NUMERICAL STUDY OF PROPAGATION AND PRECIPITATION OF RADIOACTIVE AEROSOLS IN THE CONTAINMENT IN THE EVENT OF AN ACCIDENT AT A NUCLEAR POWER STATION

A. I. Porshnev and V. P. Reshetin

UDC 621.039.58

A description of a physical NAUA model is presented. An algorithm for the numerical study of an aerosol equation by the method of trial functions has been specified. Test calculations have been conducted based on several model problems.

1. In the case of reactor active zone melting, and also in the case of interaction between the fuel melt and construction materials with a concrete building foundation, a significant release of vapor, gas and radioactive products to the containment atmosphere occur, among them, some in the form of aerosols. For a scientifically grounded choice of filtering systems, prediction of aerosol precipitation efficiency, and also for the assessment of radiation terrain contamination when a leak from the containment is present, of greatest importance is the investigation of the propagation of aerosols over condensing atmosphere under the conditions characteristic of catastrophic accidents at nuclear power stations (NPS).

At present, for analyzing aerosol behavior in gas fluxes, a number of fairly efficient mathematical models have been developed, which are successfully employed for computations in various applied problems (liquid fuel spraying, drying, Venturi tube-based filters, scribers, and the like). For analyzing aerosol propagation and precipitation after catastrophic accidents at nuclear power stations a point model has been used on a large scale, based on the assumption of a uniform composition of aerosols throughout the containment volume. This model is employed in the NAUA computer program, which is part of the STCP program complex. The NAUA point model enables one to describe with sufficient accuracy the process of aerosol precipitation in the presence of a developed small-scale convection contributing to the leveling of the aerosol composition throughout the amount of the containment. At the same time, the point model proves to be ineffective for describing aerosol behavior when a large-scale convection appears, which results in redistribution of the suspended particles over different containment zones.

2. In publications, several models have been proposed describing the process of aerosol propagation over a closed volume [1-4]. To reduce the time of computer calculations such models are based on a number of assumptions, the most important of which are to the following:

– particles are uniformly distributed over the whole controlled volume, excluding the boundary layer near the walls;

– the particles of the same size class are not distinguished in the composition;

– the properties of particles are functions of the particle size and density, which can vary depending on the composition;

- transport coefficients (and also the shape factor, boundary layers, etc.) do not depend on a particle size.

Such assumptions, which require, generally speaking, analysis and substantiation, hold, as a rule, at the containment under the conditions of active zone melting and release of a great amount of vapor, gas, and aerosols [1, 3]. Internal mixing among the molecules of the same size class occurs on account of the gravitational and Brownian (thermal) coagulation. Spatial uniformity in the reference volume is established due to convection in a condensing atmosphere. Thus, under the conditions of uniform mixing, the reactor building can be used as a reference volume. Inside the reference volume, the model describes the following physical processes:

Institute of Radiation Physicochemical Problems, Academy of Sciences of Belarus, Minsk. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 64, No. 3, pp. 363-370, March, 1993. Original article submitted March 13, 1992.

1062-0125/93/6403-0295$12.50

– disposal of aerosols (gravitational sedimentation, diffusion precipitation, thermophoresis);
– interaction of aerosol particles (Brownian coagulation, gravitational coagulation);
– vapor condensation on the particles;
– transfer (aerosol sources, leakage).

The model of aerosol disposal under mixing conditions is well known [4]. The rate of variation of the concentration n(r) of particles of radius r is defined by the relation

$$\frac{dn(r)}{dt} = -n(r)U(r)\frac{A}{V}.$$

Here, V is the volume occupied by the aerosol; A is the surface area on which its precipitation occurs; U(r) determines the aerosol precipitation rate: U(r) = FB(r), where F is the force acting on the particle, and B is its mobility. The mobility of a spherical particle is defined by the Stokes law:

$$B(r) = \frac{C(r)}{6\pi\eta r}, \tag{1}$$

where η stands for the surrounding gas viscosity; C(r) is the empirical correcting Cunningham factor, for which the following well-known expression [5] is applied:

$$C(r) = 1 + 1.246\,\mathrm{Kn} + 0.42\,\mathrm{Kn}\exp\left(-\frac{0.87}{\mathrm{Kn}}\right),$$

where the Knudsen number Kn = l/r, l is the mean free path of the surrounding gas molecules. (In numerical simulation, as the particle size it is convenient to use the radius, averaged with respect to the particle volume.)

