

Implementation of the Phase Change Model between Aerosol and Gaseous Fission Products in the GAMMA+ Code

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1. Introduction

One of the unique features of a Very High Temperature Gas Cooled Reactor (VHTR) is Vented Low Pressure Containment (VLPC) containing two separate vent paths where both have two gravity-operated relief valves in a series. Because VLPC strategy allows the release of a relatively small amount of radioactive fission products (FP) into the environment during the blowdown phase, behavior analyses of the fission products circulating in the primary coolant loop and in the containment are major consideration factors for safety evaluation.

For thermal-fluid analysis of a Very High Temperature Gas Cooled Reactor (VHTR), the GAMMA(GAS Multicomponent Mixture Analysis)+ code^[1] is under development. The multi-component analysis modules of aerosol fission products has been developed based on the MAEROS^[2,3] model and the aerosol transport model of the CONTAIN code^[4]. Furthermore, Yoo et al.^[5] developed and incorporated gaseous fission product transport phenomena into the GAMMA+ code.

In this study, the phase change of fission products between condensed and vapor forms are modeled by thermochemical equilibrium, and the thermochemical and physical data of 288 species and 26 elements are implemented as a form of look-up table in the FP module of the GAMMA+ code.

2. Implementation and Verification

2.1 Condensation/Evaporation Model by Thermochemical Equilibrium

For the chemical kinetics under the conditions of low temperatures that could be found inside the containments even in a severe accident, chemical reactions are practically modeled in a very simple way rather than in a mechanistic fashion. In this simple chemistry model, no chemical reactions occur below a certain temperature threshold and only simple phase changes occur. Above this threshold, the standard thermochemical equilibrium treatment is used. In reality, the kinetics gradually become slower as temperature is decreased, the dependence on temperature being of an Arrhenius form. Below the threshold temperature, this simple chemistry model allows change of phase for all chemical forms in the database that exist as both the vapor- and condensed-phase species. The starting point for this chemistry model is a statement of conservation

of the number of moles of a chemical species, including condensed and vapor forms. When chemical interactions are not allowed and only change of phase is considered, the following equation must be satisfied for all species:

$$C_T(i) = C_G(i) + C_C(i) \quad (1)$$

where $C_T(i)$ = total concentration of i chemical species in both the vapor and condensed phases [$\text{kg}\cdot\text{mol}/\text{m}^3$], $C_G(i)$ = concentration of i chemical species in the vapor phase [$\text{kg}\cdot\text{mol}/\text{m}^3$], and $C_C(i)$ = concentration of i chemical species in the condensed phase [$\text{kg}\cdot\text{mol}/\text{m}^3$].

Here the volume basis for calculating concentration is taken to be the entire volume that contains both vapor and condensed phases. The equilibrium vapor-phase concentration is calculated from Raoult's and the ideal gas laws as follows:

$$C_G(i) = \frac{p(i)}{RT} = \frac{p'(i)x(i)}{RT} \quad (2)$$

where $p(i)$ = partial pressure of vapor species i [Pa], R = gas constant [$\text{kg}\cdot\text{m}^2/\text{s}^2\cdot\text{kg}\cdot\text{mol}$], T = absolute temperature [K], $p'(i)$ = equilibrium vapor pressure of pure species i [Pa], and $x(i)$ = mole fraction of species i in the condensed phase in which it is soluble [dimensionless].

For a change of phase, equilibrium vapor pressures are determined according to the following relationship

$$p'(A) = \exp(-\Delta G/RT) \quad (3)$$

where, ΔG = the change in Gibbs free energy [$\text{kg}\cdot\text{m}^2/\text{s}^2\cdot\text{kg}\cdot\text{mol}\cdot\text{K}$].

The condensed-phase mole fraction is defined by the following equation:

$$x(i) = \frac{C_C(i)}{\sum_{j \in C_i} C_C(j)} \quad (4)$$

where C_i = set of condensed-phase species that are soluble in the same phase as species i [$\text{kg}\cdot\text{mol}/\text{m}^3$].

