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HORN: A COMPUTER CODE TO ANALYZE THE GAS-PHASE
TRANSPORT OF FISSION PRODUCTS IN REACTOR
COOLING SYSTEM UNDER SEVERE ACCIDENTS

November 1986

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HORN: A COMPUTER CODE TO ANALYZE THE GAS-PHASE TRANSPORT
OF FISSION PRODUCTS IN REACTOR COOLING SYSTEM UNDER SEVERE ACCIDENTS

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A computer code HORN has been developed to calculate the transport of volatile fission products in dry primary cooling circuit under severe accidents of water reactors. This report describes the analytical models of the code. As the most important factor governing the release or deposition of fission product, HORN determines the chemical forms of fission products both in the bulk flow and at the wall by the gas-liquid-solid equilibrium model. The results are used to calculate the diffusional deposition rates of vapors and to estimate the nucleation rates of aerosols. Efforts have been made to minimize the number of fission product elements for analysis by using the periodicity of elements and by treating less-volatile elements collectively as an imaginary element which always behaves as aerosols. The elements that are considered in the present version either as carrier gas or as elements for analysis are H, O, B, Kr, Xe, Cs, I, Te, Sb, Ag, Cd, and Ba.

Keywords: Fission Product Transport, HORN Code, Chemical Form, Reactor Accident, Cooling System

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HORN : 炉心損傷事故下の原子炉
冷却系における揮発性FPの移行解析コード

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(1986年10月7日受理)

炉心損傷事故時に冷却水を喪失した原子炉1次冷却系における揮発性FPの移行を解析するために、HORNコードを開発した。本報はそのモデルを解説するものである。

FPの放出または沈着を決定する最重要因子として、コードは流体中および配管表面におけるFP化学形を、気・液・固体平衡モデルにより計算する。その結果を用いて、拡散による気体FPの沈着、エアロゾルの析出等を計算する。

解析対象元素を少なくするため、周期律表上の同族元素を1つに代表させ、また揮発性の低い元素を1つの仮想元素としてまとめる等の近似を行った。現在解析対象としている元素は、非FPも含め、H, O, B, Kr, Xe, Cs, I, Te, Sb, Ag, Cd, Baである。

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1. Introduction

Fission products released from the degraded core under severe reactor accidents first travel through the primary cooling circuit before they are released into the containment or other buildings. Even if the effectiveness of the circuit as pressure boundary is lost, the circuit wall is still expected to work as deposition site for volatile fission products in some accident sequences. The postulated accident sequence taken up in the current risk analyses in which the deposition onto the primary circuit wall is particularly expected is the so-called interface LOCA (also called V-sequence) in PWRs. In this sequence, the leak path of fission products is a small-diameter tube connecting the main circuit and the outer buildings, bypassing the containment. It is obvious that bypassing of the containment increases the relative importance of the deposition onto the tube wall in the evaluation of the source term. In addition, the absolute amount of deposition will also be large in this sequence because fission products must travel over a long distance in a small tube before being released.

These circumstances will apply not only to the above particular sequence in PWR, but also to any sequence in PWRs and BWRs in which a tube penetrating the containment becomes the leak path, apart from the discussion whether such a sequence has been taken up in the current risk analysis or not. Thus it is meaningful to develop a computer code to analyze the transport of fission products in the primary cooling circuit in a general form, but still seeing the primary field of application in the analyses of containment-bypassing sequences.

The sequences in which condensed water remains in the circuit form an important field of accident analysis as the case of the TMI-2 accident implies. Involvement of aqueous chemistry, however, presents difficult problems for developing the analytical models with general applicability. If we limit ourselves to the class of accident in which the coolant is lost everywhere in the circuit, the situation is fairly simplified.

As a computer code analyzing the transport of fission products in dry circuits, TRAP-MELT⁽¹⁾ has been the best-known one. In the first version of TRAP-MELT, modeling has been made such that a single chemical form is assigned to each fission product, e.g. CsI for iodine and Ba(OH)₂ for barium, and that its deposition behavior is calculated from equilibrium vapor pressure. (Now TRAP-MELT has been improved to consider the change

of chemical forms.)

HORN is a code developed in JAERI with a similar purpose. When the development work was started, it has been well recognized that chemical form is the most important factor determining the transport of fission products. Modeling was therefore made, from the beginning, with the plan to place equilibrium chemical calculation in the center and arrange simple models for kinetic processes around it. Although the subcode for chemical calculation has a somewhat complex structure, for heat and mass transfers only the most fundamental modes were taken up and incorporated into simple governing equations.

This plan is based on the understanding that even if a code was developed with the greatest conceivable sophistication, reliability of the results would be limited by the enormous uncertainty of the conditions in 'future accidents'. What is important for the code is then to grasp the principal feature of the phenomena and to leave the users a room for understanding, extrapolating and generalizing the calculation results. In the chemical subcode, this philosophy was materialized in the limited number of species to be considered, neglecting less important species.

The code development has completed the first phase. A preliminary code assessment has been done using the data from the fission product transport experiment in LOFT. Further assessments are being planned. From the above philosophy, however, incorporation of new models will be limited to a minimum and the models will remain basically as described in the present report. Figure 1 shows the flow of calculation and hence the scope of the code too.

The most fundamental assumptions, restrictions, and simplifications in the code are as follows:

- 1) Flow path is composed of tube segments and tanks connected in series.
- 2) Volatile fission products are minor components of the gas flowing in the path, and hence their behavior does not affect the bulk flow state.
- 3) Condensed water does not exist in the tube.
- 4) Chemical forms of fission products including the nucleation of particles are determined as equilibrium composition.

- 5) Heat and mass transfer processes to the wall are described as analogous diffusional processes across the boundary layer.
- 6) Within each time step, steady flow condition is assumed.

The standard set of input data required for calculation is composed of inlet temperature, pressure distribution, wall temperatures, and flow rates of fission products and carrier gases. The results are output in terms of the amounts of fission products that was released from outlet, or deposited at each location of the flow path, fractions of gas molecules and aerosols, and dominant chemical forms of fission products.

2. Elements and Compounds for Analysis

In the periodic table, fission product elements distribute from the end of the fourth period to the beginning of the sixth period. To pay equal attentions to all these elements will cause excessive calculational load in the analysis of their transport through the primary cooling circuit. From the purpose of this code, it is necessary and possible to rank these elements according to their importance. The importance here does not always agree with the importance for the general source term evaluation. For example, less-volatile fission products may be released under the severest class of accident, and hence can be important for source-term evaluation. During the transport in the primary cooling circuit, however, they will behave as aerosols because of the low volatility and the lower temperature in the tube than the degraded core. The behavior of aerosols can be described fairly independently of their chemical forms. Hence less-volatile elements can be summed up as 'aerosol-forming elements', and need not be given as precise handling as volatile elements.

Then, the factors determining the importance here are abundance both in activity and mass, and volatility. Figure 2 shows the abundances of fission products (in logarithmic scale and arranged in the order of the periodic table) after 1000 days irradiation of ^{235}U , calculated with the DCHAIN-2 code⁽²⁾. Similarly, activities of the fission products are shown in Fig. 3. In this case, the activities after cooling time of 10^4 s were taken, considering the time for release to the environment. It is observed that the five elements from Pd to Sn have small abundances, among which, however, Ag, Cd and In can also come from the control rod, though they are nonactive. In summary, the number of elements for analysis cannot

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be reduced very much from the consideration of abundances.

Volatility of fission product is of course not an intrinsic property of element, but is determined by its chemical form. However, it is still possible to have a rough idea about the order of volatility of fission products by considering the volatility of their elemental forms. Figure 4 shows the boiling points of fission products in elemental forms arranged in the order of the periodic table. The metallic elements from Y to Rh and also lanthanoids have high boiling points, i.e. low volatilities. It is true that such low-volatility elements as Zr and Ru have highly volatile chemical forms as ZrI_4 and RuO_4 , respectively. Such species are, however, formed generally only under special conditions as high iodine or oxygen partial pressure, which is unlikely to be realized in the primary cooling circuit during accident.

The third idea to reduce the number of elements for analysis is to use the periodicity of elements. The abundant and volatile fission products are distributed in two blocks in the periodic table, i.e. one around Kr and another around Xe. Rigorously speaking, Br is different from I, and Sr is different from Ba. For fission product transport analysis, however, the differences can be neglected and the group around Xe (having I and Cs as members) can stand for the corresponding elements in the earlier period.

In any analysis of reactor accident, iodine is regarded as by far the most important fission product because of its abundance and high volatility. The chemical forms of I is one of the major issues in the source term evaluation. In the selection of elements for analysis, therefore, the chemical affinity to I was also considered. Figure 5 shows the dissociation I_2 pressure of various iodides of fission products at 1000 K, calculated from thermodynamic data. Low dissociation pressure means the stability of the iodide. It is seen that alkali metal and alkaline earth metals naturally have stable iodides. Transition metals are excluded from the important element group from this viewpoint too.

From all these considerations, the following elements were selected as the elements for analysis in the HORN code:

(fission product) Kr, Sb, Te, I, Xe, Cs, Ba

(structural elements) H, O, B, Ag, Cd

(fictitious element) Fi

The fictitious element Fi above is a concept similar to fissionium (a fictitious element made of molten fuel and core structural elements): it represents all less-volatile fission products, actinides, and structural elements that were made airborne. It is neglected in the calculation of chemical forms, but is considered in the calculations of aerosol transport and decay heat buildup in the tube wall. Fi is assumed to always behave as aerosol in the primary reactor circuit.

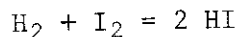
3. Chemical Forms of Fission Products

3.1 Scope

The term 'chemical form' is used here to mean not only the combination of elements but also its different phases. The purpose of estimating the chemical forms of fission products and their concentration in HORN code is, 1) to determine the equilibrium concentrations of vapors in bulk flow and at the wall surface for calculation of deposition (condensation) rate, and 2) to determine the rate of aerosol nucleation. For the first purpose, a chemical equilibrium subcode MPEC is called twice at each position of the flow path, first with the bulk fluid temperature as argument and secondly with the tube wall temperature.

The HORN code basically considers only gas phase reactions, though the concept of chemical reaction is not explicitly used in the algorithm. Surface reactions on the wall and radiation-induced reactions are neglected in the present version.

The most fundamental assumption in the HORN code is that the fission products instantaneously take their equilibrium chemical forms including phases. The validity of this assumption depends primarily on temperature. For example, in the case of the well-known first-order reaction:



characteristic time at 1000 K is about 1 ms, while it increases to about 10 s at 800 K. Considering the typical transit time of fission products in the primary cooling circuit, this temperature range will be the lower limit of using this assumption without problem.

Fundamentally the same limitation exists for the phase change of a single species. In this case, however, the delay in formation of condensed phase can be handled to some extent by the supersaturation model, which will be described in 3.4, within the framework of the equilibrium model. Thus, if the equilibrium chemical forms except phase are not so different at the upstream and downstream parts of a flow path, the HORN model can be approximately applied to the downstream part whose temperature is lower than the limit.

3.2 Equilibrium Chemical Subcode MPEC

The subcode MPEC calculates the concentrations of chemical species as a function of temperature, pressure, and gram-atoms of constituent elements. For each species, standard Gibbs partial molar free energy is given as a function of temperature. Using these data, the subcode calculates the equilibrium composition by the principle of minimum total free energy. Details of MPEC is reported elsewhere^{(3),(4)}, and the basic model is the same as that of SOLGASMIX-PV.^{(5),(6)} The algorithm of MPEC will therefore be described here only briefly.

Consider a system composed of M phases, which comprise gas phase (defined as phase 1), condensed mixtures, and pure phases. Assuming that the mixtures whose total number is MM are all ideal solutions, total Gibbs free energy of the system is given by

$$\frac{G_{tot}}{RT} = \sum_i^{IG} y_i (g_i + \ln P + \ln \frac{y_i}{Y_1}) + \sum_{m=2}^{MM} \sum_i y_i (g_i + \ln \frac{y_i}{Y_m}) + \sum_{m=MM+1}^M y_m g_m \quad (3.1)$$

where $g_i = G_i^0/RT$; G_{tot} is total Gibbs free energy (J); T is temperature (K); P is pressure (atm); y_i is moles of species i ; Y_m is total moles of phase m ; G_i^0 is standard chemical potential (partial molar Gibbs free energy) of species i (J/mole), which will be described in 3.3; R is gas constant (8.314 J/mole·K).

In equation (3.1), the first term corresponds to the gas phase; the second term corresponds to condensed mixtures whose number is $MM-1$, and the third term to pure phases. The moles of each species y_i cannot take an arbitrary value; it must satisfy the constraint determined by the gram-atoms of the constituent elements:

$$\sum_i^N A_{ij} y_i = b_j \quad (j = 1, \dots, L), \quad (3.2)$$

where b_j is gram-atoms of element j ; L is the total number of elements; A_{ij} is stoichiometry coefficient (e.g. 2 or 1 for H_2O).

The equilibrium chemical composition is a combination of y_i that makes G_{tot} (or G_{tot}/RT) minimum under the constraint of (3.2). This problem can be handled by using the Lagrange's undetermined multiplier.

Differentiating (3.1) and (3.2) by y_i , and using the undetermined multiplier π_j corresponding to element j , the condition for equilibrium is written, for gas phase,

$$\sum_j^L A_{ij} \pi_j - (g_i + \ln P + \ln \frac{y_i}{Y_1}) = 0, \quad (3.3)$$

for condensed phase m ,

$$\sum_j^L A_{ij} \pi_j - (g_i + \ln \frac{y_i}{Y_m}) = 0, \quad (3.4)$$

and for pure phases,

$$\sum_j^L A_{ij} \pi_j - g_i = 0. \quad (3.5)$$

Total moles of species in the mixtures (including gas phase) is given by

$$Y_m = \sum_{i=m_1}^{m_2} y_i. \quad (3.6)$$

Equations (3.2) to (3.6) are solved for the unknowns π_j , y_i and Y_m by the Newton-Raphson procedure. A standard procedure is to expand the nonlinear terms $\ln y_i$ and $\ln Y_m$ into Taylor series around their initial estimate values y_{i0} and Y_{m0} and to linearize the equations by taking the first-order terms. This procedure is, however, a slow convergence one. In HORN, instead, logarithms of y_i and Y_m are taken as independent variables:

$$z_i = \ln y_i \quad \text{and} \quad W_m = \ln Y_m, \quad (3.7)$$

and using these new variables, equations (3.3) and (3.4) are linearized.

