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A.M. Brener[1], A.O. Kazenova[1], V.G. Golubev[1], L.T. Tashimov[2], Sh.K.Shapalov[1], G.D. Kenzhalieva[1], D.R. Tortbayeva[1]

[1]M. Auezov South Kazakhstan State University, Shymkent, Kazakhstan,
[2]Director of the office of the NGO National Academy of Sciences of the Republic of Kazakhstan South Kazakhstan region
aikerimkazenova@mail.ru, amb_52@mail.ru, golubev_50@mail.ru, aikerkazenova@gmail.com, nursara@mail.ru, shermahan_1984@mail.ru, kafiedra.topid@mail.ru

ACCOUNTING THETIME DELAY INTHEBINARY COAGULATION MODEL

Abstract. The paper deals with the time-delayed model for cluster aggregation processes based on the Smolukhovsky equation with the help of the heat and mass transfer kernels approach. The new non-local modification of the general kinetic equation of Smoluchowski binary aggregation equation hasbeen submitted. Forthecaseof an isotropic and homogeneous medium, the model has been reduced to the ordinary differential equation. The further possible development of the proposed model with allowance for taking into account the differences in the characteristic coagulation times during the aggregation of globules of different order has been discussed.

Keywords:aggregation equation, disperse phase, polydisperse, model, coagulation process, aggregation.

Introduction. The coagulation processes of the dispersed phase in heterogeneous physicochemical systems are widespread in various chemical technologies, metallurgy, and also in natural phenomena [1].

Currently, to describe the evolution of the concentration of i-mers in polydisperse systems in the irreversible aggregation processes, the Smolukhovsky equation is commonly used for the binary coagulation process [2, 3]:

$$\frac{\partial C_i}{\partial t} = \frac{1}{2}\sum_{j=1}^{i-1}\Phi_{i-j,j}C_{i-j}C_j - C_i\sum_{j=1}^{\infty}\Phi_{i,j}C_j \quad , \qquad (1)$$

where C_i - is the concentration of i-mers; $\Phi\ i, j$ - is the rate kernels.

Todaythevariousmodel representations are used to solve the problem of the aggregation kernels forms [1, 3].

In the work [4], a model approach to this problem was also proposed, based on the use of a peculiar parameter of the order of the aggregate formed during coagulation i – and j – mers:

$$\lambda = \frac{i-j}{i+j}, \qquad (2)$$

Under a small order ofi-mers, the main role may be played by an increase in the effective capture cross section with an increase in the characteristic radius of the particles, as well as a decrease in the mobility of the particles with an increase in their size and mass. A number of numerical experiments suggested that the coagulation rate kernel may have the following phenomenological form [3].

$$\Phi_{i,j} \approx \frac{k}{(i+j)^{\beta}} + a_2\left(\frac{i-j}{i+j}\right)^2, \tag{3}$$

Using the proposed approach to the description of the coagulation process using this model of the aggregation kernels is quite promising, because opens up the possibility for the model control with a set of parameters.

A numerical experiment showed that the proposed model gives a correct qualitative description of the coagulation processes, which is consistent with the known experimental data and model analysis using the methods of asymptotic expansions [1, 2].

However, the other side of the problem of modeling coagulation processes is currently practically undeveloped. It is about taking into account the time delaying in aggregation processes. This lacune in the theory of coagulation is also noted in the science literature [1]. Indeed, without taking into account the the time delay of aggregation, the Smoluhovsky equation is internally contradictory, since it does not describe the influence of the characteristic time of aggregate formation on the kinetics of the process.

The problems of taking into account relaxation times and long-range interactions of structural elements of the medium in the mathematical modeling of transport phenomena are of great practical and theoretical interest [5, 6]. These issues are particularly relevant in cases when it comes to fast processes, which cannot be considered as a local aggregation processes. The working cycle of such processes is short, and the whole process is carried out in transition mode. Therefore, the possibilities of managing intensive processes are limited, and the correct calculation and the choice of optimal values of the control parameters are of great importance.

This article deals with the time-delayed model based on the Smolukhovsky equation with the help of the approach proposed in [5, 6]. In accordance with this approach, the temporal nonlocality of mass transfer processes can be described on the basis of the model of the transport relaxation kernels [10-12]. Relaxation transfer kernels are called the cores of integral transformations that link the flows and thermodynamic forces in the statistical theory of dissipative processes [5, 6].

