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QUALITATIVE ANALYSIS OF THE PROCESSES OF AEROSOL PROPAGATION IN A CONTAINMENT

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Characteristic times of the removal of aerosols are evaluated for different processes of deposition. Gas flows in a containment in the presence of suspended particles of various concentrations have been analyzed. A mathematical model is suggested for describing the propagation and deposition of aerosols under the conditions of large-scale convection.

Introduction. During severe accidents at electric power stations (EPS), large amounts of vapor, gas and radioactive substances, including aerosols, are released to the atmosphere of the protective envelope of the reactor (containment). For scientifically justified selection of filtering systems and evaluation of the radiative contamination of the area in the presence of leakage from the containment, the study of the process of the propagation of aerosols is of great importance. For selecting a model which would adequately describe the process of deposition of aerosol particles, it is advisable to carry out a qualitative analysis of the processes of propagation and sedimentation of aerosols.

It should be noted that the majority of mathematical models used for analyzing the behavior of aerosols in severe accidents at EPS are based on the assumption of homogeneous composition of aerosols over the containment volume. They describe the process of deposition of aerosols in the presence of developed small-scale convection with sufficient accuracy (see, for example, [1]). At the same time these models turn out to be inapplicable for describing the behavior of aerosols on initiation of large-scale convection, which leads to redistribution of suspended particles between different parts of the containment. When describing the distribution of aerosols in a gas flow, it is necessary to self-consistently describe the gasdynamics of a gas mixture and the motion of suspended particles.

We shall perform a qualitative analysis of the processes of propagation and deposition of aerosol particles, including cases where suspended particles are involved by the gas flow in the process of large-scale convection.

1. Gravitational Sedimentation. The time required to establish a constant rate of deposition of particles can be determined by using the equation of motion

$$\frac{4}{3}\pi r^3\rho_s U_s = 6\pi\nu\rho_g U_s, \tag{1}$$

from which it follows that the time of establishment of $U_s = \text{const}$ is equal in order of magnitude to

$$\tau_{st} \sim \frac{2\rho_s}{9\rho_g}\frac{r^2}{\nu}. \tag{2}$$

For the characteristic values of the quantities $\rho_s/\rho_g \sim 10^3$, $r \sim 10^4$ cm, $\nu \sim 10$ cm^2/sec the time of establishment amounts to $2\cdot10^{-5}$ sec. As will be seen from what follows, such times are rather small as compared with the lifetime of a particle in the containment atmosphere and, as a rule, are much smaller than the mean time of the path of the particles between collisions. This circumstance allows one to employ an approximation in which the velocity of the particles is regarded as constant during the entire residence time of a particle in the containment atmosphere. As a rule, the mean radius of a suspended particle does not exceed in order of magnitude several microns, which is

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much higher than the mean free path at the pressure of the vapor-gas mixture $P \geq 1$ atm. In a free-molecular regime, when the particle radius is much smaller than the mean free path length of the molecules, the time of establishment of $U_s = \text{const}$ is equal in order of magnitude is equal to $\tau_s \sim r/c_s$ where c_s is the speed of sound in a gas.

The velocity of established motion in a gravity force field is equal in order of magnitude to

$$U_s \sim \frac{2\rho_s}{9\rho_g}\frac{gr^2}{\nu}.$$

For the above example $U_s = 0.2 \cdot 10^{-2}$ cm/sec.

2. Diffusional Precipitation. The characteristic rate of diffusional precipitation is

$$U_d \sim \frac{\tilde{D}}{\delta_d}.$$

The particle diffusion coefficient $\tilde{D} = kTB$ can be estimated using the expression for the particle mobility $B = U/F \sim (\nu \rho_g r)^{-1}$:

$$U_d \sim \frac{kT}{\nu\rho_g r\delta_d}.$$

For $\delta_d \sim 10^{-2}$ cm, $\nu \sim 4 \cdot 10^{-1}$ cm^2/sec, $\rho_g \sim 10^{-3}$ g/cm^3 and $r \sim 1$ μm the rate of diffusional precipitation is equal in order of magnitude to $U_d \sim 5 \cdot 10^{-4}$ cm/sec.