In return, however, linear equations (3.2) and (3.5) become nonlinear. These equations are expanded into Taylor's series around the initial (or previous) values of the new variables z_{i0} and W_{m0} . Taking the first-order terms, we have

$$\sum_i^N A_{ij} y_{i0} z_i = \sum_i^N A_{ij} y_{i0} (z_{i0} - 1) + b_j, \quad (3.8)$$

$$\sum_{i=m_1}^{m_2} y_{i0} z_i - Y_{m0} W_m = \sum_{i=m_1}^{m_2} y_{i0} z_{i0} - Y_{m0} W_{m0} , \quad (3.9)$$

where the symbols with subscript 0 denote the initial values; m_1 and m_2 are the first and the last indices of the species belonging to phase m , and N is the total number of species.

Equations (3.3), (3.4), (3.5), (3.8), and (3.9) are solved for π_j , z_i , and W_m . The obtained solutions are iteratively used as the previous-step values until convergence is obtained.

This algorithm realizes very fast convergence, but on the other hand, suffers from numerical instability in some cases. To avoid instability, the iteration is started from the initial values well approximating the final solution. The initial estimation of composition is made also based on the principle of minimum total free energy, but with only linear terms. In equation (3.1), the logarithmic terms $\ln(y_i/Y_m)$ correspond to the mixing entropy in ideal solutions and make smaller contribution to total free energy than the standard free energy term g_i^0 . Hence we can make a guess of the final solution by minimization of total free energy composed of linear terms alone:

$$\frac{G'_{tot}}{RT} = \sum_i g'_i y_i , \quad (3.10)$$

where

$$\begin{aligned} g'_i &= g_i^0 + \ln P \quad (\text{gas phase}) , \\ &= g_i^0 \quad (\text{condensed phase}) . \end{aligned} \quad (3.11)$$

Minimization of G'_{tot} given by (3.10) under the constraint equation (3.2) and another constraint that $y_i > 0$ can be carried out by the linear programming method. The Simplex method is adopted in MPEC.

The solution obtained by the Simplex process is that out of N species only L stable species take positive y_i values whereas all others take zero values. For use of this solution as the initial estimate of z_i and W_m in the free energy equation, small y_i values are given to the zero-concentration species because the logarithmic term does not allow the presence of zero y_i values.

Another important measure to prevent instability is specific to the

use of MPEC in HORN. In the analysis of fission product transport, it often happens that fission products are very dilute and the fraction of fission product gram-atoms to that of H and O is reduced to 10^{-8} or less. In such a problem, the total moles of the gas phase is determined almost completely by the amounts of H_2 and H_2O , independently of the chemical forms of fission products. The total moles of the gas phase Y_1 can be regarded as a constant in such a problem, and hence one unknown Y_1 and the corresponding equation (3.6) can be omitted from the equation set. This handling contributes greatly to increase the stability of calculation when the gram-atoms of the constituent elements are very different. In the present version, Y_1 is regarded as a constant when the total gram-atoms of fission products is smaller than 10^{-7} of the (H + O) gram atoms.

3.3 Compounds for Analysis and Pre-Selection among Them

Numerous compounds are conceivable as the chemical forms that the sixteen elements may take between them. However, the number of stable compounds in the atmosphere typical of the primary cooling circuit under accidents (steam and hydrogen mixture) is not so large. For a metallic element such as Cs or Ba, the compounds worth considering are elemental form, oxide, iodide, and telluride. For anion-forming elements such as I or Te, hydrides are important species. In principle, the species for consideration are limited to binary compounds: complex oxides such as $CsFeO_2$ are not considered. Thermodynamically, such complex oxides are considerably stable. Neglect of these oxides are not for instability, but because it is doubtful that they are formed by the gas-phase reaction in the cooling circuit. Only exception is borates: such compounds as $CsBO_2$ and $Ba(BO_2)_2$ are considered in the code. It is because boric acid can exist in the gas phase in great excess of the amount of fission product, and also the importance of borates for the fission product chemical forms has been observed in simulation experiments.

For the chemical species adopted for consideration, the thermodynamic data and molecular transport property data are stored in the code. They are shown in Appendix B. The Gibbs standard partial molar free energy data are expressed as a second order polynomial of temperature:

$$G_i^0 = A_i + B_i T + C_i T^2. \quad (3.12)$$

The definition of G_i^0 is such that it is zero for reference state of each element at 298.15 k. In other words, for a compound at temperature T , its $G_i^0(T)$ includes both the free energy change due to reaction and the integral of specific heat. The thermodynamic data were taken mostly from the Barin-Knacke table⁽⁷⁾, and for some compounds from the JANAF table⁽⁸⁾, Gmelin handbook⁽⁹⁾ and other sources. When the numerical data were presented over a temperature range greater than 800 K, the data were fitted to second-order polynomials, and in other cases, to linear functions.

In the equilibrium chemical calculation, gas-phase species are assumed to make an ideal solution. On the other hand, liquid and solid species are assumed to make each independent pure phases.

The running time of the chemical subcode depends strongly on the number of compounds considered. To lessen the calculation load, therefore, pre-selection of compounds is made at each call of the subcode such that only those compounds with considerable stabilities are picked up for chemical calculation. The pre-selection is proceeded as follows:

- 1) For a combination of elements, out of three states (gaseous, liquid and solid), the most stable state at the temperature is selected.
- 2) The Simplex process as stated in 3.2 is applied to these compounds to determine the combination with the smallest standard free energy. Denoting the chemical potential of each element under this combination μ_j , the standard free energy (chemical potential) of a compound selected by the Simplex process is expressed as

$$G_i^0 = \sum_j^L A_{ij} \mu_j, \quad (3.13)$$

or in vector form, as

$$\{G^0\} = [A] \{\mu\}. \quad (3.14)$$

Since $[A]$ is a $L \times L$ matrix, equation (3.14) can be solved for μ_j as

$$\{\mu\} = [A]^{-1} \{G^0\}. \quad (3.15)$$

Thus, the current level of chemical potential of each element is determined.

- 3) Using this set of μ_j , we can know how 'unstable' is a species 'not selected' by the Simplex process. Inserting the μ_j into (3.13), but this time taking all the compounds as i , we can define an 'instability factor' f as follows:

$$f_i = (G_i^0 - \sum_j^L A_{ij} \mu_j) / RT . \quad (3.16)$$

This factor is zero for the compounds selected by the Simplex process and is positive for other species. Those species with relatively small positive f_i values must be considered in the chemical calculation, because they are allowed to exist at small concentration in mixture phases. The threshold level of f_i depends on how dilute the airborne fission products are expected to be in the flow path. In HORN, the following thresholds are adopted:

$$\begin{aligned} f_i &< 30 \quad (\text{for gas-phase species}) , \\ &< 5 \quad (\text{for condensed-phase species}). \end{aligned} \quad (3.17)$$

The f_i factor corresponds directly to the entropy of mixing, and the above threshold value of 30 means that gaseous species having as small a concentration as 10^{-13} of steam is considered in the chemical subcode.

3.4 Supersaturation Model

The basic assumption of instantaneous equilibrium in the HORN code means, in the case of phase change, that condensed-phase species instantaneously nucleates when the partial pressure of the gaseous species has exceeded the saturation pressure. When this model is applied to aerosol nucleation, the nucleation (rate) is overestimated for two reasons. The first reason is the neglect of the Kelvin effect. Since the free energy data used in HORN are those determined from bulk chemical equilibrium, the saturation pressure calculated is essentially for gas-planar surface equilibrium. For equilibrium with extremely small particle surface, extra gas pressure is necessary corresponding to the curvature.

The second reason is the kinetic effect: when temperature is low and the concentration of pre-existing particles is small, considerably high supersaturation (sometimes greater than 10^3) is necessary for gas

molecules to condense by homogeneous nucleation. If it is allowed to postulate a unique value of supersaturation at a location of the flow path, those two effects can be considered within the framework of the chemical equilibrium model.

Degree of supersaturation of species i , f_i^S , is defined by

$$\frac{P_{i,sat}^h}{P_{i,sat}} = 1 + f_i^S, \quad (3.18)$$

where $P_{i,sat}$ is saturation pressure of species i in a normal sense, and $P_{i,sat}^h$ is that for homogeneous nucleation of particles.

When a gaseous species i is at saturation pressure with a planar surface, its chemical potential is expressed as

$$\mu_{i,sat} = G_i^0 + R T \ln P_{i,sat}. \quad (3.19)$$

Using (3.18), (3.19) can be rewritten as

$$\mu_{i,sat} = \{ G_i^0 - R T \ln(1 + f_i^S) \} + R T \ln P_{i,sat}^h. \quad (3.20)$$

Equation (3.20) means that if we shift the standard free energy data of a gaseous species by $- R T \ln(1 + f_i^S)$, then the saturation pressure calculated by the subcode is that corresponding to homogeneous nucleation of particles. In the HORN code, in fact, the standard free energy data of condensed-phase species are shifted by $+ R T \ln(1 + f^S)$ with a single value of f^S assumed to all species.

It is desirable that the code calculates the supersaturation factor by itself, depending on temperature, particle density and other factors. In the present version, however, the supersaturation factor is left as an input parameter.

4. Heat and Mass Transfer Models

The flow path for fission products is idealized as a combination of circular tubes and cylindrical tanks connected in series. As fission products carried by steam flow in this path, gas temperature decreases and some of the fission products are removed onto the wall. With time, the tube or tank wall temperature rises both by heat transfer from gas and release of decay heat from the deposited fission products.

These time-dependent phenomena proceed at very much different speeds: the gas mixture will stay in the flow path typically for only several seconds, while an appreciable change of wall temperature will occur over a period of the order of minutes. For these fast-proceeding phenomena as decrease of gas temperature and fission product removal, the HORN code assumes that steady state is established within each time step. Changes in gas temperature and the released amounts of fission products over a whole period for analysis are expressed as a compilation of such steady states. On the other hand, heatup of the tube wall is treated by a completely transient algorithm.

Very simple models are used both for heat and mass transfer, considering the enormous uncertainties coming from the physical and chemical complexities of fission products and from the uncertainty of the accident condition.

4.1 Heat Transfer Model

4.1.1 Governing Equation

A flow path is divided into control volumes. A tank is regarded as one control volume. The thermal state of a control volume is characterized by three bulk temperatures, i.e. inlet, middle and outlet temperatures, and a wall temperature. Wall temperature in each control volume is either given by input data, or calculated from the heat balance. In the latter case, calculation of the wall temperature is separated from that of gas-wall heat transfer; it is made at the end of each time step using the results of the gas-wall heat transfer calculation as known quantities. The wall heatup model will be described in 4.1.4; here wall temperature is treated as a fixed known quantity.

Since volatile fission products are minor components of the flowing gas, and condensation of steam is not considered in HORN, the following

mole conservation condition applies to the gas flowing in the tube in each time step:

$$F_m = \rho_m u S_f = \text{const.}, \quad (4.1)$$

where F_m is mole flow rate (mole/s); ρ_m is molar density (mole/m³); S_f is surface area (m²). Applying the ideal gas law, (4.1) is expressed as

$$u = \frac{F_m R}{S_f P} T = A T, \quad (4.2)$$

where R is gas constant (8.314 J/mole K); T is temperature (K); P is pressure (Pa). Pressure is given by input data as time and segment-dependent values, but within each time step and each segment it is regarded as a constant. Then the factor A in (4.2) is a constant and the flow velocity u is a function only of temperature.

Neglecting the potential of gravity, equation of energy balance is written as

$$dH + M u du = dQ, \quad (4.3)$$

$$dH = C_p dT, \quad (4.4)$$

$$dQ = - \frac{\pi d q dx}{\rho_m u S_f}, \quad (4.5)$$

where dH is enthalpy change (J/mole); dQ is heat input to a unit mole of the gas (J/mole); M is molecular weight of the gas (kg/mole); C_p is molar specific heat (J/mole K); d is inside diameter of the tube (m); x is distance along the tube (m); q is heat flux from the gas to the wall (W/m²). Here, direct deposition of decay heat to the flowing gas is neglected; heat flux is composed of convective and radiative parts:

$$q = h (T - T_w) + \bar{\epsilon} \sigma (T^4 - T_w^4), \quad (4.6)$$

where h is heat transfer coefficient (W/m² K); T_w is wall temperature (K); $\bar{\epsilon}$ is effective emissivity between the gas and the wall; σ is Stefan-Boltzmann constant (5.67×10⁻⁸ W/m² K).

Inserting (4.2), (4.4), (4.5) and (4.6) into (4.3), we get an explicit

expression of the heat balance equation:

$$(A^2 M T + C_p) \frac{dT}{dx} + \frac{\pi d R}{A S_f P} \{ h(T - T_w) + \bar{\epsilon} \sigma (T^4 - T_w^4) \} = 0. \quad (4.7)$$

This equation is transformed into the form

$$\frac{A^2 M T + C_p}{(T - T_w) \{ h + \bar{\epsilon} \sigma (T + T_w)(T^2 + T_w^2) \}} dT = - \frac{\pi d R}{A S_f P} dx, \quad (4.8)$$

and further into the form

$$\left(\frac{\xi}{T - T_w} + \frac{\eta}{T + T_w + \frac{h}{\bar{\epsilon} \sigma \bar{T}^2}} \right) dT = - \frac{\pi d R}{A S_f P} dx, \quad (4.9)$$

where

$$\xi = \frac{A^2 M T_w + C_p}{2 \bar{\epsilon} \sigma \bar{T}^2 T_w + h} \quad (4.10)$$

$$\eta = \frac{A^2 M (\bar{\epsilon} \sigma \bar{T}^2 T_w + h) - \bar{\epsilon} \sigma C_p \bar{T}^2}{\bar{\epsilon} \sigma \bar{T}^2 (2 \bar{\epsilon} \sigma \bar{T}^2 T_w + h)} \quad (4.11)$$

and $\bar{T}^2 = T^2 + T_w^2$, which is treated as if it is a constant within each iteration step of numerical solution, but is renewed every step.

Integrating (4.9) and using the boundary condition that $T = T_0$ at $x = 0$ (inlet temperature in each segment), we have

$$\xi \ln \frac{T - T_w}{T_0 - T_w} + \eta \ln \frac{T + T_w + \frac{h}{\bar{\epsilon} \sigma \bar{T}^2}}{T_0 + T_w + \frac{h}{\bar{\epsilon} \sigma \bar{T}^2}} = - \frac{\pi d R x}{A S_f P}. \quad (4.12)$$

The bulk gas temperature and outlet temperature of a tube segment is given by inserting $x = L/2$ and L (L is segment length), respectively into (4.12) and solving it by the Newton-Raphson process.

