MARTIN MARIETTA

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**Computer Programs for Developing
Source Terms for a UF₆
Dispersion Model to Simulate
Postulated UF₆ Releases
from Buildings**

W. R. Williams

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**Martin Marietta Energy Systems, Inc., Engineering
Process Engineering**

**Computer Programs for Developing Source Terms
for a UF₆ Dispersion Model to Simulate Postulated
UF₆ Releases from Buildings**

W. R. Williams

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MARTIN MARIETTA ENERGY SYSTEMS, INC.
for the
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FOREWORD AND ACKNOWLEDGMENTS

This report is one of a series of reports documenting computer programs used for predicting health effects that might occur as a result of postulated accidental releases of UF_6 . The reports in this series are the following:

R. A. Just and W. R. Williams, *A Computer Program for Simulating the Atmospheric Dispersion of UF_6 and Other Reactive Gases Having Positive, Neutral, or Negative Buoyancy*, K/D-5694, Martin Marietta Energy Systems, Inc., Oak Ridge, Tenn., April 1986.

W. R. Williams, *Computer Programs for Developing Source Terms for a UF_6 Dispersion Model to Simulate Postulated UF_6 Releases from Buildings*, K/D-5695, Martin Marietta Energy Systems, Inc., Oak Ridge, Tenn., April 1986.

D. D. Holt, *A Computer Program for Superpositioning Multiple UF_6 Plume Simulations to Determine Health Effects*, K/D-5696, Martin Marietta Energy Systems, Inc., Oak Ridge, Tenn., April 1986.

R. A. Just, *Report on Toxicological Studies Concerning Exposures to UF_6 and UF_6 Hydrolysis Products*, K/D-5573, Rev. 1, Martin Marietta Energy Systems, Inc., Oak Ridge, Tenn., July 1984.

The four compartment models described in this report were developed by the author for the gaseous diffusion plant safety analysis report (GDP SAR) program. M. V. Helfrich was primarily responsible for utilizing these programs in the actual analysis of postulated accidental releases of UF_6 . She modified the main programs described in this report to completely write input files for the UF_6 atmospheric dispersion (PLUME) code and also implemented modifications to divide the exhaust pathway of the cascade building models between the roof vent and wall louvers. The BATCH program was modified by Helfrich to write PLUME code input for a postulated release of UF_6 resulting from seismic activity at the Paducah GDP. The author modified BATCH to provide input to the PLUME code for an outdoor release of UF_6 from a ruptured cylinder.

ABSTRACT

Calculational methods and computer programs for the analysis of source terms for postulated releases of UF_6 are presented. Required thermophysical properties of UF_6 , HF , and H_2O are described in detail. UF_6 reacts with moisture in the ambient environment to form HF and H_2O . The coexistence of HF and H_2O significantly alters their pure component properties, and HF vapor polymerizes. Transient compartment models for simulating UF_6 releases inside gaseous diffusion plant feed and withdrawl buildings and cascade buildings are also described. The basic compartment model mass and energy balances are supported by simple heat transfer, ventilation system, and deposition models. A model that can simulate either a closed compartment or a steady-state ventilation system is also discussed. The transient compartment models provide input to an atmospheric dispersion model as output.

1. INTRODUCTION AND SUMMARY

A set of compartment models has been developed to simulate the transient behavior occurring as a result of the postulated accidental release of uranium hexafluoride (UF_6) in various process buildings of the gaseous diffusion plant (GDP) complex. This development was undertaken as a part of the safety analysis report (SAR) program. This report documents the physical and thermodynamic properties and mass and energy balance techniques used in developing the compartment models. The models have been developed in modular fashion through the use of subroutines. Therefore, various building configurations can be simulated by the development of main programs, which make extensive use of the available subroutines.

When UF_6 is released from containment into moist air, it will react with H_2O vapor in the air to form hydrogen fluoride (HF) and uranyl fluoride (UO_2F_2). In modeling postulated releases, UF_6 is encountered in solid, liquid, and vapor phases, whereas both HF and H_2O can exist in liquid and vapor phases. Self-association (polymerization) of HF also occurs. Correlations for various physical and thermodynamic properties are presented in Chap. 2, along with information on subroutines in which the correlations have been implemented.

Methods needed for analyzing releases postulated to occur indoors are discussed in Chap. 3. A number of subroutines based on simple models have been developed to simulate heat transfer, flow through ventilation system components, and deposition. A subroutine has also been written to perform mass and energy balances within a compartment given initial compartment conditions and inlet and outlet stream conditions. One main program has been written to model the transient behavior in a feed and withdrawal building, and two main programs have been written to model transient behavior in a cascade building. The two cascade building models were required to simulate the differing ventilation systems characteristic of summer and winter operation. Another main program has been written to model a release into a closed compartment or a steady release of UF_6 into a ventilation system supplied by a constant-volume blower. These main programs are described in Chap. 4. These programs or variations thereof have been utilized to provide input to the UF_6 atmospheric dispersion (PLUME) code (Just and Williams 1986) in the analysis of postulated accidental releases of UF_6 as part of the GDP SAR program.

2. PHYSICAL AND THERMODYNAMIC PROPERTIES MODELS

The modeling of UF_6 releases requires consideration of various physical and thermodynamic properties for UF_6 , HF, H_2O , UO_2F_2 , and air. The moisture in the air reacts with UF_6 to form HF and UO_2F_2 . Section 2.1 presents properties for solid, liquid, and vapor UF_6 . Because of the strong interaction between HF and H_2O , properties for these compounds in both the liquid and vapor phase are discussed together in Sect. 2.2. Properties of air and UO_2F_2 , which occur as a vapor and solid, respectively, are given in Sect. 2.3. Subroutines for the properties discussed in Sects. 2.1, 2.2, and 2.3 are discussed at the end of each section, respectively. In Sect. 2.4, the method and subroutine used for calculating mixture enthalpies are described.

2.1 URANIUM HEXAFLUORIDE

At room temperature uranium hexafluoride (UF_6) is a colorless volatile solid that forms transparent crystals of high refractive index. The crystals sublime without melting under atmospheric pressure; when heated at pressures greater than about 22 psia, they melt to form a clear, colorless, liquid of high density. The sublimation point (at atmospheric pressure) is about 133.8°F. Despite the high molecular weight of UF_6 , the properties of its vapor closely approximate those of an ideal gas. A phase diagram for UF_6 is given in Fig. 1, and some commonly used properties of UF_6 are presented in Table 1. The physical properties' correlations given in this section in engineering units are based on correlations or data reported by DeWitt (1960) unless otherwise referenced.

Sections 2.1.1 through 2.1.5 cover correlations for evaluating vapor pressure, the vapor phase compressibility factor, densities, enthalpies, and entropies, respectively. Section 2.1.6 discusses other useful properties of UF_6 . The heat of reaction of UF_6 with water is discussed in Sect. 2.1.7. Methods for performing flashing and vapor phase expansion calculations are described in Sect. 2.1.8. Subroutines for evaluating the properties of UF_6 , based on correlations described in Sects. 2.1.1 through 2.1.5, are presented in Sect. 2.1.9, along with a subroutine for evaluating the saturation temperature corresponding to a given UF_6 pressure. A subroutine for performing flashing and vapor phase expansion calculations is also presented in Sect. 2.1.9.

2.1.1 Vapor Pressure

Three vapor pressure correlations were recommended by DeWitt (1960, p. 81). The vapor pressure over the solid from 32°F to the triple point is given by

$$\ln P^\circ = 10.4443 + 9.64233 \times 10^{-3} T_F - [3907.41/(T_F + 298.149)] , \quad (1)$$

where

P° = vapor pressure, (psia),

T_F = temperature, °F.

The vapor pressure over the liquid from the triple point to 241°F is given by

$$\ln P^\circ = 12.1600 - [4668.07/(T_F + 367.533)] , \quad (2)$$

Fig. 1. Phase diagram for uranium hexafluoride. The curve separating the vapor region from the condensed regions corresponds to the vapor pressure of UF_6 .

and from 241°F to the critical point by

$$\ln P^\circ = 13.7627 - [6976.11/(T_F + 511.866)] \quad (3)$$

Estimated uncertainties reported by DeWitt (1960) for the three equations, based on a 95% confidence level, are $\pm 0.05\%$, $\pm 0.03\%$, and $\pm 0.3\%$, respectively. Equations 1 and 2 intersect at a temperature of 147.3°F and a pressure of 22.04 psia, which agrees with the reported triple point (DeWitt 1960, p. 89). Equations 2 and 3 do not intersect, but it was observed that Eq. 3 overestimated vapor pressure from 240 to 276°F, whereas Eq. 2 underestimated the reported data of Oliver et al. over the same range (DeWitt, 1960, p. 85). To obtain a continuous function for vapor pressure, a weighted function combining Eqs. 2 and 3 was developed which yielded better agreement with the reported data than either equation individually, as shown in Table 2. The new equation, used in the computer subroutine VPRUF6 for the estimation of vapor pressure between 240 and 276°F is

Table 1. Properties of uranium hexafluoride

Characteristics	Value
Chemical formula	UF ₆
Molecular weight	
²³⁵ UF ₆	349
²³⁸ UF ₆	352
Sublimation point at 14.7 psia	133.8°F
Density of solid at 68°F	318 lb/ft ³
Critical temperature	446°F
Critical pressure	669 psia
Triple point data	
Vapor pressure	22.04 psia
Temperature	147.3°F
Density of solid	303 lb/ft ³
Density of liquid	227 lb/ft ³
Density of vapor	1.25 lb/ft ³
Heat of sublimation	59.5 Btu/lb
Heat of fusion	23.5 Btu/lb
Heat of vaporization	36.0 Btu/lb

Table 2. Comparison of predicted UF₆ vapor pressure, using Eqs. 2, 3, and 4 to the data of Oliver et al.⁵

Temperature (°F)	Vapor pressures (psia)			
	Oliver et al.	Eq. 2	Eq. 3	Eq. 4
240.85	88.888	88.862	89.549	88.878
255.51	106.59	106.44	106.89	106.64
271.74	128.97	128.75	129.04	129.01

$$P^{\circ} = [(276 - T_F)P_2 + (T_F - 240)P_3]/36 \quad , \quad (4)$$

where

P_2 = vapor pressure evaluated at T_F using Eq. 2,

P_3 = vapor pressure evaluated at T_F using Eq. 3.

A plot of vapor pressure vs temperature, based on the preceding equations, is shown in Fig. 1, the phase diagram for UF₆.

2.1.2 Equation of State and Vapor Compressibility Factor

Magnuson (DeWitt 1960 pp. 24, 97-101) proposed an equation of state for UF_6 which may be expressed as

$$PV/RT_R = 1/(1 + aT_R^{-3}P) , \quad (5)$$

where

P = pressure, psia,

V = molar volume, ft^3/mole ,

R = ideal gas constant = $10.73 \text{ psia-ft}^3/\text{lb mole-}^\circ\text{R}$,

T_R = absolute temperature, $^\circ\text{R} = T_F + 459.67$,

a = nonideality parameter = 4.8923×10^5 .

The stated range over which the nonideality parameter was evaluated is 127 to 270°F .

The compressibility factor, Z , is defined by

$$Z = PV/RT_R . \quad (6)$$

Thus, by comparison with Eq. 5,

$$Z = 1/(1 + aT_R^{-3}P) = T_R^3/(T_R^3 + aP) . \quad (7)$$

Other quantities required for sonic velocity and constant volume heat capacity calculations are,

$$Z_p = Z - P(\partial Z/\partial P)|_{T_R} = 2Z - Z^2 , \quad (8)$$

and

$$Z_t = Z + T_R(\partial Z/\partial T_R)|_P = 4Z - 3Z^2 . \quad (9)$$

2.1.3 Density

The following data on the density of solid UF_6 were reported by DeWitt (1960, p. 17).

Temperature ($^\circ\text{C}$)	Density (g/cm^3)
20.7	5.09 ± 0.06
25	5.06 ± 0.005
62.5	4.87

The above densities can be correlated within the reported experimental error by

$$\rho_s = 330.0 - 0.1800 T_F , \quad (10)$$

where

$$\rho_s = \text{density, lb/ft}^3 .$$

The liquid density correlation is modified from the correlation of Hoge and Wechsler (DeWitt 1960, p. 19, Eq. 5). Based on data reported in DeWitt (1960, p. 20), the predicted saturated liquid density is within 0.1% of experimental data between 65 and 131°C (149 and 268°F) and within 0.22% at 162°C (324°F). Unit conversion of the Hoge and Wechsler equation, which assumed a triple point temperature of 64.052°C, yields

$$\rho_l = 250.6 - 0.1241 T_F - 2.620 \times 10^{-4} T_F^2 . \quad (11)$$

where

$$\rho_l = \text{density, lb/ft}^3 .$$

At subatmospheric pressure, UF₆ vapor is nearly ideal. As pressure increases, the density of UF₆ vapor based on the Magnuson equation of state derived for temperatures from 127 to 270°F, which correspond to pressures from 12 to 130 psia, is given by

$$\rho_v = (MW/10.73)(P/T_R)Z , \quad (12)$$

where

$$\rho_v = \text{density, lb/ft}^3 ,$$

$$MW = \text{molecular weight, lb/lb mole,}$$

$$P = \text{pressure, psia.}$$

This equation, which differs by about 2.5% from ideal behavior at 122°F and 10.2 psia, is used for all vapor density calculations in the computer routines discussed subsequently. Deviation from ideal behavior decreases as temperature and pressure decrease.

The densities of solid and liquid UF₆ are plotted as functions of temperature in Fig. 2. The density of UF₆ vapor as a function of temperature and pressure assuming a molecular weight of 352 lb/lb mole is plotted in Fig. 3.

2.1.4 Enthalpy

DeWitt presented a series of heat capacity, enthalpy, and entropy correlations for solid, liquid, and vapor UF₆ (1960, pp. 56-70). Several of these correlations contained typographical errors; however, since enthalpy is the integral of the heat capacity and entropy is the integral of the quotient of heat capacity and temperature, errors were identified and corrected prior to converting the correlations to engineering units. Tabular data reported by DeWitt (1960, p. 59) beyond the range of applicability of the equations he presented imply the reference state for these correlations is solid UF₆ at 0 K for which the enthalpy entropy values are zero. To obtain vapor phase

Fig. 2. Density of solid and saturated liquid uranium hexafluoride.

enthalpies and entropies at pressures other than 1 atm (14.696 psia), a heat capacity departure function was incorporated based on taking the derivative of the enthalpy departure function given in Reid et al. (1977) after incorporating the compressibility factor obtained from the Magnuson equation of state (see Sect. 2.2).

