Numerical simulation of the particle size distribution function evolution of an ultradispersed system in a vapor-gas medium

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Abstract. To control the process of nanopowders generating in order to ensure the required characteristics, it is necessary to solve the general physical problem of vaporization and condensation of steam in a gas-phase synthesis plant. A model of the physical problem should describe besides the formation of condensed particles, the hydrodynamic features of the flow, as well as heat exchange processes with other objects inside the working chamber. The questions of interaction of particles within an ultradispersed system through coagulation were considered. A mathematical model that describes the process of coagulation of a monodisperse system inside the evaporation-condensation chamber was formulated. A variant of the optimization of a coagulation numerical model in an ultradispersed system by reducing the number of sections corresponding to particle fractions of the mass scale was proposed. The use of an reduced scale made it possible to optimize the information received, and also gave advantages in the speed of calculations, which made it possible to reduce the time spent in problems with coagulation of systems.

1. Introduction
The phenomenon of gas-phase deposition appears in many fields of technology. In some areas, like evaporation-condensation units, that process is implemented for powders obtaining; in others, it occurs as a side effect, for example, when various gases or vapors are compressed in a turboexpander that leading to the formation of a condensed phase. In all cases, it becomes necessary to monitor the condensed phase particles formation process, which implies monitoring the parameters in the condensation zone. Because of complexity of the processes, it is necessary to coordinate the choice of one or another operating regime of the device being operated with a huge number of factors. Particles are formed by nucleation, condensation-evaporation on droplets and coagulation, which provides evolution of the dispersed system. To provide the required parameters of particles, the control of the gas-phase deposition processes characteristics is needed, so the most effective approach in such case is to create a numerical mathematical model that could take into account not only these processes, but also the hydrodynamic flow aspects, as well as heat exchange processes with other objects. Such a model makes it possible to connect all the specified parameters of particle generation, and also allows one to obtain all the necessary information about the particle size distribution function – the key characteristic of a dispersed system. The physics of particle formation by nucleation and condensation growth is satisfactorily described thermodynamically [1]. However, the describing of the interaction between particles arise many questions, since it becomes necessary to take into account a huge number of chaotically moving
objects (which can reach up to \(10^{23}\) per m\(^3\)), that is further complicated by the lack of a clear understanding of what actually happens during the interaction of nanosized particles. It is quite important how coagulation behaves under different hydrodynamic conditions. The dynamics of coagulation can also depend on hydrodynamic conditions, which significantly affects the final distribution of particles over the region.

In this paper, the optimization of numerical simulation of particle coagulation inside an ultradispersed system was considered. For calculating particles coagulation, the system of equations is too cumbersome. A method that allows one to reduce the number of equations was proposed and also the optimization of the scale of the types of particles involved in the calculations was carried out.

2. Statement of the problem

The aim of the study is to optimize the numerical calculation method of the mathematical coagulation model. The term of the interest is how the optimization of the computational process of coagulation will change the calculation of the distribution function and how much the result of calculation will differ from the one of the previous calculation scheme.

Other forms of interaction, such as droplet fragmentation, weren’t considered in this work. The so-called Brownian coagulation was considered, its principle based on the chaotic motion of particles of the condensed phase. There are several regimes of Brownian coagulation, which are depending on the transport conditions of the coagulating system – free-molecular, transition and near-continuum. The coagulation mode can be determined by the Knudsen number: \(Kn = \lambda / r\), where \(\lambda\) is the free path, \(r\) is the radius of the particles \([2]\).

In our case, it was assumed that the Knudsen number was in the range from 50 and higher, i.e. the considered coagulation mode corresponds to the free-molecular mode.

To find the size distribution evolution, a system of equations describing the process of coagulation of nanoparticles inside a vapor-gas medium of glycerol vapor was compiled. As the initial parameters, the concentration in the system was set less than \(10^{23} 1 / \text{m}^3\).

3. Model description

The problem of coagulation of glycerol nanodroplets inside the chamber of an evaporation-condensation unit at a constant temperature of the system and no flow was considered.

