IN SITU VITRIFICATION MODEL DEVELOPMENT
AND IMPLEMENTATION PLAN

Revision 1.0

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ABSTRACT

This document describes the In Situ Vittrification (ISV) Analysis Package being developed at the INEL to provide analytical support for (ISV) safety analysis and treatment performance predictions. Mathematical models and features which comprise this analysis package are presented and the proposed approach to model development and implementation is outlined. The objective of this document is twofold: (1) to define preliminary design information and modeling objectives so that ISV modeling personnel can effectively modify existing models and formulate new models which are consistent with the objectives of the ISV treatability study and (2) to provide sufficient technical information for internal and external reviewers to detect any shortcomings in model development and implementation plans.

EXECUTIVE SUMMARY

The Idaho National Engineering Laboratory (INEL) In Situ Vittrification (ISV) treatability study has the objectives of (1) determining the site and public safety of implementing the ISV process at the INEL and (2) determining the suitability of the ISV process for remediating the wastes buried at the INEL. To accomplish these objectives the ISV program has developed a safety strategy that integrates analysis, testing, process design, probabilistic risk assessment, safety assessment, and process evaluation. One component of this strategy is a proposed analysis package that simulates the ISV process by a set of model equations which are derived from fundamental physical and chemical principles. This analysis package will provide quantitative relationships between important process parameters and play a key role in the planning and interpretation of experiments, process design, and safety analysis.

The ISV analysis package will be aimed at approximating the important physical phenomena that occur during the ISV process, both subsurface and above ground. To this end the INEL has initiated development and implementation of a suite of computer models which will comprise the ISV analysis package. The models are divided into seven major categories.

- Electrical resistance heating
- Thermal energy transport with melting
- Viscous flow in the melt zone
- Process chemistry
- Off-gas release from the melt
- Gas transport in the soil surrounding the vitrified zone
- Flow and combustion in the off-gas confinement hood

This report presents the primary models and features, including the governing equations and their numerical approximations, which will comprise the ISV analysis package and outlines the proposed approach to model development and implementation. The objective of this document is twofold:

- To define preliminary design information and modeling objectives so that ISV modeling personnel can effectively modify existing models and formulate new models which are consistent with the objectives of the ISV treatability study and
- To provide sufficient technical information for internal and external reviewers to detect any shortcomings in model development and implementation plans.

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ISV IMPLEMENTATION PLAN NOMENCLATURE

<u>Symbol</u>	<u>Description of Symbol</u>
a, b, c, d, e	coefficients in equilibrium constant
$b_{n,L}$	mass of gas species n in bubbles of size L
A, B, C	finite element matrices
c_p	heat capacity at constant pressure
c_m	mixture volumetric heat capacity
D	diffusion coefficient
E	activation energy
f	fugacity
F	Gibbs free energy
F_i	finite element vector
g	gravitational acceleration
G	rate of generation of species
h	heat transfer coefficient
H	enthalpy
J_i	current density vector
j_i	diffusional mass flux vector
k	thermal conductivity
k^e	electrical conductivity
k'	reaction rate coefficient
k^g	gas permeability
K_c	concentration equilibrium coefficient
L	latent heat of fusion
m	number of moles
M	finite element matrix
n_k^{ij}	vector normal to surface Γ_{ij} , pointing from Ω_i to Ω_j
n_L	number concentration of bubbles of size L
N	number of nodes in mesh
p	pressure
P	electrical power source
q_i	heat flux vector
Q	chemical heat source
r	reaction rate

<u>Symbol</u>	<u>Description of Symbol</u>
R	universal gas constant
S	rate of gas generation
S_L	rise speed of bubbles of size L
t	time
T	temperature
u_i	velocity vector
U	internal energy
V_i	velocity of melting interface
v	specific volume
w, w'	finite element weight functions
W	molecular weight
x_i	position vector
y	mole fraction
Z	generic chemical symbol

Greek

α	coefficient of thermal expansion
β	isothermal compressibility
γ	density ratio of solid to liquid soil
δ	characteristic length scale
δ_{ij}	unit tensor
Δ	increment
ϵ	emissivity
ζ	temperature exponent
η	heat of formation
θ	time integration parameter
κ	Stefan-Boltzman constant
λ	lagrange multiplier
μ	dynamic viscosity
μ_2	second coefficient of viscosity
ν	rate coefficient
π	porosity
ρ	density
σ_{ij}	stress tensor
τ_{ij}	deviatoric stress tensor

<u>Symbol</u>	<u>Description of Symbol</u>
Γ_i	1D electrode line
Γ_{ij}	2D surface separating Ω_i and Ω_j
ϕ	electrostatic potential
ϕ^j	electrostatic potential at electrode j
ψ, χ	finite element basis functions
ω	boundary mass fluxes
Ω	3D region
<u>Mathematical</u>	
$\rightleftharpoons$	forward and backward reactions
$\cup$	union (combined with)
$:$	double tensorial contraction
<u>Subscripts</u>	
b	backward (reaction)
f	forward (reaction)
i	vector quantity
ij	2nd rank tensor
r	reaction r
<u>Superscripts</u>	
c	convective component
k	species
n	time level
R	radiation component
s	surface component
T	matrix transpose
<u>Overlines</u>	
$\wedge$	approximate value
$\dot{}$	time derivative

IN SITU VITRIFICATION MODEL DEVELOPMENT AND IMPLEMENTATION PLAN

1. INTRODUCTION

The Idaho National Engineering Laboratory (INEL) In Situ Vitrification (ISV) treatability study has the objectives of (1) determining the site and public safety of implementing the ISV process at the INEL and (2) determining the suitability of the ISV process for remediating the wastes buried at the INEL. To accomplish these objectives the ISV program has developed a safety strategy that integrates analysis, testing, process design, probabilistic risk assessment, safety assessment, and process evaluation. A schematic representation of this strategy is shown in Figure 1.1. One component of this strategy is a proposed analysis package that simulates the ISV process by a set of model equations which are derived from fundamental physical and chemical principles. This analysis package will provide quantitative relationships between important process parameters and play a key role in the planning and interpretation of experiments and in process design. Also, the analysis package will play an important role in predicting hazard initiating events and quantifying probable consequences.

The ISV analysis package will be aimed at approximating the important physical phenomena that occur during the ISV process, both subsurface and above ground. To this end the INEL has initiated development and implementation of a suite of computer models which will comprise the ISV analysis package. The models are divided into seven major categories.

- Electrical resistance heating
- Thermal energy transport with melting
- Viscous flow in the melt zone
- Process chemistry
- Off-gas release from the melt

Figure 1.1 Integrated safety assessment, design, and process evaluation.

- Gas transport in the soil surrounding the vitrified zone
- Flow and combustion in the off-gas confinement hood

This report presents the primary models and features which will comprise the ISV analysis package and outlines the proposed approach to model development and implementation. The objective of this document is twofold:

- To define preliminary design information and modeling objectives so that ISV modeling personnel can effectively modify existing models and formulate new models which are consistent with the objectives of the ISV treatability study and
- To provide sufficient technical information for internal and external reviewers to detect any shortcomings in model development and implementation plans.

Detailed requirements for these models are described in Modeling Requirements For In Situ Vitrification.¹ Programmatic requirements have also been established for effective ISV model implementation and include:

- Prioritization of the phenomena and processes to be modeled according to their importance with respect to safety and environmental concerns,
- Incremental construction of the analysis package so that separate effects analyses can be conducted before the final version is complete,
- Incorporation and modification of computer codes available from various DOE laboratories to form a modular ISV analysis package,
- Documentation of the analysis package to include a Requirements Document and Implementation Plan, a Code Validation and Verification Plan, and a Code Description Manual, and

- Review of the ISV model implementation by internal and external peers.

1.1 Approach to Model Development and Implementation

Models of the physical and chemical processes which are likely to be dominant in determining the response of the ISV process are noted in the foregoing section. In the first stage of model development, simplified versions of these models will be considered. These simpler models are:

- 3D electrostatic field model
- 3D heat conduction/convection model including melting
- 3D Newtonian viscous fluid flow model
- Equilibrium chemistry model
- 1D gas release model
- 3D single-phase, single-component gas diffusion model, and
- 2D off-gas combustion and laminar flow model.

As ISV technology assessment efforts progress, capabilities of these models will be expanded as needed. Partial differential equations and auxiliary relations describing these models, along with associated simplifying assumptions, have been formulated and are presented in Section 2. The numerical approximations of the model equations from Section 2 are described and discussed in Section 3.

