COEFFICIENT OF INTERNAL MASS TRANSFER IN ELECTRICAL-GRADE CELLULOSICS UNDER VACUUM AND UNDER ATMOSPHERIC PRESSURE

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The hygrodiffusivity of electrical-grade cellulosics has been calculated from test data on sorption kinetics under vacuum and under atmospheric pressure and on the diffusivity as a function of the state variables.

The desiccation of high-voltage paper insulation for electrical apparatus constitutes one of the most important and lengthy stages of the manufacturing process.

Cellulosics for electrical insulation are dried under atmospheric pressure and under vacuum, the process rate being essentially limited by the internal moisture transport.

For this reason, in order to design the desiccation process on a scientifically sound basis, as well as to analyze the moisture absorption and removal mechanisms, it is necessary to know the mass transfer coefficients over wide and practical ranges of moisture content and specimen temperature under atmospheric pressure and under vacuum.

We note that only scarce information is to be found in the technical literature pertaining to coefficients of mass transfer in cellulosic insulation materials [1], while no data are available at all on how these coefficients depend on the state variables of the material.

The authors have made an experimental study to determine the diffusivity of electrical-grade cellulosics widely used in the electrical industry, namely: grade K-120 cable paper (density $\rho = 0.76$ g/cm^3, thickness $\delta = 120\ \mu$), grade KON-P capacitor paper ($\rho = 1.23$ g/cm^3, $\delta = 10\ \mu$), and cardboard grades ÉMTs ($\rho = 1.05$ g/cm^3, $\delta = 1.5$ mm and 3 mm), A ($\rho = 0.93$ g/cm^3, $\delta = 3$ mm), and B ($\rho = 0.95$ g/cm^3, $\delta = 3$ mm).

Electrical-grade cellulosics are complex colloidal capillary-porous materials. Along with a rather high porosity (varying from $\varepsilon = 0.51$ in grade K-120 cable paper to $\varepsilon = 0.21$ in grade KON-P capacitor paper), they also have an extremely complex internal structure with random distributed amorphous regions and micropores as well as macropores of most diverse shapes and sizes.

The polymer molecule chains contain hydroxyl groups which attract water molecules from the air, and this explains why these materials are so highly hygroscopic and absorb so much adsorption-bonded moisture. For instance, the amount of adsorbed moisture in a monomolecular layer, calculated by the BÉT method from the authors' earlier sorption isotherms [2, 3], was found to be in K-120 paper 4.38%, in KON-P paper 4.22%, and in EMTs cardboard 4.6% of the total moisture (referred to the dry weight of material at 296°K).

Moisture and high temperature may effect reversible and irreversible changes in the skeleton structure: shrinkage, dilatation, breakdown, etc. It is well known that the mechanism of transporting a sorbed substance can be different, depending on the sorbent structure and on the conditions under which the sorption process is taking place. Generally, a pressure gradient can produce a laminar flow of vapor through large pores. Within a certain pressure range, where the length of the mean-free path is approximately equal to the diameter of the capillary, there is a possibility of molecular vapor flow. Other possible modes

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TABLE 1. Diffusivity of Electrical Insulation Materials

