DIRECT AND INVERSE PROBLEMS OF THE DYNAMICS OF SORPTION IN THE ABSENCE OF EQUILIBRIUM AT THE PHASE BOUNDARY

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The solutions of the direct and inverse problems of nonequilibrium kinetics and dynamics of sorption were obtained. Simple methods of obtaining numerical values of kinetic and dynamic parameters are indicated.

The direct problem of the kinetics and dynamics of sorption under certain constraints has been considered by a number of authors [1-14]. The inverse problem for a given initial distribution of concentrations in an unbounded infinite column (unbounded problem) was examined in [9].

We will consider below the direct and inverse problems of the nonequilibrium kinetics and dynamics of sorption for a bounded column with a zero initial distribution and given temporal distribution of the concentration at the column boundary. This problem is of interest for chromatography and a number of processes in industrial chemistry where the processes occur in bounded columns. The dynamics of sorption in a cylindrical column filled with homogeneous symmetric porous grains is described by the following system of equations:

kinetics of acts of sorption

$$\frac{\partial q^0}{\partial t} = k_1 c^0 - k_2 q^0, \tag{1}$$

material balance for symmetric porous grains

$$\frac{\partial q^0}{\partial t} + \frac{\partial c^0}{\partial t} = D_i \left(\frac{\partial^2 c^0}{\partial r^2} + \frac{\nu}{r} \cdot \frac{\partial c^0}{\partial r} \right), \tag{2}$$

material balance for a cylindrical column filled with sorbent grains

$$\frac{\partial c}{\partial t} + u \frac{\partial c}{\partial z} + \delta_0 \gamma_0 (c - c^0 |_{r=a}) = D \frac{\partial^2 c}{\partial z^2} \tag{3}$$

with zero initial and boundary conditions of:

continuity of the external and internal flows at the grain boundary

$$\beta_0 (c - c^0 |_{r=a}) = D_i \left. \frac{\partial c^0}{\partial r} \right|_{r=a}, \tag{4}$$

symmetry at the grain center

$$\left. \frac{\partial c^0}{\partial r} \right|_{r=0} = 0, \tag{5}$$

for frontal dynamics of sorption

$$c_f (z, t) |_{z=0} = c_0 = \text{const}, \tag{6}$$

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for elution dynamics of sorption

$$c_e(z,\ t)\,|_{z=0} = c_0\delta(t), \tag{7}$$

$$c_e^*(z,\ t)\,|_{z=0} = c_0\,[1-\eta(t-t_0)]. \tag{8}$$

The effective longitudinal mixing for a nonstationary concentration field is described not by the diffusion coefficient but by the dispersion coefficient

$$D = D_d + D_1 u + D_2 u^2. \tag{9}$$

The first term in (9) corresponds to molecular diffusion in the narrow channels between the sorbent grains [15]; the second term is due to convective mixing [16] occurring long before the appearance of turbulent fluctuations in the percolating flow, turbulent mixing [17], and presence of velocity fluctuations of the percolating flow in the porous medium [15]. The third term is governed by the nonuniformity of the distribution of the percolation velocity of the flow with respect to the cross section of the cylindrical column [18] ("Taylor diffusion") and presence of stagnant zones between the sorbent grains [19-23], which are most substantial for a liquid percolating flow. Using the Laplace integral transform with respect to time with consideration of conditions (4), (5), we find the solution of system (1), (2) for $1 < \nu \leq 2$

$$\tilde{c}^0 = \left(\frac{r}{a}\right)^{1/2(1-\nu)} \tilde{c}\,\frac{I_{1/2(\nu-1)}(\lambda r)}{I_{1/2(\nu-1)}(\lambda a)}\left(1+\frac{D_i}{a\beta_0}B\right)^{-1},$$
$$B = \lambda a\,\frac{I_{1/2(\nu-3)}(\lambda a)}{I_{1/2(\nu-1)}(\lambda a)} + 1 - \nu. \tag{10}$$