It should be noted that the rate of diffusional precipitation of aerosols is inversely proportional to the particle radius ($U_d \sim 1/r$). Since for gravitational sedimentation $U_s \sim r^2$, then, starting from a certain size $r > r^*$ (where r^* corresponds to the condition $U_s(r^*) = U_d(r^*)$), gravitational sedimentation is the basic process of removal of aerosols. This statement is valid only in the case where the height and diameter of the containment base coincide in order of magnitude. Otherwise r^* should be determined from the condition $U_s(r^*)A = U_d(r^*)A_t$, where A is the area of the horizontal projections of all the containment areas; A_t is the total area of the internal surface of the containment. In order of magnitude the particle size r^* is determined by the following expression:

$$r^* \sim \left(\frac{kT}{\rho_s\delta_d g}\right)^{1/3}.$$

In the containment, under conditions characteristic of severe accidents at an EPS, $r^* \sim 0.3$ μm, and thus, to adequately describe the process of removal of particles of size $r \sim 0.1$-1 μm, it is necessary to take into account both gravitational and diffusional (Brownian) sedimentation. Fine particles of size $r \ll r^*$ precipitate on the walls due to diffusion, whereas large particles ($r \gg r^*$) settle on horizontal and inclined surfaces due to gravitational sedimentation.

We shall evaluate the time in which an aerosol is removed from the containment due to its deposition on the walls. For a containment with the characteristic dimension $L \sim 10$ m and particles with a mean radius of about 1 μm the time of removal τ_{dep} is

$$\tau_{\text{dep}} \sim L/(U_s + U_d) \sim 10^5\,\text{sec} \sim 25\ \text{h}.$$

3. Coagulation of the Particles. Collisions of the particles that lead to coagulation of the latter considerably alter the function of the size distribution of the particles, which influences the deposition of aerosols. Using the expressions

$$K_b(r_i, r_k) = 4\pi kT(r_i + r_k)(B(r_i) + B(r_k)),$$
$$K_g(r_i, r_k) = \pi\varepsilon(r_i + r_k)^2 |U_s(r_i) - U_s(r_k)|$$

for collision frequencies in Brownian K_b and gravitational K_g coagulation, it is possible to obtain the following estimates:

$$K_b \sim \frac{kT}{\nu\rho_g}, \quad K_g \sim \frac{2}{9}\frac{gr^2}{\nu}\frac{\rho_s}{\rho_g}\frac{\Delta r}{r}r^2,$$

where Δr is the variance of the radius of the particles. Assuming that the variance Δr coincides in order of magnitude with the mean particle radius ($\Delta r \sim r$) and assuming that $r \sim 1\ \mu m$, we obtain the following estimate for the frequencies of collision:

$$\nu \sim 10^{-1}\ \mathrm{cm}^2/\mathrm{sec};\ \rho_s/\rho_g \sim 10^3;\ T \sim 4\cdot 10^2\ \mathrm{K};$$
$$K_b \sim 5\cdot 10^{-10}\ \mathrm{cm}^3/\mathrm{sec};\ K_g \sim 10^{-10}\ \mathrm{cm}^3/\mathrm{sec}.$$

The above estimates show that at a large enough number density n for particles with a mean radius of about 1 μm, it is necessary to take into account collisions that occur as a result of both Brownian and gravity-induced ordered motion.

However, it is of interest to note that because of the sharp dependence of the collision frequency K_g on the particle size ($K_g \sim r^4$; $\Delta r \sim r$), gravitational coagulation becomes predominant already for particles with a radius $\geq 3\ \mu m$. In contrast to K_g, the Brownian coagulation frequency K_b is independent of the particle radius.

Comparing the mean time between collisions of particles ($\sim (Kn)^{-1}$) and the time of removal of particles from the containment τ_{dep}, it is possible to estimate the concentration of particles n^* starting from which the collisions between particles substantially influence the function of the size distribution of the particles

$$n \geqslant n^* = \frac{1}{k\tau_{dep}} \sim \frac{1}{5\cdot 10^{-10}\cdot 10^5} = 2\cdot 10^4\ (\mathrm{cm}^{-3}).$$

Thus, already at a comparatively small number density of the particles $n \sim 10^4$ particles/cm^3, Brownian coagulation leads to a considerable evolution of the function $n(r)$. At a concentration ofparticles $n \gg n^*$, the mean time between their collisions turns out to be much smaller than the time of deposition of particles on the containment walls.

