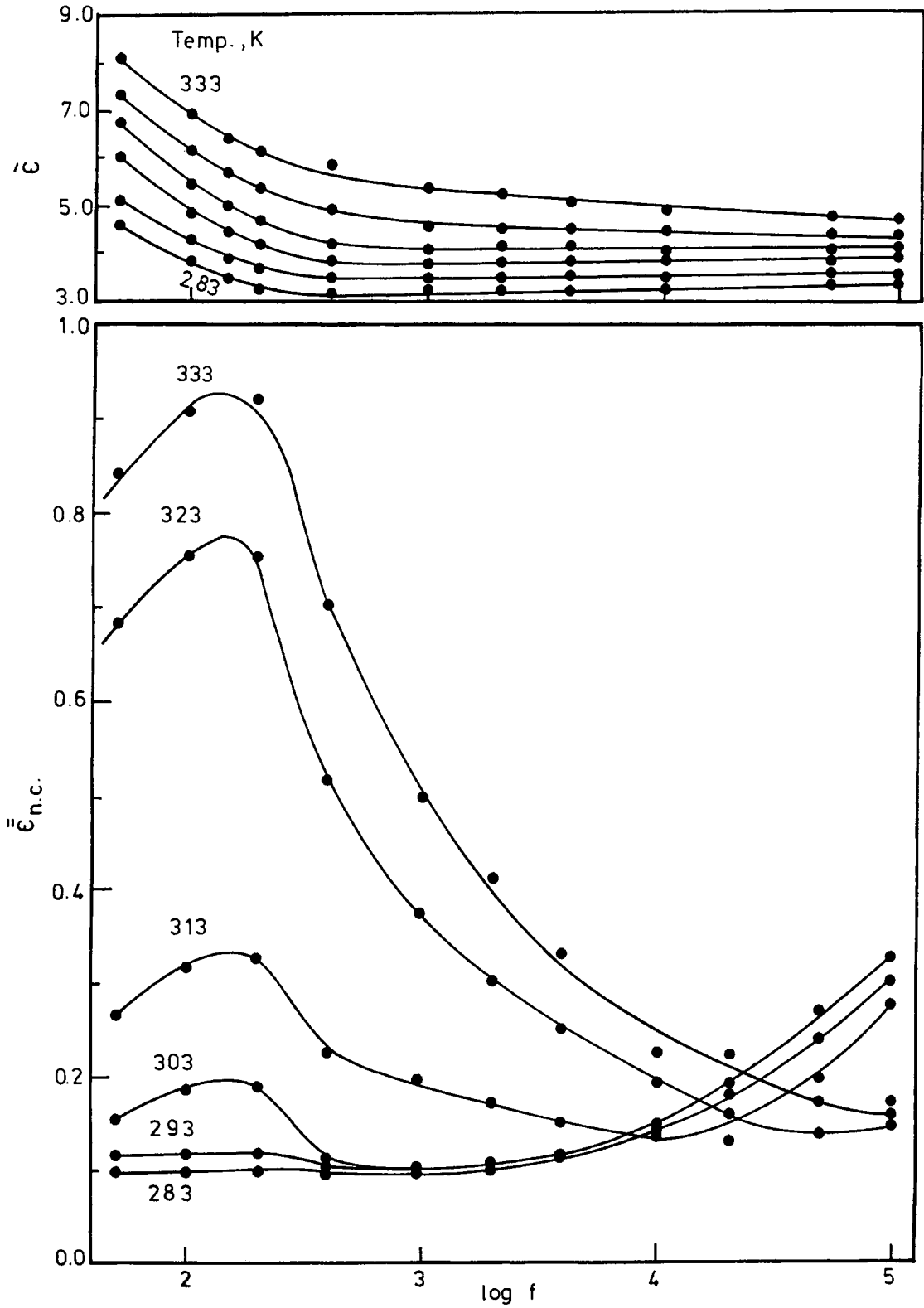
the dielectric constant,  $\bar{\epsilon}$ , and the nonconduction loss,  $\bar{\epsilon}_{NC}$ , at different temperatures for samples (A). The same relations are given in Figures 2–5 for samples (B). Although the conduction current loss and in turn the specific conductance and its relation to the fine structure and inorganic impurities of cellulosic materials has been studied previously,<sup>7</sup> a comparison between the effect of percent ash contents on  $\bar{\epsilon}_{DC}$  and  $\bar{\epsilon}_{NC}$  losses seems of importance. Figure 6 gives the change with percent ash contents of these parameters at 313.15 K and 100 c/s for the different samples studied here, as well as those examined previously.<sup>6,7</sup>