Heat transfer in tank elements is treated by a very simple model. Gas temperature in a tank is represented by a single value of bulk gas temperature T except that gas flows into the tank at a temperature T_0 . It is assumed that outlet temperature is equal to the bulk temperature. Then the governing equation for heat transfer is written as

$$\begin{aligned} C_p F_m (T_0 - T) &= q S_w \\ &= S_w \{h(T - T_w) + \bar{\epsilon} \sigma (T^4 - T_w^4)\} \end{aligned} \quad (4.13)$$

where S_w is the whole surface area of the tank wall (m^2). Equation (4.13) can be readily solved for T by the Newton-Raphson process.

Use of (4.3) within each control volume satisfies the energy conservation condition. At the boundary of two control volumes, however, outlet temperature of the upstream volume must not be used as the inlet temperature of the downstream volume. It is because discrete values of pressure are assigned to the control volumes and hence pressure is regarded to change suddenly at the boundary. Then the resultant sudden change of $m du$ term in (4.3) must be compensated by the change of gas temperature. It is achieved by assuming an instantaneous adiabatic expansion of the gas at the boundary.

Neglecting the contribution from hydrogen, temperature change due to adiabatic expansion of low-pressure steam can be expressed by⁽¹⁰⁾

$$P_1 V_1^{1.135} = P_2 V_2^{1.135} \quad (4.14)$$

Combining (4.14) with the ideal gas law:

$$\frac{P_1 V_1}{T_1} = \frac{P_2 V_2}{T_2} \quad (4.15)$$

we get an expression of the inlet temperature of the downstream control volume as

$$T_2 = T_1 \left(\frac{P_2}{P_1} \right)^{0.119} \quad (4.16)$$

4.1.2 Convective Heat Transfer Correlations

For convective heat transfer in tubes, heat transfer coefficient is given by the Dittus-Boelter correlation⁽¹¹⁾ for turbulent flow:

$$h = 0.023 \frac{\kappa}{d} Re^{0.8} Pr^{0.4} \quad (4.17)$$

where

$$Re \text{ (Reynolds number)} = \frac{F_m M d}{S_f \mu} \quad (4.18)$$

$$Pr \text{ (Prandtl Number)} = \frac{C_p \mu}{\kappa}, \quad (4.19)$$

and μ is viscosity (kg/m s) and κ is thermal conductivity (W/m K).

For heat transfer in tank elements which is predominantly by natural convection, a correlation specially derived for HORN is used:

$$h = 7.06 \frac{\kappa}{d} Gr^{0.2033}, \quad (4.20)$$

where

$$Gr \text{ (Grasshof Number)} = \frac{g \beta (T - T_s) \bar{L}^3}{\nu^2} \quad (4.21)$$

and g is acceleration of gravity (9.8 m²/s); β is coefficient of volumetric expansion of gas (K⁻¹) which is equal to $1/T$ by ideal gas law; ν is kinematic viscosity (m²/s); \bar{L} is a geometric average of the tank diameter and height (m).

The correlation (4.20) was derived from a series of thermohydraulic calculations performed on a model tank with PHOENICS code⁽¹²⁾. For natural convection heat transfer, many correlations of the (4.20) type have been already proposed. The PHOENICS code calculation was made considering the application of the correlation to the upper plenum part of pressure vessel. In the upper plenum during severe accidents, the flow and heat transfer patterns are expected to be very complex: upward steam flow from the core and dominance of radiative heat transfer will add to natural convection. Hence use of the correlation obtained from a purely natural convection experiments may not be valid. Since PHOENICS is an orthodox hydrodynamic code that rigorously solves the three conservation equations (mass, momentum, and energy), a series of calculations with the code will work as a computer experiment.

A model tank for calculation was defined as a vertical cylinder in Fig. 6 simplifying the upper plenum of PWR by neglecting the internal structures. The bottom of the cylinder was assumed to be open, at which uniform upward flux of steam was assumed as inlet flow. In a typical vessel of PWR, several outlet nozzles are located at about mid-height of the side wall. To avoid three-dimensional calculation, however, the nozzles were substituted by a circular slit on the side wall with the same flow

area.

The whole volume was separated into 22 radial and 25 axial meshes which are fine near the wall and coarse in the interior, and the flow equation was solved.

Totally twelve calculation runs were performed in which inlet flow rate, inlet temperature, pressure and wall temperature were changed in the following ranges: flow rate (0.09 to 9 kg/s), pressure (0.1 to 10 MPa), inlet temperature (700 to 2000 K), and wall temperature (500 to 1500 K). For most cases, heat transfer takes place predominantly by radiation. The convective part of heat flux, which is locally determined from temperature gradient and surface friction factor, were integrated over the whole surface and expressed as a kind of heat transfer coefficient between the volume-average steam temperature and wall temperature. It was further expressed in terms of the nondimensional quantities as shown in Fig. 7 and the results were fit into the correlation of (4.20).

Correlation (4.20) thus obtained gives larger heat transfer coefficient in most cases than the conventional natural convection heat transfer correlations found in literature⁽¹⁹⁾. It will be because in the present case larger radiative heat transfer due to higher steam temperature enhances natural convection and hence the convective part of heat transfer too. Then the applicability of (4.20) will naturally be limited to high temperature cases. In addition, neglect of internal structures and approximation by axisymmetric flow field will limit its applicability even in the upper plenum of PWRs from which the model tank was defined.

On the other hand, the good fitting to a simple correlation observed in Fig. 8, which is rather surprising considering the wide range of the parameters and predominance of radiative heat transfer, implies some general applicability beyond the dimensions of the model tank assumed in the calculation. The effect of inlet mass flow rate was not observed in the calculation in spite of the change of the rate by two orders of magnitude. The open bottom face and upward inlet flow there assumed in the calculation are not, therefore, critical restrictions in the applicability of (4.20).

4.1.3 Effective Emissivity and Kinetic Properties of Gas

The effective emissivity between the gas and the wall is given by the formula for emissivity between two parallel planes:

$$\bar{\epsilon} = \frac{\epsilon_g \epsilon_s}{\epsilon_g + \epsilon_s - \epsilon_g \epsilon_s}, \quad (4.22)$$

where ϵ_s is emissivity of the wall surface which is an input constant. ϵ_g is the emissivity of the 'surface' of the gas, which is not an intrinsic material property of the gas, but is dependent on the size. It is determined by

$$\epsilon_g = 1 - \exp(-K L_m), \quad (4.23)$$

$$L_m = f L_r, \quad (4.24)$$

where K is total energy absorption coefficient of the gas (m^{-1}); L_m is the length of average radiation flux (m); L_r is representative dimension of the gas lump (m). For gas in an infinite cylinder, when L_r is taken equal to the diameter of the tube, f is taken to be 0.94. For total absorption coefficient, the data for pure steam at atmospheric pressure⁽¹³⁾ were fitted to the following correlation between 550 K and 2800 K:

$$K_p^0 = \exp \left\{ 4.635 - 3.465 \left(\frac{T}{1000} \right) + 0.563 \left(\frac{T}{1000} \right)^2 \right\}, \quad (4.25)$$

where T is temperature (K). For mixture gas at a pressure of p (atm), absorption coefficient is given by

$$K_p = K_p^0 p F_{H_2O}, \quad (4.26)$$

where F_{H_2O} is mole fraction of steam in the mixed gas. Since steam makes much larger contribution to absorption of radiation than other monoatomic or diatomic gases, contribution from other gases was neglected in (4.26).

Material properties of the mixed gas (viscosity, thermal conductivity and specific heat) are determined from the properties of five component gases, H₂O, H₂, O₂, Kr, and Xe: contribution from the volatile fission products is neglected. Since the effect of pressure on these properties is very small as long as pressure is not so high, these data were expressed as a function of temperature alone in the form

$$\ln Y = a + b \ln T + c (\ln T)^2, \quad (4.27)$$

where Y is either viscosity (kg/m s), or thermal conductivity (W/m K), or constant-pressure specific heat (J/mole K), and T is temperature (K). The numerical values of a , b , and c for the three properties and five gases are listed in Table 1 together with the source of original data.

In the case of the two kinetic properties, viscosity and thermal conductivity, property of mixed gas is determined from the properties of the component gases by the semi-empirical Wilke's equation⁽¹⁷⁾:

$$Y_{mix} = \sum_i^n \frac{x_i Y_i}{\sum_j x_j \Phi_{ij}}, \quad (4.28)$$

in which

$$\Phi_{ij} = \frac{1}{\sqrt{8}} \left(1 + \frac{M_i}{M_j} \right)^{-\frac{1}{2}} \left[1 + \left(\frac{\mu_i}{\mu_j} \right)^{1/2} \left(\frac{M_j}{M_i} \right)^{1/4} \right]^2. \quad (4.29)$$

Here Y_i is either viscosity or thermal conductivity of a component gas i and x_i is its mole fraction; M_i is its molecular weight. Note that fraction of viscosity is used in the definition of Φ_{ij} regardless of whether Y_i is viscosity or thermal conductivity.

On the other hand, specific heat of mixed gas is given simply as an average of component gas properties weighted by mole fractions:

$$C_{p,mix} = \sum_i x_i C_{p,i}, \quad (4.30)$$

where $C_{p,mix}$ is molar specific heat of gas i (J/mole K).

4.1.4 Tube Wall Heatup Model

HORN has an option to calculate the temperature change of tube or tank wall, considering the heat transfer from the interior gas flow and to the external atmosphere. Calculation of wall temperature by this option is separated from the calculation of gas temperature: wall temperature at the end of a time step is calculated using the heat flux from the gas flow during the step as a known quantity, and the calculated wall temperature is used in the next time step.

To solve the heat conduction equation using approximations of planar geometry and representation of thermal state by one point, an effective heat transfer coefficient \bar{h} is defined as follows:

$$\frac{1}{\bar{h}} = \frac{d_w}{k_w} + \frac{1}{h_{ex}}, \quad (4.31)$$

in which d_w is wall thickness of the tube or tank (m); k_w is thermal conductivity of the material (W/m K); T_{ex} is heat transfer coefficient with the external atmosphere (W/m² K). These three quantities, d_w , k_w , and h_{ex} are all input data.

If wall (inner surface) temperature at the beginning of a time step is T_0 and the external temperature is T_{ex} (constant), then the wall temperature after a time step size of Δt (s) is given by

$$T = T_0 \exp\left(-\frac{\bar{h} \Delta t}{\rho C_p d_w}\right) + \left(T_{ex} + \frac{\phi}{\bar{h}}\right) \left\{1 - \exp\left(-\frac{\bar{h} \Delta t}{\rho C_p d_w}\right)\right\}, \quad (4.32)$$

where ρ and C_p are density (kg/m³) and specific heat (J/kg K) of the wall, respectively, and ϕ is heat source term per unit area of the inner surface (W/m²). The heat source ϕ is composed of three terms:

$$\phi = \phi_c + \phi_r + q_A, \quad (4.33)$$

where ϕ_c and ϕ_r are convective and radiative heat fluxes (W/m²) from the interior, respectively, and q_A is decay heat per unit area from the deposited fission products.

When the thermal condition of the wall is approximated as adiabatic to the exterior, the wall temperature at the end of a time step is given simply by

$$T = T_0 + \frac{\phi \Delta t}{\rho C_p d_w}. \quad (4.34)$$

Decay heat q_A is calculated by

$$q_A = \sum_j n_j^d E_j^\beta / S_w, \quad (4.35)$$

where n_j^d is the number of j -th fission product element deposited in the control volume (gram-atom); E_j^β is average beta energy release from the element (W/gram-atom); S_w is inner surface area of the control volume (m²). Since the specific beta energy release E_j^β depends strongly on the accumulated burnup before the accident, it is left as input data.

4.2 Mass Transfer Model

4.2.1 Concentration of Gaseous Species in the Stream

There are various mechanisms by which gaseous fission products and aerosols are trapped by the wall as they flow in the flow path. For aerosols the relative importances of those mechanisms are discussed in 4.2.4. For fission products in gaseous forms, conceivable trap mechanisms are condensation, physisorption and chemisorption. Physisorption, however, cannot be an important removal mechanism for a large amount of fission product released at severe accident because the amount of physisorbed gas is generally an order of several atoms layer. Chemisorption may be more important under some conditions. However, for determining the deposition rate by chemisorption, not only the combination of a specific gaseous species and the wall material, but also the co-existing other species must be considered. Although the chemisorption rate of e.g. CsOH on stainless steel is now being investigated⁽¹⁸⁾, general data base does not exist yet.

Condensation is a mechanism by which a great deal of gas molecules can be removed from the stream and whose rate can be estimated by a general way if the equilibrium pressure of the gas species is known. For these reasons, HORN considers only condensation as deposition mechanism of fission products in gaseous forms.

As in the heat transfer model, concentration of a gaseous species i in a control volume is represented by two values, i.e. bulk concentration $C_{b,i}$ and concentration near the wall $C_{w,i}$ (mole/m³). The deposition rate is given by the molecular flux due to the concentration gradient. If the thickness of the boundary layer is clearly defined, the flux is given by

$$J_i = D_i \frac{C_{b,i} - C_{w,i}}{\delta}, \quad (4.36)$$

where J_i is flux of species i to the wall (mole/m²), D_i is diffusion constant (m²/s), and δ is the thickness of boundary layer (m). In fact, rather than explicitly using the concept of boundary layer as in (4.36), the flux is calculated by using the group velocity to the wall obtained from empirical correlations, as described in 4.2.2:

$$J_i = u_{t,i} (C_{b,i} - C_{w,i}), \quad (4.37)$$

where $u_{t,i}$ is group velocity of species i to the wall (m/s).

The two concentrations $C_{b,i}$ and $C_{w,i}$ are calculated from the results of the chemical subcode MPEC. In this case, MPEC calculation is made under constant-pressure condition. Hence MPEC requires amounts of elements, pressure, and temperature as input data, and gives the amounts of chemical species:

$$n = n(w, P, T) , \quad (4.38)$$

where the amounts of chemical species n_i and the amounts of elements w_j were expressed collectively in vector forms n and w , respectively. The two concentrations are calculated as follows:

$$C_{b,i} = \frac{n_{b,i}}{V} = \frac{1}{V} n_i(w^f, P, T_b) , \quad (4.39)$$

and

$$C_{w,i} = \frac{n_{w,i}}{V} = \frac{1}{V} n_i(w^f + w^d, P, T_w) , \quad (4.40)$$

where w^f is the amounts of elements that have flowed into the control volume during the time step Δt ; w^d is the amounts of elements that are deposited on the surface; V is the volume of the gas mixture that has flowed into the control volume during Δt (m^3).

The expression (4.40) means that the concentration at the wall is calculated from the moles of chemical species corresponding to the sum of flowing fission products and deposited ones. Since T_b is higher than T_w and the effect of temperature on vapor pressure is very large, $n_{b,i}$ is generally larger than $n_{w,i}$ for gas species even when w^d is considerably large. However, when the wall temperature is continuously rising, it can happen that $n_{w,i}$ is larger than $n_{b,i}$. In that case, the concentration gradient in (4.37) becomes negative and revaporization of already-deposited fission products is automatically calculated.