4. Thermophoresis. In the presence of a temperature gradient in the containment atmosphere, the aerosol particles experience the action of a force directed against the temperature gradient. When the size of the particles greatly exceeds the free path length of the surrounding gas molecules and, consequently, the continuous-medium model can be employed, the gas velocity is equal in the order of magnitude to

$$U_{TF} \sim \nu\left(\frac{1}{T}\frac{\partial T}{\partial x}\right),$$

where the quantity $L_T \sim (1/T\cdot\partial T/\partial x)^{-1}$ defines the characteristic dimension of temperature change. Substituting Eq. (2) into the equation for the Stokes force acting on a particle in a gas stream, we obtain the following estimate for the thermophoresis force:

$$F_{TF} \sim 6\pi\nu^2\rho r\left(\frac{1}{T}\frac{\partial T}{\partial x}\right).$$

For the characteristic values of the quantities

$$\nu \sim 0{,}1\ \mathrm{cm}^2/\mathrm{sec}\ \text{ and }\ L_T \sim \left(\frac{1}{T}\frac{\partial T}{\partial x}\right) \sim 10\ \mathrm{m}$$

the rate of deposition of aerosols under the action of thermophoresis forces amounts to $U_{TF} \sim 10^{-4}$ cm/sec. This, as is seen from the above estimates, is much smaller than the rate of gravitational deposition for particles with radius $r > 0.1\ \mu m$ and the rate of diffusional deposition for particles with radius $r < 5\ \mu m$. Thus, in the most important region of the parameters in practice $r \sim 0.1$-$50\ \mu m$ the deposition of particles due to the phenomenon of thermophoresis is negligibly small.

5. Qualitative Analysis of Particle-Laden Gas Flows. In the case where large-scale motions of gas arise in the containment under the action of natural convection, it is necessary to self-consistently describe the gasdynamics of the vapor-gas mixture and the motion of suspended particles [2-5]. The relative volume or mass of the suspended particles can be conveniently characterized by the volumetric fraction and the volumetric density, i.e., by the density per unit volume of the mixture. The ratio of the volumetric density of particles to the volumetric density of gas, which represents the relative concentration of particles, is equal to the ratio of the mass of the particles to the mass of the gas phase. For a containment atmosphere containing suspended particles with mean radius $r_m \sim 1$ μm, a relative concentration of the particles equal to unity corresponds to the number density of the aerosol

$$n^* = \rho_g/(4/3\pi r^3\rho_s) \sim 3\cdot 10^8 \text{ (cm}^{-3}\text{)}.$$

In this case the volumetric fraction of the particles amounts to about 10^{-3}, whereas the volumetric fraction of the gas phase is equal to about 0.999. It is customary to assume that under these conditions the volumetric density of air is equal to its true density.

It should be borne in mind that for calculating the volumetric density it is necessary to carry out averaging over the volume. A volume involving about 10^4 molecules ensures a variation in density of less than 1%. For a gas under normal conditions, this volume is equal to about 0.1 μm^3 [3]. At a relative concentration of the suspended particles equal to unity, a side of a cube containing 10^4 particles is determined by the condition

$$L/(2r) \sim \left(\frac{\rho_s}{\rho_g} 10^4\right)^{1/3} \sim 10^2.$$

Thus, the dimension of the region over which the averaging should be performed when introducing the volumetric density of the particles amounts to $L \sim 10^2$ μm at a mean radius of the suspended particles equal to 1 μm. When analyzing the behavior of an aerosol in the containment, this dimension (L) is much smaller than the characteristic dimensions of the flow and it can be considered as a point (which, as a rule, cannot be done when analyzing Venturi-pipe-based filters).

