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Механика Механика Mechanics

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Non-local mathematical models for aggregation processes in dispersive media

Particles aggregation is widespread in different technological processes and nature, and there are many approaches to modeling this phenomenon. However, the time non-locality effects with witch these processes are often accompanied leave to be none well elaborated at present. This problem is justified especially in reference to nano-technological processes. The paper is devoted to the non-local modification of Smoluchowski equation that is the key point for describing influence of synchrony and asynchrony delays in aggregation processes for clusters of different orders. The main scientific contribution consists in deriving the non-linear wave equation describing the evolution of different orders clusters concentration under aggregation processes in polydispersed systems with following for the mentioned non-locality. The practical significance lies in the fact that the results obtained can serve as the basis for the engineering calculation of the kinetics of aggregation in polydisperse nano-systems. The research methodology is based on mathematical modeling with the help of the relaxation transfer kernels approach. Succeeding analysis of aggregation processes on the base of submitted ideology can be directed to generalizing master equations with allowing for space non-locality too. The submitted approach opens up fresh opportunities for detailed study of influence of relaxation times hierarchy on the intensity of aggregation and gelation processes in non-crystalline media containing dispersed solid phase.

Key words: aggregation, dispersive systems, non-local model, kinetic equation, relaxation times.

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Дисперсті ортадағы агрегация процестерінің локальды емес математикалық модельдері

Бөлшектердің агрегациясы әртүрлі технологиялық процестер мен табиғатта кең таралған және бұл құбылысты модельдеудің көптеген тәсілдері бар. Алайда, бұл процестер жиі жүретін уақытқа жергілікті емес әсері қазіргі уақытта жеткіліксіз зерттелген. Бұл мәселе әсіресе нанотехнологиялық процестерге қатысты негізделген. Мақала Смолуховский теңдеуінің локалды емес модификациясына арналған, ол әр түрлі ретті кластерлер үшін агрегация процестеріндегі синхрондылық пен асинхрондылықтың кідірістерінің әсерін сипаттаудың кілттік мезеті болып табылады. Негізгі ғылыми үлес - бұл жергілікті емес жағдайды ескере отырып, полидисперсті жүйелердегі агрегация процестеріндегі әртүрлі тапсырыс кластерлерінің шоғырлану эволюциясын сипаттайтын сызықты емес толқындық теңдеуді шығару. Практикалық маңыздылығы - алынған нәтижелер полидисперсті наносистемалардағы агрегация кинетикасын инженерлік есептеу үшін негіз бола алады. Зерттеу әдістемесі релаксация ядросының тәсілін қолдана отырып, математикалық модельдеуге негізделген. Ұсынылған идеологияға негізделген агрегаттау процестерін кейінгі талдау кеңістіктің жергілікті еместігін ескере отырып, негізгі теңдеулерді жалпылауға бағытталуы мүмкін. Ұсынылған тәсіл релаксация иерархиясының дисперсті қатты фазасы бар кристалды емес ортадағы агрегация және гель түзілу процестерінің қарқындылығына әсерін егжей-тегжейлі зерттеуге жаңа мүмкіндіктер ашады.

Түйін сөздер: агрегация, дисперсиялық жүйелер, локальды емес модель, кинетикалық теңдеу, релаксация уақыты.

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Нелокальные математические модели процессов агрегации в дисперсивных средах

Агрегация частиц широко распространена в различных технологических процессах и природе, и существует множество подходов к моделированию этого явления. Однако эффекты нелокальности во времени, с которыми часто сопровождаются эти процессы, в настоящее время недостаточно проработаны. Эта проблема оправдана, особенно применительно к нанотехнологическим процессам. Статья посвящена нелокальной модификации уравнения Смолуховского, которая является ключевым моментом для описания влияния задержек синхронности и асинхронности в процессах агрегации для кластеров разного порядка. Основной научный вклад заключается в выводе нелинейного волнового уравнения, описывающего эволюцию концентрации кластеров различных порядков при процессах агрегации в полидисперсных системах с учетом указанной нелокальности. Практическая значимость заключается в том, что полученные результаты могут послужить основой для инженерного расчета кинетики агрегации в полидисперсных наносистемах. Методология исследования основана на математическом моделировании с помощью подхода ядер переноса релаксации. Последующий анализ процессов агрегирования на основе представленной идеологии может быть направлен на обобщение основных уравнений с учетом также нелокальности пространства. Представленный подход открывает новые возможности для детального изучения влияния иерархии времен релаксации на интенсивность процессов агрегации и гелеобразования в некристаллических средах, содержащих дисперсную твердую фазу.