It is clear from Figure 1 that in spite of the difference in the fine structure of samples (A), as shown by the variation of their crystallinity from 0–83% and their degree of polymerization from 510–2,305, no anomalous dielectric behaviour is observed in the temperature and frequency range studied. When, however, one compares these results with those found before<sup>6</sup> with medicated cotton of ash contents of 0.3% and viscose rayon of ash contents of 1.4%, it results that the dielectric dispersion and loss peak observed with these contaminated samples with inorganic impurities disappear when the commercial fibers have been “cleaned up” from such contaminations. Therefore, the anomalous dielectric behaviours observed before<sup>6</sup> in the frequency range 0.1–1 kc/s with medicated cotton and viscose rayon are not intrinsic but extrinsic in nature, and that the inorganic impurities are the primary source for such dielectric behaviours. Accordingly, the conclusion drawn before<sup>6</sup> claiming that the observed loss and dielectric dispersion is due to the polarization of the methylol groups that originate in the low fractions or the rotationally disordered phase of these fibers must be reconsidered.

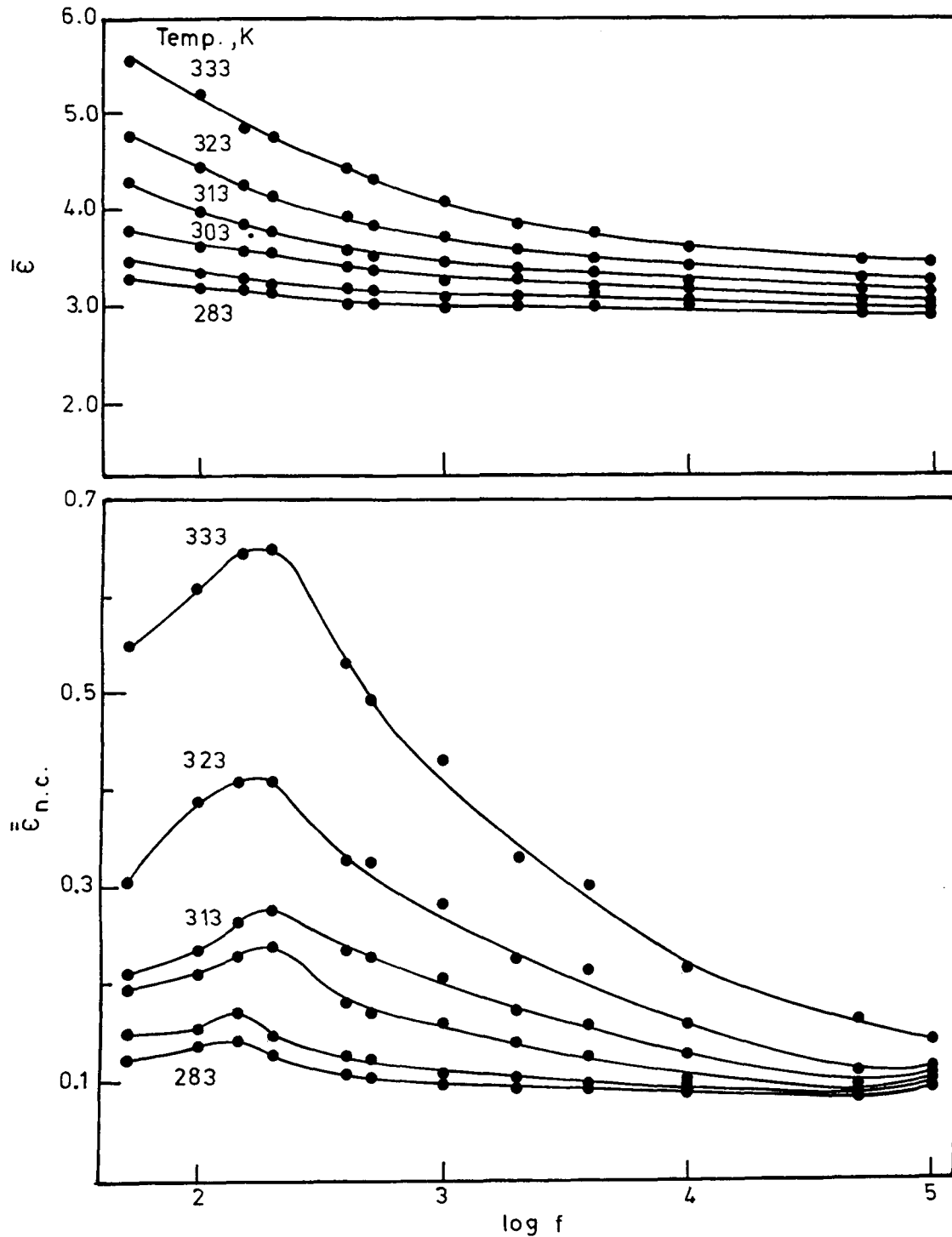
That the appearance of the low-frequency dielectric dispersion and dielectric loss is totally due to the presence of electrolytic contaminations has been strongly and directly confirmed in the present work by the results obtained with samples (B) as shown in Figures 2–5. These figures reveal that when the purified samples (A) have been recontaminated with water-soluble inorganic salts, the anomalous dielectric behaviours reappear again with the resulting “cellulose-salts” admixture samples, and that the magnitude of the dielectric constant and the intensity of the loss peak (see Figs. 2–5), as well as the DC loss (see Tables I and II), are all dependent on the amount of the salt contents. When comparing the characteristic features of the curves shown in Figures 2–5 and those found previously<sup>6</sup> with commercial fibers, it can be seen that (1) the loss peak

