

# Effect of the Fine Structure and Electrolytic Contaminations on the Specific Conductance of Some Cellulosic Materials

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## Synopsis

From dielectric loss measurements the specific conductance of some cellulosic materials of different fine structure free of and contaminated with electrolytic impurities in the temperature range of 283.15–333.15 K was determined. Also the activation energy of conductance for the different samples examined was calculated. Based on the data obtained and the discussion given, it is now possible to view the conductivity of the complex structure cellulosic substances in a much clearer light.

## INTRODUCTION

The outcome of several investigations<sup>1–10</sup> reveals that the magnitude of electrical conductivity of polymeric materials is not strictly definite and invariable, but depends on several factors, mainly the presence of impurities, moisture content, and chemical composition. The problems pertaining to the dependence of the electrical insulating substances on each of these factors have been discussed, yet there remains an open question about the role played by the arrangement of elementary units and forces between them, especially when systems of complicated structure and of highly hygroscopic nature such as cellulosic fibers are concerned. To answer this question and to shed more light on the electrical conductivity of these materials, the conductivity of some cellulosic substances free of and contaminated with electrolytic impurities of different nature and magnitude will be determined. For the sake of comparison, the specific conductance of cellobiose, a 100% crystalline material and that of starch, a 100% accessible substance, will be also determined. In addition, some characteristic properties of the fine structure of the examined samples will be determined by measurements of x-ray diffraction, iodine adsorption, and degree of polymerization.

## EXPERIMENTAL

### Materials

The present investigation will be carried out on two sets of samples, namely A samples free from electrolytic impurities as judged from their undetectable ash contents and B samples with different nature and magnitude of electrolytic contaminations. The different samples investigated were prepared as given below.

**SAMPLES A:** Cotton cellulose (I): was prepared by steeping medicated cotton cellulose in 5 *N* HCl solution for 2 h at room temperature, then washed with redistilled water till free from acid. Under such experimental conditions, it has been pointed out<sup>11,12</sup> that the acid removes all the inorganic contaminations from cellulosic fibers as well as from hydrocelluloses.

Mergerized cellulose (II) was prepared from cotton cellulose following the previous work.<sup>13</sup>

Viscose rayon (III) was prepared by purification of commercial viscose fibers following the same procedure as in case of sample.<sup>1</sup>

Hydrocelluloses (IV, V, and VI) were prepared from cotton cellulose, mergerized cellulose, and viscose rayon, respectively, as before,<sup>14</sup> in which the fibers were subjected to acid action by the use of 1 *N* HCl at 100°C for 4 h, then washed with water until free from acid.

Starch (VII) was a B.D.H. analar-soluble starch.

Cellobiose (VIII) was an Eastman Kodak analar sample in the form of crystalline powder.

**SAMPLES B:** Cotton cellulose (IX) composed of ash content of 0.3%, was prepared from medicated cotton cellulose as before.<sup>10</sup>

Viscose rayon (X, XI, XII, and XIII) samples, of ash content of 1.4, 1.1, 0.8, and 0.5%, respectively, were prepared from commercial viscose fibers by steeping the fibers in HCl solutions of various concentrations of 0.1–1.5 *N* for 2 h at room temperature. The fibers were then filtered and thoroughly washed with redistilled water till free from acid.

Viscose-iron (XIV and XV), two samples were prepared by mixing 20 g of viscose fibers, free from electrolytic impurities, at 25°C for 2 h with 500 mL of either 0.1 *M* or 0.01 *M* FeCl<sub>3</sub> solution, respectively. The fibers were then filtered and washed with redistilled water until free from both chloride and ferric ions.

Percentage ash contents of the examined samples A and B were determined following the previous procedure.<sup>15</sup>

Drying technique: before carrying out the dielectric loss measurements all samples studied were dried as before,<sup>16</sup> under such conditions of rigorous drying it has been reported<sup>16–18</sup> that the dried samples still retained some residual water amounting to  $\approx 1\%$ , and that this moisture content may change from one sample to another depending on their adsorption capacity.