In the process of gravitational precipitation, the steady-state velocity of particle motion can be defined by the equation

$$U_s(r) = \frac{2\rho_{ef}g}{9\eta} r^2 C(r), \tag{2}$$

where ρ_{ef} is the efficient density of a spherical particle. In the case of gravitational precipitation, the area of the surface A represents the sum of the projected areas of the top views of all containment platforms. For all other precipitation processes, the orientation of the platforms does not make any difference.

Since large particles (r $\geq$ 7 μm), under conditions when simulated mixing is absent, do not at all precipitate on the vertical walls, the inertia precipitation on the plane walls of the containment is extremely small. Thus, in the case under consideration, the main mechanism of aerosol precipitation on vertical walls is a diffusional one, which along with gravitational sedimentation also leads to aerosol precipitation on the horizontal and inclined walls and containment platforms.

Since the particles are delivered to a wall by convective diffusion and precipitate due to molecular diffusion in a thin wall layer, the rate of diffusional precipitation of the particles can be estimated in terms of the equation [4]

$$U_D \sim D(r)\nabla_{\mathbf{n}} n(R, r, t),$$

where **n** is the external normal to the wall surface; D(r) is the diffusion coefficient of the particle of size r. Since the law governing the change of the convective diffusion coefficient in approaching the wall is unknown, one has to strongly simplify the problem and consider that in the near-wall layer of thickness δ_d only molecular diffusion is active, whereas outside this layer, convective diffusion is so intense that aerosol concentration outside the layer is constant [4]. In this case, the rate of aerosol precipitation is expressed by the equation

$$U_d(r) = \tilde{D}(r)/\delta_d,$$

where $\tilde{D}$(r) is the molecular diffusion coefficient, related to particle mobility by the relationship $\tilde{D}$(r) = kTB(r).

In the NAUA program (mod 3-mod 4), δ_d is a constant quantity, the value of which in most cases is assumed equal to δ_d = 0.01 cm.

Collisions between particles determine the nonlinear dynamics of aerosol precipitation in the case of a large concentration of particles. A description of particle collisions requires larger computer time calculations when a detailed division with respect to particle sizes is required. Analyzing aerosol behavior in the containment atmosphere, one should consistently describe the collisions and condensation (evaporation) of vapor on the particles.

The frequency of particle collisions in Brownian motion is defined by the equation

$$K_b(r_i,\ r_k) = 4\pi kT(r_i + r_k)(B(r_i) + B(r_k)), \tag{3}$$

where r_i and r_k are the radii of colliding particles. The collision results in the fusion of particles and the formation of a new particle, the mass and size of which equals the sum of the masses and sizes of the colliding particles. Assuming that the newly formed particle is of spherical form, the laws of conservation for the process of coagulation can be presented in the form

$$m_j = m_i + m_k,\ V_j = V_i + V_k,\ r_j^3 = r_i^3 + r_k^3,$$

where the index j denotes the newly formed particle.