In accordance with the basic assumption of instantaneous equilibrium, the amount of flowing elements w^f is determined not only from those fission products that have existed in gas form in the upstream flow segment, but also those that existed as aerosols. Concentration of aerosols in the present control volume is determined by (4.39) taking i to

be condensed-phase species. In other words, aerosols imaginarily decompose to elements, take particular chemical forms, and nucleate again at every control volume. It is of course merely for convenience of calculation: ageing of aerosols (coagulation growth) is separately calculated from the travelling time from the segment in which first nucleation has taken place.

The basic concept of the mass transfer model is pictured in Fig. 8. The algorithm of using the results of equilibrium chemical calculation for mass transfer calculation is illustrated in Fig. 9.

4.2.2 Mass Transfer Governing Equation

For a gas mixture flowing in a tube, the equation for concentration change as it flows in a segment is given from the flux of (4.37) as

$$\frac{dC_{b,i}}{dt} = -\frac{4}{d} u_{t,i} (C_{b,i} - C_{w,i}) \quad (4.41)$$

in which d is inside diameter of the tube (m). Solving (4.41), moles of species i that has deposited on the wall in the segment during a time step span of Δt is given by

$$\begin{aligned} \Delta N_i &= V (C_{b,i}^0 - C_{w,i}) \left\{ 1 - \exp\left(-\frac{4 u_{t,i} L}{d u}\right) \right\} \\ &= (n_{b,i}^0 - n_{w,i}) \left\{ 1 - \exp\left(-\frac{4 u_{t,i} L}{d u}\right) \right\}, \quad (4.42) \end{aligned}$$

where L is the length of the tube segment (m); u is velocity of the gas mixture parallel to the axis (m/s); $C_{b,i}^0$ is the concentration at the inlet of the segment and $n_{b,i}^0$ is the total moles of species i in volume V which is given by

$$V = \frac{\pi}{4} d^2 u \Delta t \quad (4.43)$$

Even simpler governing equation is used for mass transfer in tank elements. As in heat transfer, a gas species i is assumed to enter a tank at concentration $C_{b,i}^0$, have an average concentration $C_{b,i}$ in the tank, and leave the tank also at the same concentration $C_{b,i}$. Then the mass-balance equation is simply an arithmetic equation:

$$(C_{b,i}^0 - C_{b,i}) V = S_w u_{t,i} \Delta t (C_{b,i} - C_{w,i}) \quad (4.44)$$

in which S_w is the surface area of the tank (m^2) and V is, corresponding to that given by (4.43), total volume of gas mixture that flow into the tank during the time span of Δt . Solving (4.44) for $C_{b,i}$, the total moles of species i that transfers to the wall is given by

$$\begin{aligned} \Delta N_i &= (C_{b,i}^0 - C_{b,i}) V \\ &= \frac{(n_{b,i}^0 - n_{w,i}) S_w u_{t,i} \Delta t}{V + S_w u_{t,i} \Delta t} \end{aligned} \quad (4.45)$$

Equations (4.42) and (4.45) apply primarily for diffusional deposition of gas molecules. However, the same forms of equation apply to the deposition of aerosols because deposition rates of aerosols by various mechanisms are also expressed by group velocities associated to the mechanisms. When (4.42) and (4.45) are used for deposition of aerosols, the aerosol concentration at the wall $n_{w,p}$ is taken to be zero, which means that accommodation factor of the wall for impinging aerosols is unity (no reflection).

4.2.3 Mass Transfer Correlations and Diffusion Constant of Gas Molecule

The diffusional group velocities of gas molecules normal to the wall are determined by analogy to heat transfer correlations. For turbulent flow in a tube, the normal velocity is given by the mass-transfer version of the Dittus-Boelter equation (4.17):

$$u_{t,i} = 0.023 \frac{D_i}{d} Re^{0.8} Sc^{0.4} \quad (4.46)$$

where

$$Sc \text{ (Schmidt Number)} = \frac{\mu}{\rho D_i} \quad (4.47)$$

and D_i is diffusion constant of species i (m^2/s) and ρ is density (kg/m^3). Note that although the Reynolds number (defined in (4.18)), viscosity μ and density ρ are determined from the state of bulk gas mixture, the Schmidt number is determined independently for each gas species.

For predominantly natural convection in a tank, a correlation analogous to (4.20) is used:

$$u_{t,i} = 7.06 \frac{D_i}{d} Gr^{0.2033} Sc^{0.25} . \quad (4.48)$$

Defference of (4.48) from (4.20) is that Schmidt number appears in the right-hand side of (4.48), whereas (4.20) does not have a corresponding non-dimensional quantity Prandtl Number. Correlation (4.20) was determined from a series of heat transfer calculations by PHOENIX code in which Prandtl number was always very near to unity. In application too, Prandtl number is nearly unity as long as predominantly steam atmosphere is concerned. On the other hand, Schmidt number can be considerably different from unity, so that Schmidt number dependency in (4.48) was added referring to a natural convection correlation in a literature⁽¹⁹⁾.

The Grashof number Gr is defined as in (4.21). There is another way of defining Grashof number in mass transfer, based on concentration gradient. In the present case, however, natural convection is driven by temperature gradient and the diffusion process is merely superimposed on the convection. Therefore, the Grashof number for heat transfer must be used.

Diffusion constant of gas molecules is determined by the Chapman-Enskog equation⁽¹⁷⁾. Diffusion constant D between two kinds of gas A and B (m^2/s) is not dependent on the mole fractions and is given by

$$D = 0.01882 \frac{\sqrt{T^3 (1/M_A + 1/M_B)}}{P \sigma_{AB}^2 \Omega} \quad (4.49)$$

where T is temperature (K); P is pressure (Pa); M_A and M_B are molecular weights of the two gases (g/mole). The two quantities σ_{AB} and Ω are parameters describing the interaction of two molecules. They are determined from the property of each molecule as

$$\sigma_{AB} = \frac{1}{2} (\sigma_A + \sigma_B) , \quad (4.50)$$

$$\Omega = f\left(\frac{\epsilon_{AB}}{kT}\right) , \quad (4.51)$$

$$\frac{\epsilon_{AB}}{k} = \sqrt{\left(\frac{\epsilon}{k}\right)_A \left(\frac{\epsilon}{k}\right)_B} , \quad (4.52)$$

where σ_A and σ_B are parameters related to the size of the potential wells of the molecules (Å). The function of (4.51) is an integral function whose values are derived by referring to a table; ϵ_A and ϵ_B are parameters related to the depth of the potential well (J); k is Boltzmann's constant (1.38×10^{-23} J/K).

Equations (4.50) to (4.52) apply to mutual diffusion in a binary gas mixture. In the present case, however, we must take individual fission product vapor as gas A, and the carrier gas mixture (steam, hydrogen, oxygen, Kr, and Xe) as gas B. In other words, we must treat a case of pseudo-binary gas mixture. Properties of gas B is determined as averages of the properties of the constituent gases:

$$\sigma_B = \sum_i^5 x_i \sigma_i, \quad (4.53)$$

$$\epsilon_B = \prod_i^5 (\epsilon_i)^{x_i}, \quad (4.54)$$

where x_i is mole fraction of gas i .

For common permanent gases as H_2 , H_2O , I_2 , the σ and ϵ (or ϵ/k) values are found in literatures⁽²⁰⁾. For most species that vaporize only at high temperature, however, these values must be determined by empirical correlations. Either of the following correlations⁽¹⁷⁾, which in fact apply only to non-polar gases, are used also for polar gases:

$$\epsilon/k = 0.77 T_c, \quad \sigma = 0.841 \tilde{V}_c^{\frac{1}{3}}, \quad (4.55)$$

$$\epsilon/k = 1.15 T_b, \quad \sigma = 1.166 \tilde{V}_{b,liq}^{\frac{1}{3}}, \quad (4.56)$$

$$\epsilon/k = 1.92 T_m, \quad \sigma = 1.222 \tilde{V}_{m,sol}^{\frac{1}{3}}, \quad (4.57)$$

in which T is temperature (K); \tilde{V} is molecular volume (ml/mole) and the three subscripts c , b , and m denote critical point, boiling point and melting point, respectively.

To check the applicability of the correlations (4.55) to (4.57) to polar gases, the molecular force constants ϵ/k and σ of selected polar gases were calculated by these correlations and the results were compared

with the literature values. As shown in Table 2, agreement is fairly good.

For some dimeric vapors such as $(\text{CsI})_2$ or $(\text{CsOH})_2$, molecular force constants cannot be estimated by the above correlations because the constants derived by these correlations are applicable to monomeric forms. For dimeric gases whose constants are unknown, the constants are estimated from the values for monomeric gas based on Stogryn and Hirschfelder's model⁽²¹⁾. They estimated the force constants for monomer-dimer interaction from those for monomer-monomer interaction as

$$\epsilon_{2A-A} = 1.32 \epsilon_{A-A} \quad , \quad \sigma_{2A-A} = 1.04 \sigma_{A-A} \quad . \quad (4.58)$$

Then, using (4.50) and (4.52), the force constants of dimers are related to those of monomers as

$$\epsilon_{2A} = 1.74 \epsilon_A \quad , \quad \sigma_{2A} = 1.02 \sigma_A \quad . \quad (4.59)$$

The molecular force constants thus estimated or referred for gaseous species are shown in Appendix B together with Gibbs free energy data. The diffusion constants of some important fission product chemical forms in 1 atm. steam were calculated from these data using the Chapman-Enskog equation and shown in Table 3. The diffusion constants of aerosols due to Brownian motion are also shown for comparison. It is observed that vapors diffuse much faster than aerosols.

4.2.4 Aerosol Growth Model

The behavior of aerosol is strongly dependent on particle size. The conditions characterizing the aerosol behavior in the primary cooling circuit under accidents are: 1) nucleation process is unseparable from growth, transport and removal processes, and 2) residence time is generally very small for sufficient growth process to occur. These circumstances lessen the meaningfulness of performing complex calculation of particle size distribution by counting the rate of collisions.

In HORN, log-normal size distribution is assumed for aerosols. Assuming that particles are all spherical, probability function for a particle of diameter d_p (m) is written as

$$df = \frac{1}{\sqrt{2\pi} \ln \sigma_g} \exp\left[-\frac{(\ln d_p - \ln \bar{d}_{cm})^2}{2(\ln \sigma_g)^2}\right] d(\ln d_p), \quad (4.60)$$

in which \bar{d}_{cm} is count-median diameter (CMD) of the distribution (m): σ_g is geometric standard deviation (GSD). For most aerosols, GSD is below 2.0 and the GSD for self-preserving size distribution is close to 1.5⁽²²⁾. Therefore, a constant GSD value of 1.5 is used in HORN unless other value is input.

Under log-normal size distribution, CMD is related to the diameter of average mass \bar{d}_{am} (m) by Hatch-Choate transformation:

$$\bar{d}_{am} = \bar{d}_{cm} \exp(1.5 \ln^2 \sigma_g) . \quad (4.61)$$

The diameter of average mass \bar{d}_{am} is determined from the mass concentration G_p (kg/m³) and number density N_p (m⁻³) of the aerosols by

$$G_p = \frac{\pi}{6} \rho_p N_p (\bar{d}_{am})^3 , \quad (4.62)$$

where ρ_p is density of particles (kg/m³) which is assumed to be independent of particle size.

From the equilibrium chemical calculation, the average mass concentration G_p is determined as concentration of condensed-phase species in the gas mixture, and \bar{d}_{am} and N_p are left as unknowns. It is known that in aerosols in which growth has not so extensively proceeded increase of \bar{d}_{am} can be described by the increase of particle size in a single-variance aerosol (uniform-size aerosol)⁽²²⁾. Decrease of number density of single-variance aerosol with time is given by

$$N_p = \frac{N_{p0}}{1 + N_{p0} K t} \quad (4.63)$$

in which N_{p0} is initial number density (m⁻³) and K is a rate constant of coagulation (s⁻¹). When Brownian motion is the mechanisms for coagulation (other mechanisms such as inertial motion or gravitational coagulation are not important because aerosols in the primary circuit remain submicron), the rate constant is given by

$$K = \frac{4 k T C_c}{3 \mu} \quad (4.64)$$

where C_C is Cunningham's correction factor for slip and determined from the ratio between the mean free path of molecules λ and particle size d_p as

$$C_C = 1 + \frac{\lambda}{d_p} \left\{ 2.514 + 0.800 \exp\left(-0.55 \frac{d_p}{\lambda} \right) \right\} \quad (4.65)$$

The mean free path of gas molecules is determined by kinetic theory of gas⁽²³⁾ as

$$\lambda = \frac{1}{\sqrt{2} \pi d_c^2 n} = \frac{k T}{\sqrt{2} \pi d_c^2 P} \quad (4.66)$$

where n is number density of gas molecules (m^{-3}); P is pressure (Pa); d_c is collision diameter of molecules (m). The collision diameter can be expressed in terms of the Lennard-Jones molecular force constants σ and Ω described in the previous section. For this purpose, viscosities of a gas that given by kinetic theory and an empirical equation using the Lennard-Jones parameters are compared. From kinetic theory, viscosity of a gas μ (kg/m s) is given by⁽²²⁾

$$\mu = \frac{2}{3} \frac{\pi^{3/2}}{\pi} \frac{\sqrt{m k T}}{d_c^2} \quad , \quad (4.67)$$

where m is mass of a molecule (kg).

From the Lennard-Jones potential model⁽¹⁷⁾,

$$\mu = 2.6693 \times 10^{-6} \frac{\sqrt{M T}}{\sigma^2 \Omega} \quad , \quad (4.68)$$

in which M is molecular weight (g/mole). Equating the two viscosities given by (4.67) and (4.68), we have an expression of d_c as

$$d_c^2 = 6.793 \times 10^{-21} \sigma^2 \Omega \quad . \quad (4.69)$$

Inserting (4.69) into (4.66), we have

$$\lambda = 4.576 \times 10^{-4} \frac{T}{\sigma^2 \Omega P} \quad , \quad (4.70)$$

where the units of the variables are λ [m], T [K], σ [Å], P [Pa].

Going back to the equation of aerosol number density (4.63), the

number density N_p does not depend so much on the choice of initial number density N_{p0} as long as N_{p0} is sufficiently large (initial size $\bar{d}_{am,0}$ is sufficiently small). In HORN, initial particle size can be either input as time-dependent values, or a small default value of 1×10^{-8} m is used. Then equation (4.62) is used to determine N_{p0} from the initial particle size $\bar{d}_{am,0}$ and initial aerosol mass density G_{p0} which is determined from the result of chemical calculation.