Based on integration of the heat capacity equations from Dewitt, as modified, the following correlations for the enthalpy of UF_6 solid, liquid, and vapor are obtained.

$$H_s = 50.4460 - 5.70531 \times 10^{-2} T_R + 1.27509 \times 10^{-4} T_R^2 \quad (13)$$

$$- (9645.63/T_R) ,$$

$$H_l = [30.6133 + 5.10057 \times 10^{-2} T_R + 5.13165 \times 10^{-5} T_R^2 \quad (14)$$

$$- (6139.34/T_R)] + [0.18268(P - P^\circ)(1/\rho_l) ,$$

ORNL-DWG-85c-15260

Fig. 3. Density of uranium hexafluoride vapor.

$$H_v = [43.2614 + 9.21307 \times 10^{-2} T_R + 6.26265 \times 10^{-6} T_R^2 + (2951.71/T_R)] + [3.0939 \times 10^{-3} T_R(Z|_{P,T} - Z|_{14.7,T})] , \quad (15)$$

where

H = enthalpy (Btu/lb) and
 s, l, v = subscripts denoting solid, liquid, and vapor, respectively.

Equation 13 deviates from experimental values by less than 0.01% between 17°F and the triple point (147.3°F) and by less than 1% between -55 and 17°F. Equation 14, excluding the last term, is stated (DeWitt 1960, p. 59) to deviate from experimental values by less than 0.01%; however, a temperature range of applicability was not given (a range of 147.3 to 206°F might be assumed based on heat capacity correlations). The last term in Eq. 14 is a correction for supersaturated liquids, assuming an incompressible fluid. No estimate of deviation from experimental values is available for the vapor enthalpy. The last term of Eq. 15 is an enthalpy

departure function based on information given by Reid et al. (1977) after incorporating the compressibility factor based on the Magnuson equation of state (see Sect. 2.1.2). Heats of vaporization, sublimation, and fusion may be obtained by using these equations. Enthalpies of saturated UF_6 solid, liquid, and vapor are plotted in Fig. 4.

2.1.5 Entropy

Use of the modified heat capacity equations in integrating $dS = (C_p/T)dT$ leads to the following entropy equations for UF_6 solid, liquid, and vapor.

$$S_s = 0.393535 - 5.70531 \times 10^{-2} \ln T_R \quad (16)$$

$$+ 2.55019 \times 10^{-4} T_R - (4822.82/T_R^2) ,$$

$$S_l = -0.172963 + 5.10057 \times 10^{-2} \ln T_R \quad (17)$$

$$+ 1.02633 \times 10^{-4} T_R - (3069.67/T_R^2) ,$$

Fig. 4. Enthalpy of solid, liquid, and vapor uranium hexafluoride.

$$S_v = -0.332704 + 9.21307 \times 10^{-2} \ln T_R + 1.25237 \times 10^{-5} T_R + (1475.86/T_R^2) , \quad (18)$$

$$+ 3.0939 \times 10^{-3} (Z|_{P,T} - Z|_{14.7,T}) + 1.0313 \times 10^{-3} \ln[(1 - Z|_{14.7,T})/(1 - Z|_{P,T})] ,$$

where

S = entropy, Btu/lb-°R.

Equation 16 deviates from experimental values by less than 0.01% from 32°F to the triple point. Equation 17 is stated to deviate by less than 0.01% from experimental values, but no temperature range of applicability is given. DeWitt (1960, p. 70) reports that entropy values obtained using Eq. 18 (without the last two terms) are in reasonable agreement with those obtained from other sources. The last two terms in Eq. 18 represent the entropy departure function based on Reid et al. (1977). Entropies of UF_6 solid, liquid, and vapor are plotted in Fig. 5.

Fig. 5. Entropy of solid, liquid, and vapor uranium hexafluoride.

2.1.6 Other Properties

Sublimation point and triple point

UF₆ does not have a normal boiling point but reaches a sublimation point at 133.8°F when the vapor pressure over the solid is 14.7 psia. The triple point for UF₆ at which solid, liquid, and vapor phases coexist is about 147.3°F, which yields a vapor pressure of about 22.04 psia based on the preceding vapor equations.

Critical properties

At the critical point, UF₆ has a critical temperature of 906°R, a critical pressure of 669 psia, and a critical specific volume of 4.10 ft³/lb-mol. Based on a molecular weight of 352, the critical density is 85.84 lb/ft³.

Molecular weight

The molecular weight of UF₆ varies with the enrichment of the uranium and is given by,

$$MW = 352.04 - 0.03 E \quad (19)$$

where

$$E = \text{enrichment in } ^{235}\text{U} (\%).$$

The small variation in molecular weight (less than 1% between 0 and 100% enrichment) is negligible, and a molecular weight of 352 can generally be used. Most of the properties subroutines include correction factors for molecular weight, and the main programs allow a molecular weight to be read as input. The default molecular weight provided is 352.025.

2.1.7 UF₆ Reaction with H₂O

UF₆ reacts with water, including moisture in the air, to form uranyl fluoride (UO₂F₂) and hydrogen fluoride (HF). The equation for the chemical reaction is

A review of material related to this reaction led to an article by Popov et al. (1957, pp. 9-12) from which the heat of reaction (a positive heat of reaction implies an exothermic reaction) of

was found to be -7416 Btu/lb-mol of UF₆ reacted at 89.6°F. Using enthalpy correlations reported herein, a heat of reaction at 77°F for Eq. 20 was calculated to be 25,199 Btu/lb-mol of H₂O reacted. Note that, in making the conversion from one reference condition to the other, HF vapor is treated as a monomer.

2.1.8 Flashing

Upper and lower bounds on the vapor mass fraction at the point of release to the environment can be estimated from thermodynamic considerations. If the expansion process is assumed to be reversible (i.e., isentropic), a lower bound on the vapor fraction can be estimated. An upper bound on the vapor mass fraction can be estimated by assuming an adiabatic, isenthalpic process. Both assumptions are in error because heat would be transferred to the piping system and because the expansion would be irreversible; however, the error is relatively small.

If an isentropic process is assumed for the release of UF_6 , then

$$\bar{S}_i = X S_{v,f} + (1 - X) S_{c,f} , \quad (22)$$

where

- X = vapor mass fraction,
- S = entropy, Btu/lb-°F,
- v, c = subscripts for vapor and condensed phases,
- i, f = subscripts for initial and final conditions, and
- $-$ = superscript indicating average.

Rearranging yields,

$$X = (\bar{S}_i - S_{c,f}) / (S_{v,f} - S_{c,f}) . \quad (23)$$

The quantity $\bar{S}_i$ is the entropy of the UF_6 as it enters the release pathway and the average entropy along the release pathway. To evaluate X based on the preceding equation, the temperature that would yield a vapor pressure equal to the outlet pressure is determined, then that temperature is used to evaluate $S_{v,f}$ and $S_{c,f}$ assuming saturation. If $\bar{S}_i$ is based on a multiphase mixture of UF_6 being released from containment, then X based on Eq. 23 may be greater than 1. In this latter case, as well as when UF_6 vapor is released, a superheated vapor is released; therefore, no condensed phase occurs and Eq. 23 is not applicable. The final temperature may be determined by iteration using the equation for entropy of the vapor at unsaturated conditions (Eq. 18) and the criterion that the final entropy equals the average entropy.

For an adiabatic, isenthalpic process,

$$\bar{H}_i = X H_{v,f} + (1 - X) H_{c,f} , \quad (24)$$

where

- H = enthalpy, Btu/lb.

Rearranging yields,

$$X = (\bar{H}_i - H_{c,f}) / (H_{v,f} - H_{c,f}) , \quad (25)$$

where the enthalpies are determined in a manner analogous to the entropies. Again, if X is found to be greater than 1 by Eq. 25, a superheated vapor is released, and the enthalpy equation for the vapor (Eq. 15) is used to determine the final release temperature.

For the special case of the release of saturated liquid UF_6 at a given temperature to a final pressure of 14.7 psia, Eq. 23 for an isentropic release becomes

$$X = -3.383935 + 0.507936 \ln T_R + 1.02206 \times 10^{-3} T_R - (3.05691 \times 10^4)/(T_R^2) \quad (26)$$

and Eq. 25 for an isenthalpic release becomes

$$X = -0.244302 + 8.51871 \times 10^{-4} T_R + 8.57062 \times 10^{-7} T_R^2 - (1.02536 \times 10^2)/T_R \quad (27)$$

These expressions for the lower and upper bounds on the vapor mass fraction resulting from a release of saturated liquid UF_6 to a final pressure of 14.7 psia are plotted in Fig. 6. If the final

Fig. 6. Initial vapor mass fraction resulting from a release of saturated UF_6 liquid to a pressure of 1 atm. (Example: For saturated UF_6 liquid at 210°F, the initial vapor mass fraction will be between 0.537 and 0.557 when a release to a pressure of 1 atm occurs.)

pressure is other than 1 atm, Eqs. 23 and 25 should be used to bound the vapor mass fraction. Also, if the final pressure exceeds the triple point pressure, the condensed phase will be liquid rather than solid.

2.1.9 Subroutines

Six subroutines have been developed to predict UF_6 physical and thermodynamic properties. These subroutines and the purpose of each are listed in Table 3. Four of the subroutines require

Table 3. UF_6 physical and thermodynamic properties subroutines

Subroutine	Properties calculated
VPRUF ₆	Vapor pressure for a given temperature
EQTUF ₆	Equilibrium temperature for a given pressure
ZUF ₆	Vapor compressibility factor and related terms given temperature and pressure
DENUF ₆	Solid, liquid, and vapor densities given temperature, pressure, and molecular weight
HUF ₆	Solid, liquid, and vapor enthalpies given temperature, pressure, and molecular weight
SUF ₆	Solid, liquid, and vapor entropies given temperature, pressure, and molecular weight

temperature, pressure, and molecular weight as input. VPRUF6 requires temperature as input and gives vapor pressure as output; EQTUF6 gives the saturation temperature corresponding to a given UF_6 pressure supplied as input. In many cases, properties reported by DeWitt (1960) were given using moles as the unit of mass. A molecular weight of 352 was used in converting equations to engineering units, so the computer routines include molecular weight as input to adjust for other molecular weights resulting from uranium enrichment operations.

The subroutine FLASH has been developed to estimate the vapor mass fraction at a final pressure given the initial temperature, pressure, UF_6 molecular weight, vapor mass fraction, and liquid mass fraction of the nonvapor fraction, the final pressure, and whether the release is to be considered isentropic or isenthalpic. Output includes the final temperature, vapor mass fraction, and liquid mass fraction of the nonvapor fraction. If the vapor mass fraction would be greater than 1 according to Eq. 23 or 25, the subroutine sets the final vapor mass fraction equal to 1 and determines the final temperature of the superheated vapor. At the triple point, the condensed phase is assumed to be liquid.

2.2 HF-H₂O SYSTEM

Two sets of physical properties for the HF-H₂O system are of particular interest in current modeling efforts. These properties are vapor pressure and enthalpy. This section discusses the

estimation of (1) partial vapor pressures of HF and H₂O as functions of temperature and the composition of the condensed phase in equilibrium with the vapor and (2) liquid and vapor phase enthalpies. Estimation of the effective molecular weight of HF and the enthalpy effects of HF self-association in the vapor phases are incorporated into vapor pressure and enthalpy calculations.

2.2.1 Self-Association of HF Vapor

HF does not exist in a purely monomeric form but is linked into short polymers by hydrogen bonding. Beckerdite et al. (1983, pp. 287-93) report that a monomer-trimer-hexamer (1-3-6) model reasonably represents the self-association of HF. Using the data of Strohmeier and Briegleb,* which was reported by Beckerdite et al. (1983, p. 292) and which is shown in Table 4, the following equations were derived for the 1-3 and 1-6 equilibrium coefficients:

$$\ln K_3 = (23884.0/T_R) - 51.2393 \quad , \quad (28)$$

$$\ln K_6 = (40319.6/T_R) - 87.7927 \quad , \quad (29)$$

where

K_3 = equilibrium coefficient for the monomer-to-trimer association, psia⁻²,

K_6 = equilibrium coefficient for the monomer-to-hexamer association, psia⁻⁵.

Table 4. HF monomer-to-trimer and monomer-to-hexamer equilibrium coefficients

Temperature (K)	Equilibrium coefficient data of Strohmeier and Briegleb ^a		Predicted equilibrium coefficients ^b	
	$K_3 \times 10^3$ (atm ⁻²)	$K_6 \times 10^3$ (atm ⁻⁵)	$K_3 \times 10^3$ (atm ⁻²)	$K_6 \times 10^3$ (atm ⁻⁵)
299.15	210 ± 10	1620 ± 50	221	1688
305.15	120 ± 8	450 ± 30	92	387
311.15	48 ± 6	84 ± 4	40	94
317.15	9.0 ± 0.7	23 ± 0.4	18	24
329.15	5.2 ± 0.1	1.9 ± 0.3	3.9	1.8

^aTaken from Table III of Beckerdite, J. M., Powell, D. R., and Adams, E. T., Jr. 1983. "Self-Association of Gases. 2. The Association of Hydrogen Fluoride," *J. Chem. Eng. Data* 28, 287-93.

^bPredicted using Eqs. 28 and 29.

*The data of Fredenhagen were also used initially in deriving expressions for K_3 and K_6 . Subsequently, it was observed that, on extrapolation to lower temperatures potentially encountered in simulating atmospheric dispersion, use of the combined data sets would result in vapor enthalpies that were less than liquid enthalpies for high HF-to-H₂O ratios.

Note that K_n is based on the following association model:

$$n(\text{HF})_1 \rightleftharpoons (\text{HF})_n \quad (30)$$

and

$$K_n = [P_{(\text{HF})_n}] / [P_{(\text{HF})_1}]^n, \quad (31)$$

where

K-values calculated from Eqs. 28 and 29 are shown in Table 4 for comparison with the data of Strohmeier and Briegleb.

The partial vapor pressure of HF as obtained by the methods described in the next section, or the partial pressure of HF, is the sum of the partial pressures of the monomer, trimer, and hexamer; therefore,

$$P_{\text{HF}} = P_{(\text{HF})_1} + P_{(\text{HF})_3} + P_{(\text{HF})_6}, \quad (32)$$

where

P_{HF} = vapor pressure or partial pressure of HF.