The average concentration of absorbed substance with respect to the grains is $q = (1+\nu)a^{-(1+\nu)}\int_0^a q^0 r^\nu dr$. With consideration of (1) and (10) for the transforms we write

$$\tilde{q} = (1+\nu)\,a^{-(1+\nu)}\left(\frac{k_1}{p+k_2}\right)\int_0^a \tilde{c}^0 r^\nu\,dr = (1+\nu)(\lambda a)^{-2}\left(\frac{k_1}{p+k_2}\right)B\tilde{c}\left(1+\frac{D_i}{a\beta_0}B\right)^{-1}, \tag{11}$$

since $\int x^{(s+1)} I_s(x)dx = x^{(s+1)} I_{(s+1)}(x)$. We find the original (11) under the condition $c = c_0 = \text{const}$ (accordingly $\tilde{c} = c_0/p$) after repeated transformations

$$q = kc_0 - kc_0\sum_{n=0} P_n(t)\,2\,(1+\nu)(\lambda_n a)^{-2}, \tag{12}$$

where

$$P_n(t) = \xi_{1n}(\xi_{1n}-\xi_{2n})^{-1}\exp(\xi_{2n}t) - \xi_{2n}(\xi_{1n}-\xi_{2n})^{-1}\exp(\xi_{1n}t),$$
$$\xi_{1n,2n} = -1/2\,(k_1+k_2+D_i\lambda_n^2) \pm [1/4\,(k_1+k_2+D_i\lambda_n^2)^2 - k_2 D_i\lambda_n^2]^{1/2}.$$

The roots λ_n are found from the characteristic equation

$$(a\beta_0 D_i^{-1} + 1 - \nu)\,J_{1/2(\nu-1)}(\lambda a) + \lambda a J_{1/2(\nu-3)}(\lambda a) = 0.$$

The concentration of the absorbed substance in the sorbent grain

$$q^0 = kc_0 - 2kc_0\sum_{n=0}(\lambda_n a)^{-1} P_n(t)\left(\frac{r}{a}\right)^{1/2(\nu-1)} J_{1/2(\nu-1)}(\lambda_n r)\,J^{-1}_{1/2(\nu+1)}(\lambda_n a) \tag{13}$$

and the concentration of sorbate within the sorbent grain

$$c^0 = c_0 - 2c_0\sum_{n=0}(\lambda_n a)^{-1} N_n(t)\left(\frac{r}{a}\right)^{1/2(\nu-1)} J_{1/2(\nu-1)}(\lambda_n r)\,J^{-1}_{1/2(\nu+1)}(\lambda_n a), \tag{14}$$

where

$$N_n(t) = \frac{\xi_{2n}}{k_2}\left(\frac{\xi_{1n}+k_2}{\xi_{1n}-\xi_{2n}}\right)\exp(\xi_{1n}t) - \frac{\xi_{1n}}{k_2}\left(\frac{\xi_{2n}+k_2}{\xi_{1n}-\xi_{2n}}\right)\exp(\xi_{2n}t).$$

For $c = c_0[1-\eta(t-t_0)]$ (accordingly $\tilde{c} = c_0 p^{-1}[1-\exp(-pt_0)]$) we obtain from (11)

$$\bar{q} = (1+\nu)(\lambda a)^{-2}\left(\frac{k_1}{p+k_2}\right) B \left(1+\frac{D_i}{a\beta_0} B\right)^{-1} c_0 p^{-1}[1-\exp(-pt_0)]. \tag{15}$$

The solutions of (12)-(14) represent the solutions of the direct problem of nonequilibrium kinetics of sorption. To find from the usual kinetic curve (boundary conditions $c = c_0 = \text{const}$) the kinetic curve (boundary conditions $c = c_0[1 - \eta(t - t_0)]$) from which we can determine the statistical moments, we must use the relationships

$$q_*(t) = q(t) - q(t-t_0), \quad q_*^0(t) = q^0(t) - q^0(t-t_0),$$
$$c_*^0(t) = c^0(t) - c^0(t-t_0). \tag{16}$$

To solve the inverse problem of nonequilibrium kinetics of sorption we find the expression for the initial α_l and central moments μ_n from the relationships [24]

$$\alpha_n = \lim_{p\to 0}\left[\frac{(-1)^n}{\bar{q}_*(p)} \cdot \frac{d^{(n)}\bar{q}_*(p)}{dp^n}\right], \quad \mu_n = \sum_{k=0}^{n} C_n^k(-\alpha_1)^k \alpha_{n-k}. \tag{17}$$

