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IDENTIFICATION OF THE POLYRELAXATIONAL NONLINEAR DIFFERENTIAL MASS DIFFUSION OPERATOR

A. G. Temkin
Polytechnical Institute, Riga, U.S.S.R.

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Abstract - Diffusion in a non-homogeneous mixture is described by a polyrelaxational equation involving the diffusion coefficient which depends on concentration and several physical parameters. A set of the relaxation time spectra and diffusion coefficients (as functions of concentration) form a polyrelaxational nonlinear differential operator of mass diffusion. The method presented for the identification of this operator consists of the experimental determination of the relaxation time spectrum and of all the diffusion coefficient parameters. As initial information, the time derivative of the mass moment of the diffusing substance, and a substantially nonhomogeneous initial distribution of concentrations in an insulated rod of any shape but constant cross section, are used.

NOMENCLATURE

$u(x, \tau)$,	mass concentration [kg m^{-3}];
x,	abscissa [m];
τ,	time; actual physical time [s];
b,	plate (rod) thickness; rod length [m];
$M(\tau)$,	diffusing substance mass moment [kg m^{-1}];
U,	total mass content [kg m^{-2}];
D,	diffusion coefficient [m^2 s^{-1}];
Π_π,	parameter of diffusion coefficient [m$^{3\pi}$ kg$^{-\pi}$];
K,	diffusion analogue of the Kirchhoff–Varshavsky function; mass transfer potential [kg m^{-1} s^{-1}];
R_ρ,	relaxation time [s].

Subscripts

π,	number of the diffusion coefficient parameter;
p,	number of parameters of the diffusion coefficient;
ρ,	relaxation time order;
r,	number of different times of relaxation; order of polyrelaxation;
i,	number of the experiment or of the moment of measurements;
J,	number of different experiments or moments of measurements;
k,	number of the moment of measurements in the ith experiment, $k = 1$; $\kappa(i)$.

IT HAS been shown [1] that during the equalization of an initially non-uniform distribution of concentrations

$$u(x, 0) = u_i(\tau) = \text{var} \neq \text{const.,} \tag{1}$$

provided that constant concentrations

$$\begin{aligned} u(x, 0) &= u_{01} = \text{const.,} \\ u(b, \tau) &= u_{02} = \text{const.} \end{aligned} \tag{2}$$

are maintained at the insulated plate end faces 0 and b, i.e. there is no mass flux $q(x, \tau)$ through these surfaces

$$q(0, \tau) = q(b, \tau) = 0, \tag{3}$$

and that the moment of the diffusing substance mass can be measured accurately in the course of the experiment

$$M(\tau) = \int_0^b u(x, \tau)x \, \mathrm{d}x, \tag{4}$$

the first relaxation time R_1 and the constant diffusion coefficient D_0.

Clearly, equation (3) yields the constancy of the total mass content

$$U(\tau) = \int_0^b u(x, \tau) \, \mathrm{d}x = \text{const.} \tag{5}$$

In the present paper, a technique is presented for determining the relaxation time spectrum R_ρ ($\rho = 1, 2, \ldots, r$) and the variable diffusion coefficient $D(u, \Pi_\pi)$, which depends on the diffusing substance concentration, u, and p physical parameters Π_π ($\pi = 1, 2, \ldots, p$), when the initial distribution of concentrations is non-uniform (1), the plate end faces are insulated (3), the mass moment of the diffusing substance (4) is determined experimentally, and its concentrations at the plate ends are known

$$u(0, \tau) = u_0(\tau); \quad u(b, \tau) = u_b(\tau). \tag{6}$$

For polyrelaxational diffusion [2, 3], the diffusing substance concentration gradient will be represented in the form of a series expansion in terms of the flux derivatives $q(x, \tau)$ [kg m^{-2} s^{-1}] and R_ρ^ρ relaxation times

$$-D\frac{\partial u}{\partial x} = q + R_1\frac{\partial q}{\partial \tau} + R_2^2\frac{\partial^2 q}{\partial \tau^2} + \cdots + R_r^r\frac{\partial^r q}{\partial \tau^r}. \tag{7}$$