observed in both cases appears only after allowance has been made for the DC loss that apparently predominates over the absorption loss at frequencies lower than 1 kc/s in the temperature range studied with all samples examined; (2) with decreasing the ash percent, the DC loss decreases and in case of “viscose-salts” admixture sample of low ash contents of 0.23% the loss peak appears after a very small correction for the DC loss; (3) the effect of temperature on the dielectric properties is almost the same whether the samples are contaminated with commercial impurities or with water-soluble inorganic salts, and the increase of temperature is accompanied with a slight increase in the frequency at which maximum absorption takes place; and (4) the set of absorption bands observed in the frequency range 0.1–1 kc/s are independent of the nature of the inorganic contaminations.

Before discussing the characteristics of the dielectric absorption and loss of cellulosic materials contaminated with inorganic impurities, it seems desirable to outline the previous views and theories concerning water-absorption dielectric systems. It has been reported<sup>10</sup> that in such systems direct-current conduction does not take place uniformly through the material as a whole, but along paths of higher conductivity than the main part of the dielectric. In cellulosic materials, conduction takes place through water that is probably condensed in the interstices between the cellulose micelles. An important property of such “interstitial conduction” system is that ions, if present, would undoubtedly be adsorbed by the interface between the insulating units and the relatively conducting interstitial medium; since the interfacial area would be large as compared with the volume of such conduction paths, the influence of these adsorbed ions on the electrical properties of the dielectric would be expected to be very important in some cases. The ions may be regarded as free to move over the surface of the particular structural unit by which they are adsorbed, but not to migrate to the adjacent structural units. When an electric field is applied, it will produce a polarized distribution of ions adsorbed on the surface of each structural unit; the ions return to their normal distribution after the removal of the electric field. The electric effect of the adsorbed ions is therefore similar to that of a dipole, and they produce a kind of “dielectric” polarization. Intensive studies were carried out and several theories based on ionic polarization processes have been proposed to account for the anomalous dielectric behaviours of colloidal systems at low frequencies. The outcome of these studies reveals that, while the dielectric be-