In connection with the numerical implementation of the model equations, a number of factors must be considered in order to achieve acceptable simulation accuracy. First, while the spatial domain of the ISV process is large due to far-field boundary conditions, suggesting the use of a relatively coarse computational mesh, steep gradients of temperature and material properties, as well as chemical reactions, occur near the melt front, suggesting that fine

meshing be used there. Second, irregularly shaped inclusions in the ISV zone plus the transient nature of the problem requires the use of a numerical simulation scheme that can model time-varying complex geometry. Third, the governing equations have contrasting numerical requirements. For instance, while fine meshing is needed in the solid region adjacent to the melt front to resolve high temperature gradients, the melt velocity and electrostatic potential gradients will be relatively small in the vicinity of the melt front. Furthermore, the grid will remain unchanged while the melt front progresses across the numerical grid over time, requiring the use of a fine mesh wherever the melt front may pass during the simulation. This situation suggests the use of adaptive grid refinement or other techniques to conserve computer memory; such techniques will be considered in the future for implementation, if warranted. Having the flexibility of employing different grids and time steps for the various phenomena will alleviate some of the difficulties mentioned above. Moreover, this approach is advantageous in transient three-dimensional calculations where computational costs can be significant.

In view of the factors discussed above, it has been determined that the various modules comprising the computational portion of the ISV analysis package will be partially uncoupled and solved sequentially on separate grids using finite element methods. Since the various solutions are interdependent, they will be communicated between modules and interpolated between grids. The finite element method has been selected for use in the analysis package because it permits nonuniform grids, is easily modularized, accommodates phase changes easily, and naturally incorporates interpolation in its framework. Selection of the numerical methods for approximating flow and combustion in the off-gas confinement hood will be decided by the suitability of available codes.

In the phase of development following the initial selection of codes and implementation of models, the models will be tested separately for a range of conditions that represent possible ISV environments. The reasons for pursuing these numerical studies are to:

- Validate the accuracy and efficiency of the numerical methods

- Determine the sensitivity of the numerical solutions to a range of physical conditions
- Determine the range of input parameters with which the simulation is valid
- Support experimental test design and analysis, and
- Form a basis for further software design.

Note that numerical studies will be performed for each independent model. If any of the models are found to be inadequate for the range of conditions tested, they will be improved or upgraded based on the experimental and computational test results.

Numerical calculations and model validation studies will begin in the latter part of FY-90 and continue through FY-91. In the middle part of FY-91, efforts will be initiated (if needed) to interface the different modules to form the basic integrated ISV analysis package. This package will then be used to conduct integral-effects computations to support safety and environmental hazard assessments for the large 'hot' field test scheduled for late FY-91.

The parametric studies and development and implementation schedules are discussed further in Section 4. The next section reviews the adaptability of existing codes for use in the analysis package.

1.2 Adaptability of Existing Computer Codes

A review of currently available computer codes has revealed that there are a limited number of three-dimensional codes that are applicable to the ISV process. Sources checked include the National Energy Software Center, contacts at Lawrence Livermore^{2, 3} and Sandia National⁴ Laboratories, the Texas Institute for Computational Mechanics,⁵ and contacts within the INEL. Minimal screening criteria have been established for reviewing the suitability of a code. These are listed below.

1. The computer code for thermal energy transport must be a three-dimensional finite-element model with both explicit and implicit time integration options, nonlinear solution capability, and phase change capability.
2. The code for the electric potential field and single-phase gas pressure field must satisfy item 1 above with the exception of phase change capability.
3. The code for flow and combustion in the confinement hood should be at least two-dimensional and employ an explicit or semi-explicit time integration scheme.
4. The code for thermal viscous flow should be three-dimensional and employ an implicit time integration scheme. The latter criterion is specified because it is anticipated that if a full fluid flow model is required to adequately model the ISV process, it will be computationally intensive and an implicit scheme will allow large time steps to be taken. This criterion is discussed further in Section 3.3.

Other general criteria used for screening available codes are readability, maintainability, efficiency, portability, and modularity. Moreover, it is desirable to identify codes for inclusion in the ISV analysis package that provide only those models and capabilities determined to be necessary to model the ISV process. Other codes, ^{6,7} while capable of providing some of the required models, are more general purpose in nature and carry unneeded baggage that will only slow down the computations and confuse the end user who must apply the analysis package. Furthermore, it may be difficult to pare such codes down to the required sizes without introducing errors or inadvertently causing other problems. Finally, all modules incorporated into the final analysis package should be amenable to reasonable straightforward modification in order to conform to the requirements for the package.

Two codes that appear to conform to the screening criteria have been acquired. These codes, named TOPAZ3D⁸ and COYOTE,⁹ have been acquired from

contacts at Lawrence Livermore National Laboratory (LLNL). TOPAZ3D is a three dimensional finite-element heat conduction code with phase change capability. This code satisfies criteria 1 and 2 above and may be used for the electrical resistance heating, energy transport including melting in the melt zone, and single phase gas diffusion. COYOTE is a two-dimensional finite-difference code that models compressible fluid flow and chemical reactions, including combustion, and satisfies criterion 3 above.

A three-dimensional viscous flow code has not yet been identified for inclusion in the analysis package. This is because it has not yet been determined which fluid transport processes are important to the integrated ISV process. In order to make this determination, a number of scoping calculations are currently underway using available codes. Computer codes which are currently available at the INEL,^{10,11} as well as from the National Energy Software Center and other sources, will be examined for suitability when the important parameters relating to viscous flow have been identified.

The computer codes for process chemistry and off-gas release are being developed at the INEL. These auxiliary models are specific to ISV applications and must be designed to interface with each other and with the melting and gas diffusion module. The code SOLGASMIX,¹² available at the INEL, will be used as a sub-module to the process chemistry model. The off-gas release model is based on concepts developed for predicting fission product release from melted reactor fuel rods. Details of these models are presented in Sections 2 and 3.

1.3 Availability of Experimental Data

It is desirable to validate the various models that will comprise the ISV analysis package against accurate experimental data in order to gain confidence in the package. A thorough review of useful data will eventually be carried out. However, for present purposes, the review of data will be restricted to information reported for actual ISV experiments and demonstrations. These data have been obtained during ISV tests conducted by Pacific Northwest Laboratories^{4,14} (PNL) and Oak Ridge National Laboratories¹⁶ (ORNL). Reference 14 reports the results for a pilot-scale test by PNL where transuranic-contaminated soil was vitrified. The pilot-scale ISV

demonstration test by ORNL¹⁷ involved compounds of cesium and strontium buried in a seepage trench. Table 1.4.1 provides a summary of these data as they relate to the models required for the ISV analysis package. Much of the data is limited in scope and will serve only to provide information useful for broad or "ballpark"-type comparisons.

Table 1.1 Data available from past ISV tests that may be useful for validation purposes for analysis package models

Model	Data Source	Description of Data
Electrical Heating	PNL-PS ¹	Voltage, current and power input to electrodes versus time. Total energy used.
	PNL-LS ²	Current density vs. time. Average power consumption.
	ORNL-PS ³	Voltage, current and power input to electrodes vs. time. Cumulative energy consumed.
Thermal Energy Transport and Melting	PNL-PS	Total Energy Consumed.
	PNL-LS	Averaged power consumption.
Viscous Flow of Melt	PNL-PS	Mass and geometry of final vitrified mass. Decontamination factors for substances initially in the soil. Limited distribution data on final disposition of vitrified materials from core sampling.
	PNL-LS	Decontamination factors for substances in soil. Retention factors for metals in soil. Cold cap subsidence data.
	ORNL-PS	Limited data on melt front progression and temperatures in solid region. Detailed data on final disposition of substances in the vitrified mass. Limited data on final subsidence levels.
Process Chemistry and Off-gas Release	PNL-PS	Decontamination factors for substances initially in the soil. Limited distribution data on final disposition of vitrified materials from core sampling. Limited data on final disposition of substances in vitrified zone from core sampling. Limited data on release rates of certain off-gases and radionuclides vs. time.
	PNL-LS	Decontamination factors for substances in soil. Retention factors for metals in soil. Limited data on off-gas release rates.
	ORNL-PS	Overall release rate and temperature data and release rates for certain off-gas components. Detailed composition and location data from core sampling of vitrified mass.

Off-gas combustion	PNL-PS	Limited data on release rates of certain off-gases and radionuclides vs. time. Data on concentrations of certain species. Temperature history data for certain locations in the hood. Hood vacuum history data.
	PNL-LS	Limited data on concentrations of species in the hood.
Vapor Migration in Surrounding Soil	PNL-PS	Limited data on substances in the soil surrounding the vitrified mass.

