Numerical modelling of nucleation processes in gas phase

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In spite of the fact that significant body of literature is currently available (see for ex. Friedlander, 2000), developments of new theoretical models describing nucleation processes still attracting strong scientific and technological interests. Such interest is justified by the problem complexity and by the fact that it involves large number of process parameters, which are, in many situations, not clearly quantified. In addition, it is well known that the nucleation is non-equilibrium process, which is, due to complexity, is usually modeled by equilibrium thermodynamics and statistical physics with some approximations. As the result, most of current theories are rather case specific and are not able to provide accurate results for general cases. A kinetic approach is suggested to attack the problem. Following kinetic equation of coagulation, which takes into account all particles participating in the process is proposed to model the process:

$$\frac{\partial c_g}{\partial t} = I(g) + \frac{1}{2}\sum_{m=1}^{n} K(g-n,n)c_{g-n}c_n - c_g\sum_{m=1}^{\infty} K(g,n)c_n \,, \quad (1)$$

where c_g –concentration of particles containing g monomers, $I(g)$ – intensity of new particle generation in the system, $K(g,n)$ – collision frequency of particles containing g and n numbers of monomers.

It is easy to estimate that 1-micrometer particle contains $10^{10} – 10^{12}$ monomer molecules in vapor phase. It means, that to obtain accurate solution, one ought to solve the same number of Equation (1), which is obviously beyond any realistic time related computer capability. To decrease the amount of calculations, it was proposed to use logarithmic scale of particle size (Zagaynov and Lushnikov, 1988). This procedure allowed to reduce the number of equations for particle in the radius size range between 1 nm and 1 micrometer down to 30 equations. However, a drawback of this approach is related to the fact that at very early stages of nucleation processes, the results obtained for ultra fine particulates are not very accurate, which is a problem, as such particles are responsible for the entire process. To overcome these model deficiency, a new approach is proposed. In particular, for early stages of nucleation when all particles are small a linear scale could be used to account for all fine particulates. Then, when particles became large enough, the scale is changed to logarithmic to keep the number of model equations within reasonable limits. This approach allowed to advance along particle radius far enough by using very few kinetic equations and take into account non effective collisions between small particles. Moreover, this approximation makes possible to take into account chemical reactions as a sauce of new particle generation in the gas phase.

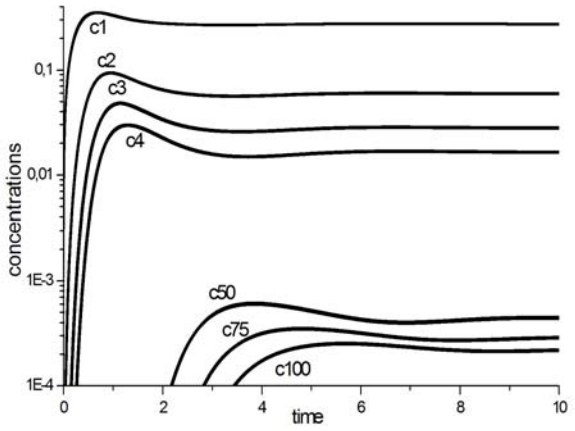

Figure 1. Results of estimation of nucleation rate for permanent sauce of monomers (*ci* is a concentration of particle containing *i* monomers for dimensionless time.

Some further model modifications were related to calculations of efficiency of collision between small particles; *i.e.* identification of a number of collisions which lead to creation of stable particle agglomerates out of total collisions' number. For this procedure power and exponential relationships were used. Exponential relation was found to be in the best agreement with experimental data. Finally, it ought to be noticed that the suggested approach enables taking into account collisions between dimers, and more complicated particles and to make nucleation calculations more accurate.

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