Rearranging Eq. 31 and substituting into Eq. 32 yields

$$P_{\text{HF}} = P_{(\text{HF})_1} + K_3 P_{(\text{HF})_1}^3 + K_6 P_{(\text{HF})_1}^6. \quad (33)$$

To find the effective molecular weight, it is necessary to recognize that, in the vapor phase,

$$\rho_i = (P_i MW_i) / RT \quad (34)$$

and

$$\rho_T = \sum \rho_i. \quad (35)$$

By using these relationships,

$$\rho_{\text{HF}} = \frac{P_{\text{HF}} MW_{\text{HF}}}{RT} = \frac{P_{(\text{HF})_1} MW_{(\text{HF})_1}}{RT} + \frac{P_{(\text{HF})_3} MW_{(\text{HF})_3}}{RT} + \frac{P_{(\text{HF})_6} MW_{(\text{HF})_6}}{RT}, \quad (36)$$

which yields

$$MW_{\text{HF}} = [P_{(\text{HF})_1} MW_{(\text{HF})_1} + P_{(\text{HF})_3} MW_{(\text{HF})_3} + P_{(\text{HF})_6} MW_{(\text{HF})_6}] / P_{\text{HF}}, \quad (37)$$

where MW is molecular weight. The quantity, MW_{HF} , is the effective molecular weight to be used in determining vapor phase densities of HF and in converting between vapor weight fraction and vapor mole fraction. It also follows from Eqs. 34 and 35 that the weight fraction of an n-mer of HF to the total HF in the vapor phase is given by

$$W_{(HF)_n} = [P_{(HF)_n} MW_{(HF)_n}] / (P_{HF} MW_{HF}) , \quad (38)$$

The partial vapor pressure of HF obtained as a function of temperature and weight fraction of HF in the condensate (see Sect. 2.2.2) appears to be the effective pressure of the associated HF. Thus, the partial vapor pressure of HF is predicted and then the effective molecular weight is determined. If no condensed phase occurs, the partial pressure of HF is determined iteratively along with the effective molecular weight of HF by assuming a partial pressure, calculating the effective molecular weight, and applying the ideal gas law to get a new partial pressure.

2.2.2 Partial Vapor Pressure of HF

The partial vapor pressure of HF is based on Fig. 7, which shows the partial vapor pressure of HF as a function of temperature for constant weight fractions of HF in the condensed HF-H₂O liquid mixture. These curves can be approximated by an equation of the form

$$\ln P_{HF} = (A/T_R) + B , \quad (39)$$

where

$$\begin{aligned} P_{HF} &= \text{partial vapor pressure of HF, mm,} \\ T_R &= \text{absolute temperature, } ^\circ\text{R.} \end{aligned}$$

The empirical coefficients A and B and the maximum error for each curve based on data extracted from Fig. 7 are given in Table 5.

Attempts were made to correlate A and B as functions of weight fraction; however, all such attempts led to greater errors in predicted partial vapor pressure for the given lines. It was finally decided that pressures over condensate compositions other than those given in Table 5 would be best obtained by interpolation. Since pressure was presented on a log scale and since consecutive concentration lines are roughly equally spaced, the following interpolation method is used (except for condensate compositions below 5 wt % HF):

$$\ln P_{HF} = \ln P_{HF,1} + \frac{\ln P_{HF,2} - \ln P_{HF,1}}{0.05} (W_{HF,I} - W_{HF,I,1}) \quad \text{for } W_{HF,I} > 0.05 , \quad (40)$$

where

$$\begin{aligned} 1, 2 &= \text{subscripts defining conditions for the known curves immediately above and below} \\ &\quad \text{the concentration of interest and} \\ W_{HF,I} &= \text{weight fraction of HF in the condensate.} \end{aligned}$$

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Fig. 7. Partial vapor pressure of HF over aqueous solutions of HF. (Source: Brian C. Rogers, Allied Chemical, personal communication to W. R. Williams, July 26, 1983).

Table 5. Coefficients for estimating the partial vapor pressure of HF by Eq. 39 over various liquid mixtures of HF and H₂O

Weight fraction of HF in liquid	A	B	Maximum error in estimate ^a (%)
0.05	-10689	17.680	3
0.10	-10536	18.069	4
0.15	-10647	18.793	5
0.20	-10675	19.409	2
0.25	-10460	19.595	3
0.30	-10362	19.951	4
0.35	-9779.5	19.429	4
0.40	-8917.7	18.948	5
0.45	-8188.2	18.256	5
0.50	-7770.0	17.975	3
0.55	-7575.0	18.054	3
0.60	-7411.4	18.178	3
0.65	-7315.7	18.365	3
0.70	-7163.4	18.457	3
0.75	-6984.3	18.492	2
0.80	-6459.7	17.857	2
0.85	-6269.7	17.812	1
0.90	-5891.7	17.391	2
0.95	-5849.8	17.555	1
1.00	-5641.0	17.364	1

^aError is estimated relative to Fig. 7. The accuracy of Fig. 7 is not known.

Note that $P_{\text{HF},1}$ and $P_{\text{HF},2}$ are evaluated at the temperature of interest for P_{HF} . Below 5 wt % HF, the partial vapor pressure of HF is calculated from

$$P_{\text{HF}} = P_{\text{HF},5 \text{ wt \%}}(W_{\text{HF},i}/0.05) \quad \text{for } W_{\text{HF},i} < 0.05 \quad (41)$$

2.2.3 Partial Vapor Pressure of H₂O

In an azeotropic system, the minimum total vapor pressure of the system (assuming constant temperature) is expected to occur at the azeotropic composition as shown in Fig. 8. When correlations for the partial vapor pressure of H₂O were derived directly from data on the partial vapor pressure of H₂O as a function of condensate composition and temperature and then summed together with the corresponding partial vapor pressure of HF, the minimum total vapor pressure of the HF-H₂O system did not occur at the azeotrope. Since the available data on HF partial vapor pressures were more extensive than those on H₂O partial vapor pressures, a decision was made to

Fig. 8. An approximate pressure-composition diagram for the HF-H₂O system at 25°C. Two-phase mixtures occur for pressure-composition combinations that fall within the two "wings" lying between those regions labeled liquid and vapor. (Source: W. H. McCulla, *Determinations of the Rate of HF Hydration and the Effects of Moisture Condensation*, K/PS-155, April 30, 1982.)

accept that HF data and to force-fit an H₂O partial vapor pressure model to yield a minimum total pressure at the azeotrope.

Figure 9 is a plot of some available partial vapor pressure data for the HF-H₂O system. The curve for HF partial vapor pressure is primarily based on data from Fig. 7 and agrees fairly well with the data of Brosheer et al. (1947, pp. 423-27), which is limited to a maximum of 30 wt % HF. The curves corresponding to H₂O partial pressure and total pressure are based on the data of Brosheer et al. Also identified on Fig. 9 is the azeotrope for the HF-H₂O system.

Figure 10(a) illustrates the inconsistency in data that led to the decision to force-fit an H₂O partial vapor pressure model. The solid curves in Fig. 10 are those shown in Fig. 9. Point 1 corresponds to the partial vapor pressure of HF calculated from the correlations of Sect. 2.2.2. By using the effective molecular weight of HF and the azeotropic composition, the partial vapor pressure of H₂O (point 2) can be determined. The total pressure, point 3, is the sum of the partial vapor pressures. Obviously, a minimum total pressure would occur at a condensate composition between 30 wt % HF and the azeotrope, which is not consistent with the expected behavior of an azeotropic system.

Since the plot of HF partial vapor pressures (see Fig. 7) appeared to represent a more extensive data base than that available for H₂O, the HF data were subsequently used and H₂O partial vapor pressure data were ignored. Thus, the current method for calculating the partial vapor pressure of H₂O proceeds as follows. First, calculate the partial vapor pressure of HF for the desired temperature at the azeotropic composition (38.26 wt % HF) and the effective molecular

Fig. 9. Available data on the partial vapor pressures of HF and H₂O at 77°F.

weight of HF using the correlations given in Sect. 2.2.1 [note point 1 in Fig. 10(a)]. Next, evaluate the vapor mole fraction of HF corresponding to the azeotropic weight fraction and the given temperature using the following equation:

$$Y_{\text{HF}} = \frac{(W_{\text{HF},v}/MW_{\text{HF}})}{(W_{\text{HF},v}/MW_{\text{HF}}) + [(1 - W_{\text{HF},v})/MW_{\text{H}_2\text{O}}]} \quad (42)$$

Fig. 10. Procedure for obtaining the partial vapor pressure of H₂O. Figure 10(a) shows the first three steps superimposed on the curves shown in Fig. 9 and demonstrates data inconsistency. Figure 10(b) shows the final two steps, which differ depending on condensate composition. Steps 4a and 5a go together above the azeotrope, while steps 4b and 5b go together below the azeotrope.

where

$$\begin{aligned} Y_{HF} &= \text{vapor mole fraction of HF,} \\ W_{HF,v} &= \text{weight fraction of HF in the vapor,} \\ MW_{H_2O} &= \text{molecular weight of H}_2\text{O.} \end{aligned}$$

The azeotropic partial vapor pressure of H_2O [point 2 in Fig. 10(a)] is given by

$$P_{H_2O, \text{azeotrope}} = P_{HF, \text{azeotrope}} [(1 - Y_{HF}) / (Y_{HF})] \quad (43)$$

Next, calculate the vapor pressure of pure H_2O (Reid 1977, pp. 629-32) from

$$\ln P_{H_2O}^{\circ} = 18.3034 - [6869.59 / (T_F + 376.64)] \quad (44)$$

where

$$\begin{aligned} P_{H_2O}^{\circ} &= \text{vapor pressure for pure H}_2\text{O, mm,} \\ T_F &= \text{temperature, } ^{\circ}\text{F.} \end{aligned}$$

To find the partial vapor pressure of H_2O over a condensate having a weight fraction of HF less than the azeotropic composition (0.3826), use

$$P_{H_2O} = P_{H_2O}^{\circ} + (P_{HF, \text{azeotrope}} + P_{H_2O, \text{azeotrope}} - P_{H_2O}^{\circ}) \quad (45)$$

$$\times (W_{HF,i} / W_{HF, \text{azeotrope}}) - P_{HF} \quad \text{for } W_{HF,i} \leq W_{HF, \text{azeotrope}} ,$$

where P_{HF} is calculated at the condensate composition of interest. Equation 45 [which corresponds to the dashed line 5b in Fig. 10(b)] assumes a linear relationship between the total pressure of the HF- H_2O system [represented by the first two terms to the right of the equals sign and line 4b on Fig. 10(b)] and the weight fraction of HF in the condensate below the azeotropic composition. To find the partial vapor pressure of H_2O over a condensate having a weight fraction of HF greater than the azeotropic composition [see line 4a on Fig. 10(b)], use

$$P_{H_2O} = P_{H_2O, \text{azeotrope}} [(1 - W_{HF,i}) / (1 - W_{HF, \text{azeotrope}})]^3 \quad \text{for } W_{HF,i} \geq W_{HF, \text{azeotrope}} \quad (46)$$

The choice of the exponent 3 in Eq. 46 is somewhat arbitrary; nevertheless, it permits a less-sharp discontinuity in the slope of P_{H_2O} versus $W_{HF,i}$ than would exponents less than 3. Higher exponents would allow P_{H_2O} to decrease too rapidly such that a total pressure minimum would occur above the azeotrope, causing numerical problems.

In analyses subsequent to development of the methods described in this section for predicting the partial vapor pressure of H_2O , it has been determined that predicted total vapor pressures for the HF- H_2O system at the azeotrope correspond to temperatures about 10°F above those specified.

It is anticipated that, if the available data on H₂O partial vapor pressures were retained and the partial vapor pressures of HF were modified between 35 wt % and about 50 wt % (see Fig. 9), a better set of correlations for the partial vapor pressure of H₂O could be derived.

2.2.4 Enthalpy of HF-H₂O Vapor Mixtures

Figure 11 is an enthalpy concentration diagram for the HF-H₂O system at 1 atm. At temperatures above 250°F, HF polymerization is negligible and the interaction between HF and H₂O in the vapor phase appears to be ideal, as evidenced by the straight-line relationship between the enthalpy of the vapor mixture and the weight percent of HF in the mixture. At lower temperatures, however, association of HF into polymers significantly reduces the enthalpy of the vapor mixture as the weight percent of HF approaches 100 wt %. This latter trend may be observed by noting the intersections of the equilibrium tie lines for various temperatures with the saturated vapor curve. These intersections, particularly above 90 wt % HF, are much lower than would be predicted by extrapolation of data above 250°F.

The enthalpy model for an HF-H₂O vapor mixture is composed of two parts: (1) a base enthalpy, assuming no polymerization, which is based on extrapolation of data above 250°F to lower temperatures, and (2) a correction term based on the polymerization of HF. The base enthalpy is given by

$$H_{\text{base}} = (1051.0 + 0.472 T_F) - (376.0 + 0.136 T_F)W_{\text{HF},v} \quad (47)$$

where

H_{base} = uncorrected enthalpy for an HF-H₂O vapor mixture, Btu/lb,

$W_{\text{HF},v}$ = weight fraction of HF in the vapor phase, lb_{HF vapor}/lb_{HF-H₂O vapor}.

The correction term is obtained from Eqs. 28 and 29 using the following basic thermodynamic relationship for chemical reactions:

$$\Delta G_i = -RT \ln K_i = \Delta H_i - T \Delta S_i \quad (48)$$

where

ΔG_i = change in Gibbs free energy,

R = ideal gas constant,

T = absolute temperature,

K_i = equilibrium coefficient,

ΔH_i = change in enthalpy,

ΔS_i = change in entropy.

Note that ΔG_i , ΔH_i , and ΔS_i are based on the sum of the product values minus the sum of the reactant values. Recognizing that Eqs. 28 and 29 are in the form

$$\ln K_n = (A_n/T) + B_n \quad (49)$$

Fig. 11. Enthalpy-concentration diagram for liquid and vapor mixtures of HF and H₂O showing equilibrium tie lines at 1 atm. (Source: Brian C. Rogers, Allied Chemical, personal communication to W. R. Williams, July 26, 1983).

and that Eq. 48 can be rearranged to a similar form, then it is found that

$$\Delta H_n = -RA_n \quad (50)$$

Using the relationship of Eq. 50, the heat of association for $(\text{HF})_3$ is -790.642 Btu/lb $(\text{HF})_3$ formed and for $(\text{HF})_6$ is -667.358 Btu/lb $(\text{HF})_6$ formed. These heats of association are applied as correction factors to Eq. 47 to obtain the following expression for enthalpy:

$$H_v = (1051.0 + 0.472 T_F) - [376.0 + 0.136 T_F + 790.642 W_{(\text{HF})_3} + 667.358 W_{(\text{HF})_6}] W_{\text{HF},v} \quad (51)$$

where

H_v = enthalpy of a vapor mixture of HF and H_2O , Btu/lb,
 $W_{(\text{HF})_n}$ = weight fraction of an n-mer of HF as defined by Eq. 38, lb_{n-mer}/lb_{HF vapor}.

2.2.5 Enthalpy of HF- H_2O Liquid Mixtures

The lower portion of Fig. 11, which corresponds to liquid mixtures of HF and H_2O , is shown in expanded scale on Fig. 12. Significant nonideal behavior is exhibited that would normally be modeled as a heat of mixing. In this modeling effort, empirical correlations have been derived to give enthalpy as a function of temperature and composition. The left side of the figure (dilute HF solutions) was used to develop the correlations required to proceed to the right side (high-concentration HF solutions). The correlations are of the form

$$H_l = A_i + B_i W_{\text{HF},l} + C_i W_{\text{HF},l}^2 \quad (52)$$

where

H_l = enthalpy of the HF- H_2O liquid mixture, Btu/lb
 $W_{\text{HF},l}$ = weight fraction of HF in the HF- H_2O liquid mixture lb HF/lb mixture.