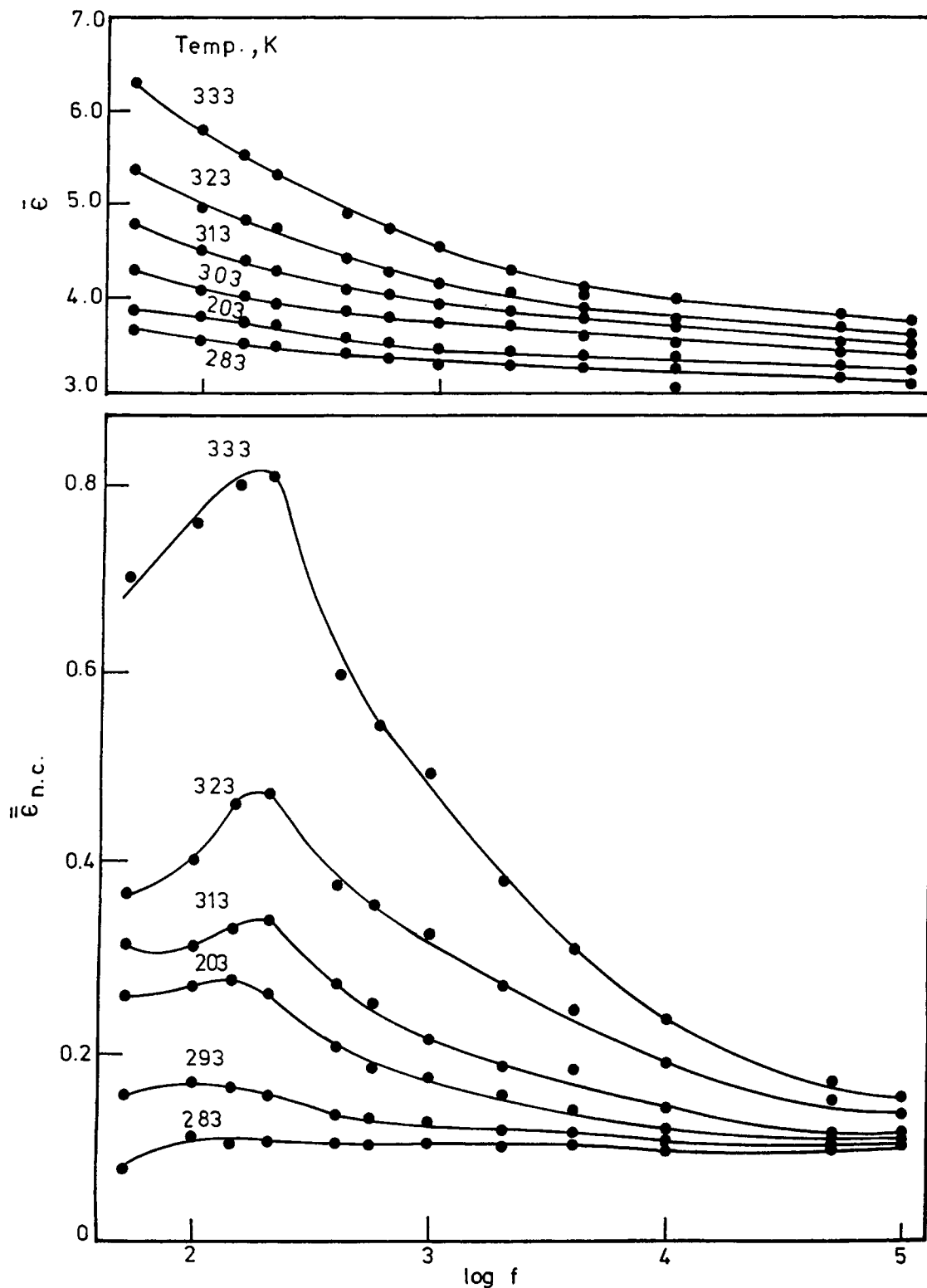
**Figure 3** Variation with  $\log f$  of  $\bar{\epsilon}$  and  $\bar{\epsilon}_{n.c.}$  for viscose salt admixture of 0.68% ash contents.



