

*Dimensionless relations, theoretical formulas, and digital filtration of the current experimental information are proposed for the express determination of the diffusion coefficients and solubility in polymers.*

The calculation of kinetic parameters of sorption in polymers (the diffusion coefficient  $D$  and solubility  $H$ ) is based on a mathematical model described by the solution of the Fourier—Fick differential equation [1]

$$\frac{\partial c(x, \tau)}{\partial \tau} = D \frac{\partial^2 c(x, \tau)}{\partial x^2} \quad (1)$$

Equation (1) is used, for example, both to find the wetness characteristics of polymers and to calculate the time of moisture protection from the penetration of moisture through the coating or packing layer. For many technical applications, this mathematical model gives satisfactory agreement of experiment and calculation, within certain limitations. However, the wide use of theoretical methods of estimating the efficiency of moisture protection is prevented by the lack of an express method of measuring the kinetic parameters of moisture sorption by polymers. The calculation of the kinetic parameters from the initial section of the sorption-kinetics curves proposed here permits the creation of such a method.

The solution of Eq. (1) by the classical method may be written in the form of an infinite series; the number of terms of the series required to achieve a specified accuracy increases sharply with decrease in time. The use of operational methods and Laplace transformation in solving Eq. (1), as shown in [1], allows convenient approximations to be obtained for both small and large values of the Fourier number. Thus, for an infinite plate of thickness  $2R$  in a medium with a partial vapor pressure  $p$ , the solution of Eq. (1) is obtained in two forms

$$\theta(x, \tau) = 1 - \sum_{n=1}^{\infty} \frac{2}{\mu_n} (-1)^{n+1} \cos \mu_n \frac{x}{R} \exp(-\mu_n^2 Fo), \quad (2)$$

$$\theta(x, \tau) = \sum_{n=1}^{\infty} (-1)^{n+1} \left\{ \operatorname{erfc} \frac{2n-1-\frac{x}{R}}{2\sqrt{Fo}} + \operatorname{erfc} \frac{(2n-1)+\frac{x}{R}}{2\sqrt{Fo}} \right\}. \quad (3)$$

Here

$$\theta(x, \tau) = \frac{c(x, \tau) - c_0}{c_{\max} - c_0}; \quad Fo = \frac{D\tau}{R^2};$$

the boundary conditions are

$$c(x, 0) = 0; \quad c(\pm R, \tau) = Hp = c_{\max}; \quad \frac{\partial c(0, \tau)}{\partial x} = 0.$$

For  $Fo \leq 0.1$  consideration may be restricted to the first term of the series in Eq. (2), and for  $Fo > 0.1$  to the first term of the series in Eq. (3). The sorption-kinetics curve may be approximated on these two sections by the following expressions for  $\theta = \Delta g_r / \Delta g_{\max}$

$$Fo \leq 0,1: \bar{\theta} = 2 \sqrt{\frac{Fo}{\pi}}, \quad (4)$$

$$Fo > 0,1: \bar{\theta} = 1 - \frac{8}{\pi^2} \exp\left(-\frac{\pi^2}{4} Fo\right). \quad (5)$$

If  $\Delta g_{\max}$  is known, the coefficient H is calculated from the formula

$$H = \frac{\Delta g_{\max} \gamma}{g_0 \rho}, \quad (6)$$

and the coefficient D from one of the formulas

$$\text{for } Fo \leq 0.1 \quad D = \frac{\pi R^2}{4\tau} \bar{\theta}^2, \quad (7)$$

$$\text{for } Fo > 0.1 \quad D = \frac{4R^2}{\pi^2 \tau} \ln \frac{\pi^2}{8} (1 - \bar{\theta}). \quad (8)$$

To calculate the kinetic parameters from Eqs. (6)-(8), the experiment must be continued up to a state of sorbate saturation, at which  $\Delta g_{\max}$  is determined. The Fourier number takes a value of 1.6 here. To reduce the experimental time, the initial section of the sorption-kinetics curve is used. If Eq. (4) cannot, in principle, be used to determine the kinetic parameters for  $Fo \leq 0.1$ , it may be done as follows on the basis of Eq. (5).

For each point of the sorptional curve, Ra is calculated

$$Ra = \frac{\Delta g'_\tau}{\Delta g_\tau} \tau. \quad (9)$$

According to Eq. (9), Ra is 0.5 on the section  $Fo \leq 0.1$  and is related to Fo as follows on the section  $Fo > 0.1$

$$Ra = 2 \frac{Fo \exp\left(-\frac{\pi^2}{4} Fo\right)}{1 - \frac{8}{\pi^2} \exp\left(-\frac{\pi^2}{4} Fo\right)}. \quad (10)$$

The graphical relation of these two numbers is shown in Fig. 1. From Ra, taking account of Eq. (10), D may be determined in the course of the sorptional experiment from the formula

$$D = R^2 Fo / \tau. \quad (11)$$

From Eq. (5),  $\Delta g_{\max}$  is calculated; H is calculated from the formula

$$H = \Delta g_\tau \gamma / \left[ 1 - \frac{8}{\pi^2} \exp\left(-\frac{\pi^2}{4} Fo\right) \right] g_0 \rho. \quad (12)$$