The condition (3) of plate protection from external effects is sufficient for the concentration gradient to be absent at the end faces

$$\partial u(0, \tau)/\partial x = 0, \quad \partial u(b, \tau)/\partial x = 0 \tag{8}$$

as follows from the definition of polyrelaxational diffusion (7). However, for substance diffusion the condition (3) is not needed. The fact that at a certain plane, $x = \text{const.}$, the concentration gradient is zero does not call for the absence of mass flux in a polyrelaxational process. This flux can exist and satisfies the following homogeneous equation:

$$\sum_{\rho=0}^{r} R_\rho^\rho \frac{\partial^\rho q}{\partial \tau^\rho} = 0. \tag{9}$$

This distinguishes mass diffusion from the diffusion of energy. In the latter case, the zero concentration or temperature gradient is necessary and sufficient for the heat flux and its time derivative (2) to be zero.

The mass conservation law for polyrelaxational and non-linear diffusion

$$\sum_{\rho=0}^{r} R_\rho^\rho \frac{\partial^{\rho+1} u}{\partial \tau^{\rho+1}} = \frac{\partial}{\partial x}\left[D(u,\Pi_\pi)\frac{\partial u}{\partial x}\right] \tag{10}$$

has a differential operator which depends on the spectrum r of the relaxation times R_ρ and p parameters, Π_p, of the diffusion coefficient $D(u,\Pi_1,\Pi_2,\ldots,\Pi_p)$. Multiplication of all the terms of equation (10) by $x\,\mathrm{d}x$ and integration over the plate thickness transforms the LHS of the diffusion equation (10) into a linear combination of time derivatives of the diffusing substance's mass moment

$$\int_0^b \sum_{\rho=0}^{r} R_\rho^\rho \frac{\partial^{\rho+1} u}{\partial \tau^{\rho+1}} x\,\mathrm{d}x = \sum_{\rho=0}^{r} R_\rho^\rho M^{(\rho+1)}(\tau). \tag{11}$$

As for the RHS of the diffusion equation,

$$\int_0^b \frac{\partial}{\partial x}\left[D(u,\Pi)\frac{\partial u}{\partial x}\right] x\,\mathrm{d}x, \tag{12}$$

to calculate its integral, it is advisable to introduce the diffusion analogue of the Kirchhoff function

$$K(u,\Pi_\pi) = \int_0^u D(u,\Pi_\pi)\,\mathrm{d}u, \tag{13}$$

the gradient of which is proportional to the concentration gradient of the substance transferred

$$\frac{\partial K}{\partial x} = D(u,\Pi)\frac{\partial u}{\partial x}. \tag{14}$$

For this reason, by virtue of equation (8), the gradient of the diffusion analogue of Kirchhoff's function on the surfaces of an insulated plate is also equal to zero,

$$\int_0^b \frac{\partial}{\partial x}\left[D(u,\Pi)\frac{\partial u}{\partial x}\right] x\,\mathrm{d}x = K[u(0,\tau),\Pi] - K[u(b,\tau),\Pi]. \tag{15}$$

Consequently, the integration by parts of equation (12) yields the difference

$$\int_0^b \frac{\partial}{\partial x}\left[D(u,\Pi)\frac{\partial u}{\partial x}\right] x\,\mathrm{d}x = K[u(0,\tau),\Pi] - K[u(b,\tau),\Pi] \tag{16}$$

between the Kirchhoff function values on the plate surfaces.

Thus, the use of the diffusing substance mass moment (2) provides an equation

$$\sum_{\rho=0}^{r} R_\rho^\rho M^{(\rho+1)}(\tau) = K[u(0,\tau),\Pi_\pi] - K[u(b,\tau),\Pi_\pi] \tag{17}$$

which interrelates the r relaxation times R_ρ and p parameters Π_π of the diffusion coefficient $D(u,\Pi_\pi)$ or of the diffusion analogue of Kirchhoff's function, $K(u,\Pi_\pi)$. For all these $(p+r)$ parameters of the polyrelaxational non-linear differential operator of the diffusion equation (10) to be determined, a minimum of

$$J \geqslant J_{\min} = p+r \tag{18}$$

independent equations will be needed.

A set of equations for the parameters R_ρ and Π_π can be obtained in a number of ways.