### Measurements

**Dielectric loss:** The apparatus and technique were previously described,<sup>10</sup> in which a WTW dekameter type DK05 covering a frequency range from 0.03 to 100 kc/s was used. The dielectric cell was kept dry during measurements by guarding it with phosphorous pentoxide and passing through it a mild stream of thoroughly dried purified air. Measurements of  $\epsilon$  were made at frequencies from 0.05 to 0.3 kc/s over the temperature range 283.15–333.15 K. The error in  $\epsilon$  was  $\pm 0.5\%$  and the constancy of temperature in the cabinet was  $\pm 0.2\%$ .

The specific conductance  $\sigma$  of each sample at any given temperature was calculated by plotting  $\epsilon f$  versus  $f$ , and then extrapolated to zero frequency,

the derived values of  $(\epsilon f)_{f \rightarrow 0}$  were substituted in the following equation to get  $\sigma$ .

$$(\epsilon f)_{f \rightarrow 0} = 1.8 \times 10^{12} \sigma \text{ Ohm}^{-1} \text{ cm}^{-1}$$

**X-ray crystallinity index:** A comparative study of x-ray crystallinity index,  $Cr_x$  of the different samples was carried out by using the empirical method of Segal et al.<sup>19</sup>

**Iodine crystallinity index:** The crystallinity index,  $CR_{I_2}$  was determined from iodine adsorption following the previous method.<sup>20</sup>

**Degree of polymerization:** The degree of polymerization, DP was determined as before.<sup>21</sup>

### RESULTS AND DISCUSSION

The calculated specific conductance  $\sigma$  at 293.15 K for the examined samples A and B is given in Table I. Also presented are some indices taken as a measure of the characteristic properties of samples A. These are, the crystallinity index determined from x-ray diffraction,  $Cr_x$  and that determined from iodine adsorption,  $Cr_{I_2}$  together with the degree of polymerization, DP. For the sake of comparison,  $\sigma$  at the same temperature for cellobiose a 100% crystalline substance and starch a 100% accessible material is cited in the same table. The variation with temperature of  $\sigma$  of some of the examined samples is shown in Figure 1.

The results as shown in Table I reveal that  $\sigma$  varies from one sample to the other and depends on the fine structure as evidenced by the different  $\sigma$  values of samples A as well as on the electrolytic contaminations shown by  $\sigma$  values of samples B. In addition, these results clearly indicate that while the change in the fine structure has little effect on  $\sigma$ , the inorganic contaminations have a

TABLE I  
Data Obtained for Samples A and B

Materials	Samples A					Samples B			
	Sample no.	$\sigma \times 10^{-13}$ at 293.15 K	$Cr_x$	$Cr_{I_2}$	DP	Sample no.	% Ash content	$\sigma \times 10^{-13}$ at 293.15 K	
Cotton cellulose	I	1.35	83	85	2305	IX	0.3	67.0	
Mercerized cellulose	II	1.8	66	69	2120	—	—	—	
Viscose rayon	III	2.6	40	42	510	X	1.4	393.0	
	—	—	—	—	—	XI	1.1	245.2	
	—	—	—	—	—	XII	0.8	114.1	
	—	—	—	—	—	XIII	0.5	70.3	
Cotton hydrocellulose	IV	5.1	87	93	196	—	—	—	
Mercerized hydrocellulose	V	4.4	80	91	150	—	—	—	
Viscose hydrocellulose	VI	11.0	70	82	45	—	—	—	
Viscose-iron	—	—	—	—	—	XIV	0.80	33.3	
	—	—	—	—	—	XV	0.14	2.5	
Starch	VII	11.1	(Taken as a 100% accessible substance)						
Cellobiose	VIII	1.25	(Taken as a 100% crystalline material)						