Ключевые слова: агрегация, дисперсионные системы, нелокальная модель, кинетическое уравнение, времена релаксации.

1 Introduction and problem set up

Particles aggregation is widespread in different chemical technological processes, metallurgy pharmaceutical industry. Because of that many approaches to modeling this phenomenon are offered [1]. However, a lot of key issues in the aggregation processes description leave to be none elaborated up to day [2, 3, 4]. One of the important and pracically non elaborated problems is time non-locality of aggregation processes [5, 6, 7]. However, it is impossible to describe the influence of characteristic relaxation times on aggregates formation kinetics without allowing for the non- locality aspect [8, 9, 10]. It is justified especially in reference to nano-technological processes.

This paper is devoted to the non-local modification of Smoluchowski equation for particles aggregation kinetics. There are not discussed some especially physical problems as, for example, particles nucleation, etc. But we try to understand and to emphasize some difficulties emerging in description of the non-locality applying to the aggregation kinetic equations.

The following non-local modification of the Smoluchowski equation for aggregation in the uniform system can be offered as the principal ansatz with allowance for the general case of asynchrony delays for clusters of different orders formation:

$$\frac{dC_i}{dt} = \frac{1}{2}\sum_{j=1}^{i-1}\int_0^t\int_0^t N_{j,i-j}C_j(t_1)C_{i-j}(t_2)dt_1dt_2 - \sum_{j=1}^{\infty}\int_0^t\int_0^t N_{i,j}C_i(t_1)C_j(t_2)dt_1dt_2 \tag{1}$$

C_i denotes the concentration of i-mers, and aggregation kernels $N_{i,j}$ are functions of the delay times $t - t_1$ and $t - t_2$.

Form (1) follows from our detail consideration of relaxation kernels method applying to heat and mass transfer problems [2].

The general linearized model for the aggregation matrix can be obtained from the model equation for transfer kernels which is submitted in [3, 4]:

$$r_i\frac{\partial N_{i,j}}{\partial s_i} + r_j\frac{\partial N_{i,j}}{\partial s_j} + \frac{\partial f^0_{i,j}}{\tau_{i,j}}N_{i,j} = 0 \tag{2}$$

where $s_i = t - t_1, \qquad s_j = t - t_2$.

In equation (2) the coefficients r_i on a level with relaxation time $\tau_{i,j}$ play a part of control parameters of clusters "inertness", the parameter f answers for media and particles characteristics.

Thus the aggregation matrix, satisfying equation (2) and coming up to the condition of fast relaxation in time $t >> \tau_{i,j}$, can be written as

$$N_{i,j} = \eta^0_{i,j}exp\Big(-\frac{f^0_{i,j}}{2\tau_{i,j}}\left(\frac{s_i}{r_i}+\frac{s_j}{r_j}\right)\Big). \tag{3}$$

The master equation reads

$$\frac{dC_i}{dt} = \frac{1}{2}\sum_1 \eta_{j,i-j}exp(-(g^{(j)}_{j,i-j}+g^{(i-j)}_{j,i-j}t)I_1I_2 - \sum_2 \eta_{i,j}exp(-(g^{(i)}_{i,j}+g^{(j)}_{i,j}t)I_3I_4 \tag{4}$$

Here

$$g^{(i)}_{m,n} = \frac{a_{m,n}}{2r_i}; \qquad g^{(j)}_{m,n} = \frac{a_{m,n}}{2r_j}; \qquad a_{m,n} = \frac{f^0_{m,n}}{\tau_{m,n}}$$

$$I_1 = \int_0^t exp(g^{(i-j)}_{j,i-j}s)C_{i-j}(s)ds; \qquad I_2 = \int_0^t exp(g^{(j)}_{j,i-j}s)C_j(s)ds;$$

$$I_3 = \int_0^t exp(g^{(j)}_{i,j}s)C_j(s)ds; \qquad I_4 = \int_0^t exp(g^{(i)}_{i,j}s)C_i(s)ds.$$

At the present time the clear way to rigorous reducing general governing equation (4) to an ODE form is unknown [8, 11].