Values for the coefficients A_i , B_i , and C_i are given in Table 6 along with the range of applicability and boundary conditions. Note that coefficients for lower intervals than the desired weight fraction of HF must be calculated to obtain coefficients for the needed interval. The preceding set of equations (Eq. 52 and Table 6) for liquid enthalpy are fairly accurate except above 90 wt % HF. Below 20 wt %, deviations between predicted and expected values are negligible. From 20 to 90 wt %, deviations up to 2 Btu/lb have been observed. Above 90 wt %, deviations of 7 Btu/lb have been observed; however, it may be worth noting that enthalpy can change that much with a 0.5% change in HF concentration.

2.2.6 HF- H_2O Vapor Liquid Equilibrium

Preceding sections have presented methods for calculating the vapor phase composition given the liquid composition. This section describes how to obtain the phase splits for HF and H_2O in a

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Fig. 12. Enthalpy-concentration diagram for liquid HF-H₂O mixtures. (Source: Brian C. Rogers, Allied Chemical, personal communication to W. R. Williams, July 26, 1983).

Table 6. Correlations for coefficient of Eq. 52
(T = temperature, °F; H = enthalpy, Btu/lb)

Interval	Limits of $W_{HF,i}$	Coefficients	Boundary conditions
1	0.0–0.2	$A_1 = -32.252 + 0.99783 T$ $B_1 = -357.75 - 0.969 T + 0.0013 T^2$ $C_1 = 0$	
2	0.2–0.6	$C_2 = (H_{20} - H_{60} + 0.4 M_{20}) / -0.16$ $B_2 = M_{20} - 0.4 C_2$ $A_2 = H_{20} - 0.2 B_2 - 0.04 C_2$	$H_{20} = A_1 + 0.2 B_1$ $H_{60} = -179.68 + 0.63569 T + 5.8429 \times 10^{-4} T^2$ $M_{20} = B_1$
3	0.6–0.8	$C_3 = (H_{60} - H_{80} + 0.2 M_{60}) / -0.04$ $B_3 = M_{60} - 1.2 C_3$ $A_3 = H_{60} - 0.6 B_3 - 0.36 C_3$	$H_{80} = -159.26 + 0.7985 T$ $M_{60} = B_2 + 1.2 C_2$
4	0.8–0.9	$C_4 = (H_{80} - H_{90} + 0.1 M_{80}) / -0.01$ $B_4 = M_{80} - 1.6 C_4$ $A_4 = H_{80} - 0.8 B_4 - 0.64 C_4$	$H_{90} = -111.54 + 0.67057 T$ $M_{80} = B_3 + 1.6 C_3$
5	0.9–1.0	$C_5 = (H_{90} - H_{100} + 0.1 M_{90}) / -0.01$ $B_5 = M_{90} - 1.8 C_5$ $A_5 = H_{90} - 0.9 B_5 - 0.81 C_5$	$H_{100} = -23.3142 + 0.870283 T$ $M_{90} = B_4 + 1.8 C_4$

multicomponent system. For the multicomponent systems of interest, the overall composition and the total enthalpy are known. For compartment models, these quantities are known with respect to the total mass of material in the compartment. The total volume of the compartment is also known. Given this information, it is possible to obtain the individual phase compositions and temperature as well as the total pressure.

The procedure for evaluating phase composition in the compartment model is to assume a temperature, evaluate the phase compositions and total pressure at that temperature given the overall composition, and then calculate the total enthalpy at these conditions. The calculated total enthalpy is then compared to the known total enthalpy of the system. Temperature is used as the variable on which to iterate until the calculated enthalpy matches the known enthalpy.

In evaluating phase compositions, the vapor phase is assumed to be ideal. As was outlined in Sect. 2.2.1, an effective molecular weight for HF can be calculated; this effective molecular weight is used to obtain the number of moles of HF vapor present. Generally, condensation is assumed to proceed independently for all components except HF and H₂O. Thus, the maximum partial pressure a component can exert is generally limited by the vapor pressure of that component. In the case of HF and H₂O, when both components are present the effective vapor pressures are reduced (i.e., the partial vapor pressure of the individual component is less than the vapor pressure of the pure component).

The first step in evaluating the phase composition of the HF-H₂O subsystem is to evaluate the maximum partial pressure of each component in the given volume at the given temperature based on the total mass of each component, assuming no condensation but including the effect of HF polymerization. Next, the weight fraction of HF in a liquid phase that would give the maximum partial pressure of HF is calculated. If a weight fraction greater than 1 is calculated, condensation will occur and the next step is skipped. If a weight fraction less than 1 is calculated, the sum of the partial vapor pressure of HF and H₂O at the given temperature and calculated weight fraction of HF in the liquid is calculated and compared to the sum of the maximum partial pressures, assuming no condensation. If the sum of the maximum partial pressures is less than or equal to the sum of the partial vapor pressures, no condensation will occur; thus, all HF and H₂O is in the vapor phase.

If condensation occurs, two special cases are to be considered: condensation of a pure component and condensation at the azeotrope. These cases occur when the weight fraction of HF in the HF-H₂O subsystem is found to be 0 (pure H₂O), 0.3826 (azeotropic mixture of HF and H₂O, implying the same composition in the liquid and vapor phases), or 1 (pure HF). For these specific cases, the partial vapor pressures are calculated for HF and H₂O along with the appropriate effective molecular weight for HF vapor; then, the mass of each component that can exist in the vapor phase at its partial vapor pressure in the given volume at the given temperature is calculated, assuming that the ideal gas law applies. The mass of each component in the liquid phase is determined by subtracting the mass in the vapor phase from the total mass for each component.

When condensation is predicted at compositions other than those discussed in the preceding paragraph, an iterative procedure is required to evaluate the phase composition. Initial and lower limits for the iteration are the azeotropic weight fraction and the weight fraction of HF in the HF-H₂O subsystem. The correct liquid weight fraction will always be between these two limits. Note that the azeotropic weight fraction is an upper limit for subsystem compositions less than the azeotropic composition and a lower limit for subsystem compositions greater than the azeotropic composition. The closure criterion for the iterative procedure is that the partial pressures of HF and H₂O must approximately equal their partial vapor pressures.

The principle relationship required, besides the partial vapor pressure and polymerization correlations, is

$$m_{\text{vap}} = \frac{[m_{\text{H}_2\text{O}}W_{\text{HF},l} - m_{\text{HF}}(1 - W_{\text{HF},l})]}{W_{\text{HF},l} - W_{\text{HF},v}} \quad (53)$$

where

m_{vap} = mass of HF and H₂O in the vapor phase,

$m_{\text{H}_2\text{O}}, m_{\text{HF}}$ = total mass of H₂O and HF, respectively,

$W_{\text{HF},l}, W_{\text{HF},v}$ = weight fractions of HF in the liquid and vapor phases, respectively.

As $W_{\text{HF},l}$ approaches the azeotrope, m_{vap} approaches infinity (provided the subsystem composition is other than azeotropic); as $W_{\text{HF},l}$ approaches the subsystem composition, m_{vap} approaches 0. Given the total amount of vapor in the HF-H₂O subsystem, the partial pressures exerted by HF and H₂O can be calculated and compared to the sum of the partial vapor pressures. Once the closure criterion is met, the individual vapor phase masses are calculated and, by calculating the difference, the liquid phase masses are obtained.

2.2.7 Computer programs for predicting HF-H₂O properties

Three computer subroutines have been developed for evaluating thermodynamic properties of the HF-H₂O system. The subroutine PHFH2O gives the individual partial vapor pressures of HF and H₂O as well as their sum along with the vapor mole fraction of HF in the HF-H₂O system given the temperature and weight fraction of HF in the HF-H₂O condensate. The enthalpies of liquid and vapor mixtures are calculated by HHFH2O given the weight fraction of HF in the liquid and vapor phases, respectively, and the temperature. The weight fractions of HF trimer and hexamer in the vapor phase (with respect to total HF in the vapor phase) are also required. The subroutine HFPOLY calculates the effective molecular weight of HF in the vapor phase as well as the trimer and hexamer fractions required by HHFH2O given the temperature and partial pressure of HF. A logical variable has been included in the call to HFPOLY to specify whether the n-mer weight fractions should be calculated.

The subroutine PHASE requires as input the compartment temperature and volume as well as the total masses of HF and H₂O present. The masses of HF and H₂O found in each phase are calculated and returned.

2.3 OTHER COMPONENTS

In addition to the properties already discussed for UF₆, HF, and H₂O, several properties are needed for UO₂F₂ and air. UO₂F₂ is a solid, while air is a vapor over the temperature range of interest for the current models. UO₂F₂ has a density of 398 lb/ft³ and has a heat capacity of 0.0821 Btu/lb-°F. The density of air is obtained by applying the ideal gas law. The heat capacity of air is 0.24073 Btu/lb-°F.

2.4 MIXTURE ENTHALPIES

In modeling a release of UF_6 within a compartment, total enthalpy becomes an important criterion for determining the final temperature and pressure in the compartment at the end of a time interval. Total enthalpy is the sum of the differences in enthalpy for each component between the current conditions and reference conditions. Reference temperature for all components is 77°F . A reference pressure of 14.696 psia is used for UF_6 because departure functions are included in UF_6 enthalpy correlations (see Sect. 2.1.4). The reference state for air, H_2O , and UF_6 is vapor, while that of UO_2F_2 is solid. The reference state of HF is a monomeric vapor.

Total enthalpy is expressed mathematically by

$$\Delta H_{\text{total}} = \Delta H_{\text{air}} + \Delta H_{\text{HF-H}_2\text{O}} + \Delta H_{\text{UF}_6} + \Delta H_{\text{UO}_2\text{F}_2} , \quad (54)$$

where the component ΔH values are determined as follows. For air,

$$\Delta H_{\text{air}} = 0.24037 m_{\text{air}}(T - 77) , \quad (55)$$

where m_i is mass of component i in the compartment, lb. For the HF- H_2O system, the weight fractions of HF in both liquid and vapor phases, as well as in the total HF- H_2O subsystem, are determined from

$$W_{\text{HF},l} = m_{\text{HF},l} / (m_{\text{HF},l} + m_{\text{H}_2\text{O},v}) , \quad (56)$$

$$W_{\text{HF},v} = m_{\text{HF},v} / (m_{\text{HF},v} + m_{\text{H}_2\text{O},v}) , \quad (57)$$

and

$$W_{\text{HF}} = \frac{m_{\text{HF},l} + m_{\text{HF},v}}{m_{\text{HF},l} + m_{\text{HF},v} + m_{\text{H}_2\text{O},l} + m_{\text{H}_2\text{O},v}} . \quad (58)$$

Also, the weight fractions of HF monomer, trimer, and hexamer must be known at the temperature of interest. The value of $\Delta H_{\text{HF-H}_2\text{O}}$ is given by

$$\begin{aligned} \Delta H_{\text{HF-H}_2\text{O}} = & (m_{\text{HF},l} + m_{\text{H}_2\text{O},l})H_l(T, w_{\text{HF},l}) \\ & + (m_{\text{HF},v} + m_{\text{H}_2\text{O},v})H_v(T, w_{(\text{HF})}, w_{(\text{HF})}, w_{\text{HF},v}) \\ & - (m_{\text{HF},l} + m_{\text{HF},v} + m_{\text{H}_2\text{O},l} + m_{\text{H}_2\text{O},v})H_v(77, 0, 0, w_{\text{HF}}) , \end{aligned} \quad (59)$$

where H_i is found from methods given in Sect. 2.2.5. H_v is given by Eq. 51. For UF_6 ,

$$\begin{aligned} \Delta H_{\text{UF}_6} = & (m_{\text{UF}_6,s} H_{\text{UF}_6,s}(T) + m_{\text{UF}_6,l} H_{\text{UF}_6,l}(T,P) \\ & + (m_{\text{UF}_6,v} H_{\text{UF}_6,v}(T,P) \\ & - (m_{\text{UF}_6,s} + m_{\text{UF}_6,l} + m_{\text{UF}_6,v}) H_{\text{UF}_6,v}(77,14.696)) \end{aligned} \quad (60)$$

For UO_2F_2 ,

$$H_{\text{UO}_2\text{F}_2} = 0.0821 m_{\text{UO}_2\text{F}_2} (T - 77) \quad (61)$$

These equations are used in the subroutine DENTHL, which makes use of the previously mentioned subroutines HUF6 and HHFH2O. The temperature, pressure, and mass of each component in each phase must be known (or assumed) before the total enthalpy can be calculated. DENTHL can also be used for flow streams by assuming that m_i refers to mass flow rate (lb/ Δt) rather than mass (lb).

3. COMPARTMENT MODELS

Accident scenarios involving the postulated release of UF_6 can occur both indoors and outdoors. For indoor releases, compartment models are needed that can handle the effects of chemical reactions and vapor liquid equilibrium, including changes in temperature and pressure within a compartment. The compartment models presented herein were developed on the assumption of homogeneous behavior; that is, at a given instant in time, the composition, temperature, and pressure within a compartment are the same; hence, there is no spatial variation. Because the characteristics of a particular site are unique, an extensive set of subroutines was written so that specialized transient models tailored to simulate specific building ventilation configurations could be quickly developed by writing only a main program. Three transient models and one batch model were developed for use in the GDP SAR program.

1. FWDTRN simulates the transient behavior of a release in a feed and withdrawal building.
2. CSCDSM simulates the transient behavior of a release in a cascade building with the ventilation system in the summer operating mode.
3. CSCDWN simulates the transient behavior of a release in a cascade building with the ventilation system in the winter operating mode.
4. BATCH evaluates final conditions in a closed room with an option (open compartment) to base final conditions on the ambient pressure while allowing the compartment volume to be changed.

The "open compartment" option identified for BATCH allows for the simulation of a steady-state release into a constant volume air flow rate ventilation system exhausting to the outside.

A simplified logic diagram for the transient models is shown in Fig. 13. The following list provides a brief description of the purpose of each subroutine as well as the section of this report in which background material and the subroutine are discussed.

1. COMPRT performs final mass and energy balances for a compartment, including reaction and heat transfer given input and output flow rates and initial conditions (see Sect. 3.5.2).
2. DENTHL calculates total enthalpy with respect to reference conditions, given composition, temperature, and pressure (see Sect. 2.4).
3. DENUF6 calculates densities of UF_6 , given temperature, pressure, and molecular weight (see Sects. 2.1.3 and 2.1.9).
4. DPFLOW calculates mass flow and enthalpy rates through a pressure-drop-controlled duct or vent given the pressure drop, a resistance term, and upstream conditions (see Sect. 3.2.2).
5. EQTUF6 calculates the temperature that would yield a given pressure as the vapor pressure of UF_6 (see Sects. 2.1.1 and 2.1.9).
6. FLASH calculates the resulting final solid-liquid-vapor split of UF_6 , given the initial composition, temperature, and pressure and the final pressure (see Sect. 2.1.8).
7. HCOEFF calculates a heat transfer coefficient required to maintain steady-state conditions, given steady-state compartment and inlet stream data (see Sect. 3.5.1).
8. HFPOLY calculates effective molecular weight and monomer, trimer, and hexamer weight fractions, given the temperature and partial pressure of HF (see Sects. 2.2.1 and 2.2.7).
9. HHHF2O calculates liquid and vapor enthalpies of HF- H_2O mixtures, given temperature and composition (see Sects. 2.2.4, 2.2.5, and 2.2.7).