K-120			KON-P			Cardboard EMTs			Cardboard A			Cardboard B		
$D\cdot10^6$, cm^2/h		$u\cdot10^2$ kg/kg	$D\cdot10^8$, cm^2/h		$u\cdot10^2$, kg/kg	$D\cdot10^4$, cm^2/h		$u\cdot10^2$, kg/kg	$D\cdot10^4$, cm^2/h		$u\cdot10^2$, kg/kg	$D\cdot10^4$, cm^2/h		$u\cdot10^2$, kg/kg
from [4]	from [5]		from [4]	from [5]		from [4]	from [5]		from [4]	from [5]		from [4]	from [5]	
a) Sorption of water vapor from the air under atmospheric pressure and at T = 296°K														
0,58	0,71	1,5				1,53	1,6	2,02	2,98	3,15	2,38	1,96	2,12	1,8
1,15	1,16	2,35		1,28	2,7	1,71	1,89	2,94	3,27	3,43	3,54	3,40	3,90	4,02
1,85	1,91	4,34		1,35	3,9	1,79	2,05	3,83	3,55	3,68	4,3	3,90	4,20	4,7
1,75	1,68	5,7		1,34	4,85	1,86	2,23	4,35	4,11	4,21	5,45	4,30	4,24	4,95
1,66	1,51	6,55		1,14	7,8	2,12	2,27	5,5	4,29	4,46	8,0	4,7	4,46	8,0
1,59	1,47	6,84		1,05	9,0	2,18	2,40	6,02	4,35	4,68	8,74	5,15	5,05	10,0
1,03	1,05	10,5		1,0	10,0	2,18	2,41	6,64	3,47	3,50	14,24	4,60	4,95	10,6
0,84	0,85	13,5		0,81	15,4	2,16	2,71	7,46	2,30	2,48	20,95	3,16	3,64	14,3
0,62	0,56	22,2				2,60	3,04	10,3				1,83	2,28	20,75
						2,33	2,65	12,37						
						1,61	1,76	19,3						
b) Sorption of water vapor under vacuum														
T=296°			T=299°			T=302°								
60	60	3,93	42	37	3,39	28	25	3,25						
68	74	4,1	70	67	6,24	36	34	4,97						
86,5	90	4,18	90	86	12,52									
119	109	5,15	76	69	14,1									
78	74	9,46												
53	63	23,0												
						T=318°								
81		1,53	72		1,36	43	1,73							
94		1,73	84		1,50	57	2,98							
101		1,8	126		3,05									
148		3,2	153		3,34									
220		4,1	208		5,28									
204		4,26												
						T=343°								
146		0,72	127		0,91	60		0,76						
151		0,96	141		0,94	102		1,40						
172		1,12	176		1,42									
256		1,92	173		1,46									
260		2,1	228		1,86									
280		2,1	236		2,05									
						T=363°								
240		0,41	222		0,52	154		0,45						
242		0,46	282		0,88	164		0,47						
379		0,98	308		0,90	175		0,93						
405		1,22	270		1,01									
			290		1,18									
			370		1,30									

of mass transport are migration of molecules along the pore surface (surface diffusion), laminar flow in the adsorption phase, displacement of capillary-condensate moisture by capillary forces, and osmotic flow of moisture. It may be assumed that there are several paths along which moisture is transported through a cellulosic material with a complex structure, as those materials under consideration here, and that the hygrodiffusivity is, in effect, the transfer coefficient.

The relation D = f(u, T) and numerical values of diffusivity were obtained for the said materials on the basis of sorption-kinetics curves according to [2, 3].

The sorption-kinetics curves for water vapor in electrical insulation materials had been plotted under atmospheric pressure by the strain-gage method and under vacuum with the vacusorption apparatus shown in [3]. The tests have revealed that pure water vapor is absorbed much faster (50 to 150 times) under vacuum than under atmospheric pressure, all other conditions remaining the same.

Meanwhile, the general trend of sorption-kinetics curves for water vapor under vacuum is similar to the trend of those for a vapor—air mixture under atmospheric pressure. After an initial steep rise, the rate of moisture absorption decreases and the process gradually approaches an equilibrium corresponding to the test conditions.

The diffusivity was calculated from the sorption-kinetics curves, by the sorption method [4] as well as by the method of equilibrium moisture level [5] and by the method shown in [6]. All these methods are

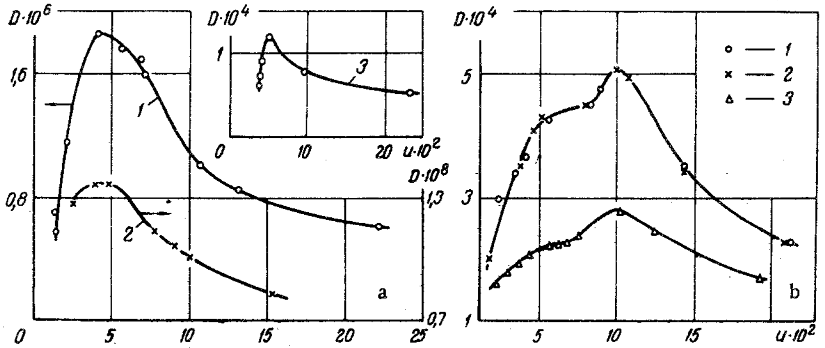

Fig. 1. · Diffusivity (cm^2/h) as a function of the moisture content (kg/kg) in a material, (a) for electrical-grade paper K-120 and KON-P: 1) K-120, sorption of water vapor from air under atmospheric pressure; 2) KON-P; 3) K-120, sorption of water vapor under vacuum); (b) for electrical-grade cardboard ÉMTs, A, and B (sorption under atmospheric pressure): 1) cardboard A; 2) cardboard B; 3) cardboard ÉMTs.