First, one can use different initial distributions of concentrations $u_i(x)$ $(i = 1,2,\ldots,J)$ to which there correspond different moments.

$$M_i(\tau) = \int_0^b u_i(x,\tau)x\,\mathrm{d}x, \tag{19}$$

since each initial distribution of $u_i(x)$ under the conditions of insulation (4) generates its own field of concentrations $u_i(x,\tau)$.

If the number of initial distributions J is at a minimum, i.e. $J = J_{\min} = r+p$, then the system of equations (17) in this, first, case has the form

$$\delta_i(\tau_i) \equiv \sum_{\rho=0}^{r} R_\rho^\rho M_i^{(\rho+1)}(\tau_i) + K[u_i(b,\tau_i),\Pi_\pi] - K[u_i(0,\tau_i),\Pi_\pi] = 0. \tag{20}$$

If $J > J_{\min}$, then the system (17) is redefined and can be very conveniently solved by the least squares method when the diffusion coefficient is approximated by the McLauren polynomial

$$D(u,\Pi_\pi) = D_0 + D_0'\cdot u + \cdots + D_0^{(p-1)}\cdot\frac{u^{p-1}}{(p-1)!} \tag{21}$$

and the Kirchhoff diffusion function is

$$K(u,\Pi_\pi) = D_0 u + D_0'\frac{u^2}{2} + \cdots + D_0^{(p-1)}\frac{u^p}{p!}. \tag{22}$$

It is understood here that the diffusion coefficient parameters

$$\Pi_\pi = D_0^{(\pi-1)}, \quad \pi = 1,2,\ldots,p \tag{23}$$

coincide with its derivatives with respect to concentration.

In order to determine the diffusion operator parameters, the mean-square deviation

$$\delta^2 = \frac{1}{J}\sum_{i=1}^{\nu}\delta_i^2 = \frac{1}{J}\sum_{i=1}^{J}\left\{\sum_{\rho=0}^{r} R_\rho^\rho M_i^{(\rho+1)}(\tau_i) + \sum_{\pi=1}^{p} D_0^{(\pi-1)}\left[\frac{u_i^\pi(b,\tau_i)-u_i^\pi(0,\tau_i)}{\pi!}\right]\right\}^2 \tag{24}$$

is minimized with respect to the spectrum of relaxation times, R_ρ, and to the mass-content derivatives of the diffusion coefficient $D_0^{(\pi-1)}$. Using the notation

$$A_{\rho j} = A_{j\rho} = \sum_{i=1}^{J} M_i^{(\rho+1)}(\tau_i) M_i^{(j+1)}(\tau_i), \tag{25}$$

$$B_{j\pi} = \sum_{i=1}^{J} [u_i(b,\tau_i) - u_i^\pi(0,\tau_i)] M_i^{(j+1)}(\tau_i)/\pi!, \tag{26}$$

$$\Gamma_{\mu\pi} = \Gamma_{\pi\mu} = \sum_{i=1}^{J} [u_i^\pi(b,\tau_i) - u_i^\pi(0,\tau_i)] \times [u_i^\mu(b,\tau_i) - u_i^\mu(0,\tau_i)]/(\pi!\,\mu!) \tag{27}$$

the system of minimization equations

$$\partial\delta^2/\partial R_\rho = 0, \quad \partial\delta^2/\partial \Pi_\pi = 0 \tag{28}$$

is reduced to the form

$$\sum_{\rho=0}^{r} R_\rho^\rho A_{j\rho} + \sum_{\pi=1}^{p} \Pi_\pi B_{j\pi} = 0, \quad j = 1,2,\ldots,r, \tag{29}$$

$$\sum_{\rho=0}^{r} R_\rho^\rho B_{\rho\mu} + \sum_{\pi=1}^{p} \Pi_\pi \Gamma_{\pi\mu} = 0, \quad \mu = 1,2,\ldots,p. \tag{30}$$

This system is not homogeneous since the zero relaxation parameter $R_0 = 1$.