*STERM IS CALLED BEFORE TRANSIENT ANALYSIS BEGINS UNLESS CODING FOR A QUASI TRANSIENT SOURCE TERM IS REQUIRED.

Fig. 13. Simplified logic diagram for transient compartment models.

10. HUF6 calculates UF_6 enthalpies, given temperature, pressure, and molecular weight (see Sects. 2.1.4 and 2.1.9).
11. HUMID calculates the molar ratio of water vapor to moist air, given the ambient temperature, pressure, and relative humidity (see Sect. 3.5.1).
12. MIXFLW combines up to four streams including reaction effects and exhausts at a given pressure, given initial stream data (see Sect. 3.2.3).
13. PHASE performs a vapor-liquid equilibrium mass balance for an $\text{HF-H}_2\text{O}$ system, given total masses of HF and H_2O , volume, and temperature (see Sects. 2.2.6 and 2.2.7).
14. PHFH2O calculates partial and total vapor pressures for the $\text{HF-H}_2\text{O}$ system, given liquid-phase composition and temperature (see Sect. 2.2.2, 2.2.3, and 2.2.7).
15. REMOVE evaluates removal rate of condensate from a compartment, given the composition in the compartment and the floor area of the compartment (see Sect. 3.4).
16. RESIST calculates a resistance term for pressure-drop-controlled vents, given either a resistance coefficient and flow area or steady-state mass flow rate, density, and pressure differential (see Sect. 3.2.1).
17. ROOM calculates the initial mass of air and water vapor in a compartment, given the ambient-water-vapor-to-moist-air ratio and the temperature, pressure, and volume of the compartment (see Sect. 3.5.1).
18. SETRAY initializes arrays (see Sect. 3.1.1).
19. SSBLOW calculates initial flow rates of air and water vapor through a constant volume blower, given the ambient-water-vapor-to-moist-air ratio, the upstream temperature and pressure, and the blower volumetric flow rate (see Sect. 3.2.1).
20. STERM sets up source term data for the transient compartment models, given input data read by main program (see Sect. 3.3).
21. SUF6 calculates entropies of UF_6 given temperature, pressure, and molecular weight (see Sects. 2.1.5 and 2.1.9).
22. TRBLOW calculates mass flow and enthalpy rates through a constant volume blower, given upstream compartment conditions and blower volumetric flow rate (see Sect. 3.2.2).
23. VPRUF6 evaluates UF_6 vapor pressure for a given temperature (see Sects. 2.1.1 and 2.1.9).
24. ZUF6 calculates UF_6 vapor compressibility, given temperature and pressure (see Sects. 2.1.2 and 2.1.9).

A general discussion of the structure of the main programs is given in Sect. 3.1; however, the specific programs (FWDTRN, CSCDSM, CSCDWN, and BATCH) are not discussed until Chap. 4. A discussion of physical and thermodynamic properties required for the compartment models has been provided earlier in Chap. 2. Ventilation system subroutines are discussed in Sect. 3.2, including a stream mixing model. A source term model which develops input values for a finite duration release is described in Sect. 3.3. Section 3.4 discusses condensate deposition. Subroutines for performing compartment mass and energy balances are presented in Sect. 3.5.

3.1 MAIN PROGRAM STRUCTURE

The general structure of a main program to perform transient compartment analyses is described in this section. This description includes certain elements of coding required in any transient program. Many elements would need to be added to develop a suitable main program for a particular application. Examples of such additional elements of coding, as well as how these

elements fit together with those presented in this section, are best seen by comparing the subsequent discussion in this section to the program listings such as FODRFT and INDRFT, which appear in Williams (1985).^{*} A generalized logic diagram for the aforementioned main programs is given in Fig. 14. The overall structure of a main program is

1. initialize arrays and read input data (Sects. 3.1.1 and 3.1.2),
2. develop initial steady state conditions existing prior to the release that are derived from input data (Sect. 3.1.3), and
3. perform the transient analysis and output results (Sect. 3.1.4).

Special features of the programs developed for inclusion in this report are discussed in Chap. 4.

3.1.1 Initial Statements

All subroutines discussed in this report are written in double precision FORTRAN. The following coding is required in all main programs.

```

      IMPLICIT REAL*8 (A-H,J-Z)
C
      LOGICAL CHECK
C
      DIMENSION FLAREA(30), ACFM(30), RATE(9)
C
      COMMON /LBMASS/ MASS(30,9), RH
      COMMON /COMPTP/ TC(30), PC(30), TSURF(30)
      COMMON /MDLWT/ WMOL(9)
      COMMON /VOLUME/ VOL(30), KRCDEF(30), DPAREA(30), DEPVEL
      COMMON /ENTHAL/ H(30), QRATE(30), QCOOL(30), HTCDEF(30),
      * HTAREA(30)
      COMMON /ISTRMS/ IIN(30,4), IDUT(30,4)
      COMMON /CONTRL/ AMINLN, TIME, DELT, MAXTIM, IFLAG, TRELS
      COMMON /POLYMR/ C1, C3, C6, WMBHF
      COMMON /MISCEL/ ITYPE, SOURCE, ISEN
      COMMON /RVFLOW/ TSTART, TSTOP
C
      EQUIVALENCE (VOL(1),ACFM(1),FLAREA(1))
C
      CALL SETRAY(INODES, INOUT)

```

Definitions and units for the variables shown in the preceding coding, as well as for other variables appearing in this chapter, are given in Table 7. Note the call to SETRAY, which initializes many of the variables to appropriate values including the array of molecular weights. As currently implemented, a system of up to 28 nodes plus an ambient node and a null node* (30 total) can be

^{*}The FORTRAN listings inserted in this chapter are taken from CSCDSM, which is based on an earlier version of the compartment models. The earlier version differs slightly from that reflected in Williams (1985). The noticeable differences are in COMMON/RVFLOW/ (see Sect. 3.1.1) and in Table 11 (see Sect. 3.3).

[†]Use of a "null node" permitted generalization of coding such that the number of inlet or outlet nodes to a given node would not need to be specified.

Fig. 14. Logic diagram for transient compartment models. (Sheet 1 of 2).

*WHEN ICOUNT EQUALS IFLAG, COMPARTMENT AND STREAM OUTPUT IS PRINTED. IFLAG IS THE PRINT INTERVAL DIVIDED BY THE SIMULATION INTERVAL.

Fig. 14. Logic diagram for transient compartment models. (Sheet 2 of 2).

Table 7. Variables required in a main program

Variable	Dimension	Definition and Units
ACFM	(30)	VOLUMETRIC FLOW RATE, FT**3/MIN
AMINLN		NATURAL LOG OF MINIMUM NUMBER ACCEPTED BY THE COMPUTER
C1		WEIGHT FRACTION OF HF MONOMER TO TOTAL HF VAPOR
C3		WEIGHT FRACTION OF HF TRIMER TO TOTAL HF VAPOR
C6		WEIGHT FRACTION OF HF HEXAMER TO TOTAL HF VAPOR
DELT		TIME INTERVAL USED FOR TRANSIENT SIMULATION, SEC
DEPVEL		DEPOSITION VELOCITY, FT/SEC
DPAREA	(30)	DEPOSITION AREA, FT**2
FLAREA	(30)	FLOW AREA FOR PRESSURE-DROP-CONTROLLED NODE, FT**2
H	(30)	NODE ENTHALPY, BTU, OR ENTHALPY RATE, BTU/(DELT)
HTAREA	(30)	HEAT TRANSFER AREA, FT**2
HTCOEF	(30)	HEAT TRANSFER COEFFICIENT, BTU/SEC-FT**2-DEG F
ICOUNT		COUNTER TO CONTROL PRINTING OF OUTPUT
IFLAG		FLAG TO CONTROL PRINTING OF OUTPUT
IIN	(30,4)	INLET STREAM NODE NUMBER
INODES		MAXIMUM NUMBER OF NODES ALLOWED BY CODING OF SUBROUTINES
INOUT		MAXIMUM NUMBER OF INLET STREAMS AND OUTLET STREAMS
IOUT	(30,4)	OUTLET STREAM NODE NUMBER
ISEN		VARIABLE TO SPECIFY ISENTROPIC OR ISENTHALPIC RELEASE
ITYPE		VARIABLE TO SPECIFY TYPE OF RELEASE
KRCOEF	(30)	RESISTANCE COEFFICIENT, --, OR RESISTANCE TERM, PSI-SEC**2/LB-FT**3
MASS	(30,9)	NODE MASS, LB, OR MASS FLOW RATE, LB/(DELT)
MAXTIM		MAXIMUM SIMULATION TIME, SEC
PC	(30)	NODE PRESSURE, PSIA
QCOOL	(30)	COOLING RATE, BTU/HR (INPUT) OR BTU/SEC (INTERNAL)
QRATE	(30)	HEAT TRANSFER RATE, BTU/SEC OR BTU/(DELT)
RATE	(9)	COMPONENT MASS FLOW RATE, LB/SEC
RATIO		RATIO OF MOLES OF WATER VAPOR TO TOTAL MOLES OF MOIST AIR
RH		RELATIVE HUMIDITY, % (AS INPUT) OR FRACTION (INTERNAL)
SOLIDS		MASS FLOW RATE OF UF ₆ SOLIDS ONTO THE FLOOR, LB/(DELT)
SOURCE		TOTAL MASS OF RELEASE, LB
TC	(30)	NODE TEMPERATURE, DEG F
TIME		CUMULATIVE SIMULATION TIME, SEC
TRELS		TOTAL RELEASE TIME, SEC
TSTART		START OF REVERSE FLOW
TSTOP		END OF REVERSE FLOW
TSURF	(30)	NODE HEAT TRANSFER SURFACE TEMPERATURE, DEG F
VOL	(30)	NODE VOLUME, FT**3
WMBHF		MOLECULAR WEIGHT OF HF MONOMER, LB/LB MOLE
WMOL	(9)	COMPONENT MOLECULAR WEIGHTS, LB/LB MOLE

accommodated. Nine "components" have been defined which are listed in Table 8. Up to four input streams can be accommodated by a compartment or mix node, and a compartment node can have four output streams. All values in the input and output arrays, IIN and IOUT, are initialized to 30

Table 8. Components incorporated into the transient models

Number	Species	Phase	Molecular weight	Foot-note
1	Air	Vapor	28.966	
2	H ₂ O	Liquid	18.016	
3	H ₂ O	Vapor	18.016	
4	HF	Liquid	20.008	
5	HF	Vapor		<i>a</i>
6	UF ₆	Solid	352.025	<i>b</i>
7	UF ₆	Liquid	352.025	<i>b</i>
8	UF ₆	Vapor	352.025	<i>b</i>
9	UO ₂ F ₂	Solid	308.025	<i>b</i>

^aThe effective molecular weight of HF vapor varies with temperature and the partial pressure of HF.

^bDefault values for UF₆ and UO₂F₂ molecular weights are shown.

(the array element number corresponding to the null node) by SETRAY and are then modified by statements of the following form,

```

C      IIN(I,J) = N
      IOUT(I,J) = N

```

where

IIN(I,J) = the J-th inlet stream to node I,
 IOUT(I,J) = the J-th outlet stream from node I,
 N = the node number corresponding to the J-th inlet/outlet stream for node I.

Values of I, J, and N are easily determined from a schematic flow sheet in which all stream and compartment nodes are numbered.

3.1.2 Input Data Requirements

The type of input data required for a compartment model and the units used in main programs written to date are shown in Table 9. In a few cases, units used internally have been different from

**Table 9. Input conditions required
for a compartment model**

Ambient conditions
Temperature, °F
Pressure, psia
Relative humidity, %
Compartment conditions
Temperature, °F
Pressure, psia
Volume, ft ³
Floor area, ft ²
Heat transfer area, ft ²
Surface Temperature, °F
Cooling rate, Btu/hr
Stream conditions
Flow area, ft ²
Resistance coefficient
or
Volume flow rate, acfm
Source-term conditions
Type of source
4 Liquid HF
5 Vapor HF
7 Liquid UF ₆ (vapor source term)
-7 Liquid UF ₆ (vapor/solid source term)
8 Vapor UF ₆
Temperature, °F
Pressure, psia
Mass, lb
Release duration, sec
Basis for flash
0 Isentropic
1 Isenthalpic
Molecular weight (for UF ₆), lb/lb-mole
Time variables
Time step for simulation, sec
Maximum time for simulation, sec
Print interval, sec

those of the input. Coding used to obtain the proper units, which must follow the READ statement supplying the data, are discussed in the next several paragraphs.

The ambient relative humidity is entered as a percentage value, whereas a fractional value is used within the code. A conditional statement has been included that allows any percentage relative humidity greater than 1 or any fractional relative humidity less than or equal to 1 to be entered. That statement is as follows:

```
IF (RH.GT.1.D0) RH = RH/1.D2
```

The input value for the cooling rate is Btu/hr; however, because the time unit used internally is the second, the cooling rate must be converted to Btu/sec. The following statement has been used:

```
QCOOL(IROOM) = QCOOL(IROOM)/3.6D3
```

Note that a heating rate can be entered as a negative cooling rate. Variables such as IROOM, which describe certain nodes, are given in Table 10.