In the case of gravitational coagulation, the frequency of collisions is defined by the equation

$$K_g(r_i,\ r_k) = \pi\varepsilon(r_i + r_k)^2 |U_s(r_i) - U_s(r_k)|. \tag{4}$$

To predict the efficiency of collisions ϵ, the well-known Pruppacher equation can be used [6], determining the dependence of this parameter on the size of colliding particles,

$$\varepsilon = 0.5\left(\frac{r_i}{r_i + r_k}\right)^2,\ r_i < r_k,$$

which allows one to obtain an acceptable accuracy when describing gravitational coagulation during the whole process of accident development at a nuclear power station. The frequency of particle collisions as a result of gravitational and Brownian coagulation ($K(r_i, r_k)$) represents the sum of frequencies defined by Eqs. (3), and (4):

$$K(r_i,\ r_k) = K_g(r_i,\ r_k) + K_b(r_i,\ r_k).$$

In the process of aerosol disposal, vapor condensation on particles is important, and since condensation is a rapid molecular process, it leads to particle enlargement and precipitation. The change in the radius of a particle resulting from vapor condensation can be described as follows:

$$r\frac{dr}{dt} = \frac{S - \exp(2\sigma M_w/(\rho_w RT_r r_k))}{\frac{L\rho_w}{KT}\left(\frac{LM_w}{RT} - 1\right) + \frac{\rho_w RT}{M_w D p_s(T)}}, \tag{5}$$

where M_w is the molecular mass of water; $\rho_w(T_r)$ is the density of water; R is a universal gas constant; $p_s(T_\infty)$ is the saturation pressure of water vapors.

It is also assumed that the gas is perfect, and the values of σ, ρ_w, L, K, and p_s, which are functions of temperature, are computed in numerical simulation for every time interval in accordance with the data reported in certain thermophysical reference books (see, for example, [7]). Equation (5) is derived from the condition of thermal equilibrium in a standard atmosphere. Experimental investigations [3], conducted at the aerosol pressure, temperature and concentrations characteristic for containment conditions, showed that a description of the condensation process based on Eq. (5) is valid without the introduction of any corrections and refinements. It should be noted, however, that such a refinement should be taken into account at a sufficiently rapid increase in temperature and pressure in the containment. (Sharp changes in the concentration and size of particles lead to the necessity of reducing the Δt step of integration in the numerical study of aerosol behavior. The usage of a large step leads to the development of numerical instability, as a result of which nonphysical oscillation relationships can appear. Thus, the requirements on the accuracy of the description of the condensation process impose restrictions on the value of the integration step with respect to the time Δt. This, in a number of cases, can lead to large expenditures of computer time).

The least radius of a thermodynamically steady liquid drop

$$r_c = \frac{2\sigma M_w}{\rho_w RT \ln S}$$

defines the laser boundary when particles are divided into size classes.

In describing the process of aerosol propagation in a closed volume, transfer processes are simulated with the help of volumetric sources and the discharge of particles.

The source of the aerosol represents the specification of the following input data: particle size distribution; density of particles; composition of nuclides; arbitrary function of time defining the evolution of the aerosol source.

The volumetric particle discharge represents a similar specification of input data. Dimensional effects in the proposed method are not considered.

The most important experimental investigations are aimed at studying the processes leading to the formation of spherical particles. Owing to the condensation–evaporation processes, with time, the particles acquire practically a spherical form. The residual deviation from the perfect sphere, as a rule, is very small and is not taken into account in the proposed physical model. A thickening of coagulating aggregates comprising primarily compact particles also occurs. Such effects have been observed in experiments with nonhydroscopic particles; they are expressed more vividly on soluble or hydroscopic particles.

It is evident that the spherical particle formation effects are to be observed also under conditions characteristic of accidents in RBMK (high-powered channel reactors) and VVÉR (water-moderated–water-cooled power reactors). Thus, it is assumed that the particles are of a spherical form at all stages of an accident in nuclear power stations, and the shape factor employed is considered to be equal to unity in all variants of computations. The following three shape factors enter the above equations:

– the dynamic shape factor, entering Eq. (1);
– the shape factor in coagulation, which is included in Eq. (3) as a multiplier;
– the shape factor in condensation, which enters the exponent of Eq. (5).