Second, one can use a rather prolonged process of concentration equalization with the common time $\tau_{\max}$. During this single experiment, at the time moments

$$0 \leqslant \tau_1 < \tau_2 < \cdots < \tau_J \leqslant \tau_{\max}, \tag{31}$$

the concentrations $u(b,\tau_i)$ and $u(0,\tau_i)$ on the plate end faces are measured and the diffusing substance's mass moment and its time derivatives are calculated. The system (29) and (30) for the determination of all the parameters of the polyrelaxational non-linear differential operator of mass diffusion remains unchanged, but its coefficients $A_{j\rho}$, $B_{j\pi}$ and $\Gamma_{\pi\mu}$ are

$$A_{j\rho} = A_{\rho j} = \sum_{i=1}^{J} M^{(j+1)}(\tau_i) M^{(\rho+1)}(\tau_i), \tag{32}$$

$$B_{j\pi} = \sum_{i=1}^{J} [u^\pi(b,\tau_i) - u^\pi(0,\tau_i)] M^{(j+1)}(\tau_i)/\pi!, \tag{33}$$

$$\Gamma_{\pi\mu} = \Gamma_{\mu\pi} = \sum_{i=1}^{J} [u^\pi(b,\tau_i) - u^\pi(0,\tau_i)] \times [u^\mu(b,\tau_i) - u^\mu(0,\tau_i)]/(\pi!\,\mu!) \tag{34}$$

which relate to the same experiment.

For the diffusion coefficient one can use, of course, a non-polynomial approximation, but then, to determine the parameters of the diffusion operator, a much more complicated and non-linear system is obtained,

$$\sum_{i=1}^{J} \left\{ \sum_{\rho=0}^{r} R_\rho^\rho M^{(\rho+1)}(\tau_i) + K u(b,\tau_i), \Pi_\pi] - K[u(0,\tau_i),\Pi_\pi] \right\} M^{(j+1)}(\tau_i) = 0 \tag{35}$$

$$\sum_{i=1}^{J} \left\{ \sum_{\rho=0}^{r} R_\rho^\rho M^{(\rho+1)}(\tau_i) + K[u(b,\tau_i),\Pi_\pi] - K[u(0,\tau_i),\Pi_\pi] \right\} \times \frac{\partial}{\partial \Pi_\mu} \{K[u(b,\tau_i),\Pi_\mu] - K[u(0,\tau_i),\Pi_\mu]\} = 0,$$
$$j = 1,2,\ldots,r; \quad \mu = 1,2,\ldots,p, \tag{36}$$

the solution of which is very difficult.

It is pertinent to emphasize that the redistribution of concentrations, no matter what the initial distribution (1) is, has one general termination

$$\lim_{\tau\to\infty} u(x,\tau) = \frac{1}{b}\int_0^b u(x,\tau)\,\mathrm{d}x = \frac{1}{b}\int_0^b u_i(x)\,\mathrm{d}x, \tag{37}$$

to which there corresponds the final moment of masses

$$\lim_{\tau\to\infty} M(\tau) = \frac{b}{2}\int_0^b u_i(x)\,\mathrm{d}x \tag{38}$$

and the equality of their concentrations on the plate end faces. This yields

$$\lim_{\tau\to\infty} [u^\pi(b,\tau) - u^\pi(0,\tau)] = 0. \tag{39}$$

Therefore, at large times the differences between the concentration degrees are commensurable with their errors, ε_0 or ε_b, while the mass moment derivatives [equation (2)], are very close to zero, i.e. also comparable with their errors (4).

These factors make it advisable to use an intermediate approach to the determination of the diffusion operator parameters, when each of the J initial distributions is used for a limited time interval, from 0 to $\tau_{i\kappa(\tau)}$, and at the time moments

$$0 \leqslant \tau_{i1} < \tau_{i2} < \cdots < \tau_{i\kappa(\tau)}, \quad k = 1,2,\ldots,\kappa(i), \tag{40}$$

measurements are made of the concentrations $u(b,\tau_{ik})$ and $u(0,\tau_{ik})$ on the plate end faces, of the diffusing substance mass moment

$$M_i(\tau_{ik}) = \int_0^b u_i(x,\tau_{ik}) x\,\mathrm{d}x \tag{41}$$

and of its higher time derivatives, which, at small τ, differ markedly from zero. If each measurement is given the weight c_{ik} and

$$\sum_{i=1}^{J} \sum_{k=1}^{k(i)} c_{ik} = 1 \tag{42}$$

then the mean square deviation of the whole experiment will be determined from the sum

$$\delta^2 = \sum_{i=1}^{J} \sum_{k=1}^{\kappa(i)} c_{ik} \left\{ \sum_{\rho=0}^{r} M_i^{(\rho+1)}(\tau_{ik}) R_\rho^\rho + K[u(b,\tau_{ik}),\Pi_\pi] - K[u(0,\tau_{ik}),\Pi_\pi] \right\}^2. \tag{43}$$