eminently suitable for determining the diffusivity in thin anisotropic fibrous materials such as our electrical-grade cellulosic specimens. It is assumed here that, during sorption under isothermal conditions, a change in the moisture content in a thin film occurs in accordance with the equation of diffusion:

$$\frac{\partial u}{\partial \tau} = D\frac{\partial^2 u}{\partial x^2}. \tag{1}$$

The moisture absorption process will now be examined under the following constraints: the specimen to be moistened is initially dry; the moisture content at the surface of a specimen corresponds to equilibrium, immediately after the sorbent volume has been filled with vapor or after the specimen has been placed in a chamber; and the moisture distribution is, at any instant of time, symmetrical at distance x within the film thickness. It will also be assumed that, while the moisture content u varies with time, within a definite rather short time period the diffusivity remains independent of the moisture content.

It is appropriate to note, at this point, that all these methods of test data evaluation have given close values for the diffusivity D.

In Table 1 are listed values of diffusivity at various temperatures and moisture levels, based on test data pertaining to sorption of water vapor in electrical-grade paper KON-P and K-120 as well as in electrical-grade cardboard ÉMTs, A, and B under atmospheric pressure (at a temperature of 296°K), also in grades KON-P, K-120, and ÉMTs under vacuum (at temperatures of 296, 318, 343, 363°K). In order to show that these methods of test data evaluation yield close results, we have included in Table 1, for illustration, numerical values of the diffusivity D according to the formulas given in [4, 5].

The values of diffusivity calculated by the method of equilibrium moisture level correspond to the same level in all cases, namely to 70% of equilibrium moisture.

The obtained values agree with those given in [1] for unsoaked electrical-grade cardboard within the range $D = (0.7\text{–}5.0)\cdot 10^{-4}$ cm^2/h.

These test data have made it possible to analyze the relation between diffusivity and the state variables of the material. The diffusivity is shown in Fig. 1 as a function of the moisture content, on the basis of the test data for electrical-grade papers KON-P and K-120, also for electrical-grade cardboard ÉMTs, A, and B in Fig. 1b. The data were obtained at a temperature of 296°K, with moisture sorption from a vapor—air mixture under atmospheric pressure.

As is evident from Fig. 1 (a, b), the functional relation between diffusivity and moisture content is a complex one in the case of our test materials.

It is well known that the mechanism of moisture transport in colloidal capillary-porous materials depends, within the hygroscopic range, on the type of bond between the moisture and the matrix.

Adsorption-bonded moisture, being the most strongly bonded kind of moisture, diffuses in the form of vapor [7]. The internal diffusivity depends on the quantity dP/du, which is proportional to the slope with respect to the sorption isotherm [7]:

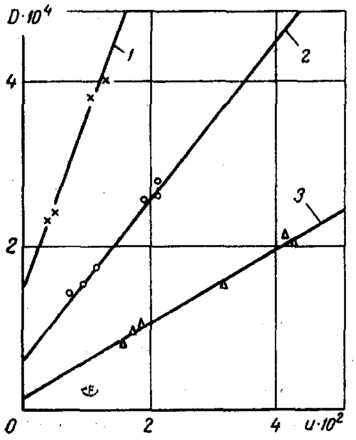

Fig. 2

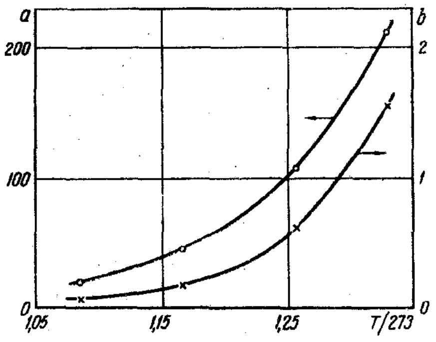

Fig. 3

Fig. 2. Diffusivity (cm^2/h) as a function of the moisture content (kg/kg), for grade K-120 paper with a moisture content within the 0.004–0.04 range (sorption under vacuum): 1) at T = 363°K; 2) T = 343°K; 3) T = 318°K.

