NONLINEAR EQUATIONS OF HEAT AND MASS TRANSFER IN DISPERSED MEDIA

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A discussion is presented of aspects of the description of heat- and mass-transfer processes in dispersed media on the basis of a single equation for the concentration of the disperse phase (temperature) in the dispersion medium when there is a nonlinear relationship between the concentrations of the disperse phase and dispersion medium at the interface.

Introduction. The development of a theory of heat and mass transfer in disperse systems has led to the gradual refinement of the basic equations of the process and the degree to which they reflect actual events. The key stages in the development of the theory have been the derivation of a single transport equation for the system as a whole [$\varepsilon = 1$ in (1)], the creation of a two-phase system of the form

$$\varepsilon \partial c/\partial t + u \partial c/\partial x = D \partial^2 c/\partial x^2 - (1-\varepsilon)\, \partial a/\partial t, \tag{1}$$

$$(1-\varepsilon)\, \partial a/\partial t = f(c) - ka \tag{2}$$

and later, in the linear variant, the return to one-phase equations which either simplify system (1-2) for $f(c) = kc$ (equivalent to the equation in [1]) or account for relaxational heat and mass transfer between the phases [2, 3]. For the sake of definiteness, we will henceforth concern ourselves with mass transfer – where use is often made of nonlinear systems of type (1-2). Since the dimensions of the problem with respect to the macroscopic space coordinates in the dispersion medium are not of central importance for the purposes of our analysis, we will consider it to be unidimensional in order to simplify the notation.

We will discuss how system (1-2) can be replaced by one equivalent equation and how the results obtained [2, 3] can be generalized to nonlinear situations for sufficiently arbitrary forms of disperse-phase particles and certain distributions of the characteristics of these forms.

1. Equivalent Equation.

The usefulness of this approach for analyzing problems in disperse systems has been discussed in several studies [1, 4, 5, etc.]. Thus, we will restrict ourselves to the observation that the equivalent equation can be used to describe processes which are large-scale in terms of time and that, as far as the physical accuracy of the model is concerned, it often proves to be sufficient to restrict the inquiry to one term in the sum which enters into the equivalent equation [1].

Following [1], we use (2) to express the quantity a through the formal expansion:

$$a = \frac{1}{k} \sum_{n=0}^{\infty} (-1)^n \left(\frac{1-\varepsilon}{k}\right)^n \frac{\partial^n f(c)}{\partial t^n}. \tag{3}$$

The inserting (3) into (1), we obtain the equivalent equation

$$\varepsilon \frac{\partial c}{\partial t} + \frac{(1-\varepsilon)}{k} \frac{\partial f(c)}{\partial t} + u \frac{\partial c}{\partial x} = D \frac{\partial^2 c}{\partial x^2} + \sum_{n=2}^{\infty} (-1)^n \left(\frac{1-\varepsilon}{k}\right)^n \frac{\partial^n f(c)}{\partial t^n}. \tag{4}$$

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If we discard all terms of the sum in (4), we obtain an equation which is frequently used [6] in the theory of sorption processes and which expresses an equilibrium relationship between the concentrations a and c — corresponding to retention of the first term of the sum in (3). Refinement of this approximation leads to Eq. (4), in which this term is kept. In this case, (4) becomes an elliptical equation [1] and must be augmented by additional conditions that address considerations of accuracy.

The above-adopted form of kinetic relation (2) is not the only form possible [6, 7]. For example, the variant in [7] can also be used:

$$(1-\varepsilon)\,\partial a/\partial t = kc - g(a). \tag{5}$$

An expansion of type (3) becomes more complicated in the present situation, but if we limit ourselves to the first two terms in the expansion, we can similarly obtain the following equivalent equation:

$$\begin{gathered}\varepsilon\frac{\partial c}{\partial t}+(1-\varepsilon)\frac{\partial}{\partial t}g^{-1}(kc)+u\frac{\partial c}{\partial x}=\\=(1-\varepsilon)\frac{\partial^2}{\partial t^2}\left[g^{-1}(kc)\frac{da}{dg}\bigg|_{a=g^{-1}(kc)}\right]+D\frac{\partial^2 c}{\partial x^2},\end{gathered}\tag{6}$$

where g^{-1} is the function which is the inverse of g and which is assumed to exist. It is easily proven that with linear functions f = kc and g = ka, we arrive at the well-known results in [1].