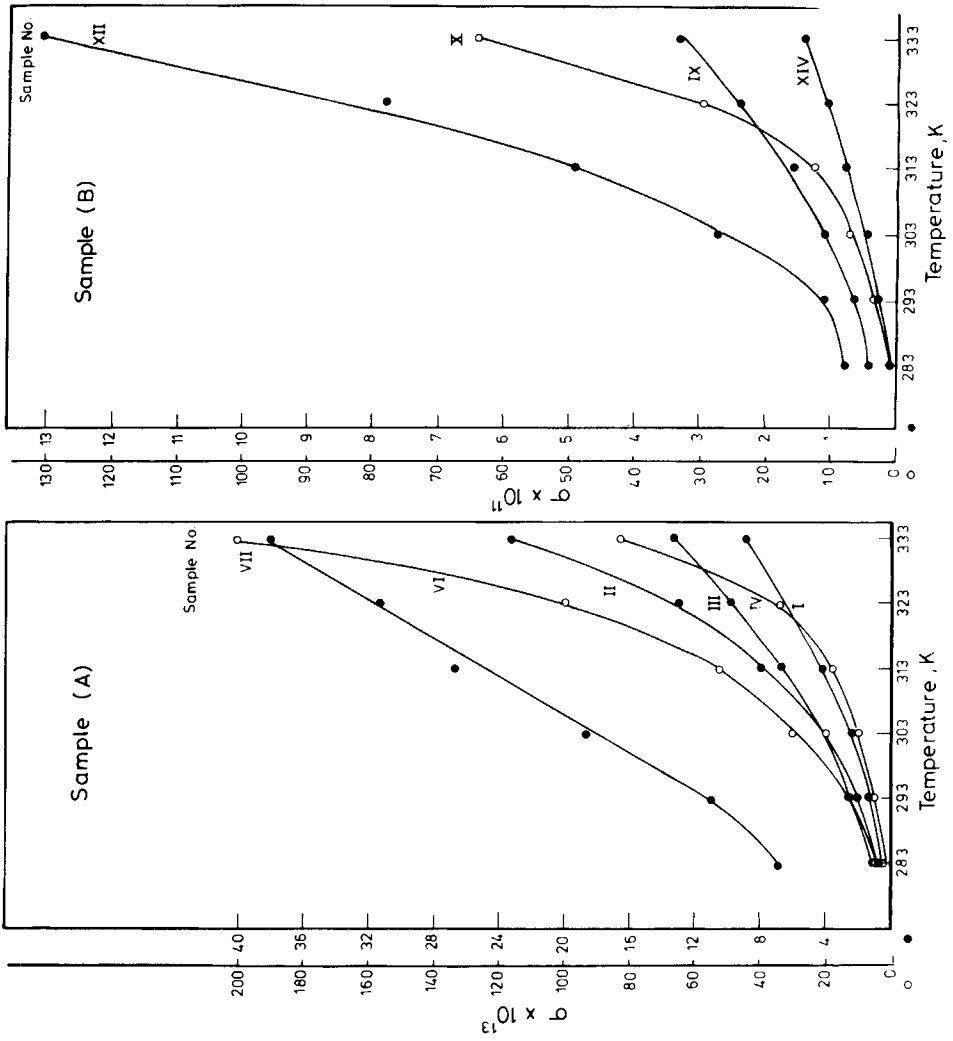


Fig. 1.

pronounced effect, depending not only on the magnitude of the ions produced from such impurities, but also on their nature. Regarding the effect of temperature, it is evident from Figure 1 that the change of  $\sigma$  with temperature takes place exponentially and that greater variations in  $\sigma$  occur on increasing temperature above 303.15 K compared with the variations below this temperature.

To analyze and interpret the significance of the variation of  $\sigma$  from one sample to another, the previous results concerning the conductivity of polymeric materials and, in particular that of cellulosic substances, should be considered.<sup>1-10</sup> These results reveal that the magnitude of conductivity of polymers depends greatly on the amount of electrolytic impurities and moisture content, and that the molecular chain proper does not participate in the transfer of electric charges, but they may only have an indirect effect on the mobility of these charges. Therefore, to test how far each of these parameters contributes to the measured conductivity of a given polymer, one has to cancel or minimize the effect of the other parameters. This is what has been done in the present work, in which the effect of water was minimized as far as possible by carrying measurements on dried samples so that the moisture content hardly exceeds 1%. However, even with such low moisture content, one cannot overlook the role played, if any, by this residual water on the conductivity of the investigated sample. Consequently, before we proceed to account for the variations of  $\sigma$  as a result of the change in the fine structure as shown by samples A and as a result of electrolytic contaminations as given by samples B, it seems important to discuss the interrelation between the water adsorption and the fine structure of cellulosic materials.