Fig. 3. Empirical relations *a* and b versus the temperature, for grade K-120 paper.

$$D = \frac{D_V \mu_V}{RT\gamma_0} \cdot \frac{\partial P}{\partial u}, \qquad (2)$$

where R denotes the gas constant R, μ_V denotes the molecular weight of vapor, and γ_0 denotes the specific weight of dry material.

It follows from Fig. 1a that the diffusivity of electrical-grade paper K-120 and KON-P with a low moisture content (u < 0.0444) increases with increasing moisture content. The $D = f(u)_{T=296°K}$ curve for cable-grade paper has a peak at u = 0.044 kg/kg which, apparently, corresponds to a completely filled monomolecular layer; then, as the material becomes moister, the diffusivity decreases. According to the sorption isotherms, dP/du also increases with increasing moisture content, until the monomolecular layer has been completely filled, and then decreases — all this being reflected in the trend of the $D = f(u)_{T=296°K}$ curve. Indeed, at moisture levels above u = 0.10 kg/kg the diffusivity decreases somewhat with increasing moisture content and, when u > 0.17 kg/kg, it becomes almost independent of the moisture content in the material. This can be explained as follows. Since the test materials are colloidal capillary-porous ones, hence one may assume that both the adsorption-bonded moisture and the microcapillary moisture will be transported after the monomolecular layer has been filled. The change in the internal diffusivity during moisture transport in microcapillaries can be analyzed on the basis of the following expression [7]:

$$D_c = \frac{2\sigma D_V \mu_V \gamma_V P_M \cos\theta}{RT\gamma_L\, gP_s r_x^2 f(r_x)\gamma_0}, \qquad (3)$$

with the coefficient of surface tension σ, the wetting angle θ, the acceleration due to gravity g, the radius of a capillary r_x, the pore size (radius) distribution function $f(r_x)$, the specific weight of vapor and liquid γ_V and γ_L respectively, the vapor pressure in microcapillaries P_M, and the saturation vapor pressure at a given temperature P_s.

As the moisture content in a material increases, the vapor pressure in capillaries P_M increases and capillaries with larger radii r_x fill up while, as the radius r_x increases, the distribution function $f(r_x)$ decreases fast, especially when there are many microcapillaries of one kind. All these factors contribute to an increase in diffusivity during the transport of capillary-bonded moisture.

On the whole, the $D = f(u)_{T=296°K}$ curve for cable paper is similar to a typical curve for a colloidal material with predominantly adsorption-bonded vapor, but it is noteworthy that D decreases slower here than along such a typical curve [7], as u increases, and this is due to the microcapillaries. At a moisture level above 0.17 kg/kg, one may assume, the transport of capillary and osmotic water predominates.

The $D = f(u)_{T=296°K}$ curve for KON-P capacitor paper follows the same trend, except that it is smoother. On the whole, the diffusivity of this denser KON-P paper depends on the moisture content less than the more porous K-120 paper. The trend of the diffusivity, as a function of the moisture content is different in the case of electrical-grade cardboards.

According to Fig. 1b, the $D = f(u)_{T=296°K}$ curves for cardboards A and B, both of about equal density, do almost coincide. The $D = f(u)_{T=296°K}$ curve for the denser cardboard ÉMTs is, on the whole, similar to the respective curves for cardboards A and B, but the absolute values of the diffusivity are approximately one half the corresponding values of D for cardboards A and B.

At a low moisture content ($u < 0.05$) the diffusivity D increases with increasing moisture content u and then stabilizes within the $u = 0.06-0.08$ kg/kg range, beyond which it increases sharply until $u = 0.10$ kg/kg. Then, as the moisture content increases further, D decreases. At a moisture level $u < 0.06$ kg/kg the transport of moisture is, apparently, governed by the adsorption-bonded moisture, at $u = 0.06-0.08$ kg/kg there occurs transport of the absorption-bonded moisture and partial filling of microcapillaries, while at $u \approx 0.10$ kg/kg microcapillaries and pores of one kind become filled at a fast rate.