Equations (4) and (6) are valid only for sufficiently large values of time. Along with this shortcoming — due to the method by which the equations were derived — they suffer from certain inaccuracies inherent in kinetic relations of types (2) and (5) [2, 3]. This leads to different values for the coefficients of the expansions in the sums of Eq. (4), etc. These problems can be eliminated by taking a more detailed approach to the description of mass exchange between particles of the disperse phase and the dispersion medium.

2. Formulation of Equations with Allowance for Relaxation Phenomena. In a number of cases in which heat and mass transfer processes are being described, the condition that the parameter k and the function f(a) be independent of time is not fully acceptable [2, 3]. This is true especially in the case of "abrupt" disturbances of concentration, when the characteristic time of the process is comparable to the time of the interphase exchange relaxation. As is known, two relaxation processes take place simultaneously in a heterogeneous disperse system: one in the dispersion medium, one in the disperse phase. The relationship between the corresponding relaxation times t_1, t_2 and the characteristic time of the process is important for simplifying the problem. Here, we will restrict ourselves to analyzing the case when the inequality $t_1 \ll t_2$ is satisfied. This makes it possible to simply formulate the conditions required to close the problem for an isolated "sample" particle. For the other particles, we assume that the disperse system satisfies the usual postulates of the mechanics of heterogeneous media [2, 3, 8].

The equation of heat and mass transfer has been derived many times by different methods. For the dispersion medium, we take this equation in the form

$$\varepsilon\partial c/\partial t + u\partial c/\partial x = D\partial^2 c/\partial x^2 - Q, \tag{7}$$

where the term Q expresses the total flow of matter into (out of) the dispersion medium from the disperse phase per unit of time in a unit volume of the system. In (7), we omitted the term corresponding to pulsative mass transfer [9]. This can be done for low values of the local Peclet number. Sometimes this component is incorporated into the diffusion term (7). In deriving (7) by averaging over the ensemble, the authors of [9] assumed that the composition of the spherical particles was monodisperse. However, it is clear that the form of the particles may remain the same in more general situations as well for the first terms of the equation. There may also be no change in the physical significance of the source term. We will give special emphasis to this subject below.

As was shown in [2, 3], under the condition $t_1 \ll t_2$, the effect of the dispersion medium on heat and mass transfer in the neighborhood of the isolated particle is manifest in the imposition of the temperature (concentration) of the dispersion medium on the boundary of the particle. Here, changes in these quantities occurring over distances on the order of the size of the particle can be ignored. Thus, the temperature (concentration) can be assumed to depend only on time for the problem of heat and mass transfer inside the particle. The dependence on the macroscopic coordinate x is assumed to be parametric. When mass transfer processes occurring at a phase boundary are studied, investigators often make use of a condition of local equilibrium of the Langmuir isotherm type. In this general case, this establishes a nonlinear relationship between the variables at the boundary. Taking this into account, we formulate the problem in the neighborhood of the isolated particle in the following form:

$$\Delta\bar{a} = \partial\bar{a}/\partial\tau, \quad \tau = t/t_2, \quad t_2 = l^2/D_2, \tag{8}$$

$$\bar{a}|_S = f[c(\tau)], \quad \bar{a}|_{\tau=0} = 0, \quad l = V^{1/3}, \tag{9}$$

where ℓ is the characteristic dimension of the particle; S is its boundary. For simplicity, we adopt a zero initial condition, since its role is insignificant on the macroscopic time scale.