Substituting equations (2) and (4) into (1) yields

$$C_T(i) - C_C(i) - \frac{p'(i)}{RT} \frac{C_C(i)}{\sum_{j \in C_i} C_C(j)} = R_i = 0 \quad (5)$$

where R_i = residual equation for chemical species i , which is equal to zero when chemical equilibrium is attained [$\text{kg}\cdot\text{mol}/\text{m}^3$].

Since equation (5) is nonlinear, the Newton-Raphson procedure is applied to obtain a solution. The Jacobian matrix is obtained by taking the derivatives of the residual equations with respect to each of the unknown concentrations.

$$\frac{\partial R_i}{\partial C_c(i)} = -1 - \frac{p'(i)}{RT} \left[\frac{1}{\sum_{j \in C_i} C_c(j)} - \frac{C_c(i)}{\left(\sum_{j \in C_i} C_c(j)\right)^2} \right] \quad (6)$$

$$\frac{\partial R_i}{\partial C_c(i)} = \frac{p'(i)}{RT} \left[\frac{C_c(i)}{\left(\sum_{k \in C_i} C_c(k)\right)^2} \right] \quad \text{for } i \neq j \quad (7)$$

The Jacobian matrix is defined simply as

$$J_{ij} = \frac{\partial R_i}{\partial C_c(j)} \quad (8)$$

The Newton-Raphson iteration is implemented in the standard fashion:

$$R_i + \sum_j J_{ij} \delta[C_c(j)] = 0 \quad (9)$$

where $\delta[C_c(j)]$ = correction to the concentration of condensed-phase species j [kg-mol/m³].

The system of equations is considered to have converged when the largest correction in concentration, on a relative basis, is less than 10⁻³. In practice, convergence is usually achieved in three or four iterations.

2.2 Diffusion coefficients for binary gas mixture

From the VICTORIA^[6] database, thermochemical and physical data of 288 species and 26 elements are implemented as a form of look-up table. 288 species consist of 178 vapor species and 110 condensed-phase species.

The diffusion coefficients of binary gas mixtures at low pressure was estimated and compared with three implemented correlations. Chapman-Enskog kinetic theory, Fuller's empirical correlation, and the GA model(PADLOC code) are used to calculate the diffusion coefficients for a binary gas mixture of helium and iodine at a pressure of 0.1 ~ 100 atm (Fig. 2). Here, mixture temperature was assumed as 590°K. Chapman-Enskog kinetic theory and the GA model give well matched results with differences less than 1%. Fuller's empirical correlation over-estimates the diffusion coefficients over the pressure range. A noteworthy fact is that the GA model uses helium as a base gas and is not suitable for a containment analysis. Another fact is that Fuller's empirical correlation has some restrictions due to the limited data set of atomic diffusion volumes. Therefore, Chapman-Enskog kinetic theory will be used to calculate the diffusion coefficients of binary gas mixtures, based on newly implemented thermochemical and physical database.

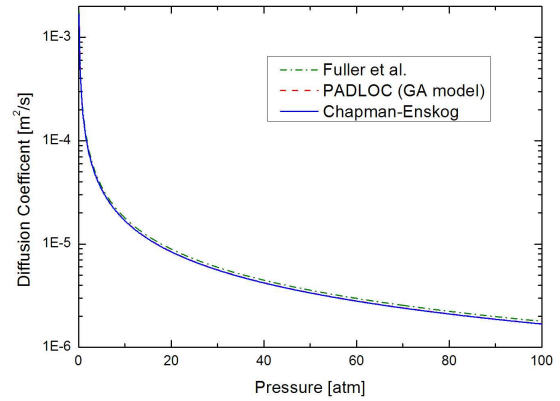


Fig. 1. Comparison of the diffusion coefficients for Helium and Iodine gas mixture at 0.1 ~ 100 atm.

3. Conclusions

The phase change model between aerosol and gaseous fission products was implemented in the GAMMA+ code and successfully verified. The thermochemical and physical data of 288 species and 26 elements were implemented as a form of look-up table, with Chapman-Enskog kinetic theory for estimating the diffusion coefficients of binary gas mixtures. In the future study, this model will be validated against available experimental data and improved for condensation onto structural surfaces.

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