Comparison of Different Methods Used in Integral Codes to Model Coagulation of Aerosols

A. I. Beketov\textsuperscript{a,b}, A. A. Sorokin\textsuperscript{a,c}, V. M. Alipchenkov\textsuperscript{a,c}, and N. A. Mosunova\textsuperscript{a}

\textsuperscript{a} Institute for Safe Development of Nuclear Power Engineering, Russian Academy of Sciences, Bol'shaya Tul'skaya ul. 52, Moscow, 115191 Russia

\textsuperscript{b} Moscow Institute of Physics and Technology, Institutskii per. 9, Dolgoprudny, Moscow oblast, 141700 Russia

\textsuperscript{c} United Institute of High Temperatures, Russian Academy of Sciences, Izhorskaya ul. str. 2, Moscow, 125412 Russia

Abstract — The methods for calculating coagulation of particles in the carrying phase that are used in the integral codes SOCRAT, ASTEC, and MELCOR, as well as the Hounslow and Jacobson methods used to model aerosol processes in the chemical industry and in atmospheric investigations are compared on test problems and against experimental results in terms of their effectiveness and accuracy. It is shown that all methods are characterized by a significant error in modeling the distribution function for micrometer particles if calculations are performed using rather “coarse” spectra of particle sizes, namely, when the ratio of the volumes of particles from neighboring fractions is equal to or greater than two. With reference to the problems considered, the Hounslow method and the method applied in the aerosol module used in the ASTEC code are the most efficient ones for carrying out calculations.

Keywords: aerosols, integral codes, coagulation, modeling, distribution function, kinetic equation

DOI: 10.1134/S0040601513090024

Simulation of the formation and alteration of aerosols in the primary coolant circuit of a nuclear power station (NPS) reactor during a hypothetical beyond-design-basis accident involving release of fission products (FPs) from fuel into the coolant volume is a necessary condition for estimating the consequences from possible escape of radioactive particles to beyond the reactor pressure vessel boundaries and radioactive contamination of the environment \cite{1, 2}. Fuel rods are heated to high temperatures in the course of such accident, causing the coolant in the reactor core (in VVER reactors this is pressurized water) becoming overheated and transferring from liquid state into vapor. It should be noted that the major fraction of finely dispersed aerosol particles is generated in the primary coolant circuit as a result of nucleation of FP vapors and uranium oxides contained in the fuel matrix \cite{3}. Coagulation is one of the most important mechanisms governing further evolution of the generated particles. It is exactly the process that accounts for the major part of the computation time when an aerosol module is incorporated into an integral code.

In the general case, FP aerosol particles have a multicomponent composition due to nucleation and condensation of vapors generated from FPs of different compositions followed by coagulation of mixed particles. Attempts to take into account the multicomponent composition of aerosol particles result in that the computer code structure becomes significantly more complex and that the computation time increases exponentially \cite{3, 4}. Therefore, development of more efficient calculation methods can be regarded as an important factor helping to improve the existing codes.

STATEMENT OF THE PROBLEM

The kinetic equation for the particle distribution function, e.g., over the radius \( n(r, t) \), where \( n(r, t)dr \) is the number of particles in the interval \((r, r + dr)\), written in the approximation of a single-component composition, has the form

\[
\frac{\partial n(r, t)}{\partial t} = S(r, t) - \frac{\partial}{\partial r} \left[ G(r, t)n(r, t) \right] - R(r, t)n(r, t) + \frac{1}{2} \int_0^r K(s, r - s)n(s, t)n(r - s, t)ds - n(r, t) \int_0^r K(r, s)n(s, t)ds
\]

(1)

with the boundary and initial conditions \( n(0, t) = 0 \) and \( n(r, 0) = n^0(r) \) \cite{3}. The term \( S(r, t) \) corresponds to the action of all possible sources of particles with the radius \( r \), e.g., nucleation [in this case, \( S(r, t) = J(t)\delta(r - r_{cr}) \)], where \( \delta(r) \) is the delta function and \( J(t) \) is the nucleation rate of particles of the critical size \( r_{cr} \), emission (inflow) or particles, or detachment of particles from the surface. The term
\( G(r, t) = \frac{dr}{dt} \) is the particle radius variation rate due to condensation and evaporation of different FP vapors, steam (e.g., in the containment premises), or due to adsorption of FPs from the gas phase. The flow-off \( R(r, t) \) is the removal rate of particles of radius \( r \) (during their sedimentation on the surface or flow-over together with steam–gas medium). The coagulation rate \( K(r, s) \) between particles of radiiases \( r \) and \( s \) depends on a few mechanisms governing the interaction of particles. The first integral in (1) corresponds to generation of particles with radius \( r \) when two particles of a smaller radius collide with each other, and the second integral in (1) corresponds to their vanishing when they collide with any other particle.