¹Pacific Northwest Laboratories, Pilot Scale Test¹⁴
²Pacific Northwest Laboratories, Large Scale Test^{15,16}
³Oak Ridge National Laboratories, Pilot Scale Test¹⁷

2. MATHEMATICAL FORMULATIONS FOR IN SITU VITRIFICATION

The governing equations that describe the ISV process include the equations of energy, momentum and mass transport. Additional relations for chemical reactions quantify the chemical composition of gases, reactants, and products generated by the ISV process. In this section, we present the governing equations and appropriate boundary conditions. These equations are given in terms of variables that describe the state of the system as a function of position and time. These variables are temperature, pressure, velocity, species concentration, and electrostatic potential. The phenomena to be modeled and the associated variables needed to describe them are shown in Table 2.1. Important secondary quantities (electric current, phase change rate, heat flux and gas flow rate) are also included in Table 2.1.

As the equations are presented, we state the major assumptions with which the equations were derived. Because of the complexity of the ISV process, it is not feasible to include in the governing equations all possible phenomena. Thus it is necessary to discard terms which are found to be unimportant. Based on a preliminary assessment of the ISV process, we have proposed an essential set of phenomena that are included in the present formulation. This will establish an initial simulation capability and provide a basis from which additional phenomena may be considered. As additional important phenomena are identified (by non-dimensional analysis or other means), we will include them in the ISV computer code and therefore broaden the scope of the numerical simulations.

Data for the governing equations are provided in the form of initial and boundary data, operating conditions and material properties. Because soil is a mixture instead of a pure substance, it is described by "effective" properties that account for the presence of multiple substances. The properties will be updated as we gain information about the soil and the waste forms contained within it.

Table 2.1. Phenomena and associated variables (primary and selected secondary variables) in the ISV process.

Phenomena	Temperature	Current	Pressure	Velocity	Species Concentration	Electrode Voltage	Gas Flow Rate	Phase Change Rate	Heat Flux
Electrostatic field in soil	I ^a	0 ^b				I			
Energy transport in soil	0	I		I				0	0
Flow in liquid soil	I		0	0				I	
Combustion in confinement hood	I 0		0	0	I 0		I		I
Gas release to atmosphere	I		I	I 0	I		0		
Vapor and particulate transport	I		I 0	0	I 0		0		
Process Chemistry	I		I		I 0				
<p>a. "I" indicates that the variable is an input to the governing equation that describes the phenomenon. b. "0" indicates an output.</p>									

The governing equations are expressed in a three-dimensional Cartesian coordinate system. Tensor notation is used to represent vectors, tensors and their derivatives. A subscript indicates the set of coordinate directions. For example, x_i is understood as (x_1, x_2, x_3) . A comma placed before a subscript indicates differentiation and repeated subscripts indicate summation.

A two-dimensional cross-section of an idealized ISV process is shown in Figure 2.1. The domain is partitioned into three zones; the melt Ω_1 , the solid Ω_2 and the combustion zone Ω_3 . Boundaries are indicated by Γ_{ij} , where i and j denote adjacent zones. The moving boundaries are Γ_{12} and Γ_{23} . Electrodes are identified by Γ_k , for k ranging from one to the number of electrodes. The vector n_k^{ij} is defined as the unit vector normal to Γ_{ij} that points from Ω_i to Ω_j .

2.1 Electrical Resistance Heating

In general, the electric and magnetic fields are governed by Maxwell's equations. In the initial model development phase, we will represent a time-varying AC voltage by its RMS value. The reasons for adopting this approximation concerns the time scales involved. The relaxation times for diffusion and convection in the material are very large relative to the period of the alternating current. A simple analysis of the relaxation times for the material involved (using average capacity and conductivity) bears this out. Similarly, transport of momentum in the viscous fluid has a large relaxation time relative to the high frequency alternating current. Assuming 60 Hz for the AC, many alternating cycles would pass before any pertinent response can be measured in the heat transfer, which is really the dominant part of the process. The high frequency fluctuation due to the alternating current is not significant insofar as heat transfer and motion are concerned.

To track the varying AC in time would require several timesteps per cycle. If we assume 20 timesteps per cycle then this would imply that the numerical simulation would require of the order of 2×10^6 timesteps per hour for the simulation. This is a conservatively low estimate since, in reality,

more time steps per cycle would probably be required in view of the nonlinear dynamics of the fluid flow and heat transfer. Since the process will in fact take days, and simulation should be designed for the entire time period, the numerical results would be too intensive for even the best supercomputers.

The current density J_i is given by Ohm's Law

$$J_i = -k_{ij}^e \phi_{,j} \quad (2.1)$$

where

ϕ = the electrostatic potential and

k_{ij}^e = the electrical conductivity tensor.

For a static electric field, in the absence of a magnetic field and free charge, Maxwell's equations reduce to

$$(k_{ij}^e \phi_{,j})_{,i} = 0 \text{ in } \Omega_1 \cup \Omega_2 \quad (2.2)$$

Appropriate boundary conditions for the potential [Equation (2.2)] are

$$\phi = 0 \text{ on } \Gamma_{22} \cup \Gamma_{23} \quad (2.3)$$

$$J_i n_i^{13} = 0 \text{ on } \Gamma_{13} \quad (2.4)$$

$$\phi = \phi^j \text{ on } \Gamma_j \quad (2.5)$$

where

ϕ^j = the applied potential at electrode j .

Figure 2.1. Idealized schematic of the ISV process.

The power per unit volume is given by

$$P = (k_{ij}^e \phi_{,j}) \phi_{,i} . \quad (2.6)$$

Integrating over the region $\Omega_1 \cup \Omega_2$, the total power becomes

$$\bar{P} = \int (k_{ij}^e \phi_{,j}) \phi_{,i} d\Omega . \quad (2.7)$$

To maintain a constant total power, we may adjust the applied potential ϕ^j . We will investigate control strategies to adjust ϕ^j (either continuously or incrementally) when the difference between the actual and desired power exceeds an allowable level.

2.2 Thermal Energy Transport with Melting

The governing equation of energy transport is given by

$$C_m T_{,t} + C_m u_i T_{,i} = (k_{ij} T_{,j})_{,i} + P + S \text{ in } \Omega_1 \cup \Omega_2 . \quad (2.8)$$

where T is the temperature, C_m is the mixture volumetric heat capacity, u_i is the velocity (equal to zero in Ω_2), k_{ij} is the mixture thermal conductivity tensor, S is the heat energy due to chemical reactions and P is the electrical heat energy. Heat energy generated by viscous dissipation is negligible.

There are two phases, solid (Ω_2) and liquid (Ω_1), which are separated by a moving phase boundary Γ_{12} . Appropriate boundary conditions are given by

$$T = T_m \quad \text{on } \Gamma_{12} \quad (2.9)$$

$$T_{,i} n_i^{22} = 0 \quad \text{on } \Gamma_{22} \quad (2.10)$$

$$-(k_{ij} T_{ij})n_i^{13} - h_T(T-T_0) \quad \text{on } \Gamma_{13} \quad (2.11)$$

$$-(k_{ij} T_{ij})n_i^{23} - h_T(T-T_0) \quad \text{on } \Gamma_{23} \quad (2.12)$$

$$(k_{ij} T_{ij})n_i^{12} + (k_{ij} T_{ij})n_i^{21} - LV_i n_i^{12} \quad \text{on } \Gamma_{12} \quad (2.13)$$

where T_m is the melting temperature, h_T is the total heat transfer coefficient from soil to air, T_0 is the ambient air temperature, L is the heat of fusion and V_i is the velocity of the moving boundary Γ_{12} .

The total heat transfer coefficient h_T is given by

$$h_T = h_c + h_r \quad (2.14)$$

where h_c and h_r are the convective and radiative heat transfer coefficients. These coefficients may be determined approximately. For convection heat transfer from a heated surface to air, the coefficient h_c is given by

$$h_c = \frac{k}{\delta} 0.332 \text{Pr}^{1/3} \text{Re}^{1/2} \quad (2.15)$$

in which the properties are those of air. The Prandtl number $\text{Pr} = \mu c_p / k$ and the Reynolds number $\text{Re} = \rho V_a \delta / \mu$, where V_a is the air velocity and δ is a characteristic length scale that we set equal to the electrode separation.

The radiative heat transfer coefficient is given by

$$h_r = \kappa \epsilon (T^2 + T_0^2) (T + T_0) \quad (2.16)$$

where κ is the Stefan-Boltzmann constant and ϵ is the emissivity of soil. Equation (2.16) is based on the assumption that the soil radiates heat energy to an infinite medium. This expression may be modified to account for the presence of a confinement hood.

The initial condition is given by

$$T(x_i, 0) = T_\alpha \quad (2.17)$$

where T_α is the ambient soil temperature.