In view of the previous results<sup>22</sup> on the sorption-desorption behaviors of cellulosic fibers, it has been pointed out that, once the "less difficultly" accessible regions are disrupted by swelling the water molecules adsorbed in them would be the last to leave upon decreasing the vapor pressure. At low temperature this water may not be totally desorbed even after outgasing for fairly long periods and that it is usually removed only at high temperature. Almost complete drying has been established by subjecting cellulosic fibers to outgasing at 65°C for 15 h.<sup>18</sup> Accordingly, the residual water corresponding to 1% moisture content in our investigated samples is most probably retained by the "less difficultly" accessible regions and the highly disordered fractions would be almost free from adsorbed water. This is because (i) at 80°C, the temperature at which the examined samples were dried, the highly accessible regions desorb water easily compared with the inaccessible ones and (ii) if only one molecule of water is to be attached to every three OH groups in the accessible regions, the calculated amount of water necessary for such adsorption is 4% in the case of cotton cellulose ( $\approx$  30% accessible) and 8% in case of viscose rayon ( $\approx$  60% accessible). Lower values are expected for such adsorption with the hydrocellulose samples of less hygroscopic nature compared with cellulosic fibers, whereas higher values amounting to 12% are necessary in the case of the 100% accessible starch sample.

Based on the foregoing, the low water content retained in the investigated samples would not be capable of forming the continuous conducting water-paths necessary to form a network or space pattern as previously pointed out.<sup>7</sup> If so, one may ask what is the role played by residual water confined to the

intermediate ordered regions of the test samples on their electrical conductivity. These regions between crystalline and amorphous components may be regarded as carriers of the required flexibility and elastoplasticity for a given cellulosic substance. At low temperature, even these regions "freeze," thus all the fibers become hard and brittle; the same happened in the very dry state. Even a low water content in these regions is expected to act as a "softener" in cellulose and promote internal mobility of the amorphous component, particularly at high temperature.<sup>17</sup> The increased flexibility of cellulose chains will allow other units such as end groups and ions if present to be polarized and also will enhance the mobility of charge carriers responsible for the passage of electric current. Furthermore, under the influence of the electric field, the high permittivity of water can aid in the dissociation of some of the OH groups in the accessible regions of cellulosic substances.<sup>8</sup>

In light of the above views, the different  $\sigma$  values so obtained with the different samples studied as shown in Table I could be attributed to (i) the amount of current charge carriers which may be inorganic ions produced from electrolytic contaminations or from ionization of the polar OH groups of cellulose molecules in the accessible regions and (ii) the mobility of the concerned ions which depends on the nature of ions themselves as well as on the flexibility of cellulose chains.

Therefore, the higher  $\sigma$  values of samples B than those of samples A and the pronounced effect of the electrolytic impurities on the specific conductance of the former samples compared with the effect of the fine structure on  $\sigma$  of the latter ones is to be expected. This is because, in absence of the inorganic ions in the examined samples A, the only source of electric charge carriers responsible for their conductivity are produced from the ionization of the polar OH groups of cellulose molecules, and under our experimental conditions, the ionization of cellulose molecules is expected to be very small. This is justified by the very small difference in  $\sigma$  values between cellobiose a 100% crystalline substance and starch a 100% accessible material, and in spite of the relatively high  $\sigma$  value of starch compared with cellulose samples A, this value is still too low compared with  $\sigma$  values of samples B of the lowest commercial ash contents.

Now to account for the variation of  $\sigma$  of samples A with the change in their fine structure as shown by the crystallinity indices given in Table I, trials were conducted to determine whether a relation could exist between  $\sigma$  and each of  $Cr_x$  or  $Cr_{I_2}$ . The outcome of these trials revealed that no relation does exist between  $\sigma$  and these indices taken as a measure of the molecular packing of cellulose chains in the examined samples A. This conclusion could also be drawn from the results given in Table I which show that, although there is a pronounced difference between each of the given crystallinity indices of the three fibers, namely cotton cellulose, mercerized cellulose, and viscose rayon, close  $\sigma$  values are found for the first two samples and a small difference between these values and that of the latter one. Furthermore, when these fibers were subjected to acid action, the resulting residues which are of more compact structure compared with the corresponding fibers, show an increase of  $\sigma$  of the order of 2-5-fold, depending on the fiber examined. These results are contrary to what is to be expected if we consider that an increase in molecular packing and thus a decrease in the cohesive forces between cellulose