So, let's assume at the beginning $r_i = r_j = 1$ and $\frac{f^0_{i,j}}{\tau_{i,j}} \equiv a_{i,j} = a = const.$

Thus we have

$$\frac{dC_i}{dt} = \frac{1}{2}exp(-at)\sum_1 \eta_{j,i-j}I_1I_2 - exp(-at)I_3\sum_2 \eta_{i,j}I_2. \tag{5}$$

Here $\sum_1$ means $\sum_{j=1}^{i-1}$; $\sum_2$ means $\sum_{j=1}^{\infty}$; $I_1 = \int_0^t exp(as/2)C_{i-j}(s)ds$;

$$I_2 = \int_0^t exp(as/2)C_j(s)ds; \quad I_3 = \int_0^t exp(as/2)C_i(s)ds$$

.

The main contribution of this work lies in that the features of the influence of the nonlocality on the aggregation process in cases of synchronous and asynchronous delays in the formation of different orders clusters have been highlighted and separately considered.

2 Materials and methods

2.1 Asymptotic analysis for asynchrony case

In order to simplify the problem it is supposed that for small relaxation times the Laplace method in the neighborhood of the time point t can be used. Immediate substitution of the integrals expansions into equation (5) requires multiplying asymptotic sequences. Such procedure is badly conditioned, as it may lead to utter loss while checking orders of approximation.

Therefore, we rearrange the equations to the form which is free from a product of integrals:

$$\frac{d^2C_i}{dt^2} + a\frac{dC_i}{dt} = \frac{1}{2}exp\left(-\frac{at}{2}\right)\sum_1 \eta_{j,i-j}(C_jI_1 + C_{i-j}I_2) -$$
$$-exp\left(-\frac{at}{2}\right)\left[C_i\sum_2 \eta_{i,j}I_2 + I_3\sum_2 \eta_{i,j}C_j\right]. \tag{6}$$

Using then Laplace method we obtain the asymptotic relations in which the orders of equations and approximations are concerted:

$$I_1^{(1)} = \frac{2}{a}\left[exp\left(\frac{at}{2}\right)C_{i-j}(t) - C_{i-j}(0)\right] - \frac{4}{a^2}\left[exp\left(\frac{at}{2}\right)\frac{dC_{i-j}}{dt} - \frac{dC_{i-j}(0)}{dt}\right], \tag{7}$$

$$I_2^{(1)} = \frac{2}{a}\left[exp\left(\frac{at}{2}\right)C_j(t) - C_j(0)\right] - \frac{4}{a^2}\left[exp\left(\frac{at}{2}\right)\frac{dC_j}{dt} - \frac{dC_j(0)}{dt}\right], \tag{8}$$

$$I_3^{(1)} = \frac{2}{a}\left[exp\left(\frac{at}{2}\right)C_i(t) - C_i(0)\right] - \frac{4}{a^2}\left[exp\left(\frac{at}{2}\right)\frac{dC_i}{dt} - \frac{dC_i(0)}{dt}\right]. \tag{9}$$