2.3 Melt Flow

The molten fluid mixture is assumed to be Newtonian and incompressible, and fluid motion is assumed to be laminar. In the present formulation, free convection due to buoyancy forces is modeled by the Boussinesq approximation. The applicability of this approximation to ISV will be examined further. Other effects, such as electromagnetic forces and surface tension, which may contribute to fluid motion, are neglected. These effects will be studied and later included if they are found to be important.

The flow of liquid soil is described by the incompressible Navier-Stokes equations

$$\rho u_{j,t} + \rho u_i u_{j,i} = \sigma_{ij,i} + \rho g_j - \rho g_j \alpha (T - T_{ref}) \text{ in } \Omega_1 \quad (2.18)$$

$$u_{i,i} = 0 \text{ in } \Omega_1 \quad (2.19)$$

where u_j is the velocity, ρ is the mixture density, g_j is the gravity vector, α is the coefficient of thermal expansion, T_{ref} is a chosen reference temperature and σ_{ij} is the stress tensor.

The stress tensor for a Newtonian fluid is given by

$$\sigma_{ij} = -p\delta_{ij} + \mu (u_{i,j} + u_{j,i}) \quad (2.20)$$

where p is the static pressure, μ is the viscosity and δ_{ij} is the unit tensor ($\delta_{ij} = 1$ for $i=j$ and $\delta_{ij} = 0$ otherwise).

The mixture density is given by

$$\rho = \sum_{k=1}^{N_p} \rho_k \quad (2.21)$$

where ρ_k is the density of the k -th component in the soil mixture and N_p is the number of components in the mixture.

Boundary conditions for the Navier-Stokes equations are given by

$$u_j n_j^{12} = (1-\gamma) V_j n_j^{12} \text{ on } \Gamma_{12} \quad (2.22)$$

$$\sigma_{ij} n_j^{13} = -p_o \quad \text{on } \Gamma_{13} \quad (2.23)$$

where V_j is the moving boundary velocity, γ is the ratio of bulk soil density to liquid soil density and p_o is the ambient air pressure. The presence of a volume change due to the solid-liquid phase change, as expressed in boundary condition (2.22), will account for subsidence at the boundary Γ_{13} . The initial condition is given by

$$u_j(x_i, 0) = 0 \text{ in } \Omega_1 \quad (2.24)$$

2.4 Process Chemistry

The waste sites at the INEL contain a variety of organic and inorganic compounds. During the ISV process, these compounds may vaporize, react with one another, or pyrolyze to simpler compounds. The purpose of the chemical process models is to simulate these potential reactions. These reactions must be described so that the quantity and composition of the compounds leaving the melt, those retained in the melt, and those going to the off-gas system can be determined with confidence. Also the chemical process models are needed to

evaluate the potential for uncontrolled fires and explosions. The chemical process models will be contained in a module that is a separate functional unit of the main code. This section will describe how the chemical models will be implemented in the code.

It is anticipated that the chemical process models will be implemented sequentially in levels of increasing complexity. The first level will be the pyrolysis model. The second level will be the full chemical reaction model. In the pyrolysis model, no intermediate reactions or reactions between different species will be modeled but the reactants will be assumed to go to simple compounds or elemental forms. In the case of organic compounds, the final compounds will be such compounds as C, CO, CO₂, H₂, H₂O, and N₂. Inorganic compounds will be assumed to go to their most appropriate oxide. The pyrolysis model should be sufficient for bounding calculations. The chemical reaction model will include reactions between different compounds and will lend itself to best estimate calculations where more detail is required. The pyrolysis model will become a submodel of the chemical reaction model in the final code. This two level approach will make it possible to obtain calculational ability in a timely and efficient manner.

It is anticipated that the pyrolysis and chemical reaction models themselves will be implemented stepwise in increasing detail and complexity and will parallel the needs of the proposed experiments and code applications. In turn, comparison of the code calculations with experimental results will help establish the degree of detail required in the computer model. Initially it will be assumed that all reactions will proceed to equilibrium. This assumption is thought to be realistic since the temperatures involved are high and the melt front moves slowly. This assumption will greatly simplify the modeling effort, since reaction rates will not have to be considered. Reaction kinetics will be added to the model if the experimental results or future analyses indicate they are required.

In order to parallel the experimental program and provide meaningful results in a timely manner, the number of compounds and potential reactions will be restricted in the initial versions of the model. As analysis needs expand, experiments become more complex, and application needs are

established, the number of compounds and reactions will be increased accordingly.

To simplify the analysis in the initial version of the chemistry model, it will be assumed that the heated region can be divided into three separate temperature zones. These three zones are depicted and described in Figure 2.2. Zone 1 (the melt region) is characterized by high temperatures and relatively small temperature gradients. Since most reactions occur at lower temperatures, it will be assumed that reactions do not occur in the region and only the solubility and transport of the reaction products will have to be considered. Zone 2 encompasses the melt front. From previous analysis and experiments, it appears that the temperature gradients are very steep near the melt front and most chemical reactions are anticipated to occur in this region. Here the models for chemical reactions, pyrolysis, and decomposition will be implemented. Zone 3 is the heat up zone where the temperature is essentially the ambient soil temperature except near the Zone 2 boundary. The chemical reaction models may or may not be required in Zone 3. The necessity of modeling chemical reactions in this zone will be assessed as part of the early code development process.

It is assumed that very near the melt front, chemical species are in equilibrium since the temperatures are so high and the front moves so slowly. In these regions the reaction will be assumed to go to completion and thus no reaction rates will have to be considered. As part of the implementation, the validity of this assumption will be examined. Initially, we will consider only gaseous reaction products. However, the methodology may be extended to non-gaseous reaction products if necessary.

Calculation of chemical equilibrium involves determining the system composition that minimizes the Gibbs free energy. The total Gibbs free energy for a gaseous mixture can be written as:

Figure 2.2. Temperature zones for the process chemistry model.

$$F = \sum_{i=1}^{N_c} m_i \left(F_i^\circ + RT \ln \frac{f_i}{f_i^\circ} \right) \quad (2.25)$$

where

F = Total Gibbs free energy

F_i° = Standard State Gibbs free energy of component i

m_i = number of moles of component i

R = Gas Constant

T = Absolute Temperature

f_i = fugacity of component i

f_i° = Standard State fugacity of component i

N_c = Number of components in the mixture

At a specified temperature RT is a constant, therefore:

$$\frac{F}{RT} = \sum_{i=1}^{N_c} m_i \left(\frac{F_i^\circ}{RT} + \ln \frac{f_i}{f_i^\circ} \right) \quad (2.26)$$

The problem is to find the set of m_i that minimize F/RT . The problem is further complicated, since the mole numbers must also satisfy the following relationship:

$$\sum_{i=1}^{N_e} a_{ji} m_i - b_j \text{ for } 1 \leq j \leq N_e \quad (2.27)$$

where

a_{ji} = number of gram atoms of element j in a mole of component i

b_j = total gm atoms of element j

N_e = number of elements.

The mole numbers which minimize the free energy (2.26) and satisfy the mass balance constraint (2.27) may be obtained by the method of Lagrange multipliers. We form the function

$$F' = \frac{F}{RT} + \sum_{j=1}^{N_e} \lambda_j \left(b_j - \sum_{i=1}^{N_e} a_{ji} m_i \right) \quad (2.28)$$

where λ_j ($j=1, N_e$) are the Lagrange multipliers. Minimizing F' is equivalent to minimizing F subject to the constraint (2.27).

The solution to the minimization problem are the mole numbers m_i and multipliers λ_j which satisfy

$$\frac{\partial F'}{\partial m_i} = 0 \quad i = 1, N_e \quad (2.29)$$

$$\sum_{i=1}^{N_e} a_{ji} m_i - b_j = 0 \quad j = 1, N_e \quad (2.30)$$

The volume of gas generated may be obtained from the mole numbers of the reaction products. Assuming the gases are ideal, the volume of gas produced is given by

$$V = \frac{RT}{P} \sum_{i=1}^{N_g} m_i \quad (2.31)$$

N_g = number of gaseous compounds formed.

In the case where chemical equilibrium is not justified, the chemical reaction rates will be calculated by the reaction rate equation:

$$r_i = f(c, p, T) \quad (2.32)$$

where

r_i = reaction rate of component i

c = concentration

p = pressure

T = temperature.

The functional form of f depends on the type and order of the reaction. We assume that the effect of temperature can be separated out so that

$$r_i = f_1(T) f_2(p, c) = k_i' f(p, c) \quad (2.33)$$

where k_i' is the reaction rate coefficient of component i .

The temperature dependence of the rate coefficient is given by the Arrhenius' law

$$k_1' = k_{10}' \exp(-E/RT) \quad (2.34)$$

where E is the activation energy and k_{10}' is independent of T .

Energy production will be calculated from standard heats of reaction.