**Figure 4** Variation with  $\log f$   $\bar{\epsilon}$  and  $\bar{\epsilon}''_{n.c.}$  for cellulose-salts admixture of 0.42% ash contents.

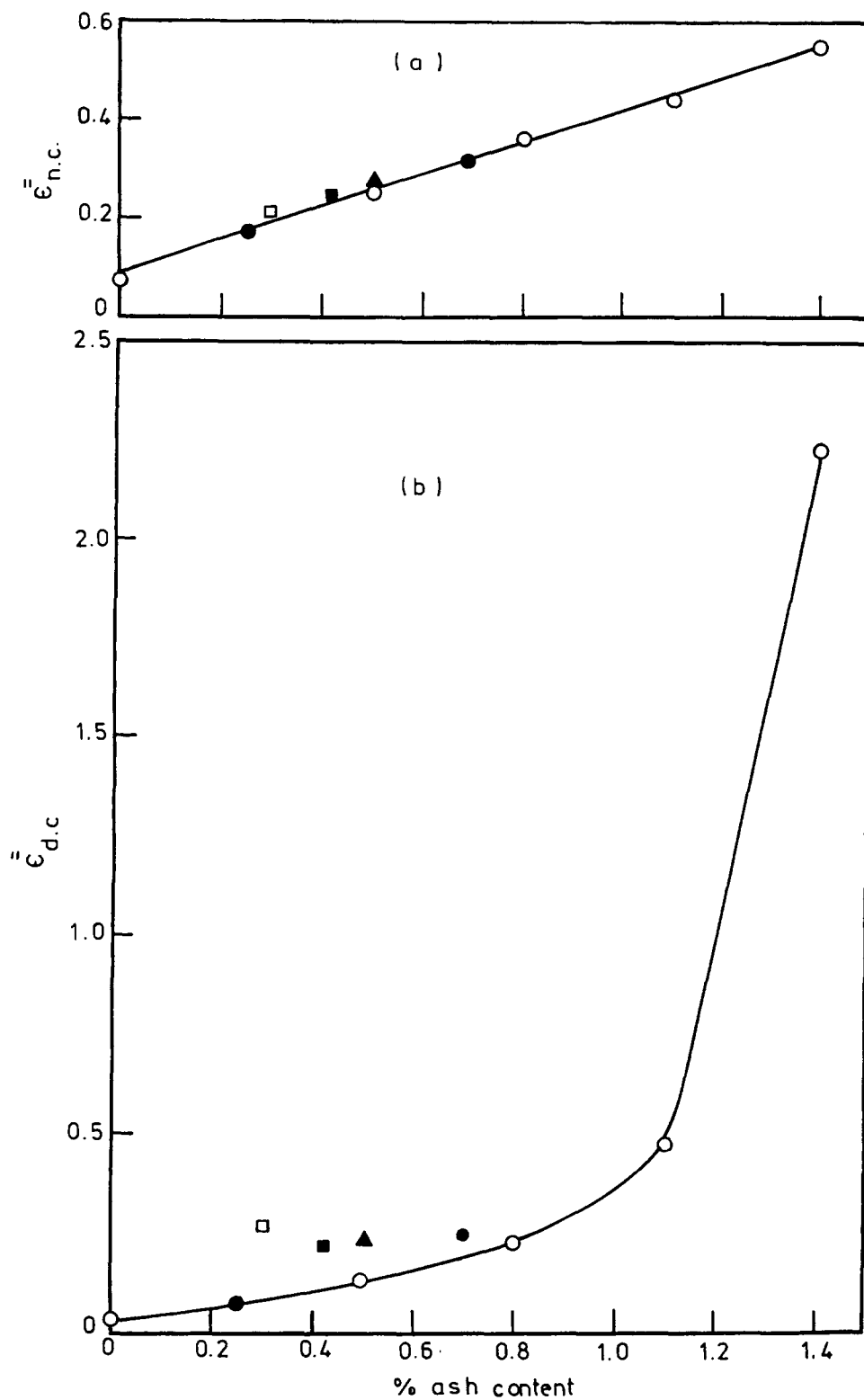
haviours of protein systems is mainly associated with dipole orientation process,<sup>29-33</sup> this does not appear to be the case with nucleic acids and other related polyelectrolytes, where ionic processes are very im-

portant.<sup>11-14</sup> Ionic polarization due to adsorbed ions have been suggested to be responsible for the displacement current and dielectric loss found previously<sup>8-10</sup> in case of cotton contaminated with



**Figure 5** Variation with  $\log f$  of  $\epsilon'$  and  $\epsilon''_{n.c.}$  for ground cellulose-salts admixture of 0.53% ash contents.