Table 10. Definitions of variables representing node numbers to be specified in subroutine cells

Variable	Definition
IROOM	Compartment node number
IAMB	Ambient node number
IBLOW	Blower node number
IVENT	Vent or duct node number
ISOURC	Source node number
IINLET	Array element corresponding to a specific inlet stream
IDEP	Deposition node number
ISTRM	Stream node number
IFLOOR	Floor node number

The default molecular weight for UF_6 may need to be changed, particularly for high enrichments of ^{235}U . If the molecular weight for UF_6 is changed, then that of UO_2F_2 must also be changed. Appropriate coding for modifying the molecular weights previously set by the call to SETRAY is the following:

```

C      IF (MW.LT.100.D0) GO TO 10
      WMOL(9) = WMOL(9) - WMOL(6) + MW
      WMOL(6) = MW
      WMOL(7) = MW
      WMOL(8) = MW
C 10 CONTINUE
```

The print interval for output data should be input as an integer multiple of the interval input for simulating the transient. Internally, ICOUNT is used as an index that permits output to be printed when ICOUNT equals IFLAG (see subsequent listing under Sect. 4.1.4). To avoid potential rounding errors resulting in a print interval other than that desired, the following statement is used to calculate IFLAG:

```
IFLAG = IDINT(PRINT/DELT+0.01D0)
```

3.1.3 Steady-State Conditions

Before the transient analysis of a postulated release can begin, it is necessary that all initial conditions existing prior to the release be established. The next section of coding must establish these conditions. The mole ratio of water vapor to moist air, which is assumed to exist both outside and inside the compartment, is established by a call to HUMID. An "ambient volume" is defined* and component masses and a total enthalpy for that volume are evaluated by a call to ROOM for subsequent use when material is drawn into the compartment from the environment. The following statements establish ambient conditions:

```

C      CALL HUMID(IAMB, RH, RATIO)
C      VOL(IAMB) = 1.D9
C      CALL ROOM(IAMB, RATIO)

```

Next, calls to ROOM and SSBLOW are made to establish initial conditions in compartments and constant volume blowers, respectively. ROOM determines the masses of air and water vapor as well as the total enthalpy of the compartment atmosphere. SSBLOW establishes mass and enthalpy flow rates based on the temperature and pressure of the upstream node as well as on the water-vapor-to-moist-air ratio and the time increment to be used in the subsequent transient analysis. These calls take the following form:

```

C      CALL ROOM(IROOM, RATIO)
C      CALL SSBLOW(IBLOW, RATIO)

```

*The ambient volume specified is arbitrary; it need not be 1 billion (ft³) as subsequently shown.

When mass flow rates are known for constant-volume blowers that feed or draw air from the compartment, a mass balance must be incorporated into the coding to determine mass flow rates through those openings through which flow occurs by pressure differential. The mass flow rates established by SSBLOW are based on the simulation interval, DELT (i.e., flow rates are in lb/sec only if DELT = 1.); likewise, the mass flow rates obtained from user-supplied coding must be based on DELT. Once these flow rates are established, the subroutine RESIST is used to calculate a resistance term for determining mass flow rates occurring during the transient analysis. A call to DENTHL must be made to determine the enthalpy flow rate for the steady-state pressure-drop-controlled flow. Representative statements for calls to these subroutines follow.

```

CALL RESIST(IVENT)
C  CALL DENTHL(TC(IVENT), PC(IVENT), IVENT, H(IVENT))

```

As a transient simulation continues after a release is stopped, it is expected that the conditions in the compartment will return to their initial steady-state values. When temperatures of the compartment and ambient surroundings are different, a heat transfer model may be required. A call to HCOEFF can be made to determine a heat transfer coefficient. The subroutine call takes the following form:

```
CALL HCOEFF(IROOM)
```

In many simple models, a constant source term is used. If this is the case, source term characteristics may be established before the transient analysis begins. Coding for more complex sources is discussed in Sect. 4.3. The subroutine call to establish source term characteristics is the following:

```
CALL STERM(ISOURC, IROOM, SOLIDS)
```

3.1.4 Transient Analysis

The transient analysis is performed by (1) determining the flow rates into and out of a compartment at the beginning of a time interval, (2) adding and removing material and energy based on these flow rates over the time interval, (3) reacting components to the extent possible within the compartment after material has been added and removed, (4) determining heat transfer based on the temperature of the compartment and heat transfer surface at the beginning of the time step, and (5) determining the final-phase equilibrium, temperature, and pressure in the compartment at

the end of the time step. Several subroutines are used for the first item, whereas the subroutine COMPRT performs the last four functions.

Coding for the transient analysis section of the main program begins with the following statements:

```

C      TIME = 0.00
C      ICOUNT = IFLAG
C      80 CONTINUE
C      IF (TIME.GE.TRELS) IIN(IROOM,IINLET) = INODES

```

By setting ICOUNT equal to IFLAG, the initial steady-state conditions will be printed. The 80 CONTINUE statement is the point to which the program returns to begin the next time step. If the cumulative time exceeds the source release time, the value of IIN corresponding to the source term inlet node into the compartment node is set equal to INODES so that the source term subsequently corresponds to the null node.

Next, a series of subroutine calls is made to determine transient flow rates. The subroutines TRBLOW and DPFLOW evaluate flows through a constant volume blower and a pressure-drop-controlled pathway, respectively. Deposition rates of condensed phases are calculated by REMOVE. The following statements show the form of these subroutine calls:

```

C      CALL TRBLOW(IBLOW)
C      CALL DPFLOW(IVENT)
C      CALL REMOVE(IROOM,IDEF)

```

Once flow rates have been determined at the beginning of a time interval, which time corresponds to the end of the previous interval, data corresponding to that time can be output. It should be noted that flow rate data must be converted to a per-second basis before being output because those data are internally treated on a per-"time interval" basis.* Coding for that purpose, as well as for determining whether or not to output data for that time, is shown in the following sequence of statements:

*The "time interval" is DELT.

```

      IF (ICOUNT.LT.IFLAG) GO TO 150
C     WRITE ( . . .                *** output compartment data ***
      :
      :
C     DO 130 I130=1,9
      RATE(I130) = MASS(ISTRM,I130)/DELT
C     130 CONTINUE
C     HRATE = H(ISTRM)/DELT
C     WRITE ( . . .                *** output data for node ISTRM ***
      :
      :
      ICOUNT = 0
C     150 CONTINUE
C     TIME = TIME + DELT
C     IF (TIME.GT.MAXTIM) STOP
C     ICOUNT = ICOUNT + 1

```

After conditions for desired streams and compartments have been output for a given time, the program stops execution if the maximum time for the simulation has been reached, as indicated in the preceding coding. Otherwise, compartment conditions at the end of the time interval should be evaluated, and condensed materials being deposited on the floor should be accumulated. Note that compartment models developed to date simply accumulate condensed materials on the floor and that they are removed from any further material or energy-balance considerations. The following sequence of statements concludes all required coding for a main program for a compartment model using the methods described herein:

```

      CALL COMPT(IROOM, INOUT, INODES)
C
C     DO 160 I160=1,9
C         MASS(IFLOOR,I160) = MASS(IFLOOR,I160) + MASS(IDEP,I160)
C     160 CONTINUE
C     IF (ITYPE.EQ.7 .AND. TIME.LE.TRELS) MASS(IFLOOR,6) =
      MASS(IFLOOR,6) + SOLIDS
C
C     GO TO 80
C
C     END

```

3.1.5 Other Considerations

The four preceding sections have outlined the structure of a main program for a transient compartment model. Multicompartment models can also be assembled following the preceding outline. The transient models discussed in Chap. 4 include additional coding to provide specialized output. Following sections in this chapter will discuss the various aspects of the compartment models in greater detail.

3.2 VENTILATION SYSTEM SUBROUTINES

The ventilation system is modeled in a simplistic manner. Four subroutines, which are discussed in Sects. 3.2.1 and 3.2.2, have been written to evaluate steady-state and transient conditions. A mixing subroutine was also written to combine flows from several sources; it is discussed in Sect. 3.2.3.

3.2.1 Steady-State Analysis

The subroutine SSBLOW is used to establish the initial component mass flow rates and the enthalpy rate for moist air flowing through a constant-volume blower. The total moles of moist air passing through the blower over the time interval used for subsequent transient analysis are evaluated using the ideal gas law and the temperature and pressure of the inlet node. Thus,

$$\Delta n_T = (P_i \dot{V}_b \Delta t) / (RT_{R,i} 60) , \quad (62)$$

where

- Δn_T = total moles passing through blower in time interval Δt ,
- P_i = inlet pressure, psia,
- $\dot{V}_b$ = volumetric flow rate of blower, acfm,
- Δt = time interval for transient simulation, sec,
- R = ideal gas constant, psia-ft³/lb mole-°R
= 10.73,
- $T_{R,i}$ = inlet temperature, °R.

The ratio of moles of water vapor to moles of moist air calculated by HUMID (see Sect. 3.5.1) is then used to determine the mass flow rate of water vapor and dry air. The mass of water vapor passing through the blower is given by

$$\Delta m_{H_2O} = \Delta n_T r MW_{H_2O} , \quad (63)$$

where

- Δm = mass passing through the blower in time interval Δt , lb,
- r = ratio of moles of water vapor to moles of moist air,
- MW = molecular weight,
- H_2O = subscript referring to water.

Similarly, the mass of dry air passing through the blower is given by,

$$\Delta M_{\text{air}} = \Delta n_T (1 - r) MW_{\text{air}} , \quad (64)$$

where

air = subscript referring to air.

The enthalpy rate is calculated by a call to DENTHL (see Sect. 2.4).

A general expression for pressure drop is,

$$\Delta P = -K(\dot{m}^2/9266.1 \rho A^2) , \quad (65)$$

where

ΔP = pressure drop, psi,

K = resistance coefficient,

$\dot{m}$ = mass flow rate, lb/sec

= $\Delta m/\Delta t$,

ρ = density, lb/ft³,

A = flow area, ft².

A resistance term that remains constant throughout the analysis can be defined by

$$\phi = K/(9266.1 A^2) , \quad (66)$$

where

ϕ = resistance term, psi-sec²/lb-ft³.

This term is directly evaluated by the subroutine RESIST if a positive resistance coefficient and flow area are specified as input. Otherwise, ϕ is evaluated using the mass flow rate, inlet density, and pressure difference without specific knowledge of the flow area or resistance coefficient by,

$$\phi = -\Delta P \rho / \dot{m}^2 . \quad (67)$$

3.2.2 Transient Analysis

The mass flow and enthalpy rates of streams are assumed to be proportional to the total mass and enthalpy of the node (compartment) feeding the stream. The transient subroutines TRBLOW and DPFLOW calculate volume and mass ratios, respectively, to obtain stream flow rates based on the simulation time interval. In the case of TRBLOW, the volume ratio is given by

$$f_v = (\dot{V}_b \Delta t / 60) / V_i , \quad (68)$$

where

$$\begin{aligned} f_v &= \text{volume ratio,} \\ V_i &= \text{volume of inlet node (e.g., compartment volume), ft}^3. \end{aligned}$$

The mass of each component in the compartment node, as well as the total enthalpy of that node, is multiplied by this volume fraction to obtain the stream rates for the blower node. Thus,

$$\Delta m_{j,b} = f_v m_{j,k} \quad (69)$$

and

$$\Delta H_b = f_v H_i, \quad (70)$$

where

$$\begin{aligned} j &= \text{subscript referring to a particular component,} \\ b, i &= \text{subscripts referring to blower (or later, vent) and inlet nodes, respectively,} \\ \Delta H &= \text{enthalpy of mass passing through the blower in the time interval } \Delta t, \text{ Btu/lb,} \\ H &= \text{enthalpy, Btu/lb.} \end{aligned}$$

The temperature and pressure of the blower node are taken to be same as the compartment node from which it draws.

The overall approach for determining stream rates resulting from pressure-drop-controlled flow is similar. In this case, Eq. 67 is rearranged, and the mass flow rate term is split into a mass term and time term to yield

$$\Delta m = \Delta t \sqrt{|\Delta P| \rho / \phi}, \quad (71)$$

where

$$\begin{aligned} \Delta m &= \text{total mass flowing through a pressure drop controlled flow node over the time interval } \Delta t, \text{ lb,} \\ \Delta P &= \text{pressure difference, psi,} \\ \rho &= \text{density, lb/ft}^3, \\ \phi &= \text{resistance term calculated by RESIST, psi-sec}^2/\text{lb-ft}^3, \\ \Delta t &= \text{time interval used for transient simulation, sec.} \end{aligned}$$

The mass ratio, which is applied in DPFLOW in an analogous fashion to the volume fraction in TRBLOW, is then obtained from

$$f_m = \Delta m / m_{T_i}, \quad (72)$$

where

$$\begin{aligned} f_m &= \text{mass ratio,} \\ m_{T_i} &= \text{total mass in the inlet node.} \end{aligned}$$

Thus, Eqs. 69 and 70 apply if f_v is replaced by f_m . Note that, if the normal direction of flow is reversed by either an increase in the normal downstream pressure or by a decrease in the upstream pressure, then the mass fraction, f_m is made negative and is applied to the normal outlet node. This change of sign and use of the outlet node as the effective inlet makes an inlet stream act as an outlet stream and vice versa. The temperature and pressure of the stream node are those of the effective inlet node.

3.2.3 Stream Mixing

Stream mixing is handled in several ways in the compartment models. When several streams combine and the combined stream enters another compartment, it is reasonable to skip the mixing step by making each individual stream an inlet stream to the downstream compartment. If the combined streams exhaust to the atmosphere, however, the streams need to be mixed such that reactions are allowed to occur and phase equilibrium is established at atmospheric pressure. Such a well-mixed stream can then become a source term for atmospheric dispersion modeling.

The subroutine MIXFLW is used to combine up to four streams and exhaust them at the pressure of the node to which the combined streams exhaust. All streams should originate from blowers because no criteria have been established or implemented in this modeling effort to address flow splitting or the occurrence of backflow in a multipath, induced-draft system (except for a single stream as described in Sect. 3.2.2). The total enthalpy and individual component masses found in the mixed stream before reactions are considered* are calculated by using the following summations:

$$H_m = \sum H_i \quad (73)$$

and

$$m_{j,m} = \sum m_{j,i} \quad (74)$$

where

m, i = subscripts referring to the mixed stream and inlet streams, respectively.

Phase composition is determined by iterating on volume while calling the subroutine COMPRT (see Sect. 3.5.2) which calculates temperature, pressure, and phase composition in a volume, given the total enthalpy and component masses. Volume is varied until the predicted pressure corresponds to the known outlet pressure.

3.3 SOURCE-TERM SUBROUTINE

A simplified source-term subroutine is currently used in the compartment models. Because the coding being developed for the compartment models was required to handle HF, in addition to UF₆, the source-term subroutine, STERM, was written to supply either UF₆ or HF data. For UF₆, three

*Reaction effects are evaluated by COMPRT.

types of source terms can be specified, two for liquid UF_6 and one for vapor. If liquid is released from containment, it is allowed to flash—either isentropically or isenthalpically—to the pressure of the compartment in which the release occurs; however, use of an isenthalpic flash is recommended. The difference in the two liquid source terms is that, in one case, the solids formed are assumed to immediately fall to the floor, while in the other case the solid particles are assumed to be dispersed in the compartment atmosphere. Thus, in the latter case, the solids are subsequently considered in the mass and energy balances of the compartment, whereas in the former case they are treated as inert solids which accumulate on the floor. If UF_6 vapor is released from containment, it is allowed to expand with a resulting increase in temperature. The subroutine FLASH (see Sect. 2.1.8) is called from STERM to perform the flashing and expansion calculations.

While information on the results of flashing or expansion are of interest, such information is not necessarily a requirement when such processes are assumed to occur isenthalpically within a compartment. Under this assumption, all the energy released with the source material, whether liquid or vapor, becomes a part of the total energy within the compartment. Such an assumption is applied to HF releases. The subroutine STERM determines the release rate and the enthalpy rate of the material being released, based on the initial temperature and pressure and whether the HF is specified to be liquid or vapor.