2.5 Gas Release from the Melt

The model discussed in this section is capable of considering the release of noncondensable fission products and other gases released to the melt by melting solid phase material, by evaporation of volatile species, or by chemical reaction of non-volatile species. The model treats gas transport in the melt by bubble rise. In the present formulation, it is assumed that horizontal melt velocities, u_1 and u_2 , are small and have negligible effect on bubble rise and coalescence. Vertical bubble rise velocities are measured relative to the vertical melt velocity, u_3 .

Horizontal velocities may be important for small bubbles having low rise velocities. For that aspect of the transport calculations, these small bubbles will be treated as though they move with the melt (zero velocity relative to the melt). Later, as they coalesce and become large, they will be treated as a field with velocity components relative to the melt. The principal limitation of the model is its requirement for detailed source rates to the melt. These sources are (a) porosity released by melting solid phases, (b) chemical reactions that produce species with high vapor pressures (pyrolysis, steam or water reactions with metal, desorption from surfaces), and (c) phase changes (boiling, evaporation from liquids, sublimation).

For sources due to porosity, the gas is assumed to enter the melt as bubbles having diameters proportional to the pore sizes. The initial bubble size for reaction products is more difficult to estimate. Slow reactions may produce smaller bubbles while rapid pyrolysis should produce larger sizes. An interim initial size estimate of 10^{-3} m will be used until data are available to improve the estimate. These data may not be critical to gas release modeling because the bubble coalescence model described below causes high

number densities of bubbles to coalesce rapidly to form large bubbles. Thus, as long as the assumed initial bubble size is less than the steady-state size for a given release rate and the number density of bubbles is high, the coalescence model will calculate the correct bubble size.

Sources due to phase changes will be assumed to form 10^{-3} m diameter bubbles when the equilibrium species vapor pressure is high enough to cause boiling. If the equilibrium species vapor pressure is less than the melt pressure, the sources due to phase change will be modeled by assuming that the species dissolve in the melt until saturation concentration is reached. (At temperatures near melt, diffusion rates are rapid so that the time required for solution is likely to be short.)

Once formed, bubbles rise toward the melt surface at a rate that is size dependent, with larger bubbles having faster rise speeds. The process is complicated by bubble coalescence and by very rapid evaporation/condensation of vapors into bubbles initially formed by noncondensable gases. The behavior of bubbles is modeled by representing ranges of bubble sizes with a single size equal to the geometric average volume of the range. In metallic melts, thirteen size ranges are sufficient.¹³ For ISV melts, the wide range of viscosities may require an extension to as many as seventeen sizes.

The smallest geometric average diameter will be 10^{-5} m and the ratio of any geometric average to the next smaller is taken to be $10^{0.25} = 1.778$ so that the thirteenth size classification would be represented by a bubble with 1 cm diameter and the seventeenth size classification would be represented by a 10 cm bubble.

For small bubbles (those that remain spherical), the relative rise speed of the L th bubble size is determined by balancing the drag and buoyant forces on the bubble¹⁸ in the vertical direction, so that

$$F - F_{\text{drag}} + F_{\text{buoyant}} = 0 \quad (2.35)$$

$$0 = 3\pi\mu S_L 2r_L + \frac{4}{3} \pi r_L^3 [\rho_g - \rho_l] g \quad (2.36)$$

where

μ = dynamic viscosity of liquid (kg/(m-s))

S_L = vertical speed of bubble rise of L th size classification (m/s)
relative to the fluid

r_L = radius of L th size classification (m/s)

ρ_g = density of gas in bubble (kg/m³)

ρ_l = density of liquid (kg/m³)

g = acceleration of gravity, (m/s²).

The solution of Equation (2.36) for s_L yields

$$S_L = \frac{2}{9} r_L^2 [\rho_l - \rho_g] \frac{g}{\mu} \quad (2.37)$$

Equation (2.37) is modified to treat larger, non-spherical bubbles and non-zero liquid velocities at the bubble surfaces using correlations given in Table 9.1 of Wallis. Three additional expressions for the bubble rise speed are used in an iterative process that involves several dimensionless groups.

If the bubble Reynolds number

$$Re_b = \frac{2\rho_l S_L r_L}{\mu} , \quad (2.38)$$

is less than 2 using the value of S_L calculated with Equation (2.37), no modification is necessary. If Re_b is greater than 2, a candidate value of S_L is calculated using

$$S_L = 0.6197 \left[\frac{10^{-4} \rho_l}{\mu} \right]^{0.52} [100r_L]^{1.28} \quad (2.39)$$

This candidate is tested by calculating both a revised value for Re_b using the rise speed determined with Equations (2.38) and (2.39) and a second dimensionless group

$$G_2 = \frac{g\mu^4}{\rho_l z^3} \quad (2.40)$$

where

z = surface tension between the liquid and gas phases (N/m).

If

$$Re_b < 4.02 G^{-2.214} , \quad (2.41)$$

the rise speed calculated with Equation (2.39) is used. If Equation (2.41) is not satisfied, a third candidate for S_L ,

$$S_L = 1.35 \sqrt{\frac{z}{\rho_l r_L}} \quad (2.42)$$

is tested. The third candidate correlation is tested by calculating a revised value of Re_b using the rise speed determined with Equation (2.42) and a third dimensionless group,

$$G_3 = \frac{g r_L^4 S_L^4 \rho_l}{z^3} \quad (2.43)$$

If

$$Re_b \leq 3.1 G^{-0.25} , \quad (2.44)$$

or

$$G_3 \leq 5.75 , \quad (2.45)$$

the rise speed calculated with Equation (2.42) is used.

Finally, if neither Equation (2.44) nor Equation (2.45) is satisfied,

$$S_L = 1.18 \left[\frac{gz}{\rho_l} \right]^{0.25} \quad (2.46)$$

is used to calculate rise speed.

The rise speed and source rates may be used to calculate the rate of gas transport through the melt. Assuming that the fluid velocity is small in comparison to the rise speed. The time rate of change of bubble concentration of radius r_L is given by

$$\frac{\partial n_L}{\partial t} + S_L \frac{\partial n_L}{\partial x} - n_L \frac{\partial S_L}{\partial x} + Q_L \quad (2.47)$$

where

- n_L = number concentration of bubbles of size L (bubbles/(m³ liquid))
- x = Coordinate in the vertical direction (m)
- t = Time (sec)
- Q_L = Source rate (bubbles/m³ sec)

Application of Equation (2.47) to calculate the rate of release of gas due to bubbles is complicated by the fact that there may be sources of bubbles from chemical reactions, or solid phases present in the control volume, and by the fact that bubbles may coalesce at rates comparable to their rise. The rate of coalescence is given by¹⁹

$$\begin{aligned} \frac{dn_{L_2}}{dt} = & - \sum_{L_1=1}^{L_{\max}} B(L_1, L_2) n_{L_1} n_{L_2} + \sum_{L_1=1}^{L_2} f(L_1, L_2) B(L_1, L_2) n_{L_1} n_{L_2} \\ & + \sum_{L_1=1}^{L_2-1} [1 - f(L_1, L_2-1)] B(L_1, L_2-1) n_{L_1} n_{L_2-1} \end{aligned} \quad (2.48)$$

where

- $B(L_1, L_2)$ = coalescence frequency function (m³/(particles s)), and
- $f(L_1, L_2)$ = fraction describing distribution of coalesced bubbles between size fractions that conserves mass and yields the correct number of bubbles.²⁰

The first sum in Equation (2.48) represents the loss rate from L_2 due to coalescence, and the second and third sums represent the supply rate of bubbles into L_2 when L_2 is the smaller or the larger of the two possible coalesced sizes.

Two contributions to $B(L_1, L_2)$ are considered. The first is due to turbulence and is represented by

$$B(L_1, L_2)_{\text{turb}} = 1.3 [r_{L1} + r_{L2}]^3 \left[\frac{\epsilon_d}{\nu} \right]^{\frac{1}{2}}, \quad (2.49)$$

where

$$\frac{\epsilon_d}{\nu} = \left[\frac{4}{d_{\text{liq}}} \right] \left[\frac{ff}{2} \right]^{1.5} \bar{u}^3 \quad (2.50)$$

and

ϵ_d = eddy diffusivity (m^2/s)

ν = kinematic viscosity (m^2/s)

d_{liq} = diameter of liquid region (m)

ff = fanning friction factor (0.004).

$\bar{u}$ = melt convection velocity

The second contribution to $B(L_1, L_2)$, is the coalescence due to differential bubble rise speeds in the liquid,^{21,22}

$$B(L_1, L_2)_{\text{rise}} = \pi [r_{L1} + r_{L2}]^2 [S_{L1} - S_{L2}] \quad (2.51)$$

It is assumed that the overall coalescence frequency function is the sum of Equations (2.49) and (2.50). This sum increases dramatically as the size of either of the coalescing bubbles increases. The increase expresses the fact that large bubbles sweep out a great deal of melt volume due to their large cross sectional area and large rise speed.