We also give an estimate of the number density of an aerosol corresponding to a dense set of particles. In a gas flow with a nondense set of particles their motion is determined by aerodynamic forces. In a gas flow with a dense set the motion of the particles is governed by their collisions. The difference in the description of flows with dense and nondense sets of particles is qualitatively established with the aid of the parameter $\psi = \tau_a/\tau_c$, where τ_a is the time of aerodynamic relaxation; τ_c is the time between collisions of particles. The frequency of collisions between particles depends on the concentration and is equal in order of magnitude to

$$\tau_c \sim \frac{1}{K_g + K_b} \sim \begin{cases} \dfrac{\nu\rho_g}{kTn} \text{ when } r \geqslant 1 \ \mu\text{m (gravitational coagulation)} \\[2ex] \dfrac{9\rho_g}{2\rho_s}\,\dfrac{\nu}{gnr^4}\,\dfrac{r}{\Delta r} \text{ when } r \leqslant 1 \ \mu\text{m (Brownian coagulation)} \end{cases}$$

The aerodynamic-relaxation time, which is defined as the time required for a resting particle to attain a velocity of the order of 0.5-0.7 of the flow velocity, can be estimated similarly to the time of establishment (see Eq. (1)). The equation of motion of a spherical particle exposed to the action of the force of aerodynamic resistance has the form

$$\frac{4}{3}\pi r^2 U = c\frac{\pi r^2}{2}\rho U^2,$$

where $c = 24/\mathrm{Re} + 4/(\mathrm{Re})^{1/2} + 0.4$; $\mathrm{Re} = rW/\nu$ is the Reynolds number.

Since the carrying-phase velocity $U \sim 10$-30 cm/sec, for particles with a characteristic dimension of about 1 μm the Reynolds number Re << 1, and the formula for the aerodynamic-drag force goes over into the well-known expression for the resistance force in the Stokes approximation. In this case the aerodynamic-relaxation time coincides with the time of constant-velocity establishment in gravitational sedimentation

$$\tau_a = \frac{2}{9}\,\frac{\rho_s}{\rho_g}\,\frac{r^2}{\nu}.$$

Using the above-given expressions for the aerodynamic-relaxation time and for the time between collisions, we obtain for the parameter ψ the following estimate in order of magnitude:

$$\psi \sim \begin{cases} \left(\frac{\rho_s}{\rho_g}\right)^2 \frac{g r^6 n}{\nu^2} \frac{r}{\Delta r} & \text{when } r \geqslant 1 \quad \mu\text{m}, \\ \frac{\rho_s}{\rho_g} \frac{g r^2}{\nu} \frac{kTn}{\nu \rho_g} & \text{when } r \leqslant 1 \quad \mu\text{m}. \end{cases}$$

Thus, if $\psi < 1$, there is ample time for a particle to acquire a velocity comparable with the carrying-phase velocity, and its motion is governed by aerodynamic forces.

This condition characterizes motion with a nondense set of particles. On the other hand, when $\psi > 1$, the particle does not succeed in being involved in the flow in the time between collisions, and its motion is governed by collisions with other particles. Such motion is typical for a gas with a dense set of particles.

For a containment atmosphere with suspended particles the condition $\psi = 1$ is attained when the density of the particles is equal to

$$n_\psi = \left(\frac{\rho_g}{\rho_s}\right)^2 \frac{\nu^2}{g r^6} \frac{r}{\Delta r} \quad (r \sim 1 \quad \mu\text{m}).$$

For the characteristic values $\nu \sim 10^{-1}$ cm^2/sec, $\rho_g/\rho_s \sim 10^{-3}$, $g \sim 10^3$ cm/sec^2, $r \sim 10^{-4}$ cm, $\Delta r \sim r$, the density of the particles amounts to $n \sim 10^{-14}$ cm^{-3}.

Analysis of the evolution of a severe accident at an EPS shows that in all the scenarios of the accident the number density of aerosols does not exceed values of about 10^{14} and, consequently, the set of particles is not dense.

In a nondense set of particles the information is transferred from particle to particle only along the trajectory of motion (in a dense set it is transferred by pressure waves, i.e., due to the interaction of molecules). This specific feature leads to a parabolic character of the equations that describe a nondense set of particles.