As a result, we get

$$\frac{d^2C_i}{dt^2} + a\frac{dC_i}{dt} = \frac{2}{a}\sum_1 \eta_{j,i-j}\left[C_jC_{i-j} - \frac{1}{a}\frac{d}{dt}(C_jC_{i-j})\right] - \frac{4}{a}\sum_2 \eta_{i,j}\left[C_iC_j - \frac{1}{a}\frac{d}{dt}(C_iC_j)\right] -$$
$$-\frac{1}{a}exp(-\frac{at}{2})\sum_1 \eta_{j,i-j}\left[C_j\left(C_{i-j}(0) - \frac{2}{a}\frac{dC_{i-j}(0)}{dt}\right) + C_{i-j}\left(C_j(0) - \frac{2}{a}\frac{dC_j(0)}{dt}\right)\right] +$$
$$+\frac{2}{a}exp(-\frac{at}{2})\sum_2 \eta_{i,j}\left[C_i\left(C_j(0) - \frac{2}{a}\frac{dC_j(0)}{dt}\right) + C_j\left(C_i(0) - \frac{2}{a}\frac{dC_i(0)}{dt}\right)\right] \tag{10}$$

Let's assume $\frac{1}{a} = \tau_*$. Parameter τ_* has a time dimension. So, let T be the characteristic time of the process.

Introducing the small parameter $\varepsilon = \tau_*/T$ we can pass to the dimensionless time $\theta = t/T$ and dimensionless aggregation kernels $\overline{\eta}_{i,j} = T^3\eta_{i,j}$.

Thus equation (10) can be rearranged to the following form:

$$\varepsilon\frac{d^2C_i}{d\theta^2} + \frac{dC_i}{d\theta} = 2\varepsilon^2\sum_1 \overline{\eta}_{j,i-j}\left[C_jC_{i-j} - \varepsilon\frac{d}{d\theta}(C_jC_{i-j})\right] - 4\varepsilon^2\sum_2 \overline{\eta}_{i,j}\left[C_iC_j - \varepsilon\frac{d}{d\theta}(C_iC_j)\right]$$

$$-\varepsilon^2 exp\left(-\frac{\theta}{2\varepsilon}\right)\sum_1 \overline{\eta}_{j,i-j}\left[C_j\left(C_{i-j}(0) - 2\varepsilon\frac{dC_{i-j}(0)}{d\theta}\right) + C_{i-j}\left(C_j(0) - \varepsilon\frac{dC_j(0)}{d\theta}\right)\right] +$$

$$+2\varepsilon^2 exp(-\frac{\theta}{2\varepsilon})\sum_2 \overline{\eta}_{i,j}\left[C_i\left(C_j(0) - 2\varepsilon\frac{dC_j(0)}{d\theta}\right) - C_j\left(C_i(0) - 2\varepsilon\frac{dC_i(0)}{d\theta}\right)\right] \tag{11}$$

Ignoring the fast decreasing terms at time $t >> \tau_*$ we obtain the reducing form of master equation

$$\varepsilon\frac{d^2C_i}{d\theta^2} + \frac{dC_i}{d\theta} = 2\varepsilon^2\sum_1 \overline{\eta}_{j,i-j}\left[C_jC_{i-j} - \varepsilon\frac{d}{d\theta}(C_jC_{i-j})\right] - 4\varepsilon^2\sum_2 \overline{\eta}_{i,j}\left[C_iC_j - \varepsilon\frac{d}{d\theta}(C_iC_j)\right]. \tag{12}$$

Essential difference between solutions of equations (11) and (12) may be observed at the initial period $\Delta\theta_{in}$:

$$\Delta\theta_{in} \sim -\varepsilon ln\varepsilon. \tag{13}$$

Depending on the specific correlation between values of the relaxation time and aggregation kernels we consider three different types of the aggregation process. They are the fast, moderate and slow aggregation:

$$\eta_{i,j} = O(1/\tau_*^2), \qquad \eta_{i,j} = O(1/\tau_*), \qquad \eta_{i,j} = O(1). \tag{14}$$

In any case, singularly perturbed kinetic equations should be obtained.

It's obvious that equation (12) can be reduced to the well-known Smoluchowski equation on the zero-approximation only in the case of fast aggregation.

3 Modified third order model for the synchronic delays case

Let us consider a modification of the Smoluchowski equation taking into account the synchronous time lag of the aggregation of clusters of different orders, which is designed to describe the effect of the characteristic time of aggregate formation on the kinetics of the process [2, 10].