Mathematical model. The general structure of the mass fluxes of the components in the n-component system in accordance with the method of relaxation nuclei of transfer [6] has the following form:

$$J(R,t) = J(R,t_0) + \sum_{k=1}^{n+1}\iint dt_1 dR' N_{ik}(R,R',t,t_1)F_k(R',t), \tag{4}$$

For analyzing the general structure of the transport basic equations, the following type of relaxation transfer kernels can be used:

$$N_{ik}(R,t-t_1) = \eta_{ik}(R,t)\exp\left(-(t-t_1)/\tau_{ik}\right), \tag{5}$$

where $\eta_{ik}(R,t)$ is a local function [6, 7].

Thejustificationofthis type of relaxation kernels is based on the characteristic form of the differential equation of relaxation processes [6, 7, 8]:

$$\frac{\partial}{\partial t}N(R,s) + \frac{N(R,s)}{\tau}f(R,t) = 0, \tag{6}$$

In the paper [5, 6], a similar approach was used to modeling the relaxation kernels of heat and mass transfer in a two-component system with allowance for cross-effects in the form:

$$\frac{\partial N_m}{\partial t} = -N_m \tau_m^{-1} + N_h \tau_\times^{-1}, \tag{7}$$

$$\frac{\partial N_h}{\partial t} = N_m \tau_\times^{-1} - N_h \tau_h^{-1}, \tag{8}$$

The choice of signs in equations (7), (8) is determined by the conditions of conjugation of perturbations of the temperature and concentration fields [9, 10].

In the case of aggregation processes, the role of the relaxation times is played by the characteristic times of the aggregation of i- and j-mers. The following non-local modification of the Smoluchowski equation is proposed for the aggregation process in a polydisperse system [11, 12]:

$$\frac{\partial C_i}{\partial t} = \frac{1}{2}\sum_{j=1}^{i-1}\int dt_1 \Phi_{i-j,j}(t,t_1) C_{i-j}(t_1) C_j(t_1) - \sum_{j=1}^{\infty}\int dt_1 \Phi_{i,j}(t,t_1) C_i(t_1) C_j(t_1), \tag{9}$$

The model equations for the aggregation kernels, by analogy with (6), look as follows

$$\frac{\partial}{\partial t}\Phi_{i,j} + \frac{\Phi_{i,j}}{\tau_{i,j}} f_{i,j}^0 = 0, \tag{10}$$

With accounting the system (10), the integro-differential equations (9) take the form:

$$\begin{aligned}\frac{\partial C_i}{\partial t} = \frac{1}{2}\sum_{j=1}^{i-1}\int dt_1 \Phi_{i-j,j}^0 \exp\left(-\frac{f_{i-j,j}^0}{\tau_{i-j,j}}(t-t_1)\right) C_{i-j}(t_1) C_j(t_1) - \\ \sum_{j=1}^{\infty}\int dt_1 \Phi_{i,j}^0 \exp\left(-\frac{f_{i,j}^0}{\tau_{i,j}}(t-t_1)\right) C_i(t_1) C_j(t_1)\end{aligned} \tag{11}$$

For the case of an isotropic and homogeneous medium, relations (11) can be considered as ordinary differential equations

The temporal derivatives of the integral terms read

$$\Phi_{i,j}^0 C_i(t) C_j(t) + \int_0^t dt_1 C_i(t_1) C_j(t_1) \frac{d}{dt}\exp\left(-\frac{f_{i,j}^0}{\tau_{i,j}}(t-t_1)\right), \tag{12}$$