The main quantities needed to complete the analysis and description of events are readily obtained from the results in [10]:

$$\begin{gathered} a = \langle \bar{a} \rangle = \frac{1}{V}\int\limits_V \bar{a}\,dV = \frac{1}{t_2}\int\limits_0^t f[c(\xi)]\,\Phi\left(\frac{t-\xi}{t_2}\right)d\xi, \\ q = \int \frac{\partial\bar{a}}{\partial n}\,dS = \frac{d}{dt}\int\limits_0^t f[c(\xi)]\,\Phi\left(\frac{t-\xi}{t_2}\right)d\xi. \end{gathered} \tag{10}$$

For the function Φ, we have the expansion:

$$\Phi(\tau) = \sum_{i=1}^{\infty} \lambda_i\gamma_i^2 \exp(-\lambda_i\tau) \tag{11}$$

and, accordingly, for the Laplace transform of Φ (with regard to the parameter τ):

$$\Phi^*(p) = \sum_{i=1}^{\infty} \gamma_i^2/(1 + p/\lambda_i), \tag{12}$$

where the asterisk denotes the transformed quantities; p is the parameter of the Laplace transform. Here, λ_i are eigenvalues of the problem

$$\Delta\Theta_i + \lambda_i\Theta_i = 0, \quad \Theta_i|_S = 0, \quad \lambda_i = \text{const} > 0 \tag{13}$$

for the internal region bounded above by the surface of the particle. The numbers γ_i are the volume integrals of the corresponding eigenfunctions Θ_i normalized with respect to unity.

We will assume that all of the particles have the same shape but that their characteristic dimensions ℓ are different. We introduce the particle-size distribution function $\psi(\ell)$. After this function is multiplied by $d\ell$, it will give us the number of particles whose dimensions fall within the interval $[\ell, \ell + d\ell]$ with small $d\ell$.

Then calculation of the total mass flow from all of the particles in a unit volume and insertion of the result into Eq. (7) makes it possible to obtain the basic equation for the function c in the form

$$\varepsilon\frac{\partial c}{\partial t} + u\frac{\partial c}{\partial x} = D\frac{\partial^2 c}{\partial x^2} - D_2(1-\varepsilon)\Big[\int\limits_0^\infty l^3\psi(l)\,dl\Big]^{-1} \times \frac{\partial}{\partial t}\int\limits_0^t f[c(x, \xi)]\,d\xi\int\limits_0^\infty l\Phi\left(\frac{t-\xi}{l^2/D_2}\right)\psi(l)\,dl, \tag{14}$$

where we have also used the definition of the porosity ε of the layer.

In the region of large values of time $t \gg t_2$ (slight transience), we can use an expansion of the function Φ^* in powers of p [10] to obtain the following relation:

$$\varepsilon \frac{\partial c}{\partial t} + (1-\varepsilon) \frac{\partial}{\partial t} f(c) + u \frac{\partial c}{\partial x} = D \frac{\partial^2 c}{\partial x^2} - \\ - D_2 (1-\varepsilon) \frac{1}{M_3} \sum_{j=2}^{\infty} (-1)^j n_{j-1} \frac{M_{2j+1}}{D_2^j} \frac{\partial^j}{\partial t^j} f(c), \tag{15}$$

where

$$M_j = \int_0^{\infty} l^j \psi(l)\, dl \tag{16}$$

are the moments of the distribution function; n_j are coefficients representing the volume-averaged values of the functions w_j. These functions are determined from the sequence of problems

$$\Delta w_j = -w_{j-1}, \quad w_0 = 1, \quad j = 1, 2, \ \ldots \tag{17}$$

for the region inside the particle with a zero value for the function w_j, $j \geq 1$, on the boundary of the region.