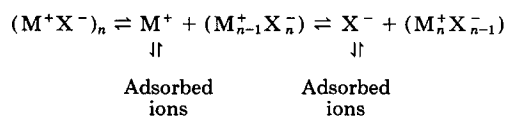
**Figure 6** Variation with percent ash contents of  $\bar{\epsilon}_{n.c.}$  loss curve (a) and  $\bar{\epsilon}_{d.c.}$  loss curve (b) determined at 313.15 K and 100 c/s for commercial samples of: ○, viscose; □, cellulose; and sample salts admixtures of: ●, viscose; ■, cellulose; ▲, ground cellulose.

soluble inorganic salts at low humidities. Based on the foregoing, it is clear that a correction of the measured loss for DC loss is not sufficient to analyse and discuss the dielectric data for cellulosic materials contaminated with inorganic impurities without taking into consideration the contribution of the adsorbed ions to the electrical properties of such systems.

In the present work, the dielectric measurements were carried out on samples of low moisture contents of the order of 1%. The residual water molecules corresponding to this low moisture content have been reported<sup>7,26-28</sup> to be strongly bound by cellulose chains, and that water in such systems will not act as a solvent for the contaminated inorganic salts as in case of the examined samples (B). In such systems of low dielectric constant, it is expected that even strong electrolytes would partially dissociate into ions and that the dissociation constant of these electrolytes would be very small. This is what has been reported<sup>34</sup> for tetra-alkyl ammonium salts when soluble in nonhydroxyl media of low dielectric constant in which only a very small fraction, e.g.,  $1 \times 10^{-4}$  and often much less has been found to be present as free ions. In harmony with these findings are the very low specific conductance determined for some of the investigated samples (B) following the previous procedure<sup>5,7</sup>; the results obtained at 293.15 and 313.15 K are given in Tables I and II. These results show that the specific conductance of the examined samples ranges from  $5.5 \times 10^{-13}$  to  $12.32 \times 10^{-11} \text{ ohm}^{-1} \text{ cm}^{-1}$  and that the change of the specific conductivity from one sample to the other depends on the amount of salt contaminations as well as temperature. Similar results have been found in a recent work.<sup>7</sup>

A quantitative analysis to determine the dissociation constant of the contaminated inorganic salts of samples (B) is not possible due to the absence of any reliable values of the ionic mobilities in cellulosic materials. Therefore, qualitatively we can fairly say that the contaminated salts in the investigated samples (B) results in a small fraction of free ions and the remaining portion is most probably present as adsorbed ions together with undissociated salt molecules or ion-pairs. An equilibrium is established between the free ions and adsorbed ions on one hand and between free ions and the undissociated ion-pairs on the other hand. In addition, these two equilibria are dependent on each other as well as on the amount of contaminated salt, the applied electric field, and temperature. Furthermore, the undissociated salt molecules will often aggregate to a range

of sizes,  $n \cdot M^+X^- = (M^+X^-)_n$ , with the possibility of  $n = 1$  or higher depending on concentration of the contaminated salts. The aggregates in turn will be in equilibrium with small concentrations of free ions. A simple picture for the equilibria between the different species could be represented by the following equation:



In view of the above equation, the electrical behaviour of such a system is expected to be quite difficult. In absence of precise values of the proportions of the different species given in the proposed equilibria and the lack of knowledge concerning the dimensions of cellulose-ions aggregates, the effect of the amount of inorganic contaminations and temperature on the dielectric properties of samples (B) will be interpreted qualitatively in view of the total ash percent and the above equilibria.