Then equations (9) can be converted to

$$\begin{aligned}\frac{d^2 C_i}{dt^2} = \frac{1}{2}\sum_{j=1}^{i-1}\Phi_{i-j,j}^0 C_{i-j}(t) C_j(t) - \sum_{j=1}^{\infty}\Phi_{i,j}^0 C_i(t) C_j(t) - \\ -\frac{1}{2}\sum_{j=1}^{i-1}\frac{f_{i-j,j}^0}{\tau_{i-j,j}}\int dt_1 \Phi_{i-j,j}^0 \exp\left(-\frac{f_{i-j,j}^0}{\tau_{i-j,j}}(t-t_1)\right) C_{i-j}(t_1) C_j(t_1) + \\ +\sum_{j=1}^{\infty}\frac{f_{i,j}^0}{\tau_{i,j}}\int dt_1 \Phi_{i,j}^0 \exp\left(-\frac{f_{i,j}^0}{\tau_{i,j}}(t-t_1)\right) C_i(t_1) C_j(t_1)\end{aligned} \tag{13}$$

In the zero approximation [13] the characteristic relaxation times are assumed to be independent of the order of the i-mers.

Then it can be denoted for all i , j :

$$\frac{f_{i,j}^0}{\tau_{i,j}} = \frac{f^0}{\tau}, \tag{14}$$

Equations (11) in this case take the form

$$\begin{aligned} \frac{d^2C_i}{dt^2} &= \frac{1}{2}\sum_{j=1}^{i-1}\Phi_{i-j,j}^0 C_{i-j}(t)C_j(t) - \sum_{j=1}^{\infty}\Phi_{i,j}^0 C_i(t)C_j(t) - \\ &- \frac{f^0}{\tau}\left(\frac{1}{2}\sum_{j=1}^{i-1}\int dt_1\Phi_{i-j,j}^0 \exp\left(-\frac{f^0}{\tau}(t-t_1)\right)C_{i-j}(t_1)C_j(t_1) + \right. \\ &\left. + \sum_{j=1}^{\infty}\int dt_1\Phi_{i,j}^0 \exp\left(-\frac{f^0}{\tau}(t-t_1)\right)C_i(t_1)C_j(t_1)\right) \end{aligned} \tag{15}$$

From equations (11) and (15) the following time-delaying modification of the Smoluchowski equation can be obtained:

$$\frac{d^2C_i}{dt^2} + \frac{f^0}{\tau}\frac{dC_i}{dt} = \frac{1}{2}\sum_{j=1}^{i-1}\Phi_{i-j,j}^0 C_{i-j}(t)C_j(t) - \sum_{j=1}^{\infty}\Phi_{i,j}^0 C_i(t)C_j(t), \tag{16}$$

A feature of equation (16) is the presence of solutions that describe the propagation of disturbances with a finite velocity [9, 10].

Further development of the proposed model may consist in taking into account the differences in the characteristic coagulation times during the aggregation of globules of different order.The estimate for the fractal dimension of the active unscreened surface can be obtained by counting the active centers arising after the capture and penetration of a new cluster into another cluster.

The number of arising reactive centers N_a can be evaluated as follows [14-16]:

$$N_a \sim R^{d_f - 1} \cdot \lambda, \tag{17}$$

where R is the characteristic radius of the cluster-recipient, λ is the characteristic penetration depth for the captured cluster (Figure 1).

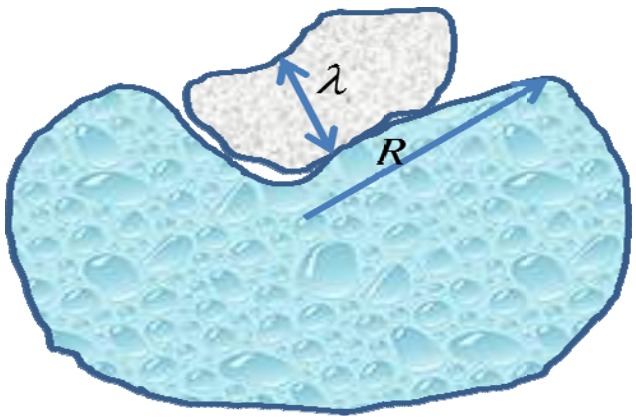

Figure 1 - Capture and penetration of clusters under the aggregation

Conclusions.It is shown that heat and mass transfer kernel approach allows for obtaining a time-delaying kinetic model of binary cluster aggregation, and to modify the Smoluchowski equation to the non-local type with reducing to an ordinary differential equation. Estimation for accounting differences between the characteristic coagulation times for clusters of different orders leads to calculations of fractal dimensions and counting the active centers arising after the capture and penetration of a new cluster into another cluster.Thisproblemneedsfurther analysis.