In downstream flow segments, growth of aerosols (decrease of N_p) is calculated by (4.63) using the travelling time from the segment of first aerosol nucleation as t . In each segment, G_p is determined first from the results of chemical calculation. Then, using G_p , N_{p0} and t as known quantities, equations (4.62) and (4.63) are solved simultaneously for N_p and \bar{d}_{am} . It means that an aerosol with mass density G_p in a segment is treated as if it remained there from the beginning with the same mass density G_p and grew from initial size $d_{am,0}$ to d_{am} during time period t .

4.2.5 Aerosol Removal Rates

Because of short residence time, the aerosols formed in the primary cooling circuit will stay sub-micron. Then the removal processes due to inertial motion in turbulent flow are unlikely to make significant contributions to overall removal rate. In HORN, therefore, diffusional deposition due to Brownian motion, thermophoresis, and gravitational settling are considered as removal processes. Diffusiophoresis can be important also for small particles. But since HORN does not treat the case in which condensation of the carrier gas occurs, driving force for diffusiophoresis is small in the cases for analysis.

Removal rates by these processes are first expressed as group velocities towards wall for each particle size class. Then they are averaged over the log-normal distribution weighted by mass:

$$\bar{u}_{t,k} = \frac{\int u_{t,k}(d_p) d_p^3 df(d_p)}{\int d_p^3 df(d_p)}, \quad (4.71)$$

where $df(d_p)$ is the log-normal probability function given by (4.60) and subscript k denotes each of the above three removal mechanisms. The overall group velocity of aerosols to the wall is determined simply as a

sum of the velocities by individual mechanisms:

$$\bar{u}_t = \sum_k^3 \bar{u}_{t,k} \quad , \quad (4.72)$$

and is used in the mass-balance equation (4.41) or (4.44). The size-dependent velocities by individual removal processes are determined as follows:

Diffusional deposition due to Brownian motion

The mean free path of aerosol is smaller than that of gas molecule. Hence the group velocity across the boundary layer to the wall can be consistently calculated by the empirical correlations as used for diffusion of gases. For turbulent flow in a tube, Dittus-Boelter equation (4.46) is used, whereas for natural convection in a tank, the correlation (4.48) is used. Diffusion constant of particles due to Brownian motion is given by⁽²²⁾

$$D_p = \frac{k T C_c}{3\pi \mu d_p} \quad , \quad (4.73)$$

in which D_p is diffusion constant (m^2/s), and Cunningham correction factor C_c is given by (4.65). D_p is used in (4.46) and (4.48) instead of D for gas molecules.

Thermophoresis

The group velocity by thermophoresis is given by Brock⁽²⁴⁾ as

$$u_{t,th} = - \frac{3 \mu C_c H}{2 \rho_g T} \frac{\partial T}{\partial r} \quad , \quad (4.74)$$

in which H is molecular accommodation coefficient and is given by

$$H = \left(\frac{1}{1 + 6 \lambda/d_p} \right) \left(\frac{\kappa_g / \kappa_p + 4.4 \lambda/d_p}{1 + 2\kappa_g / \kappa_p + 8.8 \lambda/d_p} \right) \quad , \quad (4.75)$$

where κ_g and κ_p are thermal conductivities of gas and particle, respectively ($W/m K$), and λ is mean free path of gas molecule determined as (4.70).

Thermal conductivity of particle κ_p in fact depends on the composition of

particle. However, H itself is a correction factor particularly when d_p is small, so that κ_p is assumed to be 7 W/m K independently of composition (a value typical of NaCl structure and is intermediate between the thermal conductivities of metal and oxides). In fact, even when particle size is 1 μm , difference of κ_p/κ_g between 10 and 1000 does not produce significant difference in H value.

Gravitational Settling

Terminal settling velocity of a particle against Stokes viscous force is given by⁽²²⁾

$$u_{TS} = \frac{\rho_p d_p^2 g}{18 \mu} \quad , \quad (4.76)$$

where g is acceleration due to gravity (9.8 m/s²). This terminal velocity downward is expressed as a 'group velocity to the wall' as

$$u_{t,gr} = u_{TS} \frac{S_p}{S} \quad (4.77)$$

where S and S_p are total surface area and the downward projected area (m²) of the flow segment, respectively. It is to handle the gravitational settling process in the same constitution equation as for other removal processes.

To show how these three removal mechanisms contribute to the total aerosol removal rate, group velocities to the wall by the three mechanisms were calculated for a flow of 1 atmosphere steam in 0.1 m dia. tube. Figure 10 shows the velocities as a function of particle size. Dependencies on Reynolds number (diffusional deposition) and temperature gradient across the boundary layer (thermophoresis) are also shown for the ranges realistic to the accident conditions. It is observed that gravitational settling contributes little to the removal of the aerosols in the cooling circuit tubes which are expected to be submicron.

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Table 1 Temperature coefficients of materials property
 correlations for carrier gases
 ($\ln Y = a + b \ln T + c (\ln T)^2$)

Property	Species	Coefficients			Reference
		a	b	c	
viscosity (kg/m·s)	H ₂ O	- 21.476	2.209	-0.0827	14
	H ₂	-15.458	0.672	0	15
	O ₂	-14.613	0.676	0	15
	Kr	-19.521	2.145	-0.104	15
	Xe	-19.521	2.145	-0.104	15
thermal conductivity (W/m·K)	H ₂ O	-16.128	2.760	-0.112	15
	H ₂	- 4.431	0.282	-0.0344	15
	O ₂	- 8.281	0.816	0	15
	Kr	-12.316	1.781	-0.0767	15
	Xe	-13.128	1.792	-0.0700	15
Specific Heat (J/mole·K)	H ₂ O	7.445	-1.378	0.1214	14
	H ₂	6.773	-1.092	0.0877	16
	O ₂	2.601	0.136	0	8
	Kr	3.035	0	0	($C_p = \frac{5}{2} R$)
	Xe	3.035	0	0	

Table 2 Application of empirical correlations for Lennard-Jones molecular force constants to some polar gases

compound	condensed-phase data			calculated parameters		parameters from literature		
	*	T(K)	V_m (cc/mole)	σ (Å)	ϵ/k	σ (Å)	ϵ/k	ref.
HCl	B	159	29.9	3.63	183	3.305	360	20
HI	B	238	45.7	4.18	274	4.123	324	20
HgI ₂	B	622	72.3	4.87	715	5.625	698	20
SnCl ₄	B	387	116.9	5.72	445	4.540	1550	20
Hg	B	630	15.4	2.91	725	2.898	851	20
	M	234	14.1	2.95	449			
NH ₃	B	240	25.1	3.43	276	3.15	358	25
SO ₂	B	263	43.8	4.12	302	4.04	347	25
H ₂ S	B	213	35.4	3.84	245	3.49	343	25

* When the symbol in this column is B, T is boiling temperature and V_m is molecular volume of liquid, and when it is M, T is melting temperature and V_m is molecular volume of solid.

Table 3 Diffusion constants of some gases in 1 atm. steam calculated by the Chapman-Enskog model

gas	force constants		diffusion constant (m ² /s)		
	σ (Å)	ϵ/k (K)	500 K	1000 K	1500 K
H ₂	2.92	38	2.2×10^{-4}	7.4×10^{-4}	1.5×10^{-3}
Kr	3.50	225	4.6×10^{-5}	1.7×10^{-4}	3.5×10^{-4}
HI	4.12	324	3.3×10^{-5}	1.3×10^{-4}	2.6×10^{-4}
CsOH	4.08	1050	2.5×10^{-5}	1.0×10^{-4}	2.2×10^{-4}
Te	3.34	1390	2.9×10^{-5}	1.2×10^{-4}	2.7×10^{-4}
CsI	4.71	1790	1.8×10^{-5}	7.1×10^{-5}	1.6×10^{-4}
particles d = 0.01 μ m 0.1 μ m			2.6×10^{-7} 2.9×10^{-9}	6.6×10^{-7} 6.9×10^{-9}	1.2×10^{-6} 1.2×10^{-8}

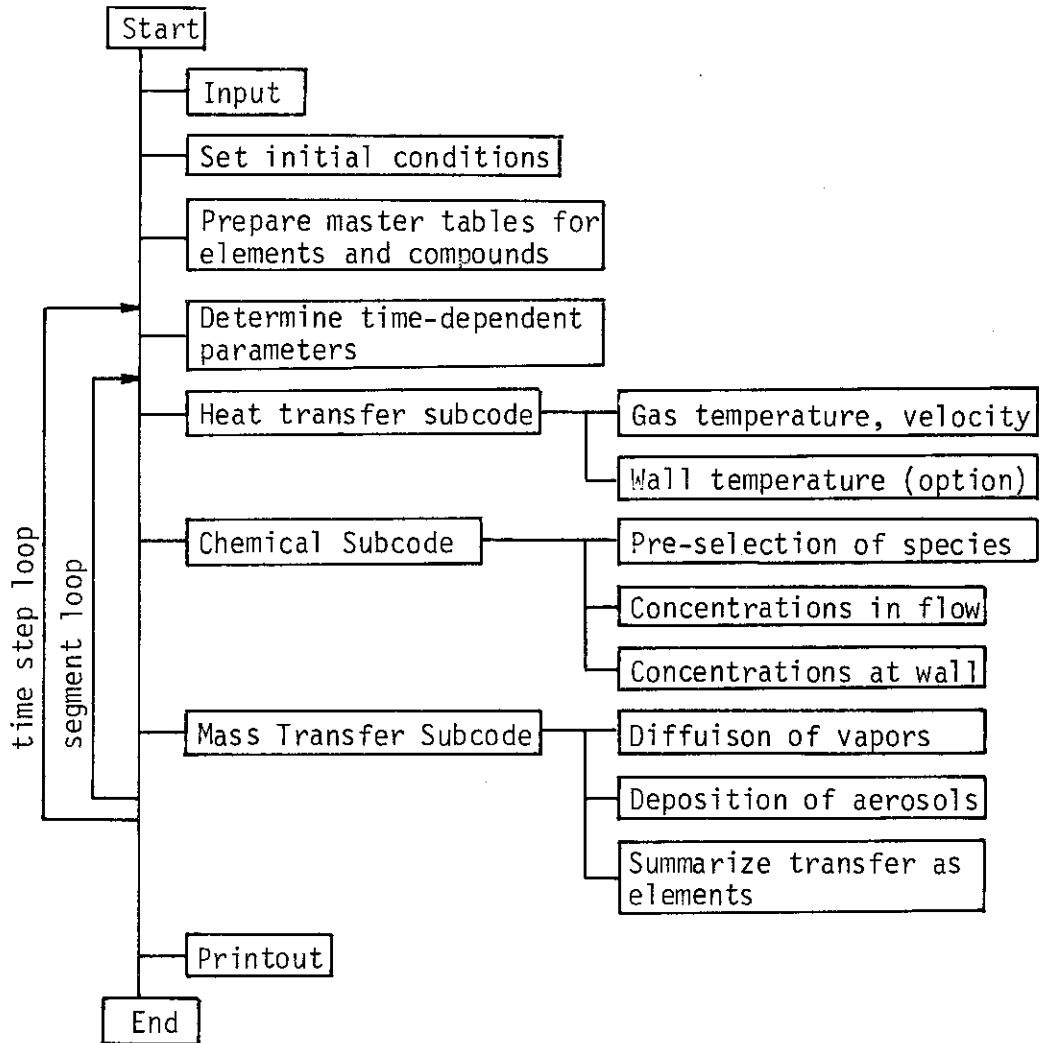
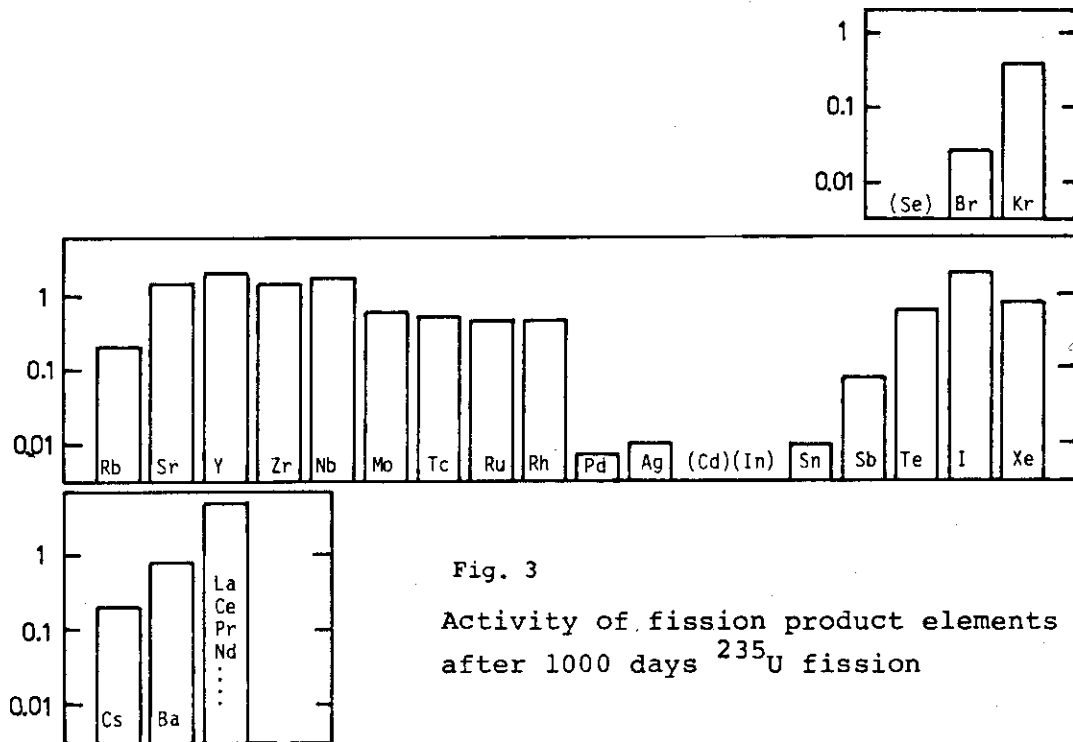
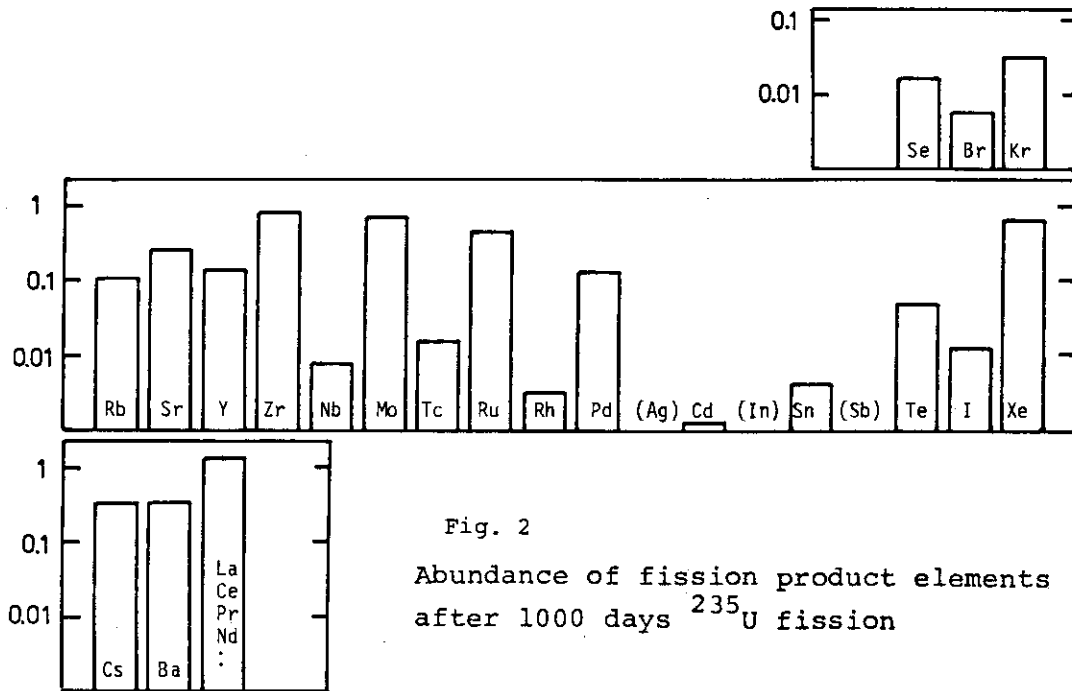