A further rise in the moisture level in the sorbent volume causes the diffusivity to decrease. The trend of a $D = f(u)_{T=296°K}$ curve within this range of moisture levels depends on the shape and the depth of pores and capillaries, on the ratio of their radii, etc. The evident difference between the $D = f(u)_{T=296°K}$ characteristics of the various electrical-grade papers and cardboards, even though the sorption isotherms of these materials are quite similar, can, it seems, be explained by the different skeleton structures of these materials. In capacitor and cable papers there are no throughfeed pores, according to the data in [8, 9], because the fibers which form pores are oriented parallel to the outside surfaces of the paper sheet. In electrical-grade cardboard which is 300 times thicker than capacitor paper, on the other hand, one may assume some amount of interconnected throughfeed pores.

The curve of diffusivity versus moisture content for grade K-120 cable paper under vacuum ($T = 296°K$, $0.039 < u < 0.23$ kg/kg) has a shape similar to that of such a curve representing the sorption of water vapor from air under atmospheric pressure.

It follows from its values listed in Table 1 that the diffusivity of the test materials during sorption of water vapor is 20-50 times higher under vacuum than under atmospheric pressure. This is, apparently, because during moisture sorption by specimens under vacuum the molecules of water vapor can penetrate unimpeded into micro- and macropores, to be then sorbed at the air-free surface of the solid cellulose skeleton.

While testing the sorption of water vapor under vacuum, we were concerned with vapor pressures up to 20 mm Hg, corresponding to the practical range of pressures for the desiccation treatment of electrical insulation materials. In order to explore the temperature characteristics of diffusivity, we performed the tests at four different temperatures T. The said range of vapor pressure corresponded to a variation of the moisture content from approximately 0.0001 to 0.015 kg/kg at 363°K and to 0.24 kg/kg at 296°K.

In Fig. 2 is shown the diffusivity as a function of the moisture content, based on the sorption of water vapor in cable paper under vacuum at temperatures of 363, 343, and 318°K and with the moisture content varying from 0.0041 to 0.04 kg/kg. According to this diagram, within the said range of moisture levels the diffusivity D increases with increasing moisture content u at each test temperature and this relation may, to the first approximation, be regarded as linear:

$$D = au + b, \tag{4}$$

with both a and b being functions of the temperature. As a specific example, we show in Fig. 3 the curves $a = f(T)$ and $b = f(T)$ based on the curve in Fig. 2 for grade K-120 paper. A quantitative evaluation of these graphs has yielded the following empirical formulas:

$$a = 7.5\,(T/273)^{11.7}, \tag{5}$$

$$b = 0.016\,(T/273)^{16}. \tag{6}$$

Also for the other test materials, characteristically, the diffusivity has been found to depend strongly on the temperature (the power exponent of T in the empirical formulas $a = f(T)$ and $b = f(T)$ for grade KON-P paper and grade ÉMTs cardboard lies within the 11-16 range).

An evaluation of the test data has yielded the following universal equation for the diffusivity, in sorption of water vapor by electrical-grade paper under vacuum (over the 296-363°K temperature range and at moisture levels up to 0.04 kg/kg). For grade K-120 cable paper we have

$$D \cdot 10^4 = 7.5\,(T/273)^{11,7}u + 0.016\,(T/273)^{16}\ \text{cm}^2/\text{h}, \tag{7}$$

and for grade KON-P capacitor paper we have

$$D \cdot 10^6 = 6.2\,(T/273)^{11,7}u + 0.013\,(T/273)^{16}\ \text{cm}^2/\text{h}. \tag{8}$$

The values obtained here for the coefficients of internal mass transfer as well as the empirical relations obtained here between hygrodiffusivity and the state variables of the material may be useful for analyzing the moisture treatment and the desiccation of electrical-insulation cellulosics, also for calculating the process parameters in the desiccation of electrical-insulation cellulosics under atmospheric pressure and under vacuum.

NOTATION

u	is the moisture content (level) in the material, $\text{kg}_{\text{moist}}/\text{kg}_{\text{dry}}$;
τ	is the time, h;
D	is the diffusivity, cm^2/h;
T	is the absolute temperature, °K;
a, b	are the empirical parameters;
P	is the vapor pressure, mm Hg.

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