The deficiency of the calculation method proposed above is the need for numerical differentiation of the sorption curve. Fluctuation of the sorption-kinetics curve on account of noise leads to very considerable error. The influence of the noise may be reduced by two methods. The first is to use Rb [2], which is calculated from Eq. (5)

$$Rb = \frac{\bar{\theta}_2}{\bar{\theta}_1} = \frac{1 - \frac{8}{\pi^2} \exp\left(-\frac{\pi^2}{4} Fo_2\right)}{1 - \frac{8}{\pi^2} \exp\left(-\frac{\pi^2}{4} Fo_1\right)}. \quad (13)$$

Here  $Fo_2 = mFo_1$ , where  $m > 1$ .

With increase in m, the accuracy of the method increases, but the time of the experiment also increases. When  $m = 2$ , the dependence  $Rb = f(Fo_1)$  takes the form in Fig. 2, which, like Fig. 1, may be used to calculate D and H from Eqs. (11) and (12).

The second method, which does not lead to increase in length of the experiment, is to use a numerical filter for the kinetic curve obtained in the automated test. Assuming that the noise in the measuring channel is a steady uncorrelated random process with dispersion  $\sigma^2$ , a sixth-order Butterworth low-frequency digital tangent filter [3] is used to increase the accuracy with which  $\Delta g_\tau$  is determined. The amplitude-frequency characteristic of the filter is determined by the expression

$$A(f) = \frac{1}{\sqrt{1 + \left( \frac{2}{\Delta \tau_d f_0} \arctg \frac{\pi f \Delta \tau_d}{2} \right)^{12}}}.$$

This type of filter is chosen because it allows linear amplitude-frequency and phase-frequency characteristics to be obtained in the region of low (working) frequencies, as well as an adequate level of suppression of the high-frequency noise signals. The linearity of the phase characteristic of the Butterworth filter permits filtration without

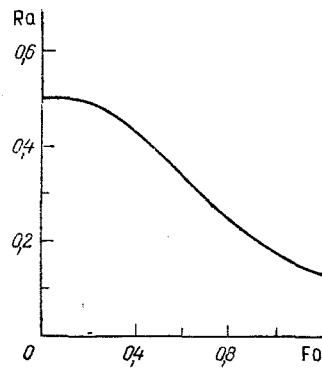


Fig. 1

Fig. 1. Graph of  $R_a = f(F_0)$ .

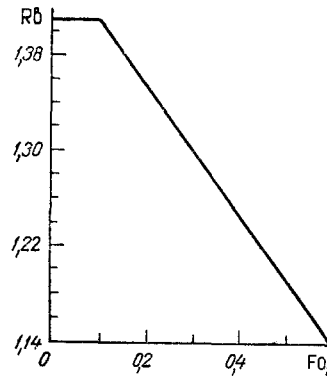


Fig. 2

Fig. 2. Graph of  $R_b = f(F_{01})$ .

distortion in form of the useful signal, and the delay time  $\tau_D$  is compensated in calculating  $R_a$ . The upper frequency limit on the transmission of the filter is determined from the condition of a minimum of the mean square error of filtration

$$f_0 = \arg \min_{f_0} \frac{1}{T} \int_0^T [\Delta \tilde{g}_{f_0, \tau} - \Delta g_\tau]^2 d\tau.$$

The expression for the mean square error of filtration takes the form

$$I = \frac{1}{\pi T} \int_{2\pi f_0}^{+\infty} |\Delta g'(j\omega)|^2 d\omega + \frac{1}{\pi T} \int_0^{2\pi f_0} N(\omega) d\omega. \quad (14)$$

Here

$$|\Delta g'(j\omega)|^2 = \frac{\Delta g_{\max}^2}{1 + \frac{R^2 \omega^2}{D}}; \quad N(\omega) = \sigma^2 \Delta \tau_d T \omega^2.$$

Minimization of the functional in Eq. (14) leads to a theoretical formula for the limiting frequency

$$f_0 = \frac{1}{R} \sqrt[4]{\frac{\Delta g_{\max}^2 D^2}{\Delta \tau_d T \sigma^2}}. \quad (15)$$

Since the true values of  $D$  and  $\Delta g_{\max}$  are unknown prior to the experiment, an a priori estimate on the basis of accumulated experience or literature data may be used in Eq. (15). The values of  $R$  and  $\sigma^2$  are measured prior to the experiment, and  $\Delta \tau_d$  is determined once in certification of the apparatus. The value of  $T$  is specified by the experimenter, taking account of the expected error.