With consideration of (15) we obtain after transformations

$$\alpha_1 = \frac{1}{k_2} + \tau_i + \frac{1}{\gamma_0^*} + \frac{t_0}{2}, \quad \mu_2 = \left(\frac{1}{k_2} + \tau_i + \frac{1}{\gamma_0^*}\right)\left(\frac{1}{k_2} + \tau_i + \frac{1}{\gamma_0^*}\right.$$
$$\left. + \frac{2}{k_1+k_2}\right) + \frac{2(\nu+1)}{(\nu+5)}\tau_i^2 + \frac{t_0^2}{12}, \quad \mu_3 = 2\left(\frac{1}{k_2} + \tau_i + \frac{1}{\gamma_0^*}\right)$$
$$\times\left(\tau_i + \frac{1}{\gamma_0^*} - \frac{2}{k_2}\right) + \frac{6(3\nu^2+2\nu+9)}{(\nu+5)(\nu+7)}\tau_i^3 + \frac{6(\nu+1)\tau_i^2}{(\nu+5)\gamma_0^*} + \frac{12(\nu+1)\tau_i^2}{(\nu+5)k_2}, \tag{18}$$

where

$$\tau_i = (1+k)a^2D_i^{-1}(\nu+3)^{-1}(\nu+1)^{-1}; \quad \gamma_0^* = \beta_0(1+\nu)(1+k)^{-1}a^{-1}. \tag{19}$$

The expressions for the moments (18) are essentially the solution of the inverse problem of the kinetics of sorption. Having determined from the experimental kinetic curves the numerical values of the moments

$$\alpha_1 = \frac{1}{\alpha_0}\int_0^\infty t q_*(t)\,dt, \quad \mu_n = \frac{1}{\alpha_0}\int_0^\infty (t-\alpha_1)^n q_*(t)\,dt, \tag{20}$$

we can find the parameters τ_i, γ_0^*, k_2 from the solution of algebraic system (18).

The solution of nonequilibrium dynamics of sorption in transforms from (1), (3), (10) with consideration of conditions (6)-(8) is found in the form

$$\bar{c}(z, p) = c_0 f(p) \exp\left\{\frac{uz}{2D}\left[1 - \sqrt{1 + 4Du^{-2}\left[p + D_i\delta_0(1+\nu)a^{-2}B\left(1+\frac{D_i}{a\beta_0}B\right)^{-1}\right]}\right]\right\}, \tag{21}$$

where

$$f(p) = p^{-1} \text{ (frontal dynamics of sorption)}, \tag{22}$$
$$f(p) = 1, \ f(p) = f^*(p) = p^{-1}[1 - \exp(-pt_0)] \text{ (elution dynamics of sorption)}. \tag{23}$$

We substitute (21) into (17) and for $f(p) = 1$ we find the expressions for the first initial and four central moments for a fixed length of the column L:

$$\alpha_1 = [1 + (1+k)\delta]Lu^{-1},$$
$$\mu_2 = 2\alpha_1\left\{\tau_i + k\delta bk_2^{-1} + (1+k)\delta b\left(\tau_i + \frac{1}{\gamma_0^*}\right)\right\},$$
$$\mu_3 = 6\alpha_1\left\{2\tau_i^2 + 2\tau_i k\delta bk_2^{-1} + 2\tau_i(1+k)\delta b\left(\tau_i + \frac{1}{\gamma_0^*}\right) + k\delta b\,(k_2^{-1}\right.$$
$$\left. + 2\tau_i k_2^{-1} + 2k_2^{-1}(\gamma_0^*)^{-1}) + (1+k)\delta b\left[2\frac{(\nu+3)}{(\nu+5)}\tau_i^2 + 2\tau_i(\gamma_0^*)^{-1} + (\gamma_0^*)^{-2}\right]\right\},$$

$$\mu_4 = 3\mu_2^2 + 24\alpha_1 \left\{5\tau_i^3 + 6\tau_i^2 k\delta b k_2^{-1} + 6\tau_i^2 (1+k)\,\delta b \left(\tau_i + \frac{1}{\gamma_0^*}\right)\right.$$

$$+ \tau_i b \left[2k\delta k_2^{-2} + 2k\delta \left(\frac{3k+2}{k+1}\right) k_2^{-1} \left(\tau_i + \frac{1}{\gamma_0^*}\right) + 2(1+k)\,\delta \left(2\tau_i^2 \frac{(\nu+3)}{(\nu+5)}\right.\right.$$