The kinetic equation can be numerically solved using different methods, the choice of which depends primarily on the method for approximating the distribution of particles by size: methods of moments and fractions, stochastic models, etc. [3, 4]. The method of fractions is the most universal modeling method. Within the framework of this method, the spectrum of particle sizes (e.g., volume) is subdivided into separate fractions with fixed or varying boundaries, and it is supposed that particles can transfer from one fraction to another as a result of various processes. In this case, integral-differential equation (1) transforms into a system of nonlinear first-order differential equations for the concentration (or mass) of particles of each fraction [3, 4]. The main drawback of the fraction method is that the distribution function by size has a blur along the size axis due to numerical diffusion. To minimize this effect in modeling coagulation processes, the number of considered fractions is increased in order to decrease the difference in the volume of particles from neighboring fractions [3, 4, 9].

At present, 20 to 50 fractions are currently used in the majority of integral codes in modeling aerosol processes, which may turn to be insufficient for achieving the required accuracy. The processes through which aerosols are generated and transferred in elements of the reactor primary coolant circuit during severe accidents are characterized by a fairly wide range of aerosol particle sizes (approximately from 1 nm to 10 μm). Hence, a hundred or more fractions may need to be considered to keep the solution accuracy.

Similar methods are applied in the basic version of the aerosol module SOPHAEROS used in the ASTEC code and in the aerosol module PROFIT used in the SOCRAT code. However, a fewer number of fractions is considered in the PROFIT module, due to which the calculations yield less accurate results. In view of this fact, further consideration will be carried out only for the ASTEC code. We assume that all particles are characterized by the same and constant density irrespective of their composition. The aerosol kinetics equations are written for the concentration \( N_i \) of particles from the \( i \)th fraction with the volume \( V_i \). By default, we will use a simplified spectrum of sizes for which the volumes of particles belonging to neighboring fractions are interconnected by the relation \( \alpha^2 - \alpha - 1 \geq 0 \) (i.e., \( \alpha = v_i/v_{i+1} \geq 1.6 \)), and the number of considered fractions does not exceed 50 [5]. In this approximation, particles of the \( i \)th fraction can be generated during coagulation only as a result of collision between particles from the \((i-1)\)th fraction and particles of the \( j \)th fraction, with \( j \leq i - 1 \).

For the case when there is only a coagulation process and \( \alpha = v_i/v_{i+1} \geq 2 \), the system of equations for the change of concentrations \( N_i \) has the following form (the default version in the SOPHAEROS module used in the ASTEC code [5]):

\[
\frac{dN_i}{dt}_{\text{coag}} = \sum_{j=1}^{i-1} \left( 1 - \frac{\beta_{i-j}}{2} \right) K_{i-j} N_j - N_i \sum_{j=1}^{M} \beta_{i,j} K_{i,j} N_j, \tag{2}
\]

\[
\beta_{i,j} = \frac{v_j}{v_{i+1} - v_i} \quad \text{with} \quad j \leq i \quad \text{and} \quad \beta_{i,j} = 1 \quad \text{with} \quad j > i,
\]

where \( M \) is the total number of fractions, and \( K_{i,j} \) is the rate of coagulation between particles from the \( i \)th and \( j \)th fractions.

The MELCOR code uses a similar spectrum of sizes with the parameter \( \alpha \geq 2 \) for particles with constant density and with the calculation scheme from the aerosol module MAEROS [6]. This method assumes that the particle distribution function by size has a constant value for each fraction. The distribution coefficients for modeling the collision of particles are determined by double integrals for each pair of particles. In carrying out this determination, the condition for conservation of only the total volume of particles is taken into account.

The system of equations for concentrations \( N_i \) (the number of particles in the \( i \)th fraction) during coagulation that is used in the model [7] is represented in the following form:

\[
\frac{dN_k}{dt} = \sum_{i=1}^{k} \sum_{j=1}^{k-1} f_{i,j,k} V_j N_i N_j - N_k \sum_{j=1}^{M} (1 - f_{k,j,k}) K_{k,j} N_j,
\]

where the coefficients \( f_{i,j,k} \) correspond to the percentage of particles with the volume \( V_j = v_i + v_j \), distributed into the \( k \)th fraction during the collision of particles from the \( i \)th and \( j \)th fractions [7]:

\[
f_{i,j,k} = \begin{cases} 
\frac{(v_k + v_j) - v_k}{v_{k+1} - v_j} V_j, & v_k \leq V_j < v_{k+1}, \quad k < M; \\
1 - f_{i,j,k-1}, & v_{k-1} \leq V_j < v_k, \quad k > 1; \\
1, & V_j \geq v_k, \quad k = M; \\
0, & \text{in all other cases}.
\end{cases}
\]