Defining these factors as depending only on the particle shape, one should also introduce the density correction for porous particles into Eq. (2) for gravitational sedimentation, and into Eq. (4) for gravitational coagulation. It is more convenient to perform the correction using the efficient density ρ_{ef}, which can be determined experimentally. Thus, in experiment [3] for the UO_2 aerosol, the value $\rho_{ef}/\rho_s = 0.47$ has been established.

Thus, excluding the specially mentioned cases, the efficient particle density recommended for computations comprises 50% of the density of the particle material. In numerical simulation, the efficient density is an input parameter.

In the physical model described aerosol processes are considered additive. The problem concerning the validity of such an assumption for conditions of the containment condensing atmosphere has been considered in [8]. Combining the equations for separate physical processes described above, one can obtain the equation

$$\frac{\partial n(r,\ t)}{\partial t} = S(r,\ t) - [\alpha_d(r) + \alpha_s(r) + \alpha_T(r)]\, n(r,\ t) + \\ + \int_0^{r/2^{1/3}} K((r^3 - r'^3)^{1/3},\ r')\, n((r^3 - r'^3)^{1/3},\ t)\, n(r',\ t) \frac{r^2}{(r^3 - r'^3)^{2/3}}\, dr' - \\ - n(r,\ t) \int_0^{\infty} K(r,\ r')\, n(r',\ t)\, dr' + \frac{dr}{dt} \frac{\partial n(r,\ t)}{\partial r}, \tag{6}$$

where $\alpha_d = kTB(r)\, A_d/(\delta_d V)$; $\alpha_s = \frac{4}{3} \pi \rho r^3 g B(r)/V$; $\alpha_T = RT\bar{K}/[(p_d M_w + p_l (M_l M_w)^{1/2})\, V]$.

3. The integral equation (6) can be solved only numerically. For this, one should present the size particle distribution n(r) in the form of a set of monodisperse fractions (in the form of histograms). This procedure allows one to reduce integral (relative to the size of the particles) equation (6) to a system of related differential (with respect to time) equations for particles in every size class:

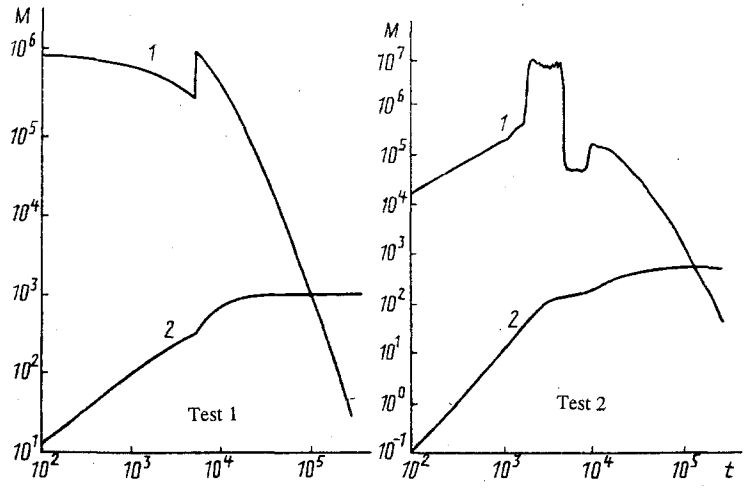

Fig. 1. Time dependence of the suspended mass of aerosol particles (curve 1) and total containment loss (curve 2). t, sec; M, g.

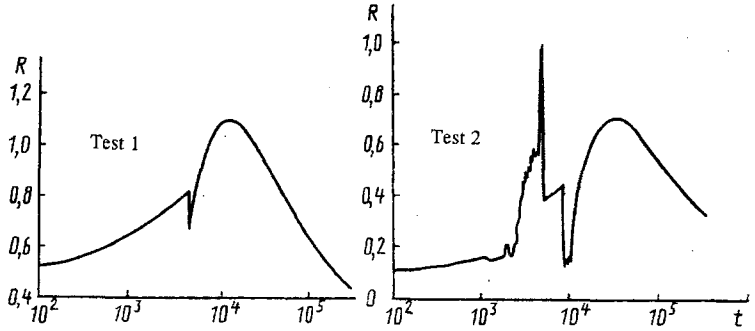

Fig. 2. Time dependence of the mean radius of aerosol particles. t, sec; R, μm.