Then the following nonlocal modification of the Smoluchowski equation is proposed for the aggregation process in a polydisperse system [10, 12]:

$$\frac{\partial C_i}{\partial t} = \frac{1}{2}\sum_{j=1}^{i-1}\int dt_1\Phi_{i-j,j}(t,t_1)C_{i-j}(t_1)C_j(t_1) - \sum_{j=1}^{\infty}\int dt_1\Phi_{i,j}(t,t_1)C_i(t_1)C_j(t_1). \tag{15}$$

Model equations for the elements of the coagulation matrix are as follows [10]:

$$\frac{\partial}{\partial}\Phi_{i,j} + \frac{\Phi_{i,j}}{\tau_{i,j}} f^0_{i,j} = 0. \tag{16}$$

Then the integro-differential equations take the form:

$$\frac{\partial C_i}{\partial t} = \frac{1}{2}\sum_{j=1}^{i-1}\int dt_1 \Phi^0_{i-j,j} exp\left(-\frac{f^0_{i-j,j}}{\tau_{i-j,j}}(t-t_1)\right) C_{i-j}(t_1)C_j(t_1) -$$
$$-\sum_{j=1}^{\infty}\int dt_1 \Phi^0_{i,j}(t,t_1) exp\left(-\frac{f^0_{i,j}}{\tau_{i,j}}(t-t_1)\right) C_i(t_1)C_j(t_1). \tag{17}$$

The time derivatives of the integral terms have the form

$$\Phi^0_{i,j}C_i(t)C_j(t) - \frac{f^0_{i,j}}{\tau_{i,j}}\Phi^0_{i,j}\int_0^t dt_1 C_i(t_1)C_j(t_1) exp\left(-\frac{f^0_{i,j}}{\tau_{i,j}}(t-t_1)\right). \tag{18}$$

Then the governing equation can be transformed to the form:

$$\frac{d^2C_i}{dt^2} = \frac{1}{2}\sum_{j=1}^{i-1}\Phi^0_{i-j,j}C_{i-j}(t)C_j(t) - \sum_{j=1}^{\infty}\Phi^0_{i,j}C_i(t)C_j(t) -$$
$$-\frac{1}{2}\sum_{j=1}^{i-1}\frac{f^0_{i-j,j}}{\tau_{i-j,j}}\int dt_1 \Phi^0_{i-j,j} exp\left(-\frac{f^0_{i-j,j}}{\tau_{i-j,j}}(t-t_1)\right) C_{i-j}(t_1)C_j(t_1) + \tag{19}$$
$$+\sum_{j=1}^{\infty}\frac{f^0_{i,j}}{\tau_{i,j}}\int dt_1 \Phi^0_{i,j} exp\left(-\frac{f^0_{i,j}}{\tau_{i,j}}(t-t_1)\right) C_i(t_1)C_j(t_1).$$

Taking the time derivative again the following equation can be derived:

$$\frac{d^3C_i}{dt^3} = \frac{d}{dt}\left(\frac{1}{2}\sum_{j=1}^{i-1}\Phi^0_{i-j,j}C_{i-j}(t)C_j(t) - \sum_{j=1}^{\infty}\Phi^0_{i,j}C_i(t)C_j(t)\right) - \frac{1}{2}\sum_{j=1}^{i-1}\frac{f^0_{i-j,j}}{\tau_{i-j,j}}\Phi^0_{i-j,j}C_{i-j}(t)C_j(t) +$$
$$+\frac{1}{2}\sum_{j=1}^{i-1}\left(\frac{f^0_{i-j,j}}{\tau_{i-j,j}}\right)^2\int dt_1 \Phi^0_{i-j,j} exp\left(-\frac{f^0_{i-j,j}}{\tau_{i-j,j}}(t-t_1)\right) C_{i-j}(t_1)C_j(t_1) + \tag{20}$$
$$+\sum_{j=1}^{\infty}\frac{f^0_{i,j}}{\tau_{i,j}}\Phi^0_{i,j}C_i(t_1)C_j(t_1) - \sum_{j=1}^{\infty}\left(\frac{f^0_{i,j}}{\tau_{i,j}}\right)^2\int dt_1 \Phi^0_{i,j} exp\left(-\frac{f^0_{i,j}}{\tau_{i,j}}(t-t_1)\right) C_i(t_1)C_j(t_1).$$