$$\left.\left. + \frac{2\tau_i}{\gamma_0^*} + \frac{1}{\gamma_0^{*2}}\right)\right] + \tau_i b^2 \left[k\delta k_2^{-1} + b\left(\tau_i + \frac{1}{\gamma_0^*}\right)\right]^2 +$$

$$+ k\delta b \left[k_2^{-3} + \left(\frac{3k+2}{k+1}\right) k_2^{-2} \left(\tau_i + \frac{1}{\gamma_0^*}\right) + 6\,\frac{(\nu+3)}{(\nu+5)}\,\tau_i^2 k_2^{-1}\right.$$

$$\left.\left. + \frac{6\tau_i k_2^{-1}}{\gamma_0^*} + \frac{3k_2^{-1}}{\gamma_0^{*2}}\right] + (1+k)\,\delta b \left[\frac{(5\nu+17)(\nu+3)}{(\nu+5)(\nu+7)}\,\tau_i^3 + \frac{(5\nu+17)\,\tau_i^2}{(\nu+5)\,\gamma_0^*} + \frac{3\tau_i}{\gamma_0^{*2}} + \frac{1}{\gamma_0^{*3}}\right]\right\},$$

$$\mu_5 = 10\mu_2\mu_3 + 120\alpha_1 \left\{14\tau_i^4 + 20\tau_i^3 k\delta b k_2^{-1} + 20\tau_i^3 (1+k)\,\delta b \left(\tau_i + \frac{1}{\gamma_0^*}\right)\right.$$

$$+ 6\tau_i^2 b \left[k\delta \left(k_2^{-2} + 2\tau_i k_2^{-1} + \frac{2k_2^{-1}}{\gamma_0^*}\right) + (1+k)\,\delta \left(2\tau_i^2 \frac{(\nu+3)}{(\nu+5)}\right.\right.$$

$$\left.\left. + \frac{2\tau_i}{\gamma_0^*} + \frac{1}{\gamma_0^{*2}}\right)\right] + 6\tau_i^2 b^2 \left[k\delta k_2^{-1} + (1+k)\,\delta \left(\tau_i + \frac{1}{\gamma_0^*}\right)\right]^2$$

$$+ 2\tau_i b^2 \left[k\delta k_2^{-1} + (1+k)\,\delta \left(\tau_i + \frac{1}{\gamma_0^*}\right)\right] \left[k\delta \left(k_2^{-2} + 2\tau_i k_2^{-1}\right.\right.$$

$$\left.\left. + \frac{2k_2^{-1}}{\gamma_0^*}\right) + (1+k)\,\delta \left(2\tau_i \frac{(\nu+3)}{(\nu+5)} + \frac{2\tau_i}{\gamma_0^*} + \frac{1}{\gamma_0^{*2}}\right)\right] + 2\tau_i k\delta b \left[k_2^{-3}\right.$$

$$\left. + k_2^{-2} \left(\frac{3k+2}{k+1}\right) \left(\tau_i + \frac{1}{\gamma_0^*}\right) + 6\tau_i^2 k_2^{-1} \frac{(\nu+3)}{(\nu+5)} + \frac{3k_2^{-1}}{\gamma_0^*} \left(2\tau_i + \frac{1}{\gamma_0^*}\right)\right]$$

$$+ 2\tau_i (1+k)\,\delta b \left[\frac{(5\nu+17)(\nu+3)}{(\nu+5)(\nu+7)}\,\tau_i^3 + \frac{(5\nu+17)\,\tau_i^2}{(\nu+5)\,\gamma_0^*} + \frac{3\tau_i}{\gamma_0^{*2}}\right.$$

$$\left. + \frac{1}{\gamma_0^{*3}}\right] + k\delta b \left[k_2^{-4} + k_2^{-3} \left(\frac{2k+1}{k+1}\right) \left(\tau_i + \frac{1}{\gamma_0^*}\right) + 3k_2^{-2} \left(\frac{2k+1}{k+1}\right) \left(2\tau_i^2 \frac{(\nu+3)}{(\nu+5)} + \frac{2\tau_i}{\gamma_0^*} + \frac{1}{\gamma_0^{*2}}\right)\right.$$

