CONF-840701--6

RAFT: A Computer Model for Formation and Transport of Fission Product Aerosols in LWR Primary System

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CONF-840701--6

DE84 006416

Summary

A computer model RAFT (Reactor Aerosol Formation and Transport) has been developed to predict the size distribution and composition of the particles (aerosols) formed from condensation of the fission-product and control rod material vapors released in LWR accidents [1,2]. The condensation calculations in RAFT are driven by the equilibrium gas phase chemistry of Cs-I-Te-Ag-In-Cd-H-O-Ar system. The formation of the particles is considered to result as the gas becomes supersaturated with Ag, In, Cd, CsI, CsOH, Te_2 or TeO_2 vapor leading to the initiation of homogeneous nucleation. The rate of change in particle size spectrum as a result of homogeneous nucleation, heterogeneous nucleation, agglomeration, convection and deposition is described by the transient population balance equation [3,4].

$$\frac{\partial n}{\partial t} + \frac{1}{A}\frac{\partial}{\partial x}(Aun) + \frac{\partial}{\partial r}(\dot{r}n) = \sum_{j=1}^{\ell} J_j\, \delta(r-r_j^*) + \left(\frac{\delta n}{\partial t}\right)_{coll} - v_d A_s n$$

In the foregoing equation n is the distribution function, t, x and r are the time, distance and particle size coordinates, A and A_s the cross-sectional and specific surface areas, u the particle velocity, $\dot{r}$ the particle growth rate, ℓ the number of condensible species (Ag, In, Cd, CsI, CsOH, Te_2, TeO_2), J the homogeneous nucleation rate, r^* the critical size of the embryo, and v_d the deposition velocity; the collision term represents the following coagulation integral.

$$\left(\frac{\delta n}{\delta t}\right)_{coll} = \int_0^{r/2^{1/2}} K(r_1,r_2)n(r_1,x,t)n(r_2,x,t)\frac{r^2}{r_2^2}\, dr_1$$

$$- n(r,x,t)\int_0^{\infty} K(r,r_1)\, n(r_1,x,t)\, dr_1$$

where

$$r_2 = (r^3 - r_1^3)^{1/3}$$

In RAFT, the rate of change of molar concentration of elements (Cs, I, Te, Ag, In, Cd, H, O, Ar) due to convection, homogeneous nucleation, vapor condensation on particles, and vapor condensation on reactor structure is given by the following transient conservation equation:

$$\frac{\partial c_i}{\partial t} + \frac{1}{A}\frac{\partial}{\partial x}(Au_g c_i) = - \sum_j a_{ij}(\dot{m}_{pj} + \dot{m}_{wj})/M_j$$

where the subscript i refers to an element and the subscript j to a species (Cs, CsI, CsOH, I, I_2, HI, Te, Te_2, TeO, TeO_2, Ag, In, Cd, H, H_2, OH, H_2O, O, O_2, Ar), u_g is the gas velocity, a_{ij} is the stoichiometric coefficient (number of atoms of element i per molecule of species j), M is the molecular weight, $\dot{m}_{pj}$ is the condensation rate of species j due to homogeneous and heterogeneous nucleation,

$$\dot{m}_{pj} = \int_0^\infty 4\pi\rho_\ell r^2 \dot{r}_j n \, dr + \frac{4}{3}\pi\rho_\ell (r_j^*)^3 J_j$$

and $\dot{m}_{wj}$ is the condensation rate of species j on the structural surfaces,

$$\dot{m}_{wj} = A_s \frac{h_{mj}}{R_j T}(P_j - P_{sj})$$

The overall composition of the particles is determined in RAFT by solving for the mass fraction (Y_j) of the condensed species.

$$\frac{\partial Y_j}{\partial t} + \frac{1}{A}\frac{\partial}{\partial x}(A\langle u\rangle Y_j \rho_a) = \int_0^\infty 4\pi\rho_\ell r^2 \dot{r}_j n dr + \frac{4\pi}{3}\rho_\ell (r_j^*)^3 J_j - Y_j \rho_a A_s v_d$$

where <u> is the mass-averaged velocity of the particles and ρ_a is the aerosol density.

Finally, given the molar concentration of the nine elements and the local pressure and temperature, the distribution of the elements among the twenty species is determined by minimizing the Gibbs free energy of the gas phase system.

RAFT is presently being applied to many ongoing experimental and analytical programs, e.g., tests being conducted on Marviken reactor, source-term experiments being planned at Argonne National Laboratory, and "V" accident comparison study being conducted at EPRI. Significant results obtained in these simulations are summarized below.

1. The existing aerosol computer codes are based on the hypothesis that homogeneous nucleation initially produces numerous Angstrom-sized particles so much so that the evolution of the final particle size spectrum is governed by the rate at which the particles can coagulate and is independent of the assumed initial number density provided it is large enough. On the basis of this hypothesis, the details of the homogeneous nucleation process have been ignored in the existing computer codes. On the other hand, RAFT, which includes homogeneous nucleation mechanism in parallel with heterogeneous nucleation, agglomeration and deposition mechanisms, gives contrary results. It shows that under most accident conditions, the particle size spectrum is determined by the competition between the homogeneous and heterogeneous nucleation mechanisms and that the final number density of particles is too small for agglomeration to play any significant role in the particle formation process. Homogeneous nucleation mechanism augments the number density by forming new particles whereas heterogeneous nucleation restricts the number density by condensing the vapor on the surface of the existing particles. For typical conditions of nuclear accidents, although the particle number density is determined by the competition between the two mechanisms, the actual mass of vapor condensed by the homogeneous nucleation process is negligibly small.

2. Parametric calculations performed using RAFT have shown that important variables influencing the formation of particles are gas pressure, concentration of fission product vapors and gas cooling rate. The particle number density is found to be inversely proportional to the gas pressure and fission product concentration and directly proportional to the gas cooling rate. Conversely, the number-averaged size of particles is directly

proportional to the gas pressure and fission product concentration and inversely proportional to the gas cooling rate. In majority of LWR accidents wherein pressure is higher than 5 atm, mole fraction of condensible species is greater than 10 ppm and the gas cooling rate is slower than 1000 K/s, the calculated number density is less than $10^7/cm^3$.

3. In the presence of sufficient quantities of control rod material vapors, Ag and In undergo self-nucleation and provide sites for condensation of CsI and CsOH. In the absence of Ag and In vapors, CsI is the species that undergoes homogeneous nucleation with CsOH condensing on the nucleated CsI particles; but at high loading, CsOH may homogeneously nucleate first and provide site for condensation of CsI.

4. The fission product vapors are removed either through condensation on particles or via condensation on the structural surfaces. The calculated breakdown of vapor condensation is sensitive to the given surface temperature. For accidents in which because of large thermal inertia of upper plenum structure, the surface temperature remains below the dew point temperature, a substantial fraction of fission product vapor is calculated to be removed via condensation on the upper-plenum structure.

5. Because of the strong coupling between the homogeneous and heterogeneous nucleation processes, the calculated particle size spectrum is somewhat insensitive to whether classical or modified nucleation theory is used for calculating the homogeneous nucleation rate.

References

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