The release rate determined by STERM may be represented by

$$\Delta m = (M/T)\Delta t \quad , \quad (75)$$

where

- Δm = mass of material released per time increment Δt , lb,
- M = total mass of source, lb,
- T = release duration, sec,
- Δt = time increment used for transient simulation, sec.

In the case of a UF_6 liquid release, Δm is subdivided into a solid term and a vapor term. For HF as well as UF_6 vapor releases, the specified phase of the source is the phase assigned to the Δm term. The subroutine DENTHL (see Sect. 2.4) is used to calculate the enthalpy rate for the source term, given the value of Δm assigned to the appropriate component of the mass vector as well as the appropriate temperature and pressure. For UF_6 releases, the appropriate temperature and pressure are those obtained after flashing or expansion, whereas the temperature and pressure supplied as input to STERM are used for HF releases.

The preceding discussion has described the evaluation of a single set of source-term characteristics. A quasi-transient source-term method can be incorporated into a main program which will allow the user to input several sets of constant conditions (mass released in a time interval and the duration of that interval) via the primary input file so that a transient release can be approximated. The required coding is outlined in Table 11. Note that the source term is reevaluated periodically within the transient section of the main program rather than prior to the transient analysis. The source-term vector is replaced by the null vector at the end of the release.

Table 11. Additional coding for quasi-transient source term

	<u>Initial Statements</u>
	DIMENSION MSOURC(5), TSOURC(5)
	<u>Read Statements</u>
	READ (40,*) ITYPE, TC(ISOURC), PC(ISOURC), IRELST,
	* ISEN, MW
C	IF (MW.LT.100.D0) GO TO 10
C	WMOL(9) = WMOL(9) - WMOL(6) + MW
	WMOL(6) = MW
	WMOL(7) = MW
	WMOL(8) = MW
C	10 CONTINUE
C	READ (40,*) (MSOURC(I), TSOURC(I), I=1,IRELST)
	<u>Transient Analysis</u>
	TRELS = 0.D0
C	30 CONTINUE
C	IF (TIME.LT.TRELS) GO TO 35
C	IF (IRELS.GT.IRELST) GO TO 35
C	TRELS = TSOURC(IRELS)
C	SOURCE = MSOURC(IRELS)
C	CALL STERM (ISOURC, IROOM, SOLIDS)
C	TRELS = 0.D0
C	DO 32 I32=1,IRELS
C	TRELS = TRELS + TSOURC(I32)
C	32 CONTINUE
C	IRELS = IRELS + 1
C	35 CONTINUE
C	IF (TIME.GE.TRELS) IIN(ICOMPT, INNUMB) = INODES

3.4 CONDENSATE DEPOSITION SUBROUTINE

A very simple condensate deposition model has been used in the subroutine REMOVE. The model assumes a constant deposition velocity for all condensed phases of 1 cm/sec, which is specified in SETRAY as 0.033 (ft/sec). All condensed material within a slab having a volume specified by the product of the deposition area, the deposition velocity, and the time interval used for the transient simulation is assumed to be removed over the time interval. Thus, the amount deposited is given by

$$\Delta m_{j,d} = [(A_f v_d \Delta t) / V_c] m_{j,c} \quad , \quad (76)$$

where

- $m_{j,d}$ = mass of component j deposited over the time interval Δt ,
- A_f = deposition (floor) area, ft^2 ,
- v_d = deposition velocity, ft/sec ,
- Δt = time interval for transient simulation, sec ,
- V_c = compartment volume, ft^3 ,
- $m_{j,c}$ = mass of component j present in compartment, lb .

Equation 76 applies only to condensed components. It is important to remember that the compartment volume from which the condensed material is deposited is assumed to be well mixed, so the amount of material removed by deposition in each time interval is (in general) continually varying.

3.5 COMPARTMENT MASS AND ENERGY BALANCE SUBROUTINES

Conditions within a compartment at any specific time are assumed to be homogeneous. In other words, the temperature, pressure, component concentrations, and phase splits are uniform throughout the compartment volume at any specific time. Within the overall compartment model structure, compartment conditions are determined for two circumstances. First, normal steady-state initial conditions must be established; second, conditions must be evaluated at the end of each time interval of the transient simulation. Sect. 3.5.1 discusses the steady-state mass and energy balance; Sect. 3.5.2 discusses evaluation of transient conditions.

3.5.1 Initial Conditions

Initial conditions within a compartment are determined on the basis of ambient temperature, pressure, and relative humidity; compartment temperature, pressure, and volume; and the assumption that the ideal gas law applies. First, the ratio of moles of water vapor to moles of moist air is calculated using ambient conditions and the following equation:

$$r = [RH(P_{\text{H}_2\text{O}}^\circ)]/P \quad , \quad (77)$$

where

- r = molar ratio of water vapor to moist air,
- RH = fractional relative humidity,
- $P_{\text{H}_2\text{O}}^\circ$ = vapor pressure of water, psia
- P = total pressure, psia .

The subroutine HUMID calculates r using the subroutine PHFH2O (see Sects. 2.2.3 and 2.2.7) to evaluate $P_{\text{H}_2\text{O}}^\circ$. Note that it is assumed that all compartments and streams, as well as the ambient surroundings, have the same composition prior to a release.

Once the molar ratio is determined, the mass quantities of air and water for each node can be determined. The procedure for evaluating these quantities for ventilation system nodes has been outlined in Sect. 3.2.1. For a compartment, the total moles of vapor contained in the compartment at a given temperature and pressure are calculated assuming the ideal gas law,

$$n_T = PV/RT_R, \quad (78)$$

where

n_T = total number of moles in a compartment,

P = compartment pressure, psia,

V = compartment volume, ft³,

R = ideal gas constant, psia-ft³/lb-moles-°R
= 10.73,

T_R = compartment temperature, °R.

The water-vapor-to-moist-air-molar ratio is then applied along with the molecular weights of water and air to obtain the component masses of air and water vapor in the compartment node using the following relationships,

$$m_{H_2O} = n_T r MW_{H_2O} \quad (79)$$

and

$$m_{air} = n_T (1 - r) MW_{air}, \quad (80)$$

where

m = mass, lb,

MW = molecular weight,

H_2O, air = superscripts referring to water and air, respectively.

A call to DENTHL (see Sect.2.4) is made to evaluate the total enthalpy of the compartment node. These calculations for a compartment are programmed into the subroutine ROOM.

To close the energy balance for the compartment, heat transfer from process equipment or other heat sources within a compartment must be considered; otherwise, in the subsequent transient simulation, the temperature will eventually approach ambient temperature rather than the initial room temperature. The subroutine HCOEFF has been written to evaluate a heat transfer coefficient, based on normal steady-state conditions, which is used in the transient simulation. The coding has been written to handle up to four inlet streams. The heat added to a compartment by heat transfer is found from

$$Q = [H_c(\dot{m}_i/m_c) - \dot{H}_i + Q_{cool}\Delta t]/\Delta t, \quad (81)$$

where

Q = heat transferred from process equipment, Btu/sec,

H_c = total enthalpy of compartment atmosphere, Btu,

$\dot{m}_i$ = total inlet mass flow rate of all inlet stream, lb/ Δt ,

m_c = total mass of compartment atmosphere, lb,

$\dot{H}_i$ = total inlet enthalpy rate of all inlet streams, Btu/ Δt ,

Q_{cool} = cooling rate, Btu/sec,

Δt = time interval for transient simulation, sec.

Note that inlet mass flow and enthalpy rates are based on the time interval used for the transient simulation and that a negative cooling rate is a heating rate. The heat transfer coefficient is then determined from

$$h = Q/[A(T_s - T_c)] , \quad (82)$$

where

- h = heat transfer coefficient, Btu/sec-ft²-°F,
- A = heat transfer surface area, ft²,
- T_s = heat transfer surface temperature, °F,
- T_c = temperature of compartment atmosphere, °F.

Of the variables h , A , and T_s , T_s is probably most important. The product of h and A is assumed to remain constant throughout the analysis.

3.5.2 Transient Behavior

The procedure for analyzing transient behavior is divided between the main program and the subroutine COMPRT. The approach is to determine various mass flow and enthalpy rates based on conditions at the beginning of the time step, then to accumulate mass and energy terms for the compartment and react components to the extent possible, and finally to determine temperature, pressure, and phase compositions at the end of the time step. Mass flow and enthalpy rates, except for heat transfer rates, are evaluated in subroutines called from the main program. The other calculations are carried out in COMPRT.

COMPRT can be divided into two sections. In the first, mass and energy terms are accumulated and components are reacted to determine the final overall composition and total enthalpy. In the latter section, the final temperature, pressure, and phase composition are determined iteratively using assumed temperatures as the basis for iteration and total enthalpy as the basis for closure. Accumulating mass and energy terms is done by adding inlet stream terms and subtracting outlet stream terms from the comparable compartment node terms. Thus, for each component (and prior to considering the effect of chemical reactions),

$$m_j|_f = m_j|_i + \sum_{in=1}^4 \Delta m_j|_{in} - \sum_{out=1}^4 \Delta m_j|_{out} , \quad (83)$$

where

- m = mass of a component in a compartment, lb,
- Δm = mass passing through a node over the transient simulation time interval Δt , lb,
- f, i = subscripts referring to final and initial conditions, respectively,
- in, out = summation indices for inlet and outlet streams, respectively.

Up to four inlet and four outlet streams can be assigned for each compartment. If reverse flow occurs in a pressure drop controlled node, the subroutine DPFLOW (see Sect. 3.2.2) assigns negative mass flow and energy rates so that when COMPRT accumulates terms the proper results are obtained.

The only reaction currently considered in COMPRT is the reaction of UF_6 and H_2O . It is assumed to go to completion, being limited only by the exhaustion of one of the reactants, according to the following equation:

The heat released by this reaction is 25,199 Btu/lb-mole of H_2O reacted based on a reference temperature of 77°F (see Sect. 2.1.7). If H_2O is the limiting reactant,

$$m_{\text{UF}_6}|_f = m_{\text{UF}_6}|_i - m_{\text{H}_2\text{O}}|_i \text{MW}_{\text{UF}_6} / (2 \text{MW}_{\text{H}_2\text{O}}) \quad (85)$$

$$m_{\text{UO}_2\text{F}_2}|_f = m_{\text{UO}_2\text{F}_2}|_i + m_{\text{H}_2\text{O}}|_i \text{MW}_{\text{UO}_2\text{F}_2} / (2 \text{MW}_{\text{H}_2\text{O}}) \quad (86)$$

$$m_{\text{HF}}|_f = m_{\text{HF}}|_i + 2 m_{\text{H}_2\text{O}}|_i \text{MW}_{\text{HF monomer}} / \text{MW}_{\text{H}_2\text{O}} \quad (87)$$

$$m_{\text{H}_2\text{O}}|_f = 0 \quad (88)$$

$$\Delta H_{\text{rxn}} = 25199 m_{\text{H}_2\text{O}}|_i / \text{MW}_{\text{H}_2\text{O}} \quad (89)$$

where

ΔH_{rxn} = heat released by the reaction of UF_6 with H_2O , Btu.

If UF_6 is the limiting reactant,

$$m_{\text{H}_2\text{O}}|_f = m_{\text{H}_2\text{O}}|_i - 2 m_{\text{UF}_6}|_i \text{MW}_{\text{H}_2\text{O}} / \text{MW}_{\text{UF}_6} \quad (90)$$

$$m_{\text{UO}_2\text{F}_2}|_f = m_{\text{UO}_2\text{F}_2}|_i + m_{\text{UF}_6}|_i \text{MW}_{\text{UO}_2\text{F}_2} / \text{MW}_{\text{UF}_6} \quad (91)$$

$$m_{\text{HF}}|_f = m_{\text{HF}}|_i + 4 m_{\text{UF}_6}|_i \text{MW}_{\text{HF monomer}} / \text{MW}_{\text{UF}_6} \quad (92)$$

$$m_{\text{UF}_6}|_f = 0 \quad (93)$$

$$\Delta H_{\text{rxn}} = 50398 m_{\text{UF}_6}|_i / \text{MW}_{\text{UF}_6} \quad (94)$$

In Eqs. 85 through 94, initial masses correspond to the final masses obtained from Eq. 83. Note that, although the compartment model in general treats each phase of a chemical component as an individual component, all phase components of a chemical component are combined prior to evaluating the effects of chemical reactions. Subsequently, the amount of a chemical component in each phase is reallocated in the process of determining the final temperature and pressure.

The amount of heat transferred from process equipment is given by

$$Q_{\Delta t} = hA(T_s - T_c)\Delta t \quad , \quad (95)$$

where $Q_{\Delta t}$ is the amount of heat transferred over the time interval used for the transient simulation. The units of $Q_{\Delta t}$ are Btu/ (Δt) , which is consistent with other enthalpy rate terms calculated internally by the various compartment model subroutines.

Once the heat of reaction and heat transferred from process equipment have been determined, the total enthalpy with respect to the reference conditions is given by,

$$H|_f = H|_i + \sum_{in=1}^4 H_{in} - \sum_{out=1}^4 H_{out} + \Delta H_{rxn} + Q_{\Delta t} - Q_{cool}\Delta t \quad , \quad (96)$$

where

H_{in}, H_{out} = enthalpy rates of inlet and outlet streams Btu/ Δt ,

Q_{cool} = cooling rate, Btu/sec,

Δt = time interval for transient simulation, sec.

Note that ΔH and $Q_{\Delta t}$ in effect have the units of Btu because Δt has already been accounted for in calculating these values.

The iterative procedure to determine the temperature, pressure, and phase composition at the end of the time step begins by assuming a final temperature. For condensible components, their respective vapor pressures are calculated, at the assumed final temperature and the amount of each component in the compartment is compared with the maximum amount of that component allowed in the compartment on the basis of the assumed temperature and calculated vapor pressure and the assumption the ideal gas law applies. If the amount of the component exceeds the maximum, the excess is placed in the condensed phase. For UF_6 , the calculation is performed in COMPRT except for the calculation of vapor pressure, which is done by a call to VPRUF6. For the $HF-H_2O$ subsystem, these calculations are performed in PHASE which is described in Sects. 2.2.6 and 2.2.7. The number of moles in the vapor phase is determined and a total pressure is obtained using the ideal gas law and the assumed temperature. Given the phase composition and pressure at the assumed temperature, a total enthalpy is calculated. If the total enthalpy calculated for the assumed temperature exceeds the total enthalpy calculated by Eq. 154, a lower "assumed" temperature is selected, or vice versa, and the process is repeated until the two values of enthalpy agree. When the enthalpies agree, then the conditions determined for the last assumed temperature are known to be the conditions for the end of the time step.

4. GDP SAR COMPARTMENT MODEL MAIN PROGRAMS

The GDP SAR compartment models are specialized main programs developed to simulate the ventilation patterns of specific buildings in which releases were postulated to occur. The structure of these programs follows the general outline presented in Sect. 3.1; additional coding provides input for PLUME code analysis of the atmospheric dispersion of released materials.