$$\left. + 4k_2^{-1} \left(\tau_i^3 \frac{(5\nu+17)(\nu+3)}{(\nu+5)(\nu+7)} + \frac{(5\nu+17)\,\tau_i^2}{(\nu+5)\,\gamma_0^*} + \frac{3\tau_i}{\gamma_0^{*2}} + \frac{1}{\gamma_0^{*3}}\right)\right]$$

$$\left. + (1+k)\,\delta \left[\frac{2(7\nu+31)(\nu+3)^2 \tau_i^4}{(\nu+5)(\nu+7)(\nu+9)} + \frac{2(7\nu+31)(\nu+3)\,\tau_i^3}{(\nu+5)(\nu+7)\,\gamma_0^*} + \frac{6(\nu+4)\,\tau_i^2}{(\nu+5)\,\gamma_0^{*2}} + \frac{8\tau_i}{\gamma_0^{*3}} + \frac{1}{\gamma_0^{*4}}\right]\right\}, \tag{24}$$

where

$$b = [1 + (1+k)\,\delta]^{-1}; \quad \tau_i = [1 + (1+k)\,\delta]\, Du^{-2}. \tag{25}$$

The expressions for moments α_1, μ_2, μ_3, μ_4 from (24) describe fully the elution curve in a form close to a Gaussian curve [26]. Using conditions (23), we find the relation between the moments α_1, μ_n (they correspond to f(p)) and α_1^*, μ_n^* (they correspond to $f^*(p)$):

$$\alpha_1^* = \alpha_1 + \frac{1}{2} t_0, \quad \mu_2^* = \mu_2 + \frac{1}{12} t_0^2, \quad \mu_3^* = \mu_3,$$
$$\mu_4^* = \mu_4 + \frac{1}{80} t_0^4 + \frac{1}{2} t_0^2 \mu_2, \quad \mu_5^* = \mu_5 + \frac{5}{6} t_0^2 \mu_3. \tag{26}$$

We see from Eqs. (21), (22), (23) that the frontal and elution dynamic curves are associated by the relationships

$$p\tilde{c}_f(z,\ p) = \tilde{c}_e(z,\ p), \quad [1 - \exp(-pt_0)]\,\tilde{c}_f(z,\ p) = \tilde{c}_e^*(z,\ p) \tag{27}$$

or

$$c_e(z,\ t) = \frac{\partial c_f(z,\ t)}{\partial t}, \quad c_e^*(z,\ t) = c_f(z,\ t) - c_f(z,\ t - t_0). \tag{28}$$

For a fixed length of the column L, the elution and frontal dynamic curves can be described by series with a small number of terms according to Hermite orthogonal polynomials

$$c_e(L,\ t) = \sum_{n=1}^{\infty} A_n H_n(y)\exp(-y^2),\quad y = (t-\alpha_1)(2\mu_2)^{-1/2},$$

$$c_f(L,\ t) = \frac{1}{2}[\operatorname{erf}(y) + \operatorname{erf}(y_0)] + \sqrt{2\mu_2}\sum_{n=3} A_n [F_n^*(y) + (-1)^n F^*(y_0)], \tag{29}$$

$$y_0 = \frac{\alpha_1}{\sqrt{2\mu_2}};\ \operatorname{erf}(y) = \frac{L}{\sqrt{\pi}}\int_0^y e^{-t^2}dt;\ F_n^*(y) = H_{n-1}(0) - e^{-y^2}H_{n-1}(y).$$

Using the orthogonality of Hermite polynomials [25], we find

$$A_n = \sum_{k=0}^{\left[\frac{n}{2}\right]} \frac{(-1)^k}{\sqrt{\pi}(n-2k)!\,k!}\, 2^{\left(-\frac{n}{2}-k-\frac{1}{2}\right)} \mu_{n-2k}(\mu_2)^{\left(-\frac{n}{2}+k-\frac{1}{2}\right)},$$

$$A_0 = \frac{1}{\sqrt{2\pi\mu_2}},\quad A_1 = A_2 = 0. \tag{30}$$

In the presence of stagnant zones between the sorbent grains [19-23], the elution dynamic curves are greatly "extended" and differ from the Gaussian curve, and therefore such curves can be described by series with a small number of terms according to generalized Laguerre polynomials