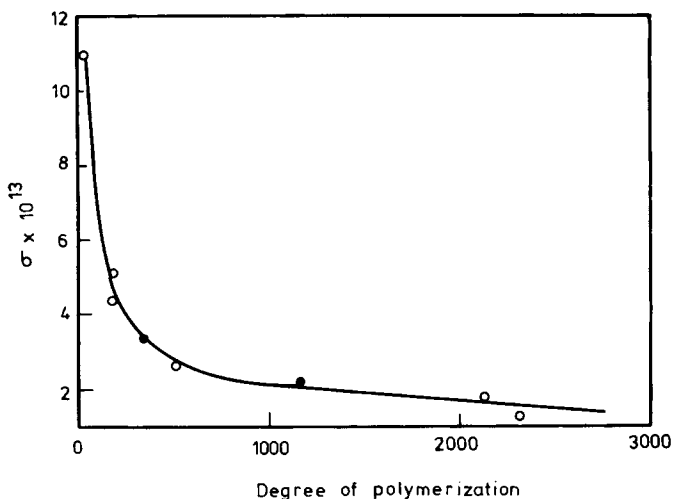


Fig. 2. Variation of  $\sigma$  with DP of:

(○) Sample (A)

(●) Sample XVI and XVII.

chains in these rigid residues will be accompanied by a decreased flexibility and dissociation of cellulose molecules and thus a decrease of their specific conductance. Together with the crystallinity index, the degree of polymerization, (DP) contributes greatly to the characteristic property of the fine structure of a given cellulosic material. It is evident from the DP results shown in Table I that the increase of  $\sigma$  of hydrocelluloses is accompanied with a much lower DP compared with the corresponding fibers. This decrease in DP results in an increase of surface area and end groups. If we accept the previous conclusion that the surface area has no effect on  $\sigma$ ,<sup>6</sup> then the increased end groups in hydrocellulose samples will result in increased ionization of cellulose molecules and thus an increase of their  $\sigma$ . Furthermore, the high  $\sigma$  values of these residues of less hygroscopic nature than the corresponding fibers, together with the very small difference between  $\sigma$  of cotton cellulose and mercerized cellulose of comparable DP, but of pronounced difference in their hygroscopic nature, could be taken as evidence that these variations in  $\sigma$  of samples A are most probably due to the ionization of cellulose molecules rather than water molecules retained in the "difficultly" accessible regions of the examined samples.

To verify that the degree of polymerization is one of the determining factors that affect the conductivity of cellulosic substances, a plot between  $\sigma$  and DP is shown in Figure 2. This figure reveals that a fairly good relation exists between  $\sigma$  and DP, and that  $\sigma$  decreases rapidly with the increase of DP and a limiting off value is attained when the DP exceeds 2300. It has been reported<sup>6,7</sup> that there is a relationship between fiber length and its resistance, but no reports concerning the dependence of  $\sigma$  on the degree of polymerization of cellulosic materials have been published.

In view of the relation given in Figure 2,  $\log \sigma$  is plotted against  $\log 1/DP$  as shown in Figure 3. This figure reveals that in the range of 45–2300 DP, and

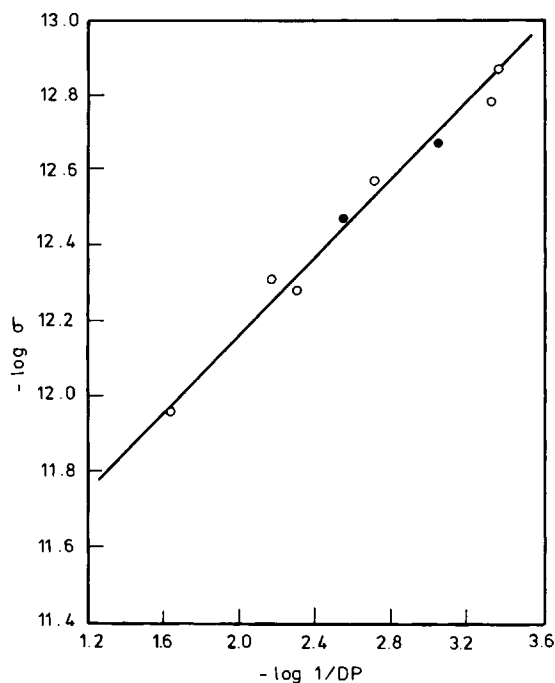


Fig. 3. Variation of  $\log \sigma$  with  $\log 1/DP$  of:

(○) Sample (A)

(●) Sample XVI and XVII

in spite of the marked difference in the origin and crystallinity indices of the examined samples, a straight line is obtained. This linear relation indicates that  $\sigma$  is an exponential function of  $1/DP$  and can be expressed by the following equation

$$\text{Log } \sigma = \text{Log } A + B \text{Log}(1/DP)$$

or

$$\sigma = A(1/DP)^B$$

where  $\sigma$  is the specific conductance in  $\text{ohm}^{-1} \text{cm}^{-1}$  and  $DP$  is the degree of polymerization of the examined sample.  $A$  is a constant which has a value of  $7.78 \times 10^{-12}$  and  $B$  is the slope of the straight line equal to 0.5263.

To test how far the experimental  $\sigma$  values deviate from the above relation, two hydrocellulose samples (XVI and XVII) were prepared from mercerized cellulose and their  $\sigma$  values at 293.15 K were determined by substituting for their  $DP$  in the above equation. The  $\sigma$  values so obtained are  $1.94 \times 10^{-13}$  and  $3.70 \times 10^{-13} \text{ohm}^{-1} \text{cm}^{-1}$  for the first and the second sample having a  $DP$  of 1156 and 325, respectively. For the same samples, their  $\sigma$  calculated from experimental measurements were  $2.10 \times 10^{-13}$  and  $3.48 \times 10^{-13} \text{ohm}^{-1} \text{cm}^{-1}$ , which shows that for a given sample, the experimental  $\sigma$  value deviates from



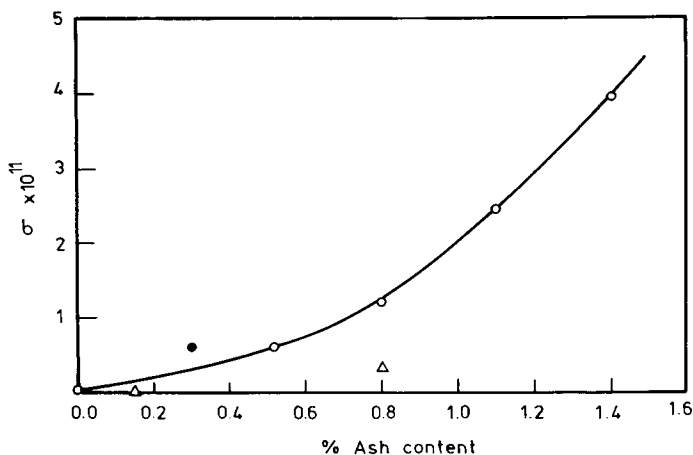


Fig. 4. Variation of  $\sigma$  with % ash contents of (o) viscose rayon, (●) cotton cellulose and ( $\Delta$ ) viscose-iron samples.

that determined from the above equation by  $\pm 0.2 \times 10^{-13}$ . The  $\sigma$  values determined from experimental results are presented graphically together with those of samples A in Figures 2 and 3.

Regarding the effect of the inorganic contaminations, a plot representing the variation of  $\sigma$  with ash contents is shown in Figure 4. It is obvious from this figure that a good relation exists between  $\sigma$  and the % ash contents in the case of viscose of different amounts of commercial electrolytic impurities. On the other hand,  $\sigma$  of cotton cellulose (sample IX) and that of viscose-iron (samples XIV and XV) deviate from such relation, being higher for the former and lower for the latter samples compared with  $\sigma$  values of viscose samples, containing commercial impurities, of comparable ash contents. As shown in Table I the specific conductance of the purified viscose (sample III) is higher than that of purified cotton cellulose (sample I). Therefore, the higher  $\sigma$  value of the contaminated cellulose (sample IX) than that of contaminated viscose of comparable ash contents may be taken as evidence that the inorganic ions produced from the electrolytic contaminations of the two fibers are of different natures. This conclusion is justified by the lower  $\sigma$  values of viscose-iron (samples XIV and XV) than those of the same fibers of comparable ash contents but of different electrolytic contaminations. In addition, the lower  $\sigma$  values of the former samples compared with the latter ones may be attributed to the difference in the nature of chemical bondings between viscose chain molecules and the inorganic ions which depend on their oxidation state as previously claimed.<sup>11</sup>

It has been reported<sup>6,7</sup> that the electrical conductivity of cellulosic materials depends on the amount of electrolyte contents, as confirmed by the present results which add that not only the magnitude of the inorganic electrolytes, but also the nature of their ions and the type of chemical bonds between them and cellulose molecules have a decided effect on such a property.