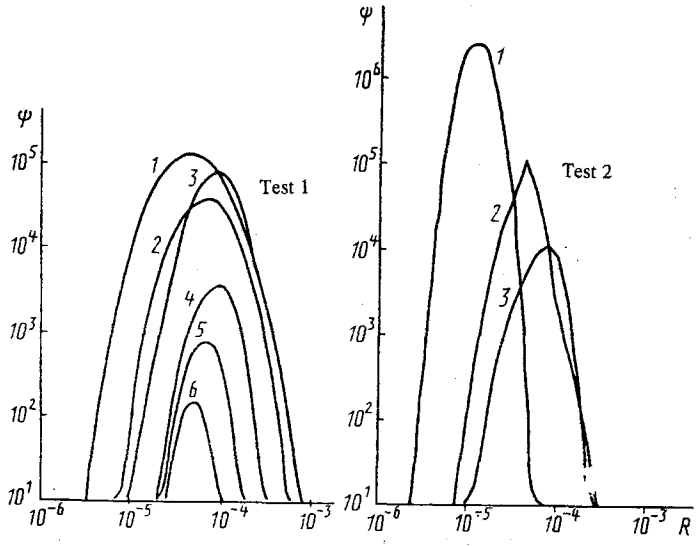

Fig. 3. Time dependence of the particle size distribution function: 1) t = 5 min; 2) 1 h; 3) 2; 4) 10; 5) 25; 6) 60 h; R, cm; ψ, cm^{-3}.

$$\frac{\partial n(r_k,\ t)}{\partial t} = S(r_k,\ t) - [\alpha_d(r_k) + \alpha_s(r_k) + \alpha_T(r_k)]\, n(r_k,\ t) -$$
$$- \sum_{i=1}^{N} [1 - 1/(2\delta_{ik})]\, K(r_i,\ r_k)\, n(r_i,\ t)\, n(r_k,\ t) +$$
$$+ \frac{1}{2} \sum_{i=1}^{N} \sum_{j=1}^{N} K(r_i r_j) B_{ij}^{k} n(r_i,\ t) n(r_j,\ t) + (1 - \delta_{1k}) \times \tag{7}$$
$$\times \frac{V_{k-1}(t)}{V_k - V_{k-1}}\, n(r_{k-1},\ t) - \frac{V_k(t)}{V_{k+1} - V_k}\, n(r_k,\ t),\ k = 1,\ \ldots,\ N.$$

Such a system of equations in numerical investigation is more stable than the initial integral equation (6). The difficulty arising in the process of coagulation, attributed to the formation of particles, the sizes of which are intermediate as compared to the nearby monodisperse fractions, can be overcome by interpolation of new particles between these fractions. It is natural that such interpolation preserves the particle number and mass. Another problem arises when describing the particles comprising both a solid fraction and also a soluble liquid one. On the one hand, the assumption about a uniform mixing throughout the whole particle size distribution does not hold because of the strong dependence of condensation and evaporation rates on the particle size, and on the other hand, introduction of a second size, describing a separate composition in addition to the size of the whole particle, requires much computer time, and, therefore, is hard to realize in practice. The particle composition, in this case, is averaged with respect to each size class, but varies from one class to another. The change in composition is computed taking into account the coagulation of various composition particles, water condensation on the particles, and also the source of new particles. Since such precipitation of the solid phase (containing radioactivity) justifies such simplification [3], the proposed model becomes applicable for analyzing the behavior of aerosols of different composition.

4. Based on the model problems [1], testing calculations have been performed, the results of which are presented in Figs. 1-3 (for a more detailed description, see [1]).