To calculate the derivative at the calculational points of the sorption-kinetics curve, the curve is approximated by the least-squares method [4] by a second-order polynomial and, using the parameters of the polynomial, numerical differentiation is undertaken. It is evident that, with a symmetric interval of approximation  $[\tau - \Delta \tau_a, \tau + \Delta \tau_a]$ , the derivative  $\Delta g'_\tau$  is equal to the coefficient in the linear term of the polynomial and is calculated from the formula

$$\Delta g'_\tau = \frac{1}{5\Delta \tau_a^2} [2\Delta g_{\tau+\Delta \tau_a} + \Delta g_{\tau+\frac{\Delta \tau_a}{2}} - \Delta g_{\tau-\frac{\Delta \tau_a}{2}} - 2\Delta g_{\tau-\Delta \tau_a}].$$

Since the intervals between calculational points of the curve  $\Delta g_\tau$  equal to  $f_0/2$  are optimal after low-frequency filtration,  $\Delta \tau_a = 1/f_0$ .

On the basis of the foregoing, an automated sorptional-experiment procedure using a measurement-computational complex may be developed. Practical experience with the complex shows the possibility of accelerating the determination of the kinetic sorptional parameters by a factor of 10-20 with increase in accuracy and reproducibility by a factor of 3-5.

## NOTATION

$c$ , moisture concentration in polymer,  $\text{g}/\text{m}^3$ ;  $\tau$ , time, sec;  $x$ , coordinate,  $\text{m}$ ;  $D$ , diffusion coefficient,  $\text{m}^2/\text{sec}$ ;  $H$ , solubility,  $\text{sec}^2/\text{m}^2$ ;  $\Delta g_\tau$ , increment in mass of polymer sample at a time  $\tau$  from the onset of sorption,  $\text{g}$ ;  $\Delta g_{\max}$ , increment in mass of sample in a state of sorptional equilibrium,  $\text{g}$ ;  $R$ , half the thickness of the sample plate,  $\text{m}$ ;  $\gamma$ , polymer density,  $\text{g}/\text{cm}^3$ ;  $g_0$ , initial mass of sample,  $\text{g}$ ;  $p$ , partial vapor pressure of sorbate,  $\text{Pa}$ ;  $A(f)$ , amplitude—frequency characteristic of filter;  $\sigma^2$ , dispersion of noise signal in measuring channel,  $\text{g}^2$ ;  $f$ , frequency,  $\text{Hz}$ ;  $\omega$ , circular frequency,  $\text{rad}/\text{sec}$ ;  $\Delta\tau_d$ , interval of discretization in filtration, determined by the length of the analog—digital-converter operating cycle and the time of digital analysis of the signal, sec;  $\tau_D$ , delay time of low-frequency filter, sec;  $f_0$ , upper frequency limit on the transmission of the low-frequency filter,  $\text{Hz}$ ;  $\Delta\bar{g}_\tau$ , increment in sample mass at point  $\tau$  after digital filtration,  $\text{g}$ ;  $T$ , time of experiment, sec;  $|\Delta g'(j\omega)|^2$ , spectral energy density of the derivative of the useful signal,  $\text{g}^2$ ;  $N(\omega)$ , spectral energy density of the derivative of the noise signal,  $\text{g}^2$ ;  $\tau$ , mean square error of filtration,  $\text{g}^2$ ;  $\Delta\tau_a$ , half the interval of approximation of the sorption-kinetics curve by a second-order polynomial, sec.

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## CONVECTIVE INSTABILITY OF A FREE-CONVECTION VORTEX

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*The stability of free-convection vortex formations obtained in the laboratory is compared with natural tropical cyclones.*

Unstable motion in rotating systems is the explanation for a wide class of phenomena such as tropical cyclones [1-3]. In the present paper we demonstrate the possibility of using laboratory experiments on a model of a free-convection vortex to determine the stability of catastrophic atmospheric vortices of the tropical cyclones type.

The subject of the laboratory study was a strong air-water-vapor vortex in a temperature-stratified flow [3, 4]. The main difference between this model and those studied in [3, 5] is that in this model the role of horizontal velocity gradients in vortex formation is isolated. A vortex with a vertical axis was created in a vortex cell with tangential windows (Fig. 1). Water was heated to nearly the boiling point (70–80°C) in the lower part of the apparatus 1. Humid air formed by evaporation fills a cylindrical column which is forced to rotate at a certain angular velocity. Its degree of twist was measured by the angle at which air masses enter the cell through the tangential windows 2 on the lateral surface. A horizontal velocity shear was created by a system of adjustable cylindrical rings 3 in the upper part of the apparatus. The experimental information was obtained with the help of optical visualization and laser-Doppler anemometry [6].

Analysis of this information [4, 5] confirms that free-convection vortex formations can be found in stable and unstable states. There are two parameters controlling the stability of these vortices. One is the flow angle  $\alpha$  (the angle between the velocity vector and the isobars). The second parameter is the relative radius  $x_0 = r_0/R_0$  at which the horizontal velocity shear is imparted. Here  $r_0$  is the radius of the opening of the cell from above and  $R_0$  is the outer radius of the device (Fig. 1).

By analyzing tables of stable and unstable states of a free-convection vortex and from the measurements of the radial distribution of the velocity a relation can be established between the quantity  $x_0$  and the parameter  $B$  introduced

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