An important parameter that characterizes the motion of suspended particles in a gas flow is the Stokes number, defined as the ratio of the time of aerodynamic relaxation τ_a to the characteristic time of flow τ_s: $\mathrm{Sk} = \tau_a/\tau_s$.

As already noted, the characteristic velocity of a gas mixture in a containment does not exceed values of about 10-30 cm/sec. In this case we may consider that the velocities of the particles and of the carrying phase coincide, and the flow can be regarded as the flow of a single-phase medium with sources.

The above analysis has demonstrated that suspended particles in the containment atmosphere form a nondense set, whereas their relative concentration does not exceed a value of the order of unity in a number of practically important cases. In this case, as already noted, the presence of particles does not change the gas velocity and temperature fields, and the vapor-gas mixture dynamics can be described in the approximation of a one-sided effect.

To describe the propagation and deposition of particles in a gas flow, an aerosol equation can be used extended to the case of nonuniform distribution of particles. Introducing into consideration the concentration of particles n(r, R, t) of size r at a fixed point of space R at time t, it is possible to obtain the following equation for the function n(r, R, t):

$$\frac{\partial n(r, R, t)}{\partial t} = S(r, R, t) - \operatorname{div}[(U + U_s)n] + \nabla(D\nabla n) +$$
$$+ \int_0^{r/(2)^{1/3}} K((r^3 - r'^3)^{1/3}, r')\, n((r^3 - r'^3)^{1/3}, R, t)\, n(r', t) \frac{r^2}{(r^3 - r'^3)^{2/3}}\, dr - \tag{3}$$
$$- \int_0^{\infty} K(r, r')\, n(r', R, t)\, dr' + \frac{dr}{dt} \frac{\partial n(r, R, t)}{dr},$$

where U_s is the velocity of particle motion under the force of gravity; U is the gas velocity. Under homogeneous thermodynamic conditions in the containment, in the absence of gas motion and with uniform distribution of aerosol, Eq. (3) can be integrated over the volume and can be reduced to the point model equation.

The simultaneous solution of the generalized aerosol equation and the gasdynamic equation for a vapor-gas mixture allows a self-consistent description of the motion of particles suspended in a gas flow. Boundary conditions for diverse physical situations are given in the well-known review [6].

When carrying out numerical investigation of the propagation and sedimentation of aerosols, we can solve Eq. (3) by the method of trial functions. This method allows one to reduce integrodifferential equation (3) to a system of three interrelated parabolic equations for the mean radius and the variance and mean concentration of the particles. Such an approach considerably shortens the time of calculations both when the point model is used and when the equations of gasdynamics are solved simultaneously with the generalized aerosol equation.

Conclusion. The use of the approximation of the point model [1] turns out to be inapplicable for describing the motion of aerosols under large-scale convection conditions, which lead to a noticeable redistribution of the particles between different parts of the containment. In the case of severe accidents at an EPS the number density of the aerosol corresponds to conditions under which the set of suspended particles is not dense and the Stokes number, which an important parameter characterizing the flow of a gas with particles, is, as a rule, much smaller than unity. For modeling such flows the literature suggests separate-trajectory methods within the scope of which the motion of the gas is described in Euler coordinates by a system of gasdynamic equations with source terms, and the motion of the particles is described in Lagrange coordinates, where the time of particle escape from the source is used as one coordinate.

The mathematical model suggested in the present work is based on the one-sided-action approximation. This allows one to self-consistently describe the motion of a gas and of particles suspended in it in the case of a small relative concentration of them. To describe the processes of propagation and deposition of aerosols the "generalized" aerosol equation is used. Just as in the case of the point model, it is suggested to use the method of trial functions to investigate the behavior of aerosols in a gas flow.

NOTATION

n, concentration of a particle; r, radius of a particle; U, particle velocity; A, surface area; ρ_s, density of a solid particle; ρ_g, density of a gas; τ, time; g, free fall acceleration; $\tilde{D}$, coefficient of molecular diffusion; δ_d, boundary layer dimension; k, Boltzmann constant; T, surrounding-gas temperature; B, particle mobility; ν, kinematic viscosity; S(r, t), source of aerosols; ψ, parameter characterizing the density of a set of particles; Sk, Stokes number.

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