The following assumptions applied:

- the system is in thermodynamic equilibrium \([1]\);
- the system is isolated from the environment;
- the temperature of all elements of the system is constant and the same;
- initial distribution of particles is monodisperse;
- only double collisions are counted;

The size distribution of particles subjected to Brownian coagulation in the free molecular regime was described by the following equation \([3]\):

\[
\frac{\partial f(v, t)}{\partial t} = \frac{1}{2} \int_0^v \alpha(v - \bar{v}, \bar{v}) f(v - \bar{v}, t) f(\bar{v}, t) \, d\bar{v} - f(v, t) \int_0^v \alpha(v, \bar{v}) f(\bar{v}, t) \, d\bar{v},
\]

where \(f(v, t)\) is the density function of the number of particles, \(t\) is time, \(\alpha(v, \bar{v})\) is the collision nucleus for two particles of volume \(v\) and \(\bar{v}\).

The expression for the collision coefficients was derived using a number of approximations \([1]\): the assumption of the sphericity of liquid nanoparticles, the model of collision of hard spheres, the hypothesis of active collisions in which all collisions lead to the merging of particles. Combined with the kernel \([3]\), we get:
\[ a(v, \bar{v}) = \left( \frac{v}{v_0} \right)^{1/3} + \left( \frac{\bar{v}}{v_0} \right)^{1/3} \right) \sqrt{\frac{v + \bar{v}}{v\bar{v}}} R_0^2 \sqrt{\frac{8\pi R T}{m}} \exp \left( -\frac{E_a}{k_B T} \right), \tag{2} \]

where \( v_0 \) is the volume of a particle in the 1st section, \( k_B \) is the Boltzmann constant, \( T \) is the absolute temperature, and \( p \) is the particle density, \( n_{\text{max}} \) is the maximum number of types of particles, \( E_a \) is the activation energy, \( R_0 \) is the size of the first section particle.

4. Numerical model

4.1. Scale optimization

The discrete Smoluchowski equation is usually solved on the mass scale, starting by the unit element \( m_0 \), then \( 2m_0 \), and then linearly up to \( n_{\text{max}} m_0 \). The system of equations that corresponds to the linear scale is written as:

\[ \frac{dN_i}{dt} = \frac{1}{2} \sum_{j=1}^{n-1} a_{i-j,j} N_i N_j - N_i \sum_{j=1}^{n} a_{ij} N_j, \quad i = 1 + n_{\text{max}}. \tag{3} \]

In this case, the particle size varies from 1 nm to 1 μm, and therefore \( n_{\text{max}} \) can reach values around \( 10^9 \), and so the same number of Smoluchowski equations must be solved. Such a number of calculations undoubtedly exerts a significant load on the computing system. To reduce it a dynamic mass scale with a progressive step is used [4]; an example is a grid from a numerical progression with a factor of 2. In this paper, we investigate the application of a grid based on the Fibonacci series; 1,2,3,5,8,13, etc.

4.2. System of equations

After a scale changing, the layout of the sections was changed according to the Fibonacci number series. The nodes of the new scale which correspond to the nodes from the linear scale, with numbers that numerically correspond to the numbers of the Fibonacci scale, are now like “separately considered” from linear scale, and so the coagulation equations was written only for these nodes. The system of equations was written in several stages. At the first stage, a table function at the initial moment of the calculation the initial particle distribution by the size is approximating. The lognormal distribution is used as an approximating function, which calculates the value of the particle density in the interval from \( m \) to \( m + dm \). It was written depending on the volume of particles \( v \):

\[ f(v, t) = \frac{N(t)}{3\sqrt{2\pi} \ln \sigma(t)} \exp \left\{ -\frac{\ln^2 \left( v/v_g \right)}{18\ln^2 \sigma(t)} \right\} \left( \frac{1}{v} \right). \tag{4} \]

where \( v_g(t) \) is the geometric mean volume of particles, \( \sigma(t) \) is the geometric standard deviation based on particle size, and \( N(t) \) is the total number of particles.

Using the distribution formulae (4) in equations (3), we obtain the equations for the rate of change in the number of particles at the nodes from the modified scale. Since the nodes of the new scale correspond to the nodes of the linear scale, which numbers numerically similar to the numbers of the Fibonacci scale, the coagulation equations should be written only for these nodes. The form of these equations corresponds to (3), where the number of particles in the nodes of the linear mass scale are calculated by the approximating function (4). The final system of equations is:

\[ \frac{dN_i}{dt} = \frac{1}{2} \sum_{j=1}^{n'-1} a_{i-j,j} f_i f_j - f_i \sum_{j=1}^{n'} a_{ij} f_j, \quad i' = 1 + n'_{\text{max}}. \tag{5} \]

Thus, the new scale significantly reduces the number of equations in the system from \( n_{\text{max}} \) to \( n'_{\text{max}} \), and the final distribution now consists of \( n'_{\text{max}} \) sections instead of \( n_{\text{max}} \).
The solution of the obtained system of equations is carried out by the Runge-Kutta method of the 4th order, which is quite often used in solving similar corresponding systems [5].