А.М. Бренер[1], А.О. Казенова[1], В.Г. Голубев[1],
Л.Т. Тәшімов[2], Ш.К. Шапалов[1], Г.Д. Кенжалиева[1], Д.Р. Тортбаева[1]

[1]М. Әуезов атындағы ОҚМУ, Шымкент, Қазақстан;
[2]Оңтүстік Қазақстан облысы бойынша ҚР ҰҒА РОО өкілеттілігінің директоры,

БИНАРЛЫҚ КОАГУЛЯЦИЛАУ МОДЕЛІНІҢ УАҚЫТТЫҚ ТОҚТАУЫН ЕСЕПТЕУ

Аннотация. Мақала жылу масса алмасу ядроларының әдісі көмегімен Смолуховский теңдеуіне негізделген кластерлерді агрегациялау процестеріне уақыт бойы тоқтаумен модельдерді жасауға арналған. Смолуховскийдің агрегациялаудың бинарлы теңдеуінің жалпы кинематикалық теңдеуінің жаңа оқшаулап шектелмеген түрлендірілуі келтірілген. Изототропты және біртектік орта жағдайы үшін модель қарапайым дифференциалды теңдеуге келтірілген. Әр түрлі ретті глобулаларды агрегациялау кезінде коагуляцияның сипаттық уақыттарының айырмасын ескере отырып, ұсынылған модельдің ары қарай даму мүмкіндігі талқыланады.

Түйін сөздер: агрегациялау теңдеуі, дисперстің фаза, полидисперсия, модель, коагуляция, агрегация.

А.М. Бренер[1], А.О. Казенова[1], В.Г. Голубев[1], Л.Т. Ташимов[2],
Ш.К. Шапалов[1], Г.Д. Кенжалиева[1], Д.Р. Тортбаева[1]

[1]Южно-Казахстанский государственный университет им.М.Ауэзова, Шымкент, Казахстан,
[2]Председатель филиала РОО НАН РК по Южно-Казахстанской области,

УЧЕТ ВРЕМЕННОЙ ЗАДЕРЖКИ В МОДЕЛИ БИНАРНОЙ КОАГУЛЯЦИИ

Аннотация. Статья посвящена разработке модели с задержкой по времени для процессов агрегации кластеров, основанной на уравнении Смолуховского с помощью метода ядер тепломассопереноса. Представлена новая нелокальная модификация общего кинетического уравнения бинарного уравнения агрегации Смолуховского. Для случая изотропной и однородной среды модель приведена к обыкновенному дифференциальному уравнению. Обсуждается возможность дальнейшего развития предложенной модели с учетом различий в характерных временах коагуляции при агрегации глобул различного порядка.

Ключевые слова:уравнения агрегации, дисперсная фаза, полидисперсия, модель, процесс коагуляции, агрегация.

Information about authors:

Kazenova Aykerim - PhD student of the Department of "Technological Machines and Equipment", M.Auezov South Kazakhstan State University, aikerimkazenova@mail.ru;

Brener Arnold - Doctor of Technical Sciences, professor of the Department of "Computing Systems and Software", M.Auezov South-Kazakhstan State University, amb_52@mail.ru;

Golubev Vladimir Grigorievich - Doctor of Technical Sciences, professor of the Department of "Oil & Gas Business", M.Auezov South Kazakhstan State University, golubev_50@mail.ru;

Tashimov Lesbekov Tashimovich - Doctor of Technical Sciences, professor, academician NAS RK, Director of the office of the NGO National Academy of Sciences of the Republic of Kazakhstan South Kazakhstan region, aikerkazenova@gmail.com;

Shapalov Shermakhan Kuttibayevich - PhD, senior teacher Department of «Life safety and environmental protection»; M. Auezov South Kazakhstan State University, shermahan_1984@mail.ru;

Kenzhalieva Gulmira - Candidate of technical science, associate professor of t Department Department of «Life safety and environmental protection», M.Auezov South-Kazakhstan State University, nursara@mail.ru;

Tortbayeva Dinara Ryskulbekkyzy - Candidate of technical science, associate professorof the Department of"Transport, transportation and traffic", M.Auezov South Kazakhstan State University, kafiedra.topid@mail.ru;

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