If all of the bubbles are formed by species of inert gas whose fraction does not vary with time or bubble size, the rates given by Equations (2.47) and (2.48) plus any source terms could be solved for n_L as a function of time. This requires an initial value n_L and a boundary value at the melt interface.

The initial condition for Equation (2.47) is

$$n_L(x, 0) = n_L^i \quad (2.52)$$

where n_L^i is the initial bubble concentration, and the boundary condition is given by

$$n_L(0, t) = \frac{q_L}{\frac{4}{3} \pi r_L^3 \rho_g S_L} \quad (2.53)$$

where q_L is the mass flux of gas bubbles of size L from solid to melt. This mass flux will be the primary source of bubbles, and will be caused by chemical reactions in the solid.

The distribution of species within bubbles is a function of bubble size and time. Vapors of volatile species like cesium, iodine, lead, and cadmium, whose vapor pressures may not be high enough to form bubbles, will evaporate into bubbles formed by species like hydrogen and steam that are above their boiling temperature. The extension of Equation (2.47) to determine the rate of removal of individual species is

$$\frac{\partial b_{n,L}}{\partial t} + S_L \frac{\partial b_{n,L}}{\partial x} - b_{n,L} \frac{\partial S_L}{\partial x} + Q_L \quad (2.54)$$

where

$b_{n,L}$ = mass of n th gas species in the L th bubble size class per unit volume (kg/m_3), and is given by

$$b_{nL} = \frac{4}{3} \pi r_L^3 \rho_n n_L \quad (2.55)$$

The extension of Equation (2.55) to form an equation for the effect of coalescence on the amount of mass of each species in each bubble size classification is

$$\begin{aligned} \frac{d b_{n,L_2}}{dt} = & - \sum_{L_1=1}^{L_{\max}} B(L_1, L_2) n_{L_1} b_{n,L_2} \\ & + \sum_{L_1=1}^{L_2} f(L_1, L_2) B(L_1, L_2) \left[n_{L_1} b_{n,L_2} \frac{r_{L_2}^3}{r_{L_1}^3 + r_{L_2}^3} + \Gamma_{L_2} b_{n,L_1} \frac{r_{L_1}^3}{r_{L_1}^3 + r_{L_2}^3} \right] \\ & + \sum_{L_1=1}^{L_2-1} [1 - f(L_1, L_2-1)] B(L_1, L_2-1) \\ & \left[\Gamma_{L_1} b_{n,L_2-1} \frac{r_{L_2-1}^3}{r_{L_1}^3 + r_{L_2-1}^3} + \Gamma_{L_2-1} b_{n,L_1} \frac{r_{L_1}^3}{r_{L_1}^3 + r_{L_2-1}^3} \right] \quad (2.56) \end{aligned}$$

It was mentioned above that the generalization to cases where the species mass fractions can vary with bubbles size and time allows treatment of species with equilibrium partial pressures below the system pressure. Analysis by Petti et. al.²¹ showed (at least for reactor materials) that these species

diffuse to a 10 cm bubble with time constants on the order of a millisecond. Based on the Petti et. al. results, these condensable species are assumed to be at equilibrium concentration in the bubbles. That is, the mass per unit volume of these species in the L th size class is assumed to be

$$b_{k,L} = M_k V_L p_k n_L V / RT = V_L C_{eqk} n_L V \quad (2.57)$$

where

M_k = kg-mole mass of k th condensable species {kg/(kg-mole)}

p_k = partial pressure of k th condensable species (Pa),

V_L = volume of L th size class bubble after diffusion of all species into the bubble, and

C_{eqk} = equilibrium concentration of k th condensable species (kg/m³).

Unfortunately, the activities of most species are not known so the partial pressure and equilibrium concentrations are assumed to follow Raoult's law.

Since there are $\#_k$ times $\#_L$ equations (where $\#_k$ is the number of condensable species and $\#_L$ is the number of size classes) and $\#_k$ times $\#_L$ plus $\#_L$ unknowns ($\{b_{k,L}\}$ and V_L), L more equations are required for a solution of Equation (2.54). The additional equations are the ideal gas law for all species originally in the L th size class,

$$\sum_{n=1}^{\# \text{ of species}} \frac{b_{n,L}}{\rho_{n, \text{ gas}}} = V_L n_L = \sum_{n=1}^{\# \text{ of species}} \frac{b_{n,L}}{M_n} \frac{RT}{P} \quad (2.58)$$

where

$\rho_{n, \text{ gas}}$ = density of n th chemical species in the gas phase (kg/m³).

The simultaneous solution of Equations (2.57) and (2.58) for each size class can be obtained by dividing the k-th Equation of (2.57) by M_k and subtracting the resulting equations for a given L from Equation (2.58) to obtain

$$\sum_{\text{i-non condensable species}} \frac{b_{i,L}}{M_i} - \left[\frac{P_k}{RT} - \sum_{\text{k-condensable species}} \frac{C_{eq,k}}{M_k} \right] n_L V_L = 0 \quad (2.59)$$

or

$$V_L = \frac{\sum_{\text{i-non condensable species}} \frac{b_{i,L}}{M_i}}{\left[\frac{P_k}{RT} - \sum_{\text{k-condensable species}} \frac{C_{eq,k}}{M_k} \right] n_L} \quad (2.60)$$

Substitution of V_L into Equation (2.57) yields $b_{k,L}$ for the condensable species and $b_{i,L}$ for the noncondensable species are not changed.

The volume V_L usually will not be one of the allowed size classifications so a fraction, fff, of each species in the new volume must be put into the first smaller classification J and the remainder must be put into the next larger classification J + 1. The fraction is determined by the requirement that the number of bubbles is not changed by the diffusion of vapor into or out of the bubbles

$$n_L = \frac{fff V_L}{V_J} + \frac{[1-fff] V_L}{V_{J+1}} \quad (2.61)$$

or

$$fff = \frac{n_L - \frac{V_L}{V_J}}{\left[\frac{V_L}{V_J} - \frac{V_L}{V_{(J+1)}} \right]} \quad (2.62)$$

Figure 2.3 is a schematic illustration that summarizes the process considered.

2.6 Gas Migration in the Surrounding Soil

During the active vitrification and cooldown phases of the ISV process heat conducts away from the melt into the surrounding soil and waste. Temperatures in this region range from the soil melt temperature 1200°C at the melt front to ambient temperature (~20°) at some distance away from the melt front (~1 m to 5 m). Wastes present in this region may pyrolyze and volatilize and generate gases. Temperature, concentration, and pressure gradients will force this multiphase multicomponent gas to migrate into the melt zone and through the surrounding soil. (Note that the gas entering the melt zone will act as a gas source for the gas release model.) In the initial phase of model development we are primarily interested in determining if significant quantities of gas flow away from the melt zone. To simplify the problem we will first treat gases not entering the melt zone as a single-phase single component ideal gas which is forced through the permeable soil and waste regions surrounding the melt by gas pressure gradients.

Figure 2.3. Processes considered in the gas release model.

By assuming Darcy flow, applying the principle of mass conservation and using the definitions of fluid compressibility and thermal expansivity, the basic equation for single phase gas flow in porous media is

$$\left(\rho_g \frac{K_{ij}^g}{\mu_g} p_{g,j} \right)_{,i} = \rho_g \pi \left(\beta_g \frac{\partial p_g}{\partial t} - \alpha_g \frac{\partial T}{\partial t} \right) + Q_g \text{ in } \Omega_1 \cup \Omega_2 \quad (2.63)$$

where P_g is gas pressure, ρ_g is gas density, μ_g is gas viscosity, K_{ij}^g and π are soil/waste permeability and porosity respectively, β_g is gas compressibility, α_g is the gas thermal expansion coefficient, T is temperature and Q_g is the gas mass source rate per unit volume. The appropriate boundary condition for the gas flow equation is

$$P_g(x_i, t) = 0 \text{ on } \Gamma_{22} \cup \Gamma_{13} \cup \Gamma_{23} . \quad (2.64)$$

The initial condition is given by

$$P_g(x_i, 0) = 0 \text{ in } \Omega_1 \cup \Omega_2 . \quad (2.65)$$

2.7 Flow and Combustion in the Confinement Hood

It has been determined that the combustion module of the ISV code will need to address the safety issues of (a) possible damage to the confinement hood due to excess thermal energy release or explosion and (b) incomplete combustion in the hood allowing combustible material to enter the treatment system where it may ignite. The physical phenomena that need to be modeled in the combustion module of the ISV code to address the above issues have been identified as mass, energy and momentum transport, chemical reactions, thermodynamics, effects of turbulence, and the possibility of achieving conditions conducive to explosions. The equations representing these physical phenomena that will be incorporated into the combustion module are presented and discussed below.