5. Results

5.1. Calculation graphs
In fig. 1 shows graphs of several distributions of particles taken at different time moments. As you can see, the distribution function gradually begins to correspond to a lognormal distribution, a shift of the peak towards larger sizes occurs, and the width of the peak increases with time. Thus, the so-called “aging” of the ultradispersed system occurs.

![Graph 1](image1)

**Figure 1.** Distribution functions with a modified scale at different times at an initial concentration of $5.0 \times 10^{17}$ per m$^3$, the maximum number of varieties, $d_0 = 2.5$ nm.

![Graph 2](image2)

**Figure 2.** Distribution functions with a linear and a modified scale at different times at an initial concentration of $5.0 \times 10^{17}$ per m$^3$, the maximum number of varieties, $d_0 = 2.5$ nm.

![Graph 3](image3)

**Figure 3.** Graphs of the mean relative deviation of the distribution graph with a modified scale from the distribution with a linear scale in time at different initial concentrations.
5.2. **Comparison of two simulation schemes**

Figure 2 shows a comparison calculating the linear scale and the modified one results. As you can see, there is some deviation in results of the modified scale from linear scale results, and the size of this can be estimated from the graphs shown in Fig. 3. In addition to the graphs of deviations of the distribution function, in Fig. 4 also shows the graphs of changes in the average radius over time for a linear scale and a modified one. Comparison shows that the value of the average radius of particles at first increases in an increasing way, and then goes on to a gradual increase. It should be noted that the results for the modified scale on the initial segment only slightly differ from the linear scale.

Figure 5 shows graphs of changes in the total number of particles for linear and modified scales at an initial concentration of particles of $5 \times 10^{17}$ /m$^3$. As you can see, the differences in the graphs are insignificant.

The reason for such deviations can be supposed by the using the approximation of the table function, that is need more accuracy in the approximating parameters determining of the function. Because of the significant nonlinearity of the distribution function, the accuracy of approximating parameters determining of the function decreases, which decreases the accuracy of coagulation equations calculating.

Nevertheless, this scale reduces the number of calculated equations by several orders of magnitude, which undoubtedly makes the reduced scale more preferable than the classical linear one. This factor becomes especially important in the unregular case, when there is an uneven distribution of the thermohydraulic indicators of the working medium, such as temperature and flow rate, and forces the computational domain to be divided into several cells.
6. Conclusion

Thus, the paper considers the issue of numerical simulation of coagulation in an ultradispersed system. The disadvantages of the classical system of coagulation equations associated with a huge number of equations were analyzed. A method for optimizing the system by means of a reduced scale and approximation of the particle distribution function, which is written in the form of a table function, is proposed. Comparison of the results of calculating systems with linear and reduced scales, which showed a slight difference in distribution graphs, as well as a slight deviation of the parameters of the distribution function, presented in the form of a lognormal distribution. These results show the feasibility of reconciling the modified design model by significantly reducing the number of equations to be solved.

7. References

[1] Samuylov E V 2009 Kinetic equation for the size distribution function of condensation aerosol droplets taking into account the coagulation of droplets J. Izvestiya Ak Nauk Energetika 5 125-30
[2] Park S H, Lee K W, Otto E and Fissan H 1999 J. Aerosol Sci. 30 3–16
[3] Adzhiev S Z, Vedenyapin V V, Volkov Yu A, Melikhov I V 2017 Generalized Boltzmann-Type Equations for Aggregation in Gas, J. Comp. Math. and Math. Phys. 57 138–51
[4] Gelbard F, Tambor Y and Seinfeld J H 1980 Sectional representation for simulating aerosol dynamics, J. Colloid Int. Sci. 76 541–36
[5] Hounslow M J, Ryall R L and Marshall V R 1988 A discretized population balance for nucleation, growth, and aggregation A. I. Ch. E. Journal. 34:1821–32.