Regarding the effect of temperature on  $\sigma$ , it can be seen from Figure 1 that  $\sigma$  varies exponentially with temperature and that the variation is more rapid above 303.15 K than below this temperature. Furthermore, the magnitude of such variation below as well as above this temperature differs from one sample to another. It has been reported that in the glassy state the conductivity of polymers is approximately  $10^{-13}$  to  $10^{-19}$   $\text{ohm}^{-1} \text{cm}^{-1}$ .<sup>9</sup> And that with increasing temperature the conductivity of polymers increases according to an exponential law:

$$\sigma = Ae^{-\Delta E_a/RT}$$

where  $A$  is a coefficient weakly dependent on temperature,  $R$  is the universal gas constant, and  $\Delta E_a$  is the activation energy of conduction. This exponential change may be explained most satisfactorily by activation jumps of charge carriers from one region to another over the dielectric barriers created by the polymer structure. By increasing temperature, the carrier concentration does not change, but the mobility increases. Therefore to shed more insight onto the mechanism by which the current leakage takes place, the activation

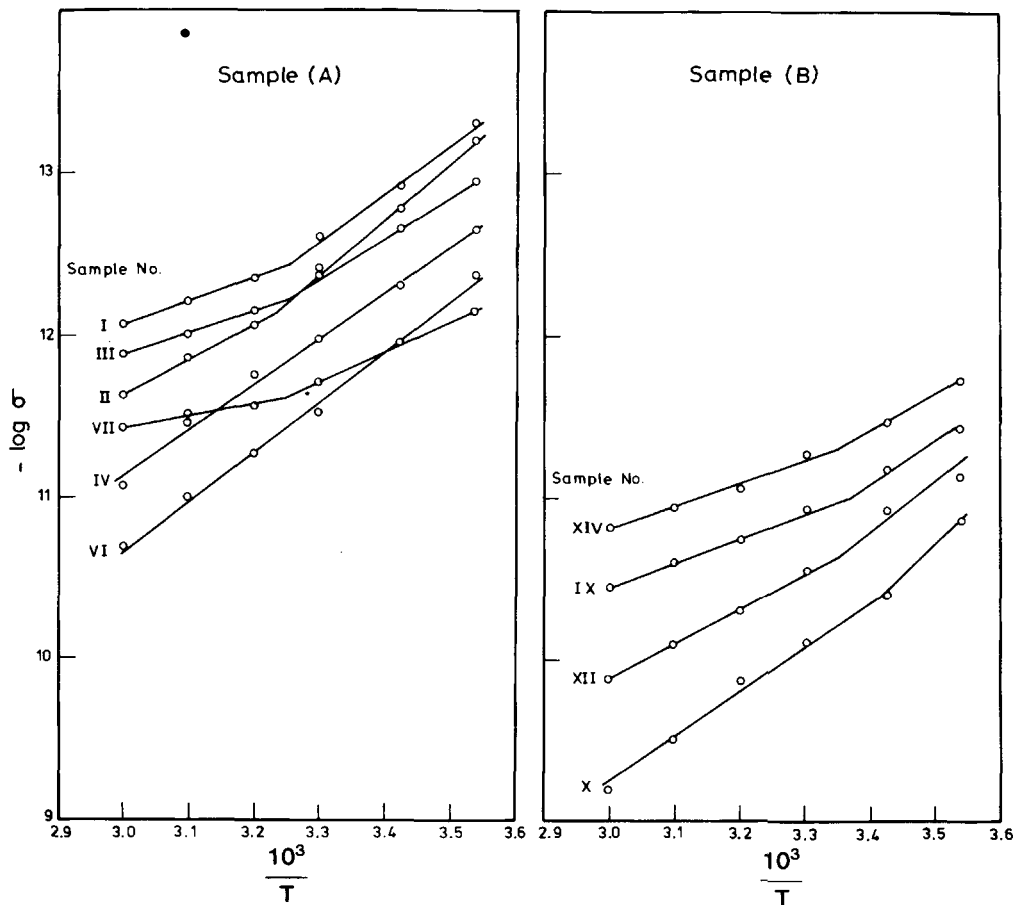


Fig. 5. Variation of  $\log \sigma$  with  $1/T$  of some of samples (A) and (B).