Now regarding the effect of the amount of salt contaminations at a given temperature on the dielectric constant, as well as on the total measured loss and its components, it is clear from Tables I and II and Figures 2-6 that all these parameters increase with increasing the percent ash contents. Furthermore, it is obvious from Tables I and II that while  $\bar{\epsilon}_{DC}$  loss predominates over  $\bar{\epsilon}_{NC}$  loss with samples of high ash percent at all temperature examined, viscose sample of low ash contents of 0.23% at low temperature shows the reverse. These results indicate that at low concentration of inorganic salts and at low temperature, where the conductivity is very low, the electrical effect due to the adsorbed ions predominates over the conduction loss due to free ions. The dependence of both components on percent ash contents is shown in Figure 6, which reveals that (1) the variation of  $\bar{\epsilon}_{DC}$  loss with ash percent takes place exponentially and the change is dependent on the nature of the contaminated inorganic impurities, and (2) in case of  $\bar{\epsilon}_{NC}$  loss, its variation with ash percent is linear and independent on the nature of the ions present. These results show that, although each loss process results from the presence of inorganic impurities, the two loss processes are of different nature. This has been justified by the pronounced difference found between their activation energy determined here by following the previous procedures.<sup>6,7</sup> The activation energy of conductance,  $E_s$  for the examined samples (B) above

and below the transition temperature of cellulosic fibers<sup>7</sup> has been found to be in the order of 7.0–13.8 kcal/mol. At the same time, the activation energy of the dielectric absorption process,  $E$ , has been found, for the same samples and in the same temperature ranges, to be in the order of 2.4–3.5 kcal/mol. These results are of importance because they confirm the above views that the total measured loss must be divisible into components and agree well with the previous opinions of Murphy and Lowry.<sup>10</sup> These authors pointed out that the existence of several components of dielectric loss provides an alternative physical interpretation of the several characteristic time constants introduced by the Maxwell–Wagner theory of dielectric absorption and loss to account for the experimental facts and attributed by Wagner to the presence of several types of impurities in dielectrics.<sup>35</sup>

Based on the above findings and in particular (1) the absence of dipole polarization process as found with the purified samples (A), (2) the appearance of the anomalous dielectric behaviour with samples of low ash percent where the  $\bar{\epsilon}_{NC}$  loss is greater than the  $\bar{\epsilon}_{DC}$  loss, and (3) the interstitial conduction systems and the adsorption of ions, which is its most essential property, one may conclude that the anomalous dielectric behaviours observed in Figures 2–5 as well as those previously reported<sup>6</sup> with commercial cellulosic samples are due to the polarization of the ions adsorbed in the inner surfaces of the interstitial conduction systems examined. When an electric field is applied to such a system, the least strongly adsorbed ions will become desorbed and join the conduction current; thus, conductivity increases. As the voltage gradient is increased, more adsorbed ions would join the conduction current; this may explain the increase of conductivity with increasing field strength, which is a prominent characteristic of solid dielectrics. However, under constant electric field, as in the present work, and at a given temperature an equilibrium is expected to be established, as shown by the above equation, and the proportions of free ions as well as adsorbed ions would depend mainly on the amount of the ash percent. With increasing temperature, dissociation of salt molecules would result in a shift of the above equilibria toward formation of more free ions and in turn more adsorbed ions would occur. This may explain the increase of the two loss processes with increasing temperature as given in Tables I and II and as shown in Figures 2–5.

Finally, the slight shift of the critical frequency at which maximum absorption takes place with in-

creasing temperature and its independence on the nature of the contaminated inorganic impurities are both in favour of the above mechanism of the adsorbed ions. This is because the dielectric polarization of the adsorbed ions results in local displacement of these ions, and thus a low activation energy is expected for this process as found in the present work and given above. Consequently, this polarization process is slightly affected by the nature of the ions adsorbed, compared with the conduction process in which free ions can move in matter over comparatively large distance. Thus, the nature of ions and their mobility play an important role on the electrical conductivity of a given system.