T e s t 1. Aerosol particles (with minimum and maximum radius 0.0025×10^{-4} and $20 \cdot 10^{-4}$, respectively) are instantaneously injected to a containment of volume 7.2×10^{10} cm^{-3}, compartment floor area 6.46×10^{7} cm^2, and total surface area 5.054×10^{8} cm^2 at the time instant t = 0 when the mass is 10 g, and t = 5000 when the mass is equal to $9 \cdot 10^{5}$ g. (The number of disperse fractions taken in the computations equals 50.) The compartment temperature is assumed equal to 130°C, the leakage rate is 1.16×10^{-7} sec^{-1}. Vapor condensation on the particles is absent.

T e s t 2. Various portions of aerosol particles are continuously injected to a containment of the same volume as in test 1 at different moments of time. The minimum particle radius is 0.0025×10^{-4} cm, the maximum $50 \cdot 10^{-4}$ cm, the number of fractions is 36. Vapor condensation on particles is predicted. The compartment temperature equals 130°C, the leakage rate is 1.15×10^{-7} sec^{-1}.

5. Under conditions of the developed small-scale convection equalizing the aerosol composition over the containment volume, to describe the aerosol propagation and precipitation processes the point model applied in the NAUA program proved to be good for studying the processes of disposal and interaction of suspended particles. The aerosol equation of the point model can be efficiently numerically investigated using the "kinetic" approximation, based on the size distribution of the suspended particles. Since, as a result of precipitation and coagulation, the distribution function varies with time, the recalculation of the particle fraction distribution should be performed at each iteration.

Note that in studying the behavior of suspended particles, the method of trial functions, which allows one to pass over from the integrodifferential aerosol equation to a system of several differential equations for adjusting parameters (for example, mean radius, dispersion and mean counter concentration of particles), can be very efficient.

NOTATION

n, concentration of a particle; r, radius of a particle; U, velocity of a particle; A, surface area; ρ_s, density of a solid particle; ρ_g, gas density; τ, time; g, free fall acceleration; $\bar{D}$, coefficient of molecular diffusion; δ_d, dimension of the boundary layer; k, Boltzmann constant; T, temperature of the surrounding gas; T_r, drop temperature; L, specific heat of vaporization; S, degree of vapor saturation; σ, surface water tension; K(T), thermal conductivity of water vapors; l,

mean free path of gas molecules; ϵ, efficiency of collisions; B, particle mobility; S(r, t), source of aerosols; η, gas viscosity; C(r), Cunningham factor; K, fraction of the vapors condensed on the wall; P_d, partial pressure of vapor; P_l, partial pressure of the air; M_l, molecular mass of the air.

LITERATURE CITED

1. H. Bunz, M. Koyro, and W. Schock, NAUA Mod 4 – A Code for Calculating Aerosol Behavior in LWR Core Melt Accidents Code Description and Users Manual, Karlsruhe (1983).
2. M. Furrer, P. Passalacqua, V. Prodi, and F. Belosi, Characterization of Simulated Accident Aerosols and Sparc Code Calculations for Sparta Pool Scrubbing Experiments, IAEA-SM (1988).
3. H. Bunz, M. Koyro, and W. Schock, NAUA Mod 3 Ein Computer Programm zur Beschreibung des Aerodolverhaltens in Kondensierender Atmosphure. Karlsruhe (1981).
4. N. A. Fuks, Mechanics of Aerosols [in Russian], Moscow (1955).
5. R. A. Milikan, Phys. Rev., **22**, 1-23 (1923).
6. H. R. Pruppacher and I. D. Klett, Microphysics of Clouds and Precipitation. D. Reidel Publishing Company (1978).
7. Tables of Physical Quantities, Reference Book. Edited by I. K. Kikoin [in Russian], Moscow (1976).
8. B. J. Mason, The Physics of Clouds, Clarendon Press (1971).