The flow, heat transfer, and combustion processes to be modeled will be based on the compressible Navier-Stokes equations for a mixture of gases in Eulerian coordinates.⁹

The conservation of mass or mass transport for each species k is represented by

$$\begin{aligned} \frac{\partial \rho^k}{\partial t} + (\rho^k u_i)_{,i} - - j_{i,i}^k + G^k & \text{ in } \Omega_3 \\ j_i^k(x_i, t \geq 0) \cdot n_i - 0 & \text{ on } \Gamma_{33} \\ j_i^k(x_i, 0) \cdot n_i - 0 & \text{ on } \Gamma_{23} \cup \Gamma_{12} \\ j_i^k(x_i, t > 0) \cdot n_i - \omega^k(x_i, t) & \text{ on } \Gamma_{23} \cup \Gamma_{12} \end{aligned} \quad (2.66)$$

where ρ^k is the species density, u_i is the average mixture velocity, j_i^k is the diffusional mass flux vector of species k , G^k is the rate of change of species k due to chemical reaction, n_i is the boundary unit normal vector, ω^k is the ground surface boundary flux of species k , and Γ_{ij} are boundaries as illustrated in Figure 2.4. The diffusional flux is a complicated function which in general depends on pressure gradient, temperature gradient, body forces and the concentration gradients of other species present in the system. However, since the only body force present is gravity, the body force term is identically zero while the dependencies on pressure and temperature gradients will be neglected. The concentration gradient term involves a matrix of diffusion coefficients that relates the diffusion of each species to the concentration of each of the other species. In view of the complexity of the concentration gradient diffusion, it is frequently assumed to be binary, that is, for the diffusion of species k , all other species are assumed to have the same diffusion coefficient with respect to species k . The result is known as Fick's Law:

Figure 2.4 Idealized problem domain for combustion in the confinement hood.

$$j_i^k = -\rho D_{12} \left(\frac{\rho^k}{\rho} \right)_{,i} \quad (2.67)$$

where D_{12} is the binary diffusion coefficient and ρ is the total density. Initially, D_{12} will be assumed to be the same for all species in the combustion module. Later, it will be determined if the binary generalization should be implemented.

Upon summing (2.66) over all species present, the continuity equation is obtained:

$$\frac{\partial \rho}{\partial t} + (\rho u_i)_{,i} = 0 \quad (2.68)$$

The momentum transport (Navier-Stokes) equations are given by

$$\begin{aligned} \frac{\partial \rho u_i}{\partial t} + (\rho u_i u_j)_{,j} &= \rho g_i - p_{,i} - \tau_{ij,i} \\ u_i (x_i, t \geq 0) \cdot n_i &= 0 \text{ on } \Gamma_{33} \\ u_i (x_i, 0) \cdot n_i &= 0 \text{ on } \Gamma_{23} \cup \Gamma_{12} \\ u_i (x_i, t > 0) \cdot n_i &= u^s \text{ on } \Gamma_{23} \cup \Gamma_{12} \end{aligned} \quad (2.69)$$

where p is the pressure, g_i is the acceleration due to gravity and u^s is the velocity of the off-gases entering the hood from the ground surface. The deviatoric stress tensor τ_{ij} is given by

$$\tau_{ij} = -\mu [u_{i,j} + u_{j,i}] - \mu_2 u_{i,j} \delta_{ij} \quad (2.70)$$

where μ is the dynamic viscosity, μ_2 is the second coefficient of viscosity, and δ_{ij} is the unit tensor. Initially, it will be assumed that the flow of gases in the confinement hood is laminar. If evidence is later given that turbulence effects are important, turbulence modeling will be implemented.

Energy conservation may be expressed in terms of the internal energy

$$\begin{aligned} \frac{\partial \rho U}{\partial t} + (\rho u_i U)_{,i} - \rho u_{i,i} - \tau_{ij} u_{i,j} + \sum_k \eta^k G^k \\ q_i (x_i, t \geq 0) \cdot n_i = q^c + q^R \text{ on } \Gamma_{33} \\ q_i (x_i, 0) \cdot n_i = 0 \text{ on } \Gamma_{23} \cup \Gamma_{12} \\ q_i (x_i, t > 0) \cdot n_i = q^s \text{ on } \Gamma_{23} \cup \Gamma_{12} \end{aligned} \quad (2.71)$$

where U is the internal energy, η^k is the heat of formation for species k and q^c , q^R , and q^s are convective, radiative, and ground surface heat fluxes. The heat flux vector in (2.71) is given by

$$q_i = -k T_{,i} + \sum_k H^k j_i^k + q_i^R \quad (2.72)$$

where T is the temperature and H^k is the enthalpy of species k .

The equation of state will be based on the ideal gas law and given by the sum of partial pressures for each species

$$p = \sum_k \frac{R \rho^k T}{W^k} \quad (2.73)$$

where R is the universal gas constant and W^k is the molecular weight of species k , and T is the absolute temperature.

The mixture internal energy is given by

$$\rho U = \sum_k \rho^k U^k(T) \quad (2.74)$$

where the species internal energy U^k will be linearly interpolated from data provided in the JANAF¹² tables for species enthalpy.

Chemical reactions occurring during the combustion process can be represented by

$$\sum_k a_r^k Z^k \rightleftharpoons \sum_k b_r^k Z^k \quad (2.75)$$

where the symbol $\rightleftharpoons$ indicates both forward and backward reactions, Z^k represents the chemical symbol for species k , and a_r^k and b_r^k are the stoichiometric coefficients for the r th reaction. The rate of change G^k of species k appearing in the species continuity equation is represented by

$$G^k = W^k \sum_r (a_r^k - b_r^k) r_r \quad (2.76)$$

where r_r is the rate of reaction of the r th reaction.

The reaction may be either an equilibrium reaction where the forward rate of reaction is equal to the backward rate, or a kinetic reaction where the rates are not equal. In the latter case, the net reaction rate is given by

$$r_r = k'_{fr} \prod_k \left(\frac{\rho^k}{W_k} \right)^{a_r^{k'}} - k'_{br} \prod_k \left(\frac{\rho^k}{W_k} \right)^{b_r^{k'}} \quad (2.77)$$

where k'_{fr} and k'_{br} are forward and backward reaction coefficients for reaction r and the exponents $a_r^{k'}$ and $b_r^{k'}$ are reaction orders. The reaction orders will be approximated⁹ by $a_r^{k'} = a_r^k$ and $b_r^{k'} = b_r^k$. A generalized Arrhenius form will be used for the forward and backward rate coefficients:⁹

$$k'_{fr} = v_{fr} T^{\zeta_{fr}} e^{(-E_{fr}/RT)} \quad (2.78)$$

$$k'_{br} = v_{br} T^{\zeta_{br}} e^{(-E_{br}/RT)} \quad (2.79)$$

where E_{fr} and E_{br} are forward and backward activation energies and v_{fr} , and v_{br} , ζ_{fr} , and ζ_{br} , are empirical constants. For equilibrium reactions, the net reaction rate is zero. Equation (2.77) then becomes

$$\frac{k'_{fr}}{k'_{br}} = \prod_k \left(\frac{\rho^k}{W^k} \right)^{b_r^{k'} - a_r^{k'}} = K_{cr}(T) \quad (2.80)$$

where $K_{cr}(T)$ is the concentration equilibrium constant for reaction r . The concentration equilibrium will be represented by the following temperature dependent relation

$$K_{cr}(T) = \exp \left[a_r \ln T_A + \frac{b_r}{T_A} + c_r + d_r T_A + e_r T_A^2 \right] \quad (2.81)$$

where a_r , b_r , c_r , d_r , and e_r are numerical coefficients related to reaction r , and $T_A = T/1000K$.

3. NUMERICAL MODELS FOR IN SITU VITRIFICATION

3.1 Electrical Resistance Heating

The finite element method is used to obtain an approximate solution to a weighted integral form of the governing equation. This approach is described in detail in Section 3.2 for the case of thermal energy transport. The finite element method leads to a set of non-linear algebraic equations to be solved for the approximate solution. This solution is defined on a mesh, which is a discretization of the domain into finite elements. Each "corner" of an element is called a node, and the solution vector consists of approximations of the dependent variable at each node in the mesh. Within each element, the finite element approximation is given by a polynomial function. This approximation is uniquely determined by its values at the nodes. A linear approximation defined on hexahedral elements will be used for the electrostatic potential.