The additional coding that provides PLUME code input data was developed on the basis of several arbitrary criteria. Because the PLUME code is a steady-state atmospheric dispersion model, time-averaged data rather than transient data are required. By using the MERGE program (Holt 1986), it is possible to approximate the behavior of a transient release by a series of steady-state releases; therefore, coding was developed to provide several input data sets for each simulation. Generally, each transient source was divided into no more than six time-averaged, "steady-state" releases. The first two releases were limited to the time of the postulated release of UF_6 from containment. The first release assumed complete reaction of all released UF_6 with excess water remaining. The second release assumed excess UF_6 and, hence, no water release. The next two releases occur with UF_6 present but after the release from containment has ceased. During these releases, all water entering the compartment is totally reacted. A flag is set at the beginning of the third release to identify the total amount of UF_6 remaining. The third release continues until the amount of UF_6 remaining is less than half the flagged value. The fourth release continues until all UF_6 has been exhausted or reacted. The final two releases exhaust no UF_6 but do exhaust H_2O , HF , UO_2F_2 , and air. The total amount of HF present at the beginning of the fifth release is flagged, and this release continues until 75% of the flagged amount has been exhausted. The sixth release continues until the specified maximum simulation time is reached or until other criteria (minimum concentrations of the total uranium and HF) stop the transient analysis. As implemented, the flags are set continuously until they are actually needed to control development of output data, at which time they are fixed. Thus, for a small release of UF_6 that does not consume all the moisture in the compartment atmosphere, only three release data sets would be generated, corresponding to releases 1, 5, and 6.

Based on the above transition criteria, component masses and exhaust temperatures are summed during the transient analysis and subsequently converted to time-averaged values. By reference to Fig. 14 in Sect. 3.1, the summations for the various plumes occur just before time is incremented ($\text{TIME} = \text{TIME} + \text{DELT}$), whereas the averaging occurs last (between $\text{TIME} > \text{MAXTIM}$ and STOP). Output includes average total mass release rate, average temperature, total and gas phase compositions as weight fractions relative to total mass, and exhaust velocity obtained by applying the ideal gas law to gas phase components to obtain the density. Exhaust area is specified as input.

It should be noted that for induced-draft systems, such as the feed and withdrawal building (see Sect. 4.1), only two release data sets are generated for potential reverse flow paths. The first release resulting from reverse flow corresponds to the first release discussed above. Transition-criterion to move from the first to the second release is the same as above; however, the second release resulting from reverse flow is not terminated until normal flow is reestablished. Reestablishment of normal flow has generally been observed to occur at approximately the end of a postulated release for containment.

The transient simulation is stopped when the maximum simulation time specified in the input is reached or earlier, if the following criteria are met. Based on toxicity data, HF and UF_6

concentrations less than 1 mg/m^3 and 0.1 mg/m^3 , respectively, are assumed to have no effect. If exhaust source concentrations drop below both of these concentrations, releases from that source are terminated. When concentrations in all rooms drop below these values, the transient simulation is terminated.

4.1 FEED AND WITHDRAWAL BUILDING TRANSIENT MODEL

A schematic of the feed and withdrawal building model is shown in Fig. 15. All GDP buildings used for UF_6 feed and withdrawal are ventilated with induced-draft systems, which exhaust air from the compartment by means of a blower or fan while air is drawn into the compartment by pressure difference. The program FWDTRN was written to simulate this system. A list of required input is given in Table 12.

Besides the rather straightforward modifications to the general outline, the modeling of an induced-draft system presented several problems. Initially, coding was added to allow the calculation of the internal compartment pressure (based on mass flow rate, specified inlet area, and resistance coefficients) or the internal pressure could be specified. Allowing INDRFT to calculate the internal pressure, though, generally leads to such a small pressure difference between the compartment and the surroundings under steady-state conditions that subsequent transient analysis is usually unstable. Specifying a pressure about 0.05 to 0.1 psi lower than ambient pressure avoids long-term instability (note that the resistance term for Node 2 would be based on Eq. 94), whereas little difference is found in results obtained over such a range of initial internal pressures. Short-

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Fig. 15. Schematic for the transient feed and withdrawal building model (FWDTRN).

Table 12. Input Data Required for FWDTRN

Ambient data (Node 7)
Temperature, °F
Pressure, psia
Relative Humidity, %
Compartment data (Node 1)
Temperature, °F
Pressure, psia
Volume, ft ³
Floor area, ft ²
Heat transfer area, ft ²
Surface temperature, °F
Cooling rate, Btu/hr
Inlet air data (Node 2)
Flow area, ft ²
Resistance coefficient
Exhaust air data (Node 4)
Volume flow rate, acfm
Source term data (Node 3)
Type of source
Temperature, °F
Pressure, psia
Mass, lb
Release duration, sec
Basis for flash
Molecular weight (for UF ₆), lb/lb-mole
Time data
Time step for simulation, sec
Maximum time for simulation, sec
Print interval, sec
Plume data
Source location, ft
Minimum output interval, ft
Maximum simulation distance, ft
Local elevation above sea level, ft
Release height above local elevation, ft
Plume source radii, ft
Atmospheric stability class
Wind speed, ft/sec

term transient instabilities resulting from reverse flow (occurring when internal pressure exceeds external pressure) were overcome by limiting the amount of material exhausted over a time step to that quantity that would reduce the compartment pressure to the ambient pressure, given the conditions existing in the compartment at the beginning of the time step. Coding for these special conditions appears between the statements 20 CONTINUE and 26 FORMAT and between 30

CONTINUE and 36 CONTINUE in the FWDTRN program listing. The bases for this latter section of coding are the following volumetric relationships

$$V_{\max} = P_i V_i / P_{\text{amb}} \quad (97)$$

$$\Delta V_{\max} = V_{\max} - \dot{V}_b \Delta t / 60 - V_i, \quad (98)$$

where

$V_{\max}$ = volume of compartment contents if expanded to ambient pressure, ft^3 ,

P_i = compartment pressure, psia,

V_i = compartment volume, ft^3 ,

P_{amb} = ambient pressure, psia,

$\Delta V_{\max}$ = maximum volume of compartment contents that can be exhausted via reverse flow through the inlet over the transient simulation time interval Δt , ft^3 ,

$\dot{V}_b$ = blower volumetric flow rate, acfm,

Δt = time interval for transient simulation, sec.

If the product of the fraction defined by the ratio of $\Delta V_{\max}$ to $V_{\max}$ and the mass of material in the compartment is less than the mass of material that would be exhausted based on the results of the call to DPFLOW, then the mass of each component exhausted by reverse flow through the inlet is limited to the product of the fraction $\Delta V_{\max}/V_{\max}$ and the component mass in the compartment. The enthalpy rate is also limited, if the mass exhausted is limited, to the product of the fraction $\Delta V_{\max}/V_{\max}$ and the enthalpy of the compartment.

Also added to the general outline was special coding to produce input to the PLUME code, based on the considerations discussed in the introductory section of this chapter. By reference to Fig. 15, up to six plume input data sets can be generated for exhaust air stream 4, whereas one or two data sets will be generated for inlet air stream 2, which becomes an exhaust stream while UF_6 is being released. Normal flow through stream 2 is rapidly reestablished once the release ends.

FWDTRN was derived for summer ventilation conditions, but it was also used to model postulated winter releases after minor modification (not described herein) because blowers are not operated in winter. In this case an exhaust rate one-tenth that of the induced rate was assumed.

4.2 CASCADE BUILDING TRANSIENT MODEL—SUMMER MODE

The ventilation system used in the GDP cascade building produces significantly different airflow patterns under summer and winter conditions. Furthermore, because of the tremendous size of these buildings, only a portion of the building is considered to be involved in a release. The main program CSCDSM was developed to model the transient behavior occurring in a cascade building during and subsequent to a release postulated to occur during the summer. A schematic for the model is shown in Fig. 16. In the summer, the ventilation system is a once-through system in which air is drawn into the operating floor by pressure difference, forced to the cell floor by large blowers, and then exhausted partially by pressure difference and partially by the motor exhaust fans. The volume of the cell floor affected by the release is assumed to be a portion of the total volume, usually one-eighth. The volume specified as input for the operating floor is not critical (as long as it

Fig. 16. Schematic for the transient, summer-ventilation-pattern cascade building model (CSCDSM).

is positive) because it does not become involved in the release. Depending on the fraction of the cell floor involved in the release, the motor exhaust stream from the contaminated volume may be diluted with air exhausted from uninvolved sections of the cell floor. Up to 18 plume data sets may be generated by CSCDSM. Up to six plumes may originate from the combined motor exhaust. Although the cell floor exhaust is represented by one pathway in the schematic, there are in fact two pathways: roof-top ventilators and louvers on the walls. Up to six plumes may result from each of these two release pathways; the amount released via each pathway is based on the ratios of the normal flows for each pathway to the total combined flow via these pressure-drop-controlled exhaust pathways. Input requirements for CSCDSM are given in Table 13.

Table 13. Input Data Required for CSCDSM

Ambient data (Node 11)	
Temperature, °F	
Pressure, psia	
Relative humidity, %	
Cell floor compartment data (Node 1)	
Temperature, °F	
Pressure, psia	
Volume, ft ³	
Floor area, ft ²	
Heat transfer area, ft ²	
Surface temperature, °F	
Cooling rate, Btu/hr	
Operating floor compartment data (Node 10)	
Temperature, °F	
Pressure, psia	
Volume, ft ³	
Supply air stream data (Node 2)	
Volume flow rate, acfm	
Contaminated motor exhaust stream data (Node 5)	
Volume flow rate, acfm	
Uncontaminated motor exhaust stream data (Node 8)	
Volume flow rate, acfm	
Cell floor exhaust stream data (Node 4)	
Flow area, ft ²	
Normal flow rate through roof vent, ft ³ /min	
Normal flow rate through wall louvers, ft ³ /min	
Combined motor exhaust stream data (Node 9)	
Flow area, ft ²	
Source term data (Node 3)	
Type of source	
Temperature, °F	
Pressure, psia	
Number of sequential releases	
Mass, lb	
Release duration, sec	} (pairs of data)
Basis for flash	
Molecular weight (for UF ₆), lb/lb-mole	
Time data	
Time step for simulation, sec	
Maximum time for simulation, sec	
Print interval, sec	

Table 13. (continued)

Plume data
Source location, ft
Minimum output interval, ft
Maximum simulation distance, ft
Local elevation above sea level, ft
Release height above local elevation, ft
Plume source radii, ft
Atmospheric stability class
Wind speed, ft/sec

Because instabilities had been previously encountered in developing the induced-draft feed and withdrawal building model (and for CSCDSM because the operating floor does not become involved in the transient analysis), initial pressures for both cell and operating floors must be specified in both CSCDSM and CSCDWN. The specified pressure for the operating floor must be below the ambient pressure, and the cell floor pressure must be greater than the ambient.

Several of the postulated release scenarios for the GDP SARs involved quasi-transient releases (i.e., several step changes in release rate). The coding for a quasi-transient source term, which was presented in Sect. 3.3, (see Table 11) was incorporated into CSCDSM.

4.3 CASCADE BUILDING TRANSIENT MODEL—WINTER MODE

The ventilation system for the cascade building recycles air between the operating and cell floors when operating in the winter mode. The winter inlet air rate is about half the summer rate, the motor exhaust being almost totally recycled (total recycle is assumed in the model). Because of the ventilation system arrangement, at least one-eighth of the building is expected to be contaminated by a release. The recirculating air contaminates a portion of the operating floor and, possibly, additional cell floor areas. A schematic for CSCDWN is shown in Fig. 17, and input requirements are listed in Table 14.

Up to 20 plume data sets are generated by CSCDWN. Six plumes may result from release pathway 2, and 12 plumes may result from release pathway 3, which is split between the roof vent and wall louvers. Two plumes may result via reverse flow through pathway 1. As mentioned previously, the coding for a quasi-transient source term has been incorporated into CSCDWN.

4.4 BATCH MODEL

The BATCH program can be used to simulate releases into a closed room or steady-state releases into a ventilation system. When UF_6 or HF is released into a closed compartment, the final pressure may increase above the ambient pressure. For steady-state releases the batch model assumes that the source term represents a constant release rate and that the specified "compartment" volume is a constant volume flow rate. The two streams are mixed and exhausted at the ambient pressure using the MIXFLW subroutine (see Sect. 3.2.3). Input conditions required for BATCH are listed in Table 15. This particular program is interactive in contrast to the main programs previously discussed which require input files.

Fig. 17. Schematic for the transient, winter-ventilation-pattern cascade building model (CSCDWN).

Table 14. Input Data Required for CSCDWN

Ambient data (Node 18)
Temperature, °F
Pressure, psia
Relative humidity, %
Operating floor compartment data (Node 14)
Temperature, °F
Pressure, psia
Volume, ft ³
Floor area, ft ²
Heat transfer area, ft ²
Surface temperature, °F
Cooling rate, Btu/hr
Cell floor nonrelease sector compartment data (Node 15)
Volume, ft ³
Floor area, ft ²
Heat transfer area, ft ²
Surface temperature, °F
Cooling rate, Btu/hr
Cell floor release sector compartment data (Node 16)
Temperature, °F
Pressure, psia
Volume, ft ³
Floor area, ft ²
Heat transfer area, ft ²
Surface temperature, °F
Cooling rate, Btu/hr
Inlet air stream data (Node 1)
Flow area, ft ²
Nonrelease sector exhaust stream data (Node 2)
Flow area, ft ²
Release sector exhaust stream data (Node 3)
Flow area, ft ²
Normal flow rate through roof vent, ft ³ /min
Normal flow rate through wall louvers, ft ³ /min
Nonrelease sector supply air stream data (Node 4)
Volume flow rate, acfm
Release sector supply air stream data (Node 5)
Volume flow rate, acfm
Nonrelease sector motor exhaust stream data (Node 6)
Volume flow rate, acfm
Release sector motor exhaust stream data (Node 7)
Volume flow rate, acfm

Table 14 (continued)

Source term data (Node 17)

Type of source

Temperature, °F

Pressure, psia

Number of sequential releases

Mass, lb

Release duration, sec

} (pairs of data)

Basis for flash

Molecular weight (for UF₆), lb/lb-mole

Time data

Time step for simulation, sec

Maximum time for simulation, sec

Print interval, sec

Plume data

Source location, ft

Minimum output interval, ft

Maximum simulation distance, ft

Local elevation above sea level, ft

Release height above local elevation, ft

Plume source radii, ft

Atmospheric stability class

Wind speed, ft/sec

Table 15. Input data required for batch

Title

Ambient data

Temperature, °F

Pressure, psia

Relative humidity, %

Compartment data

Temperature, °F

Pressure, psia

Volume, ft³

Open or closed

Source term data

Type

Temperature, °F

Pressure, psia

Molecular weight, lb/lb-mole (for UF₆)

Mass, lb

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