Fig. 1 Flow of calculation in HORN



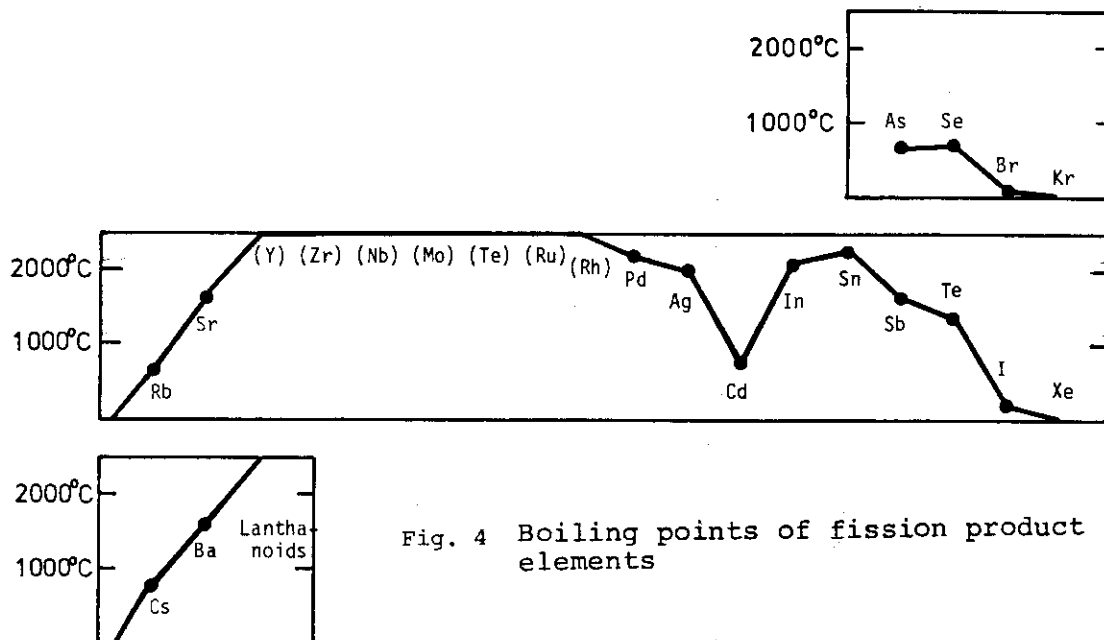


Fig. 4 Boiling points of fission product elements

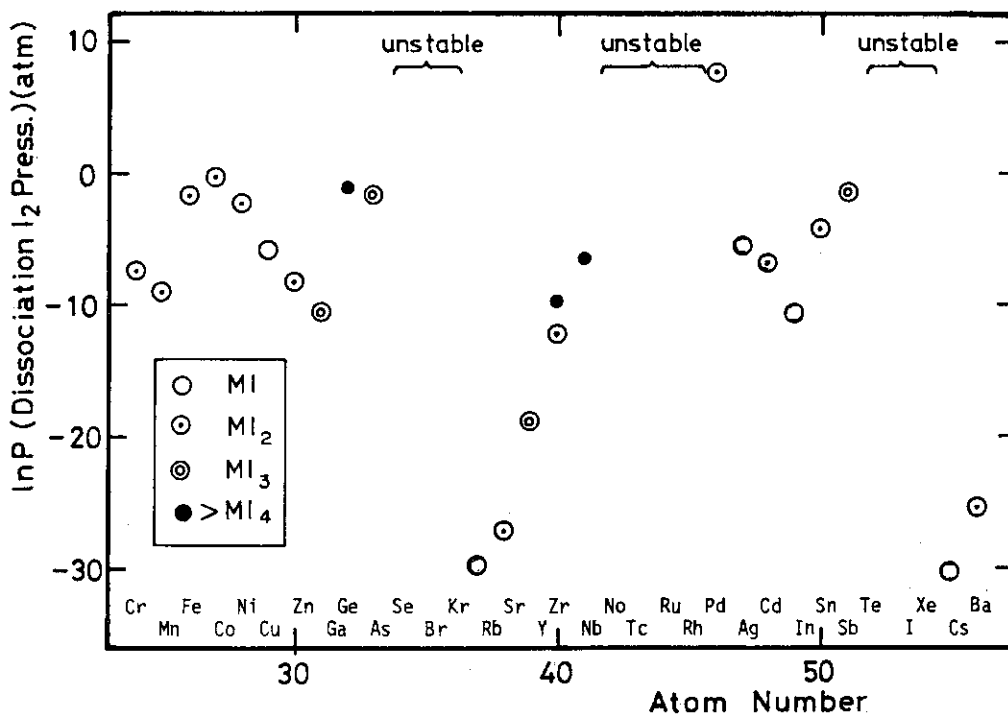


Fig. 5 Stability of iodides of elements in terms of dissociation pressure at 1000 K

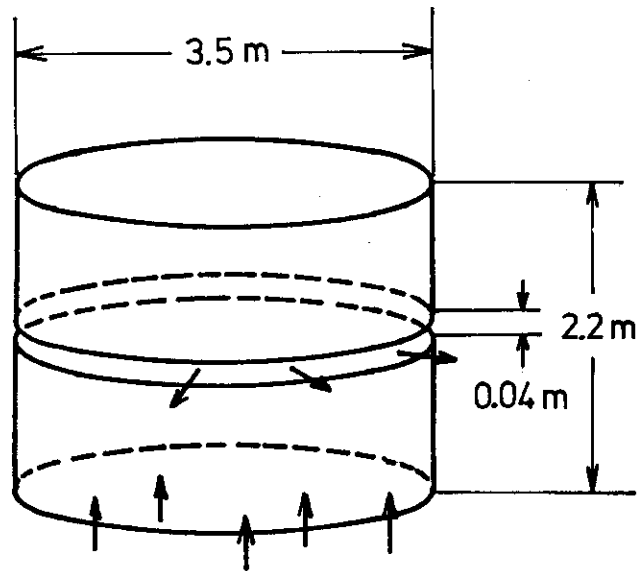


Fig. 6 Dimensions of model axisymmetric tank

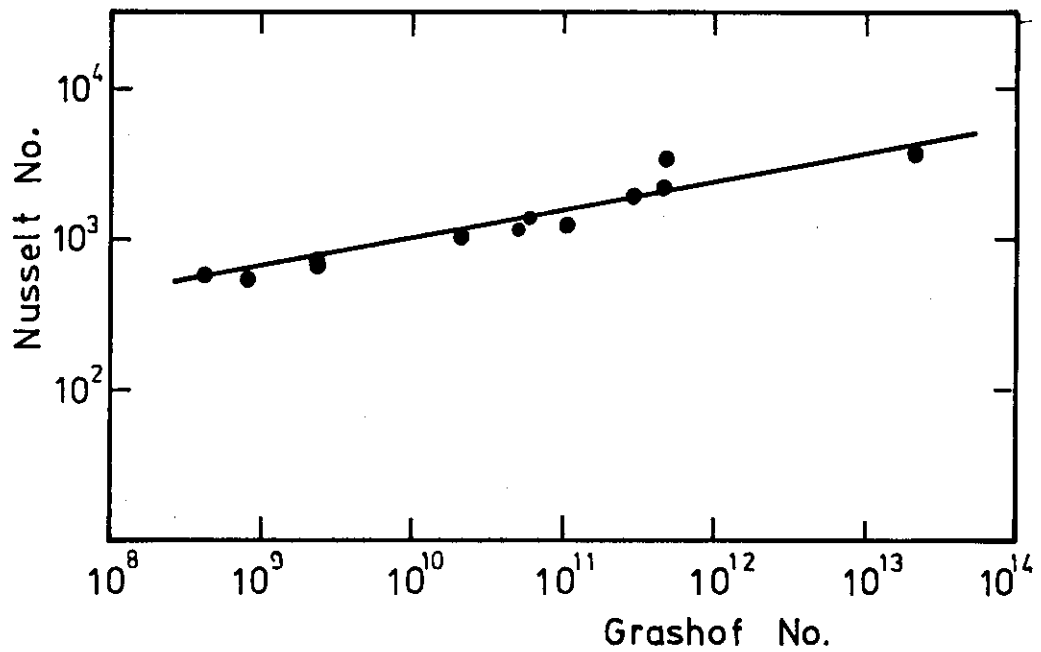


Fig. 7 Natural convection (plus radiative) heat transfer in a cylindrical tank calculated by PHOENICS code

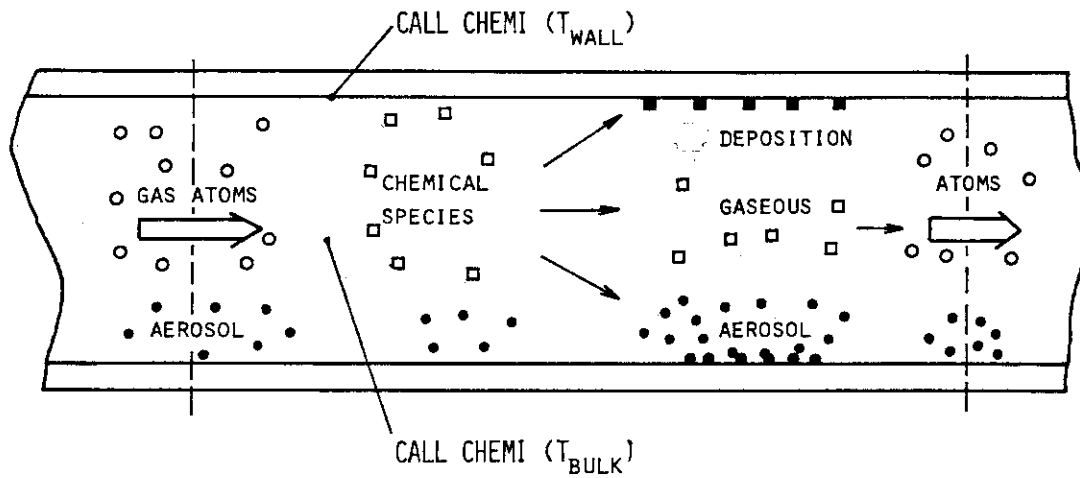
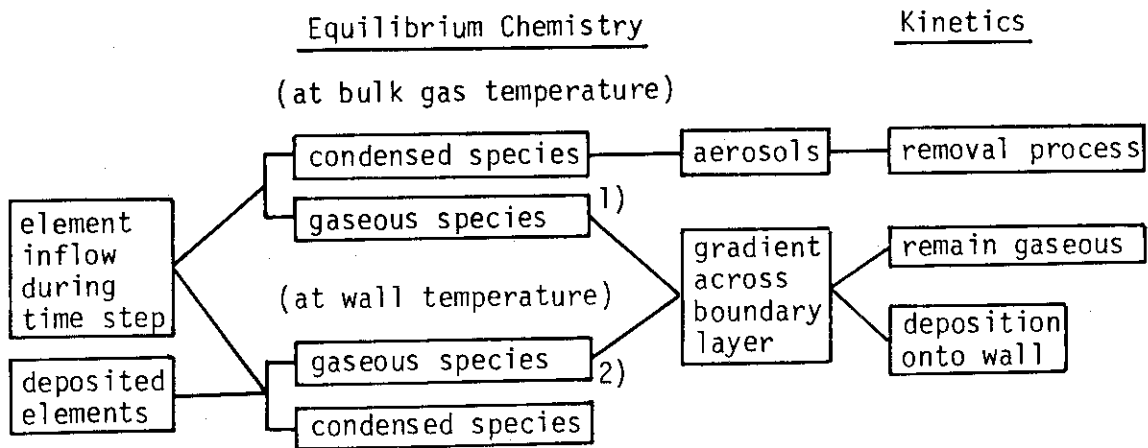


Fig. 8 HORN mass transfer model



* If 2) is greater than 1), reevaporation is automatically calculated.