$$c_e(L,\ t) = \sum_{n=0}^{\infty} N_n \exp\left(-\frac{t}{\tau}\right) L_n^m\left(\frac{t}{\tau}\right)\left(\frac{t}{\tau}\right)^m, \tag{31}$$

$$c_f(L,\ t) = \tau\sum_{n=0}^{\infty} N_n \sum_{k=0}^{n} \frac{(-1)^k\Gamma(n+1)\Gamma(m+n+1)}{\Gamma(k+1)\Gamma(n-k+1)\Gamma(m+k+1)}\gamma\left(m+k+1,\ \frac{t}{\tau}\right). \tag{32}$$

Using the orthogonality of the generalized Laguerre polynomials [27], we obtain

$$N_n = \frac{D}{8}\cdot\sum_{k=0}^{n} \frac{(-1)^k \alpha_k \tau^{-(k+1)}}{\Gamma(k+1)\Gamma(n-k+1)\Gamma(m+k+1)}.$$

Varying the independent parameters (m), we can change the leading edge and, varying the parameter (τ), the trailing edge of the elution dynamic curve. With an appropriate selection of the parameters in series (31), (32), we can restrict ourselves to two or three terms.

The expressions for the moments obtained for an unbounded column [9] are more complex and differ from (24). Thus, the expression for the first initial and second central moments is [9]

$$\alpha_1 = (1+k\delta)\left(\frac{L}{u}+\frac{2D}{u^2}\right),\quad \mu_2 = 2\alpha_1\left(\frac{1}{k_2}+\tau_i+\frac{1}{\gamma_0^*}\right) + 2\tau_i(1+k\delta)\left(\frac{L}{u}+\frac{4D}{u^2}\right). \tag{33}$$

The other central moments in [9] have an even more complex form. However, for asymptotically large lengths of columns the expressions for the moments (24) and (33) coincide. For small lengths of columns it is necessary to use (24).

Relationships (29)-(32) represent the solutions of the direct problem of nonequilibrium dynamics of sorption. The expressions for the moments (24) are essentially the solution of the inverse problem of the nonequilibrium dynamics of sorption. Having determined from the experimental elution curve (we can easily obtain the elution curve from the frontal curve by means of relationship (28)) the numerical values for the moments, we can find parameters δ, D_i, β_0, k_2, D from the solution of the algebraic system. To avoid solving a complex algebraic system it is expedient to find δ from the equation for α_1 at first, and then record the dynamic curves for large linear velocities (in this case $\tau_i \gg 1/\gamma_0^*$ [28] and find k_2, D_i, D from the expressions for μ_2 and μ_3. From the elution curve for a small flow velocity (in the case $\tau_i \sim 1/\gamma_0^*$ [28]) we can find β_0 from the expression for μ_2. The agreement of the experimental values of μ_4 with the calculated (with consideration of determination of the parameters by the method indicated above) values of μ_4 is the criterion of accuracy of determining the parameters, on one hand, and, on the other, of the correctness of the given model, which correctly describes percolation of a gas mixture through a column.

NOTATION

q^0	is the concentration of absorbed substance per unit volume of sorbent grain;
c^0	is the concentration of sorbate within free space of sorbent grains;
k_1, k_2	are the sorption and desorption coefficients, respectively;
$k = k_1/k_2$;	
D_i	is the coefficient of (internal) diffusion within narrow channels of the sorbent grains;
ν	is the symmetry parameter ($\nu = 2$ for a sphere with radius a; $\nu = 1$ for a cylinder with radius a; $\nu = 0$ for grains in the form of plates $2a$ thick);
β_0	is the coefficient of (external) mass transfer on the surface of the sorbent grains;
u	is the percolation velocity of flow of gas (liquid) mixture;
c	is the concentration of sorbate in flow;
$\delta_0 = (1-\sigma)/\sigma$;	
$\delta = \delta_0/(\nu - 1)$;	
σ	is the portion of free space of the granular column;
γ_0	is the kinetic coefficient of sorption taking into account the delivery velocity of the substance to the surface of the sorbent grains by the percolating flow and external diffusion;
$\gamma_0 = (1+\nu)\beta_0/a$;	
D	is the dispersion coefficient taking into account effective longitudinal mixing;
t_0	is the time of admission of the investigated mixture.

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