TABLE II  
Activation Energy of Conductance,  $E_{\sigma}$ , of Some Samples A and B

Materials	Samples		$E_{\sigma}$ kcal/mol	
	(A)	(B)	Before $T_r$	After $T_r$
Cotton cellulose	I	—	13.8	7.6
	—	IX	12.9	7.0
Mercurized cellulose	II	—	17.6	11.8
Viscose rayon	III	—	10.9	6.3
		X	15.9	12.6
	—	XII	15.9	12.0
Viscose-iron	—	XIV	9.0	6.0
Starch	VII	—	8.6	6.8
Cotton				
hydrocellulose	IV	—		14.0
Mercurized	V	—		14.2
hydrocellulose				
Viscose	VI	—		14.0
hydrocellulose				

energy of conductance  $\Delta E_{\sigma}$  of the different samples examined has been calculated by applying the above equation.  $\Delta E_{\sigma}$  for each sample was calculated by plotting the specific conductance against the reciprocal of the absolute temperature as shown in Figure 5 and the results obtained are given in Table II. It can be seen from Figure 5 that the change of  $\sigma$  with  $1/T$  for all cellulosic fibers examined exhibits a transition temperature,  $T_r$ , in the vicinity of 303.15 K and that in case of hydrocellulose residues this transition temperature disappears. In addition it is obvious from Table II that  $\Delta E_{\sigma}$  values differ from one sample to another and that with samples showing transition temperature  $\Delta E_{\sigma}$  is higher below than above this temperature.

The transition temperature,  $T_r$ , is too low to be considered as the glass transition of cellulose as has been established by several investigations.<sup>23-30</sup> Some of the examined properties indicate that this transition is a property of cellulose amorphous structure<sup>23-27</sup> and others reveal that some increased motion of parts of cellulose molecules takes place above this transition.<sup>28-30</sup> Hence, the greater increase of  $\sigma$  and the lower  $\Delta E_{\sigma}$  values above  $T_r$  of samples exhibiting such transition compared with the corresponding changes below this transition could be attributed to the increased flexibility of cellulose chain molecules which enhances the mobility of the concerned ions.

Comparing  $\Delta E_{\sigma}$  values given in Table II, it is clear that the difference between such values reveals that the activation energy of conductance can be fairly precisely related to both the nature of current charge carriers and the fine structure of cellulose samples as well. This is because the higher  $\Delta E_{\sigma}$  values of samples B than those of samples A could be attributed to the different nature of their inorganic impurities and the same could be said for the variation of  $\Delta E_{\sigma}$  from one sample to another in the case of the former samples. On the other hand in case of samples A, the electric charge carriers are mainly  $H^+$  and  $OH^-$  ions, the difference in their  $\Delta E_{\sigma}$  could be due to the change in chain flexibility from one sample to another as confirmed from the decrease of  $\Delta E_{\sigma}$  in a descending order parallel with the increased chain

flexibility from cellulose possessing the lowest flexible chain molecules to starch that has the highest flexible structure. That the chain flexibility of polymeric materials has a decided effect that contributes to the electrical conductivity of a given sample is justified by the high  $\Delta E_0$  values of hydrocellulose residues compared with the values of  $\Delta E_0$  of the corresponding fibers. As stated above, these residues are of strong cellulose inter-H-bonding, which is manifested by the high crystallinity index of these samples and the disappearance of  $T_r$  as shown by their  $\sigma - 1/T$  relations. The decreased flexibility of the chains of these residues results in an increase of the activation energy of conductance required for the concerned ions to overcome the potential energy barriers over their rigid structure.

Finally one can fairly conclude that in addition to the degree of polymerization, the nature and magnitude of the current charge carriers, the chain flexibility of cellulosic materials has a decided effect on the conductivity of these substances particularly at high temperatures.

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Received July 14, 1988

Accepted September 29, 1988