Fig. 9 Algorithm of using the equilibrium chemical compositions for kinetic calculations

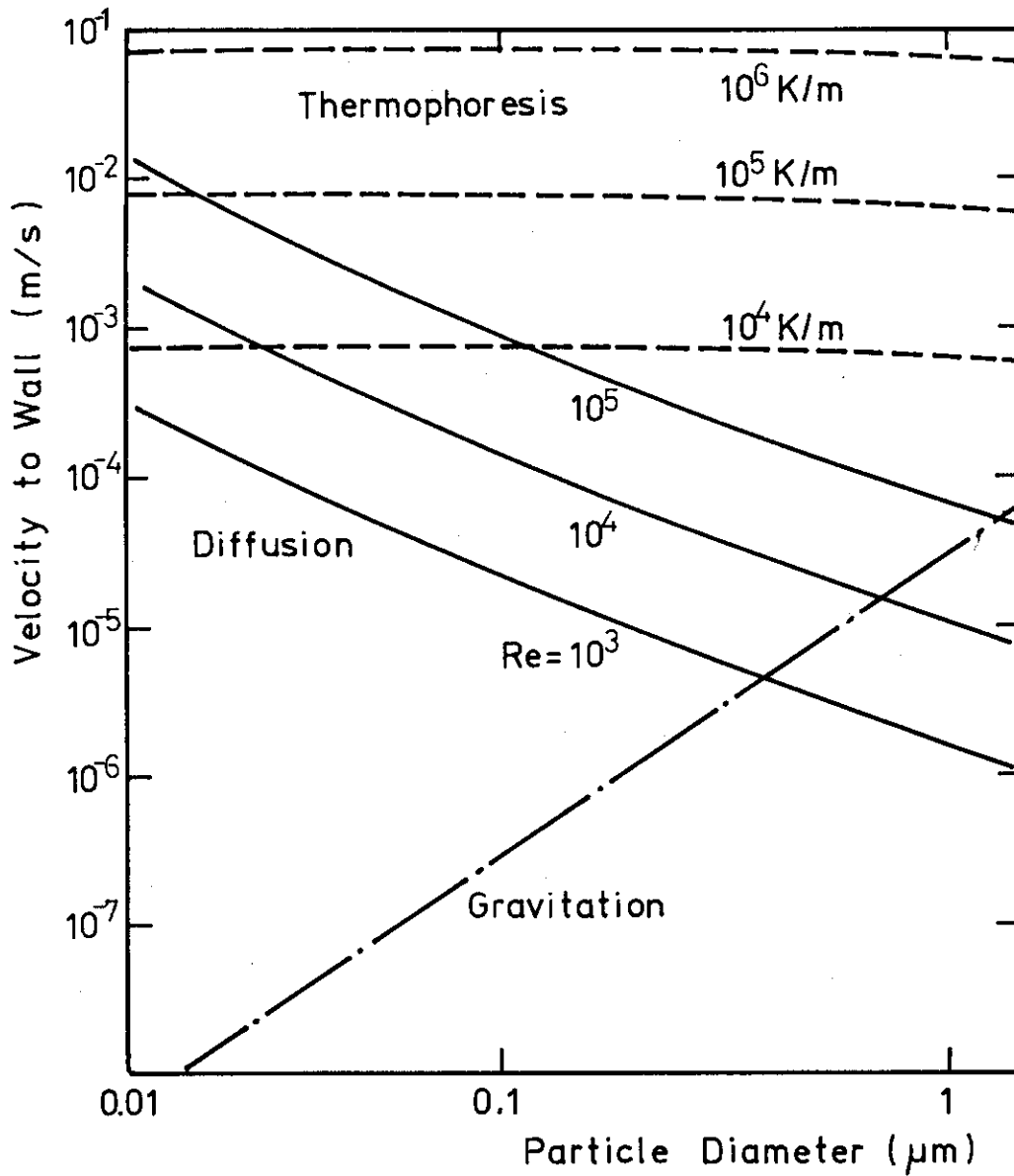


Fig. 10 Group velocities of submicron particles to the wall by various deposition mechanisms (gas flow in a 0.1 m dia. tube is assumed.)

Appendix A Input Manual

(1) Input Format

Input data of HORN are given by namelist. For a run, input data should be arranged in the following order:

Title Card (70A1)

Namelist /INPl/

The description of /INPl/ is given in Table A1.

(2) Format of Time-Dependent Data

In namelist /INPl/, many arrays describing time-dependent data such as TVPRS, TVFP appear. These time-dependent parameters can be specified by the values at arbitrary number of periods. First, the number of such periods is specified by an integer as NPRS, NTVFP, or NTVDT. If such an integer is unity, the parameter corresponding to it is regarded as constant, and the only one input data is used throughout the transient. If such data are given at multiple periods, the value at arbitrary time is derived by linear interpolation from the two input values at just before and after the time. As an example, the case of TVTS, inlet temperature, with three input periods is shown in Fig. A1.

Some data such as wall temperature or pressure are functions of two independent variable, i.e. space and time. Such data are also input through two-dimensional arrays, and the first argument is used both for specifying time and location. In the case of gas pressure, for example, array TVPRS is used as follows:

TVPRS(1,K) = time (s) of the k-th input period,
 TVPRS(2,K) = pressure in the first segment at k-th period,
 TVPRS(j+1,K) = pressure in the j-th segment at k-th period.

Thus, the data in the j -th segment is shifted into the $(j+1)$ -th component because the first component is used for specifying the input time.

Similarly, TVFP describing the flow rate of fission product is a two-dimensional array of element number and time. TVFP(1,K) is used for describing the input time and the flow rate data for i -th element is written in TVFP($i+1$, K).

(3) Element Number

For describing flow rates of elements, the following indices are used for the elements considered in HORN:

- | | | | | |
|--|--------|------------------|-------|--------|
| 1. H | 2. O | 3. Kr | 4. Xe | 5. Cs |
| 6. I | 7. Te | 8. B | 9. Ag | 10. Cd |
| 11. Sb | 12. Ba | 13.-- 19. (void) | | |
| 20. Fi (imaginary element representing low-volatility elements). | | | | |

(4) Maximum Size of Input Data

In the present version of HORN, the maximum sizes of input data are limited as follows:

- | | |
|--|-----|
| - number of flow segments | 50, |
| - number of printout times | 50, |
| - number of periods at which
a single time-dependent
variable is specified | 50. |

(5) Model Input Data

A model input data set is shown in Table A2.

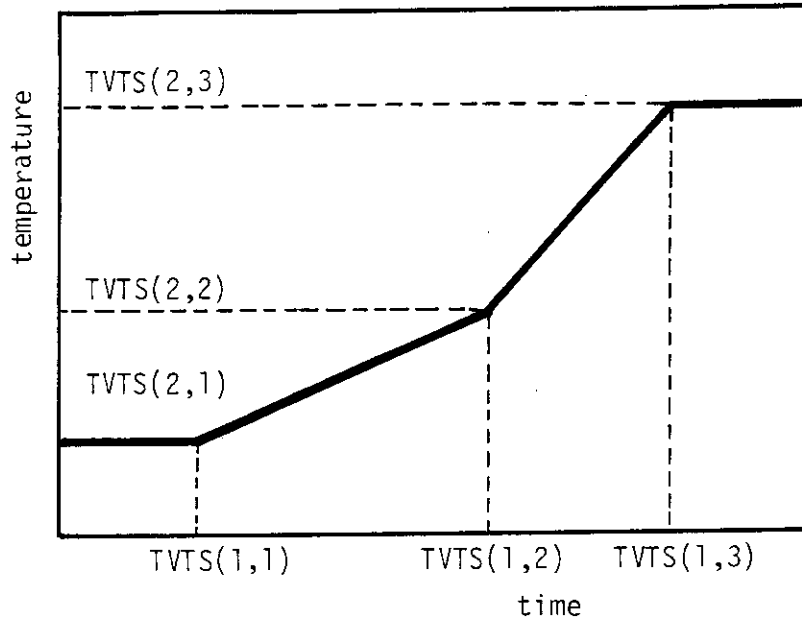


Fig. A1 Linear interpolation of time-dependent input data (case of inlet temperature with three input periods)

Table A1 Namelist /INP1 /

Variable Name	Description	Default Value
NSEG	number of flow segments	
MSEGF(I) (I=1,NSEG)	classification of the segments (= 0: tube, = 1: tank)	50*0
DDE(I) (I=1,NSEG)	inside diameter of tube or tank (m)	50*0.0
DSEGL(I) (I=1,NSEG)	length of tube segment or tank height (m)	50*0.0
DFAREA(I) (I=1,NSEG)	flow area of flow segment; when DFAREA equals zero, flow area is calculated from DDE. (m ²)	50*0.0
LEVEL(I) (I=1,NSEG)	orientation of tube segment or tank (= 0: vertical, = 1: horizontal)	50*1
SEMI(I) (I=1,NSEG)	emissivity of tube or tank wall	50*0.9
TIMEØ	initial time of transient (s)	0.0
TMAX	end time of transient (s). HORN calcu- lation is made between TIMEØ and TMAX.	0.0
NTVDT	size of time step size table TVDT	1
TVDT(J,K) (J=1,2, K=1,NTVDT)	time step size table TVDT(1,K) = time (s) TVDT(2,K) = time step size (s)	
NTVQS	size of flow rate table	1
TVQS(J,K) (J=1,2, K=1,NTVQS)	flow rate table for mixed gas (steam, H ₂ and O ₂) TVQS(1,K) = time (s) TVQS(2,K) = flow rate (mole/s) (Unnecessary when flow rates of H and O are input through TVFP data.)	
NTVTS	size of inlet temperature table TVTS	

Table A1 Namelist /INP1 / (continued)

Variable Name	Description	Default Value
TVTS(J,K) (J=1,2, K=1,NTVTS)	inlet gas temperature table TVTS(1,K) = time (s) TVTS(2,K) = temperature (K)	
NTVTT	size of wall temperature table	1
TVTT(J,K) (J=1,NSEG+1, K=1,NTVTT)	wall temperature table TVTT(1,K) = time (s) TVTT(2,K) = TVTT(NSEG+1, K) = wall temperature (K) TVTT data are required for all the flow segments and selected periods NTVTT.	
NPRS	size of gas pressure table	1
TVPRS(J,K) (J=1,NSEG+1, K=1,NPRS)	gas pressure table TVPRS(1,K) = time (s) TVPRS(2,K) = TVPRS(NSEG+1, K) = gas pressure (N/m ²)	
NHYDR	size of hydrogen fraction table	1
HYDFR(J,K) (J=1,2, K=1,NHYDR)	hydrogen (or oxygen) fraction table HYDFR(1,K) = time (s) HYDFR(2,K) = H ₂ /(H ₂ + H ₂ O) mole ratio When HYDFR(2,K) is negative, HYDFR(2,K) gives fraction of O ₂ . Flow rates of elements H and O can be specified, as fission products, also by TVFP table. In that case, TVQS and HYDFR data are unnecessary. Combination of TVQS and HYDFR data is used only when the TVFP data for H and O are both zero.	
NTVFP	size of element flow rate table	1
TVFP(J,K) (J=1,21, K=1,NTVFP)	element flow rate table TVFP(1,K) = time (s) TVFP(2,K) = H flow rate (g-atom/s) (3,K) = 0 (4,K) = Kr	

Table A1 Namelist /INPl / (continued)

Variable Name	Description	Default Value
TVFP(J,K) (continued)	TVFP(5,K) = Xe flow rate (g-atom/s) (6,K) = Cs (7,K) = I (8,K) = Te (9,K) = B (10,K) = Ag (11,K) = Cd (12,K) = Sb (13,K) = Ba (21,K) = Fi(imaginary element) If TVFP(2,K) and TVFP(3,K) are both zero, flow rates of H and O are calculated from TVQS and HYDFR.	
NTVDIM	size of aerosol initial size table	0
TVDIMØ(J,K) (J=1,2, K=1,NTVDIM)	initial size (AMD) of aerosol at inlet TVDIMØ(1,K) = time (s) TVDIMØ(2,K) = aerosol diameter (m) This table is used when some aerosols are considered to exist already at inlet. If NTVDIM=0, aerosol size is calculated from the concentration of condensed species.	
SPGRV	density of aerosol particles (kg/m^3)	2000.
STDEV	logarithmic standard deviation of aerosol size distribution	1.5
OVSAT	supersaturation for aerosol nucleation	1.0
FCONF	fraction of the length of average radiation flux to diameter of flow segment	0.94
IOPTI	option flag for tube heatup calculation (= 0: wall temperature is input, = 1: wall temperature is calculated from heat balance.)	0
DSTEN(I) (I=1, NSEG)	thickness of wall (m) (Unnecessary when IOPTI = 0)	1.0×10^{50}

Table A1 Namelist /INP1 / (continued)

Variable Name	Description	Default Value
HEX(I) (I=1, NSEG)	heat transfer coefficient at the tube outer surface ($W/m^2 K$) (Unnecessary when IOPTI = 0)	5.024×10^2
CPSTEN(I) (I=1, NSEG)	specific heat of tube wall (J/kg K) (Unnecessary when IOPTI = 0)	502.44
ROSTEN(I) (I=1, NSEG)	density of tube wall (kg/m^3) (Unnecessary when IOPTI = 0)	8.0×10^3
WKSTEN(I) (I=1, NSEG)	thermal conductivity of tube wall (W/m K) (Unnecessary when IOPTI = 0)	20.935
NTVEX	size of decay heat table	1
TVEX(J,K) (J=1, 21, K=1, NTVEX)	elementwise decay heat table (Unnecessary when IOPTI = 0) TVEX(1,K) = time (s) TVEX(6,K) = decay heat of Cs (W/g-atom) TVEX(7,K) = I TVEX(13,K) = Ba TVEX(21,K) = Fi (imaginary element)	
NTPRIN	number of printout periods (max. 50) (Initial and end states are printed out even when NTPRIN = 0.)	2
TPRINT(J) (J=1, NTPRIN)	periods for printout (s)	0.0, $49 \times 1.0 \times 10^{10}$
NSEPRI(K) (K=1, NSEG)	flow segment numbers for which calculation results should be printed out (ex.) NSEG=10, NSEPRI=1,2,3,5,10, (Printout is made at five points.) If NSEPRI is not input, printout is made only at the first and last segment.	50*0

Table A1 Namelist /INPl / (continued)

Variable Name	Description	Default Value
NSPPRI(J) (J=1,160)	indices of chemical species to be edited Standard output of fission product behavior is made on each element, not on individual chemical species. However, for species specified by NSPPRI, release or deposition fractions as chemical species (such as CsI, TeO) are also output. (Ex.) NSPPRI=16, 23, 55, 57, For species indices, see Table B1.	160*0
IDEBUG(I) (I=1,30)	subroutinewise flag for debug print IDEBUG(I) = 0: no print except for standard output, IDEBUG(I) = 1: printout of detailed information, IDEBUG(I) > 1: debug print	3*0, 1, 26*0,
IDBOUT(I) (I=1,NSEG)	flow segmentwise flag for debug print Used in combination with IDEBUG. IDBOUT specifies, when IDEBUG \geq 1, the flow segments whose debug informations are to be printed out. IDBOUT(I) = 0: no print IDBOUT(I) = 1: print.	50*0

Table A2 Model input data

CARD NO.	INPUT DATA LIST						
	1	2	3	4	5	6	7
1.	5	0	5	0	5	0	5
2.	0	5	0	5	0	5	0
3.	5	0	5	0	5	0	5
4.	0	5	0	5	0	5	0
5.	5	0	5	0	5	0	5
6.	0	5	0	5	0	5	0
7.	5	0	5	0	5	0	5
8.	0	5	0	5	0	5	0
9.	5	0	5	0	5	0	5
10.	0	5	0	5	0	5	0
11.	5	0	5	0	5	0	5
12.	0	5	0	5	0	5	0
13.	5	0	5	0	5	0	5
14.	0	5	0	5	0	5	0
15.	5	0	5	0	5	0	5
16.	0	5	0	5	0	5	0
17.	5	0	5	0	5	0	5
18.	0	5	0	5	0	5	0
19.	5	0	5	0	5	0	5
20.	0	5	0	5	0	5	0
21.	5	0	5	0	5	0	5
22.	0	5	0	5	0	5	0
23.	5	0	5	0	5	0	5
24.	0	5	0	5	0	5	0
25.	5	0	5	0	5	0	5
26.	0	5	0	5	0	5	0
27.	5	0	5	0	5	0	5
28.	0	5	0	5	0	5	0
29.	5	0	5	0	5	0	5
30.	0	5	0	5	0	5	0

Appendix B Molecular Data

The thermodynamic properties of the species considered in HORN and molecular force constants of gaseous species are stored in the BLOCKSATA as a kind of library. A data set on a species consists of the atom numbers of the constituent elements, stoichiometry coefficients for them, molecular weight, phase, force constants, and the temperature coefficients of Gibbs free energy. The order of storage is random. Therefore new species can be easily added. The code judges which species are necessary from the combination of flowing elements according to the stored atom numbers, picks them up, and sorts them in the order of gaseous, liquid and solid species.