The minimum of δ corresponding to the r relaxation times and p parameters of the diffusion coefficient is also found from the conditions (28) at the values of R_ρ and Π_π which are the solutions of the linear system of equations $(r+p)$ of the order of (29)–(30). But the coefficients A_{ij}, $B_{j\pi}$ and $\Gamma_{\pi\mu}$ of this system are calculated as the sums

$$A_{\rho j} = A_{j\rho} = \sum_{i=1}^{J} \sum_{k=1}^{\kappa(i)} c_{ik} M_i^{(\rho+1)}(\tau_{ik}) M^{(j+1)}(\tau_{ik}), \quad (44)$$

$$B_{j\pi} = \sum_{i=1}^{J} \sum_{k=1}^{\kappa(i)} c_{ik} M^{(j+1)}(\tau_{ik}) \times [u^{\pi}(b, \tau_{ik}) - u^{\pi}(0, \tau_{ik})]/\pi!, \quad (45)$$

$$\Gamma_{\mu\pi} = \Gamma_{\pi\mu} = \sum_{i=1}^{J} \sum_{k=1}^{\kappa(i)} c_{ik}[u^{\pi}(b, \tau_{ik}) - u^{\pi}(0, \tau_{ik})] \times [u^{\mu}(b, \tau_{ik}) - u^{\mu}(0, \tau_{ik})]/(\pi!\, \mu!)$$

$$\pi = 1, 2, \ldots, p, \quad \mu = 1, 2, \ldots, p, \quad j = 1, 2, \ldots, r. \quad (46)$$

For each experiment, which is characterized by a set of distributions at the start of the process $u_i(x)$ and by persistence $\tau_{i\kappa(i)}$ of corresponding fields, the values of the parameters R_ρ and Π_π depend completely on the choice of the numbers r and p, i.e. on the magnitude of the relaxation time spectrum $[R_\rho]$ and the order of the polynomial (21) which approximates the diffusion coefficient D or, in general, on the p number of its parameters Π_π. The choice of the numbers r and p is associated with optimization of the polyrelaxational nonlinear differential mass diffusion operator and is an independent problem.

Each of the sub-experiments i, together with the numbers r and p, provided the insulation conditions (4), the initial conditions

$$\partial^\rho u_i(x, \tau)/\partial\tau^\rho = 0, \quad \rho = 1, 2, \ldots, r \quad (47)$$

or any other type of conditions are fulfilled, makes it possible to show the solution of the diffusion equation (10)

$$u_i(x, \tau) = u(x, \tau, b, i, R_1, \ldots, R_r, \Pi_1, \ldots, \Pi_p). \quad (48)$$

This calculation field depends on the axis x, the plate thickness b, the time τ, the initial distribution of concentrations $u_i(x)$, and the parameters R_ρ and Π_π. The inner transport parameters R_ρ and Π_π are independent of the initial diffusing substance mass distribution and are determined, for all of the calculation fields, from the same system of equations (29) and (30) involving the coefficients (44), (45) and (46). But in this system, until optimization of the differential operator of the diffusion equation (10) is performed, the numbers r and p are selected quite arbitrarily and their choice predetermines both the magnitude of the coefficients and the very parameters of the polyrelaxational nonlinear differential operator of equation (10). In other words, since the values of R_ρ and Π_π depend on the choice of r and p, the calculation field of concentrations

$$u_i(x, \tau) = u(x, \tau, b, i, r, p) \quad (49)$$

depends also on these two numbers. The calculation field of mass contents (48) or (49) can be used to calculate the mass moment

$$M_{\text{cal}}(\tau, b, i, r, p) = \int_0^b u(x, \tau, b, i, r, p) x \, \mathrm{d}x. \quad (50)$$

This calculation moment differs from the experimental one (2)

$$M_{\text{exp}}(\tau, b, i) = \int_0^b u_i(x, \tau) x \, \mathrm{d}x, \quad (51)$$

the measurement of which is associated with the uncertainty ε_M.