For small values of time – more accurately, for $t_1 \ll t \ll t_2$ – it is also possible to simplify the expression for the function Φ. Using the results in [10], we obtain the following equation:

$$\varepsilon \frac{\partial c}{\partial t} + u \frac{\partial c}{\partial x} = D \frac{\partial^2 c}{\partial x^2} - \frac{(1-\varepsilon) D_2^{1/2} M_2 S_*}{M_3 \sqrt{\pi}} \frac{\partial}{\partial t} \int_0^t \frac{f[c(x, \xi)]\, d\xi}{\sqrt{t-\xi}} - \frac{D_2 (1-\varepsilon)}{2M_3} \varkappa_* S_* M_1 f(c), \tag{18}$$

where S_* is the proportionality factor in the expression for the area of the surface bounding a particle of the dimension ℓ: $S = S_* \ell^2$; κ_* is the analogous coefficient in the expression for the mean curvature of the particle surface

$$\langle \varkappa \rangle = \varkappa_* / l = \frac{1}{S} \oint \varkappa(S)\, dS. \tag{19}$$

Here, we assume that the particle surface is sufficiently smooth (see [10] in regard to allowance for the presence of dihedral angles, etc. on the surface). It should be noted that by the curvature of the surface, the author of [10] meant a quantity half as great as the quantity used here.

As an example of the determination of the parameters S_* and κ_* in (18), we will examine a particle of a bed in the form of an ellipsoid of revolution:

$$z^2/b^2 + (x^2 + y^2)/d^2 = 1, \quad l^3 = 4\pi b d^2/3. \tag{20}$$

Having designated $\gamma = b/d$ and performing the necessary quadratures, we arrive at the following relations:

$$S_* = 2\pi \left[1 + \frac{\gamma}{\sqrt{\gamma - 1}} \arcsin \sqrt{1 - \frac{1}{\gamma}} \right], \tag{21}$$

$$\varkappa_* = \frac{2\pi}{S_*} \left[\frac{1}{\sqrt{\gamma^2 - 1}} \ln \left(\frac{\gamma + \sqrt{\gamma^2 - 1}}{\gamma - \sqrt{\gamma^2 - 1}} \right) + 2\gamma \right]. \tag{22}$$

The formulas for S_* and κ_* obviously yield real values at $\gamma > 1$. It is easy to see that they are also valid at $\gamma < 1$. In this case, it is best to replace the inverse trigonometric functions by logarithmic functions and vice versa in accordance with established [11] relations. It should be noted that the point $\gamma = 1$ (corresponding to a spherical particle) is not a singular point in Eqs. (21) and (22). The value of the first coefficients n_j in (15) was found in [10] for this example.

It is interesting to note that the given case is that of a particle whose geometric characteristics are completely determined by the two parameters ℓ and γ. The description of the problem could be generalized by examining particles with two, three, or more parameters characterized by a certain scatter and by introducing distribution functions for the particles in accordance with the corresponding number of variables. Although such a generalization would not complicate the basic problem being studied, the functions S_* and κ_* in (18) would depend on the parameters analogous to γ [see (21-22), for example] and would be located under the integral sign for these parameters in the final formula.

One interesting feature of Eqs. (15) and (16) is that they contain only "moment" characteristics of the distribution function $\psi(\ell)$. Meanwhile, if the customary procedure is followed and one term of the sum is kept in (15), then it will be sufficient to know the moments M_j up to the fifth moment, inclusively. Production processes are often organized so as to consist of several specific stages (operations). Thus, equations such as (14), (15), and (18) are used to describe the removal of undesirable impurities from the end product (crystals). Previously, crystals were usually obtained in crystallizers, with the liquid phase subsequently being removed in filters or centrifuges. Moment characteristics of the distribution functions were also used in this case to model the process. In describing the crystallization operation, it often turns out to be quite difficult to obtain complete information on the size distribution of the crystals. However, a number of crystallization models make it possible to use the less informative but simpler "moment" approach [12, 13, etc.]. Thus, if the problem being examined here is addressed with consideration of the corresponding stages of the production process, then it becomes clear that the "tightening" of the requirements established for the description of the crystallization process in accordance with models (15) and (16) may prove to be a useful simplification of the modeling of the entire production cycle and may make it easier to attain given optimization targets and other production goals.