Table B1 shows the species before such sorting together with their force constants and temperature coefficients of free energy.

Table B1 Chemical species considered in HORN and their properties

(SIGMA and EPS/K are molecular force constants σ (A) and ϵ/k (K), respectively. A,B,C are temperature coefficients of Gibbs free energy (unit in J/mole and K). Species not considered in the present version are also included in this table.)

NO	SPECIES	STATE	SIGMA	EPS/K	A	B	C
1	AG(S)	S			1.97E+04	-6.59E+01	0.0
2	AG(L)	L			6.54E+04	-1.03E+02	0.0
3	AG	G	2.63	2590.00	3.32E+05	-2.12E+02	0.0
4	AGI(S)	S			-1.81E+04	-1.62E+02	0.0
5	AGI(L)	L			3.51E+04	-2.26E+02	0.0
6	AGI	G	4.14	1590.00	2.08E+05	-3.23E+02	0.0
7	AG2O(S)	S			3.14E+04	-1.42E+02	0.0
8	AG2TE(S)	S			1.21E+04	-1.33E+02	-6.48E-02
9	BA(S)	S			2.73E+04	-9.14E+01	0.0
10	BA(L)	L			5.57E+04	-1.41E+02	0.0
11	BA	G	4.15	2080.00	2.40E+05	-2.38E+02	0.0
12	BAI2(S)	S			-5.35E+05	-2.24E+02	0.0
13	BAI2(L)	L			-5.31E+05	-1.74E+02	-5.41E-02
14	BAI2	G	5.17	1940.00	-2.45E+05	-3.56E+02	-2.85E-02
15	BAO(S)	S			-4.97E+05	-7.15E+01	-2.90E-02
16	BAO(L)	L			-4.40E+05	-9.58E+01	-2.91E-02
17	BAO	G	3.60	4220.00	-7.20E+04	-2.39E+02	-1.80E-02
18	BA(OH)2S	S			-8.09E+05	-1.72E+02	0.0
19	BA(OH)2L	L			-8.07E+05	-1.32E+02	-6.86E-02
20	BA(OH)2	G	4.11	1148.00	-5.02E+05	-3.22E+02	-4.52E-02
21	BATE(S)	S			-2.33E+05	-9.56E+01	-2.91E-02
22	CD(S)	S			1.84E+04	-6.18E+01	0.0
23	CD(L)	L			3.54E+04	-9.03E+01	0.0
24	CD	G	2.87	1195.00	1.51E+05	-2.01E+02	0.0
25	CDI2(S)	S			-1.43E+05	-1.97E+02	0.0
26	CDI2(L)	L			-8.46E+04	-2.85E+02	0.0
27	CDO(S)	S			-2.12E+05	-5.00E+01	-3.09E-02
28	CDTE(S)	S			-7.14E+04	-8.20E+01	-4.06E-02
29	CS(S)	S			2.24E+04	-7.50E+01	0.0
30	CS(L)	L			3.41E+04	-1.14E+02	0.0
31	CS	G	5.06	1188.00	1.05E+05	-1.79E+02	-1.02E-02
32	CS2	G	5.46	2067.00	1.58E+05	-2.80E+02	-2.43E-02
33	CSH	G	4.15	1300.00	1.84E+05	-2.81E+02	-2.67E-02
34	CSI(S)	S			-3.07E+05	-1.14E+02	-3.73E-02
35	CSI(L)	L			-2.85E+05	-1.42E+02	-3.52E-02
36	CSI	G	4.71	1786.00	-1.12E+05	-2.77E+02	-1.97E-02
37	(CSI)2	G	5.09	3108.00	-3.70E+05	-4.34E+02	-4.26E-02
38	CSOH(S)	S			-3.35E+05	-1.21E+02	0.0
39	CSOH(L)	L			-3.21E+05	-1.30E+02	-4.00E-02
40	CSOH	G	4.08	1046.00	-1.79E+05	-2.60E+02	-2.69E-02
41	(CSOH)2	G	4.41	1820.00	-5.29E+05	-3.63E+02	-5.36E-02
42	CS2O(S)	S			-2.23E+05	-1.72E+02	0.0
43	CS2O	G	4.90	1465.00	-7.28E+03	-3.21E+02	-2.96E-02
44	CS2O3(S)	S			-3.99E+05	-3.12E+02	0.0
45	CSO2(S)	S			-2.19E+05	-1.88E+02	0.0
46	CS2TE(S)	S			-2.21E+05	-1.69E+02	0.0
47	CS2TEO3L	S			-7.87E+05	-2.28E+02	-2.50E-02
48	FE(S)	S			8.00E+03	-2.03E+01	-2.19E-02
49	FEI2(S)	S			-4.60E+04	-2.21E+02	0.0
50	FEI2(L)	L			5.94E+04	-3.43E+02	0.0

Table B1 Chemical species considered in HORN and their properties

(continued)

<u>NO</u>	<u>SPECIES</u>	<u>STATE</u>	<u>SIGMA</u>	<u>EPS/K</u>	<u>A</u>	<u>B</u>	<u>C</u>
51	FEI2	G	4.73	864.00	1.34E+05	-3.52E+02	-3.23E-02
52	(FEI2)2	G	5.11	1503.00	1.02E+05	-5.50E+02	-6.86E-02
53	FE0(S)	S			-2.32E+05	-5.72E+01	-3.24E-02
54	FE304(S)	S			-9.65E+05	-1.37E+02	-1.11E-01
55	FE203(S)	S			-7.14E+05	-7.53E+01	-8.08E-02
56	FE(OH)2S	S			-4.43E+05	-1.65E+02	0.0
57	FE(OH)3S	S			-6.47E+05	-1.96E+02	0.0
58	FETEO.9	S			1.17E+04	-1.25E+02	0.0
59	FETE2(S)	S			-1.87E+04	-1.54E+02	0.0
60	H2	G	2.92	38.00	4.12E+04	-1.34E+02	-1.50E-02
61	HI	G	4.12	324.00	6.47E+04	-2.07E+02	-1.65E-02
62	H2O	G	2.47	776.00	-1.69E+05	-1.86E+02	-2.15E-02
63	H2TE	G	4.30	520.00	1.53E+05	-2.21E+02	-2.74E-02
64	I2(S)	S			3.68E+04	-1.32E+02	0.0
65	I2	G	4.98	550.00	1.02E+05	-2.66E+02	-1.86E-02
66	I	G	4.61	316.00	1.26E+05	-1.82E+02	-1.07E-02
67	KR	G	3.50	225.00	0.0	0.0	0.0
68	O2	G	3.43	113.00	6.34E+04	-2.08E+02	-1.67E-02
69	SB(S)	S			1.87E+04	-6.28E+01	0.0
70	SB(L)	L			4.20E+04	-7.81E+01	-1.16E-02
71	SB2	G	3.47	3020.00	2.61E+05	-2.57E+02	-1.91E-02
72	SB4	G	3.75	5260.00	2.66E+05	-3.55E+02	-4.24E-02
73	SB13(S)	S			-2.43E+04	-2.37E+02	0.0
74	SB13(L)	L			2.14E+04	-3.40E+02	0.0
75	SB13	G	5.70	509.00	1.13E+05	-4.76E+02	0.0
76	SB203(S)	S			-5.72E+05	-2.17E+02	0.0
77	SB203(L)	L			-4.02E+05	-4.00E+02	0.0
78	SB205(S)	S			-8.16E+05	-1.66E+02	0.0
79	SBO2(S)	S			-3.63E+05	-1.19E+02	0.0
80	SB2TE3-S	S			4.14E+04	-3.33E+02	0.0
81	SB2TE3-L	L			2.11E+05	-5.23E+02	0.0
82	TE (S)	S			1.91E+04	-6.39E+01	0.0
83	TE (L)	L			5.27E+04	-1.10E+02	0.0
84	TE	G	3.34	1392.00	2.15E+05	-1.88E+02	-9.51E-03
85	TE2	G	3.61	2422.00	1.90E+05	-2.56E+02	-2.34E-02
86	TEO	G	3.58	500.00	1.21E+05	-2.37E+02	-2.08E-02
87	(TEO)2	G	3.87	870.00	-1.67E+04	-3.18E+02	-4.92E-02
88	TEO2(S)	S			-2.32E+05	-1.30E+02	0.0
89	TEO2(L)	L			-2.03E+05	-1.59E+02	0.0
90	TEO2	G	3.67	1930.00	2.30E+04	-2.92E+02	-1.67E-02
91	XE	G	4.06	299.00	0.0	0.0	0.0
92	BAB204-S	S			-1.89E+06	-6.87E+01	-9.94E-02
93	B4C(S)	S			-5.97E+04	-2.30E+01	-4.93E-02
94	BI3	G	5.50	1510.00	1.29E+05	-3.50E+02	-4.13E-02
95	B203(S)	S			-1.16E+06	-9.35E+01	0.0
96	C (S)	S			1.57E+03	-2.15E+00	-1.04E-02
97	CO	G	3.59	110.00	-7.67E+04	-1.98E+02	-1.66E-02
98	CO2	G	4.00	190.00	-3.29E+05	-2.11E+02	-2.64E-02
99	CS2CO3	S			-1.01E+06	-1.44E+02	-1.26E-01
100	FECO3(S)	S			-6.23E+05	-1.49E+02	0.0

Table B1 Chemical species considered in HORN and their properties

(continued)

<u>NO</u>	<u>SPECIES</u>	<u>STATE</u>	<u>SIGMA</u>	<u>EPS/K</u>	<u>A</u>	<u>B</u>	<u>C</u>
101	IN(S)	S			1.88E+04	-6.30E+01	0.0
102	IN(L)	L			2.43E+04	-6.99E+01	-1.39E-02
103	INI(S)	S			-7.50E+04	-1.53E+02	0.0
104	INI(L)	L			-3.20E+04	-2.20E+02	0.0
105	INI	G	4.32	674.00	8.34E+04	-3.37E+02	0.0
106	IN2O3(S)	S			-9.21E+05	-1.06E+02	-6.56E-02
107	IN2TE(S)	S			-1.96E+04	-1.94E+02	0.0
108	INTE(S)	S			-2.93E+04	-1.42E+02	0.0
109	INTE(L)	L			3.91E+04	-2.12E+02	0.0
110	IN2TE3(S)	S			-8.63E+04	-3.28E+02	0.0
111	IN2TE3L	L			7.53E+04	-4.99E+02	0.0
112	MN(S)	S			9.16E+03	-2.39E+01	-2.30E-02
113	MN(L)	L			9.03E+04	-1.12E+02	0.0
114	MN	G	2.40	2720.00	3.23E+05	-2.12E+02	0.0
115	MNI2(S)	S			-1.83E+05	-2.03E+02	0.0
116	MNI2(L)	L			-8.72E+04	-3.09E+02	0.0
117	MNI2	G	4.83	1750.00	3.18E+04	-4.01E+02	0.0
118	MNBR2	G	4.46	1860.00	-7.49E+04	-4.01E+02	0.0
119	MNO(S)	S			-3.42E+05	-6.29E+01	-2.58E-02
120	MN3O4(S)	S			-1.23E+06	-1.37E+02	-1.03E-01
121	MN2O3(S)	S			-8.47E+05	-8.27E+01	-8.31E-02
122	MNO2(S)	S			-4.39E+05	-8.98E+01	0.0
123	MG(S)	S			1.51E+04	-5.07E+01	0.0
124	MG(L)	L			4.28E+04	-8.07E+01	0.0
125	MG	G	2.94	1590.00	1.84E+05	-1.84E+02	0.0
126	MGCO3	S			-9.78E+05	-1.19E+02	0.0
127	MGI2(S)	S			-3.06E+05	-1.82E+02	0.0
128	MGO(S)	S			-5.59E+05	-2.56E+01	-2.57E-02
129	MG(OH)2S	S			-8.07E+05	-9.11E+01	0.0
130	MGTE(S)	S			-1.84E+05	-7.01E+01	-2.72E-02
131	HBO2(S)	S			-7.16E+05	-6.61E+01	0.0
132	(HBO2)3	G	3.56	2240.00	-2.02E+06	-3.25E+02	-1.19E-01
133	CSBO2(S)	S			-9.35E+05	-9.10E+01	-5.08E-02
134	CSBO2(L)	L			-9.23E+05	-8.41E+01	-6.43E-02
135	(CSBO2)2	G	5.45	1930.00	-1.21E+06	-6.21E+02	-7.68E-02
136	B2O3(L)	L			-1.13E+06	-1.04E+02	-4.88E-02
137	NA2CO3	S			-9.88E+05	-2.04E+02	0.0
138	SN(S)	S			1.77E+04	-5.93E+01	0.0
139	SNI2(S)	S			-8.60E+04	-1.99E+02	0.0
140	SNI2(L)	L			0.0	0.0	0.0
141	SNI2	G	5.04	1140.00	0.0	0.0	0.0
142	SNO(S)	S			-2.41E+05	-4.44E+01	-3.52E-02
143	SNO	G	3.38	1950.00	6.86E+04	-2.33E+02	-1.84E-02
144	SNO2(S)	S			-5.03E+05	-4.60E+01	-4.35E-02
145	SNTE(S)	S			-1.81E+04	-1.41E+02	0.0
146	SNTE(L)	L			7.37E+04	-2.26E+02	0.0
147	SNTE	G	0.0	0.0	2.42E+05	-3.28E+02	0.0