Deviation of the predicted moment from the experimental one

$$F[\varepsilon_{\text{cal}}(r, p)] = \sum_{i=1}^{J} \sum_{k=1}^{\kappa(i)} c_{ik} \times F[M_{\text{cal}}(\tau_{ik}, b, i, r, p) - M_{\text{exp}}(\tau_{ik}, b, i)] \quad (52)$$

(here F is an arbitrary function) allows one, in case of the same thickness of specimens, to attribute the discrepancy between the predicted and experimental moments only to the numbers r and p. These numbers can be regarded as optimum when the difference between the instrumentation error of measuring the mass moment, ε_M (its value depends only on the technique and culture of experiment), and the predicted one, $\varepsilon_{\text{cal}}(r, p)$, is minimal

$$|\varepsilon_{\text{cal}}(r, p) - \varepsilon_M| = \min. \quad (53)$$

In other words, the parameters of the differential operator of the equation of polyrelaxational and nonlinear diffusion can be regarded as chosen optimally, if for all the J sub-experiments the deviation of the predicted moment from the experimental one coincides roughly with the experimental error. When this approximate coincidence is observed for two pairs of the numbers (r_1, p_1) and (r_2, p_2), then the operator with fewer parameters seems to be more preferable

$$r + p = \inf(r_1 + p_1\,;\, r_2 + p_2). \quad (54)$$

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IDENTIFICATION DE L'OPERATEUR DE DIFFUSION MASSIQUE DIFFERENTIEL NON-LINEAIRE ET POLYRELAXATIONNEL

Résumé—La diffusion dans un mélange non-homogène est décrite par une équation polyrelaxationnelle contenant le coefficient de diffusion qui dépend de la concentration et d'autres paramètres physiques. Un système de spectres de temps de relaxation et de coefficients de diffusion fonctions de la concentration forme un opérateur différentiel non-linéaire polyrelaxationnel de diffusion massique. La méthode présentée pour l'identification de cet opérateur consiste en la détermination expérimentale du spectre des temps de relaxation et de tous les paramètres du coefficient de diffusion. En première information sont présentés un changement en temps du moment de la masse diffusante et une distribution initiale fortement non homogène de concentration dans un cylindre isolé de section quelconque mais à aire droite constante.

IDENTIFIKATION DES NICHTLINEAREN DIFFERENTIELLEN POLYRELAXATIONS-OPERATORS DER DIFFUSION

Zusammenfassung—Die Diffusion in einem nicht-homogenen Gemisch wird durch eine Polyrelaxations-Gleichung beschrieben, die den Diffusions-Koeffizienten enthält. Dieser hängt von der Konzentration und von mehreren physikalischen Parametern ab. Ein Satz von Relaxations-Zeit-Spektren und Diffusions-Koeffizienten (als Funktion der Konzentration) bilden den nichtlinearen differentiellen Polyrelaxations-Operator der Diffusion. Das Verfahren, das für die Identifikation dieses Operators vorgestellt wird, beruht auf der experimentellen Bestimmung des Relaxations-Zeit-Spektrums und sämtlicher Parameter des Diffusions-Koeffizienten. Als Anfangsinformation werden eine zeitliche Verschiebung des Augenblicks, in dem die Substanz diffundiert, und eine stark nicht-homogene Anfangsverteilung der Konzentrationen in einem isolierten Bereich mit beliebiger Form aber konstantem Querschnitt verwendet.

ИДЕНТИФИКАЦИЯ ПОЛИРЕЛАКСАЦИОННОГО НЕЛИНЕЙНОГО ДИФФЕРЕНЦИАЛЬНОГО ОПЕРАТОРА ДИФФУЗИИ МАССЫ

Аннотация—Диффузия в неоднородной смеси описывается полирелаксационным уравнением с коэффициентом диффузии, который зависит от концентрации и нескольких физических параметров. Совокупность спектра времен релаксации и функциональной зависимости коэффициента диффузии от концентрации образуют полирелаксационный нелинейный дифференциальный оператор диффузии массы. Излагается метод идентификации этого оператора, состоящий в экспериментальном определении спектра времен релаксации и всех параметров коэффициента диффузии. В качестве исходной информации используется изменение во времени момента массы диффундирующего вещества и существенно неоднородное начальное распределение концентраций в изолированном стержне постоянного сечения любой формы.