## REFERENCES

1. Y. Ishida, M. Yoshino, and M. Takayangi, *J. Polym. Sci.*, **I(2)**, 227 (1959).
2. W. N. Stoops, *J. Am. Chem. Soc.*, **56**, 1480 (1934).
3. H. G. Shinouda, A. A. Hanna, and A. Kinawi, *J. Polym. Sci.*, **15**, 1991 (1977).
4. H. G. Shinouda, A. A. Hanna, and A. Kinawi, *J. Polym. Sci.*, **16**, 2019 (1978).
5. S. Boutros and A. A. Hanna, *J. Polym. Sci.*, **16**, 89 (1978).
6. M. M. Abdel Moteleb, M. M. Naoum, H. G. Shinouda, and H. A. Rizk, *J. Polym. Sci.*, **20**, 765 (1982).
7. M. M. Abdel Moteleb and H. G. Shinouda, *J. Appl. Polym. Sci.*, **38**, 2079 (1989).
8. E. J. Murphy, *J. Phys. Chem.*, **33**, 200 (1929).
9. E. J. Murphy, *J. Phys. Chem.*, **33**, 509 (1929).
10. E. J. Murphy and H. H. Lowry, *J. Phys. Chem.*, **34**, 598 (1930).
11. G. A. Johnson and S. M. Neale, *J. Polym. Sci.*, **54**, 229 (1961).
12. G. A. Johnson and S. M. Neale, *J. Polym. Sci.*, **54**, 241 (1961).
13. S. M. Neale and D. A. Weyl, *Proc. Roy. Soc. (London) A*, **291**, 368 (1966).
14. D. A. Weyl and S. M. Neale, *Dielectrics*, **1**, 22 (1963).
15. H. P. Schwan, G. Schwarz, J. Maczuk, and H. Pauly, *J. Phys. Chem.*, **66**, 2626 (1962).
16. G. Schwarz, *J. Phys. Chem.*, **66**, 2636 (1962).
17. M. Mandel, *Mol. Phys.*, **4**, 489 (1961).
18. C. T. O'Konski, *J. Phys. Chem.*, **64**, 605 (1960).
19. D. J. Allen, S. M. Neale, and P. J. T. Tait, *J. Polym. Sci., Part A-2*, **10**, 433 (1972).
20. L. Segal, J. J. Creely, A. E. Martin, and C. M. Conrad, *Text. Res. J.*, **29**, 786 (1959).
21. L. Büchs and F. Mertes, *Melliand Text. Ber.*, **45**, 275 (1964).
22. Y. Ogiwara and H. Kubota, *J. Polym. Sci.*, **7**, 2087 (1969).
23. M. Dubois, K. A. Giltes, J. K. Halmillon, P. A. Rebers, and F. Smith, *Anal. Chem.*, **28**, 350 (1956).

24. F. Sadv, M. Korchagin, and A. Matetsky, *Chemical Technology of Fibrous Materials*, Mir Publishers, Moscow, 1978.
25. C. Doree, *The Methods of Cellulose Chemistry*, 2nd ed. Reprinted, Chapman and Hall, London, 1950.
26. C. E. Boesen, *Cellulose Chem. Technol.*, **4**, 149 (1970).
27. Y. Ogiwara, H. Kubota, S. Hayashi, and N. Mitomo, *J. Appl. Polym. Sci.*, **13**, 1689 (1969).
28. Y. Ogiwara, H. Kubota, S. Hayashi, and N. Mitomo, *J. Appl. Polym. Sci.*, **14**, 303 (1970).
29. S. Takashima, *J. Am. Chem. Soc.*, **80**, 4478 (1958).
30. S. Takashima, *J. Polym. Sci.*, **56**, 257 (1962).
31. R. Lumry and R. H. S. Yue, *J. Phys. Chem.*, **69**, 1162 (1965).
32. P. Moser, P. G. Squire, and C. T. O'Konski, *J. Phys. Chem.*, **70**, 744 (1966).
33. S. Takashima, *J. Polym. Sci., A*, **1**, 2791 (1963).
34. N. E. Hill, W. Vaughan, A. H. Price, and M. Davies, *Dielectric Properties and Molecular Behaviour*, Van Nostrand Reinhold Company, London, 1969.
35. K. W. Wagner, *Arch. Elektrotech.*, **2**, 371 (1914).

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