Performing a separate averaging over the groups of indices for the terms describing the formation and destruction i- measures, the following system has been obtained

$$\frac{d^3C_i}{dt^3} = \frac{d}{dt}\left(\frac{1}{2}\sum_{j=1}^{i-1}\Phi^0_{i-j,j}C_{i-j}(t)C_j(t) - \sum_{j=1}^{\infty}\Phi^0_{i,j}C_i(t)C_j(t)\right) - \frac{1}{2}A_1\sum_{j=1}^{i-j}\Phi^0_{i-j,j}C_{i-j}(t)C_j(t) +$$

$$+\frac{1}{2}B_1^2\sum_{j=1}^{i-1}\int dt_1\Phi^0_{i-j,j}exp\left(-\frac{f^0_{i-j,j}}{\tau_{i-j,j}}(t-t_1)\right)C_{i-j}(t_1)C_j(t_1)+ \tag{21}$$

$$+A_2\sum_{j=1}^{\infty}\Phi^0_{i,j}C_i(t)C_j(t)-B_2^2\sum_{j=1}^{\infty}\int dt_1\Phi^0_{i,j}exp\left(-\frac{f^0_{i,j}}{\tau_{i,j}}(t-t_1)\right)C_i(t_1)C_j(t_1).$$

After transformations, a more compact form of the system has been obtained

$$\frac{d^3C_i}{dt^3}+(B_1+B_2)\frac{d^2C_i}{dt^2}+B_1B_2\frac{dC_i}{dt}=$$

$$=\left(B_1+B_2+\frac{d}{dt}\right)\left(\frac{1}{2}\sum_{j=1}^{i-1}\Phi^0_{i-j,j}C_{i-j}(t)C_j(t)-\sum_{j=1}^{\infty}\Phi^0_{i,j}C_i(t)C_j(t)\right)- \tag{22}$$

$$-\frac{1}{2}A_1\sum_{j=1}^{i-1}\Phi^0_{i-j,j}C_{i-j}(t)C_j(t)+A_2\sum_{j=1}^{\infty}\Phi^0_{i,j}C_i(t)C_j(t).$$

A feature of the obtained equation (22) is the presence of solutions describing the propagation of perturbations with a finite velocity in the form of single waves [8].

At the same time, the analysis of the obtained equation shows that for small values of the parameter τ/τ_c, the use of the local form of the Smoluchowski equations with aggregation matrices obeying equations of the form (3) is quite correct, since the correction to the local form has no less than the second order of smallness [10].

4 Conclusion

In this paper a brief introduction to the problem of time nonlocality applying to aggregation process kinetics has been submitted. The main result is that the relaxation kernels approach may be advantageous for deriving master equations with accounting of hierarchy of relaxation times.

Comparing the obtained equations with equations submitted in [8] we notice that account of different time delays for clusters of different orders essentially changes the form of kinetic equations. This circumstance can especially show itself at the initial time when the master equation must be considered in extended form (11). Need for the information about derivatives of clusters production at the initial time manifests, in our opinion, more profound physical content of the submitted model. Of course, this information is out of the competence of the model as such. A separate description of synchronous and asynchronous delays in the formation of clusters of different orders has shown significant differences in the fundamental models of the kinetics of aggregation processes in these situations.

Succeeding analysis of aggregation processes on the base of submitted ideology can be directed to generalizing master equations with allowing for space non-locality too. In our opinion the submitted approach opens up fresh opportunities for detailed study of influence of relaxation times hierarchy on the intensity of aggregation and gelation processes in non-crystalline media containing dispersed solid phase. This opens up new possibilities not only for calculating kinetics, but also for developing new approaches to controlling fine technological processes.

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