The changeover from Eq. (14) to Eqs. (15) and (18) presupposes that it is valid to regard the corresponding asymptotic stage as realized for all particles of the bed. This can always be achieved if the granulometric composition of the particles is bounded from above (which it always is) and below: $\ell_- \leq \ell \leq \ell_+$. Then the necessary inequalities will be satisfied immediately for the entire particle spectrum for certain time intervals. For example, the inequalities will be satisfied at $t \gg t_2^+ = \ell_+^2/D_2$ in the case of Eq. (15). Similar aspects of the problem were discussed in [14]. This and other studies (such as [15, 16]) examined adsorption in bidisperse porous structures, along with other topics. Both the mathematical and the physical features of the problems investigated in these studies turn out to be similar to the analogous features of our problem. For example, the authors of [15] presented expressions for functions Φ corresponding to certain particles of simple form. Dealing with the quasisteady case ($t \gg t_2$), the authors of [14, 17] proposed a changeover to a system of equations of the type (1-2). Here, the function Φ^* for a sphere, of the form $(\sqrt{p}\,\mathrm{cth}\sqrt{p}-1)/p$, is replaced by a simple fraction with expansion of the hyperbolic functions of the numerator and denominator into Taylor series with small values of p. A more exact expression in the form of a simple fraction is obtained if we first exapnd Φ^* into a series in powers of p to terms on the order of $O(p^2)$ and then replace the last expression by a fraction with the same degree of accuracy. In the general case, we have the expression

$$\Phi^*(p) = 1/(1 + n_1 p) + O(p^2),$$

which with the aid of (10) leads us to a relation of type (2):

$$n_1 t_2 \partial a/\partial t = f(c) - a \tag{23}$$

for a monodisperse bed or

$$(n_1 M_5/M_3 D_2)\,\partial a/\partial t = f(c) - a \tag{24}$$

for a polydisperse system. Here, the quantity a corresponds to the concentration of the end product in the particles within a unit volume of the solid phase. Having made the corresponding transformations, it is not hard to reduce Eqs. (23) and (24) to the form (2) – where the function f can be included as a constant coefficient if necessary. This was already taken into account in the formulation of boundary condition (9).

The author of [14] proposed dividing the entire particle spectrum into two parts. The mass-transfer relations for small values of time are satisfied in the first part, while the analogous relations for large values of time are satisfied in the second part. It is clear that if the particle-size distribution function is not carefully chosen, there can be no clear boundary between these parts – even more so, since the location of the boundary may depend on time. If the particle spectrum is limited to $0 < \ell_- \leq \ell_+ < \infty$, then in the general case, with a fixed time interval, the particles can be divided into three groups by adding an intermediate group in which mass transfer cannot be described by either of the above-mentioned asymptotic stages. The question of the efficacy of simplifying the kernel Φ arises in this case. At attempt could be made to keep several initial terms in expansion (11). However, it would be better to first make use of Eq. (12) for Φ^* and the fact that the intermediate stage corresponds to moderate values of time and, thus, intermediate values of the parameter p. Under such circumstances, the successful approximation of Φ^* in the time interval of interest to us depends on the rate of decrease in γ_i and rate of increase in λ_i with an increase in the number i, i.e., it is determined by i rather than p, as was the case earlier at $t \to \infty$ and $t \to 0$. We have the corresponding theorems in [18] to evaluate the increase in λ_i with i. The use of the simple example of spherical particles [10, 15] shows that the rate of decrease in the terms of series (12) will be on the order of i^{-2}, i.e., the convergence is not very good. In this case, we can use the methods available for accelerating the convergence of series [19]. This allows us to obtain the following expression, for example:

$$\Phi^*(p) = 1 - n_1 p + p^2 \sum_{i=1}^{\infty} \gamma_i^2 / \lambda_i (\lambda_i + p),$$

the terms of the sum of this expression decreasing as i^{-6} with i in the case of spherical particles. This procedure can obviously be continued further. In the given case, the first term of the sum may be sufficient for describing the intermediate time interval. Let us pursue this and for the sake of simplicity assume that the given stage is realized for the entire particle spectrum (this is not a fundamental change and means only that there is a change in the limits of integration over ℓ in Eq. (25) and that a correction is thus made to certain moments (16)). We therefore obtain the following simplified equation (instead of (14)):

$$\begin{aligned}
&\varepsilon \frac{\partial c}{\partial t} + (1-\varepsilon) \frac{\partial}{\partial t} f(c) + u \frac{\partial c}{\partial x} = D \frac{\partial^2 c}{\partial x^2} + \\
&+ \frac{(1-\varepsilon) n_1 M_5}{D_2 M_3} \frac{\partial^2}{\partial t^2} f(c) - \frac{(1-\varepsilon)\gamma_1^2}{\lambda_1 D_2 M_3} \frac{\partial^3}{\partial t^3} \times \\
&\times \int_0^t f[c(x, \xi)]\, d\xi \int_0^\infty l^5 \psi(l) \exp\left[\frac{(t-\xi) D_2 \lambda_1}{l^2}\right] dl.
\end{aligned} \qquad (25)$$

It should be noted that representation of Φ as the sum of exponents (11) might prove useful for specific distribution functions $\psi(\ell)$, since it leads to one type of integral over ℓ. The form of this integral, shown in (25), permits its analytic or asymptotic investigation, depending on the specific situation. It should also be noted that in the case of a monodisperse bed, $\psi(\ell) = N\delta(\ell - \ell_0)$, where $\delta(z)$ is the delta function and N is the number of particles of the size ℓ_0, Eq. (25) can be localized (i.e. reduced to a form not involving integral operations) by making use of the fact that the kernel has the form of an exponent and differentiating (25) can be carried out with respect to t, etc.

Conclusion. It should also be noted that the proposed equations can be also be used to analyze several other processes, such as the filtration of flows in cracked-porous rocks (media with dual porosity). This and other mathematically similar systems are characterized by the existence of linear relations between the sought functions at the phase boundaries, etc. Nonlinear effects are often due to factors other than those investigated here (see [20], for example). Thus, within the framework of the subject matter of the present study, the use of size distribution functions for particles (blocks, etc.) is of generalizing value. The author of [21] examined only the simplest variants for the form of filtration blocks in an analysis of relaxation phenomena. In practical situations, this approach may turn out to be too idealized.

The practical value of the questions examined here lies not only in modeling certain unit operations (adsorption, washing of sediments, etc.), but also in finding parameters which describe various phenomena and which may be encountered in the formulation of problems for other stages of the complete production cycle. This makes it possible to determine the degree of detail with which the corresponding processes need to be described, which may in turn make it possible to simplify the modeling of the production cycle as a whole (to discuss the relationship between the operations of crystallization and sediment washing, for example).

NOTATION

a, $\bar{a}$, concentrations of the end product in the disperse phase (average and local values, respectively); c, concentration of the component in the dispersion medium; D, coefficient of effective diffusion in the dispersion medium; D_2, coefficient of diffusion in particles of the disperse phase; $f(c)$, $g(a)$, functions linking the concentrations of the components of the phases at equilibrium; k, proportionality factor; $\ell_{\pm}$, largest and smallest particle size in the spectrum; q, dimensionless mass flux per particle; t, time; u, transport (filtration) rate; V, particle volume; x, coordinate along the system; $\langle\ \rangle$, averaging sign.

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