The finite element method, applied to the electrostatic potential equation, leads to a set of non-linear algebraic equations. A predictor-corrective method, described fully in Section 3.2, is used to solve these equations. This method is an iteration method in which the equations are linearized at each iteration, and the iteration process continues until convergence is obtained. For each solution of the linearized equations, a direct elimination solver for banded, symmetric matrices will be used. Iteration methods like conjugate gradient may be more efficient for problems with a large number of nodes, and this option will be investigated as the computer code is developed.

The finite element approximation to the electrostatic field problem is given by

$$A\hat{\phi} = 0 \quad (3.1)$$

where $\hat{\phi}_j$ is the approximation of ϕ at node j in the computational mesh and

$$A_{\ell m} = \int_{\hat{\Omega}} k_{ij}^e \psi_{\ell,i} \psi_{m,j} dx \quad (3.2)$$

where Ψ_ℓ is the finite element weight function for node ℓ and $\hat{\Omega}$ is the approximation of the domain $\Omega = \Omega_1 \cup \Omega_2$. Boundary conditions (2.3) and (2.5) are enforced by setting $\hat{\phi}_j = 0$ at nodes j on $\hat{\Gamma}_{22} \cup \hat{\Gamma}_{23}$ and $\hat{\phi}_j = \phi^k$ at nodes j on $\hat{\Gamma}_k$ (electrodes $\hat{\Gamma}_k$, $k = 1, 2, 3, 4$). Boundary condition (2.4) is the natural boundary condition in the finite element formulation, and is satisfied approximately by the solution of equation (3.1).

An approximation to the gradient of potential may be obtained by differentiating the weight functions, from which we obtain

$$\hat{\phi}_{,j}(x) = \sum_{\ell=1}^N \hat{\phi}_\ell \psi_{\ell,j}(x) \quad (3.3)$$

where N is the number of nodes in the mesh.

Introducing equation (3.3) into equation (2.6), we obtain the approximation

$$\hat{P}(x_j) = (k_{ij}^e \hat{\phi}_{,j}(x_j)) \hat{\phi}_{,i}(x_j) \quad (3.4)$$

Continuous nodal values of $P(x_i)$ at node j may be approximated by a weighted average given by

$$\hat{P}_j(x_i) = \frac{\int_{\hat{\Omega}} \hat{P}(x_i) \psi_j(x_i) dx}{\int_{\hat{\Omega}} \psi_j(x_i) dx} \quad (3.5)$$

3.2 Thermal Energy Transport With Melting

Let $\hat{T}$ be an approximation to the temperature T , and w be a finite element weight function. The Galerkin finite element approximation to Equation (2.8) is defined to be: find $\hat{T} \in H^h$ satisfying the essential boundary condition (2.9) and initial condition (2.17), such that

$$\begin{aligned} \int_{\Omega} C_m \hat{T}_{,t} w dx + \int_{\Omega} C_m u_i \hat{T}_{,i} w dx = & - \int_{\Omega} K_{ij} \hat{T}_{,j} w_{,i} dx \\ & + \int_{\Gamma_{22}} K_{ij} \hat{T}_{,j} n_i^{22} w ds + \int_{\Gamma_{13}} K_{ij} \hat{T}_{,j} n_i^{13} w ds \\ & + \int_{\Gamma_{23}} k_{ij} \hat{T}_{,j} n_i^{23} w ds + \int_{\Gamma_{12}} (k_{ij} \hat{T}_{,j} n_i^{12} + k_{ij} \hat{T}_{,j} n_i^{21}) w ds \\ & + \int_{\Omega} (P + S) w dx \end{aligned} \quad (3.6)$$

for all $w \in H^h$. H^h is a finite dimensional subspace of H^1 , where H^1 is an infinite dimensional function space comprised of functions having square integrable first derivatives in domain Ω for $t \geq 0$.

Introducing boundary conditions (2.10) - (2.13) into (3.6), we obtain: find $\hat{T} \in H^h$ such that

$$\int_{\Omega} C_m \hat{T}_{,t} w dx + \int_{\Omega} C_m u_i \hat{T}_{,i} w dx = - \int_{\Omega} k_{ij} \hat{T}_{,j} w_{,i} dx$$

$$\begin{aligned}
& - \int_{\Gamma_{13} \cup \Gamma_{23}} h_T (\hat{T} - T_0) w \, ds + \int_{\Gamma_{12}} L V_i n_i^{12} w \, ds \\
& + \int_{\Omega} (P+S) w \, dx
\end{aligned} \tag{3.7}$$

for all $w \in H^h$.

The approximation $\hat{T}$ and weight function w are defined by

$$\hat{T}(x_i, t) = \sum_{k=1}^N \hat{T}_k(t) \psi_k(x_i) \tag{3.8}$$

and

$$w(x_i) = \psi_k(x_i) \tag{3.9}$$

where N is the number of nodes in the mesh, $k = 1, 2, \dots, N$, and functions ψ_k form a basis of subspace H^h . In the initial phase of model development we will choose $\psi_k(x_i)$ to be C^0 piecewise-bilinear functions.

Note that the term

$$\int_{\Gamma_{12}} L V_i n_i^{12} w \, ds \tag{3.10}$$

in (3.7) represents the weighted average amount of energy consumed in melting the soil and buried solid materials. Various fixed mesh techniques have been formulated to account for or approximate this term. These techniques will be examined to determine the one most suitable for ISV applications, in terms of both computational efficiency and required accuracy.

Introducing (3.8) and (3.9) into (3.7), and the approximate domain $\hat{\Omega}$ and boundary $\hat{\Gamma}$ in place of Ω and Γ , we obtain the set of semi-discrete equations

$$M \frac{d\hat{T}}{dt} + C\hat{T} + A\hat{T} = F \quad (3.11)$$

with initial condition

$$\hat{T}(0) = T_a \quad (3.12)$$

where

$$M_{ij} = \int_{\hat{\Omega}} c_m \psi_i \psi_j dx \quad (3.13)$$

$$C_{ij} = \int_{\hat{\Omega}} c_m u_m \psi_{j,m} \psi_i dx \quad (3.14)$$

$$A_{ij} = \int_{\hat{\Omega}} k_{mn} \psi_{i,n} \psi_{j,m} dx + \int_{\hat{\Gamma}_{13} \cup \hat{\Gamma}_{23}} h_T \psi_j \psi_i ds \quad (3.15)$$

$$F_j = \int_{\hat{\Omega}} (P+S) \psi_j dx + \int_{\hat{\Gamma}_{13} \cup \hat{\Gamma}_{23}} h_T T_o \psi_j ds + \int_{\hat{\Gamma}_{12}} L V_i n_i^{12} \psi_j ds \quad (3.16)$$

$$\hat{T}_j - \hat{T}(x_i^j) \quad (3.17)$$

where x_i^j is the coordinate of node j . To solve equations (3.11)-(3.12), we introduce an approximation to the time derivative

$$\frac{d\hat{T}}{dt} \approx \frac{1}{\Delta t} (\hat{T}^{n+1} - \hat{T}^n) \quad (3.18)$$

where the superscript denotes the time level (n is a non-negative integer) and Δt is the time step. Using a generalized trapezoidal method, we obtain

$$\left(\frac{1}{\Delta t} M^{n+\theta} + \theta C^{n+\theta} + \theta A^{n+\theta} \right) \hat{T}^{n+1} - \left(\frac{1}{\Delta t} M^{n+\theta} + (\theta-1) C^{n+\theta} + (\theta-1) A^{n+\theta} \right) \hat{T}^n + F^{n+\theta} \quad (3.19)$$

$$\hat{T}^0 = T_a \quad (3.20)$$

where θ is a numerical parameter that varies from 0 (explicit method) to 1 (implicit method). Because the material properties are functions of temperature, equation (3.19) is non-linear except for the case $\theta = 0$. To "relax" the non-linearity for $\theta \neq 0$, we may set $\theta = 1$ and evaluate the matrices M , C , A , and F at time level n to obtain the semi-implicit method

$$\left(\frac{1}{\Delta t} M^n + C^n + A^n \right) \hat{T}^{n+1} - \frac{1}{\Delta t} M^n \hat{T}^n + F^n \quad (3.21)$$

$$\hat{T}^0 = T_a \quad (3.22)$$

The non-linear Equation (3.19) is solved by a predictor-corrector method using the linearized form (3.21).

3.3 Melt Flow

The discrete approximations to the Navier-Stokes equations (2.18) and (2.19) are a system of non-linear algebraic equations for nodal point velocities and pressures. These equations are written symbolically as

$$M\dot{u} + Cu + Au + Bp = F \quad (3.23)$$

$$B^T u = 0 \quad (3.24)$$

where matrices M , C , and A represent mass, convection, and diffusion terms, respectively. B is a linear gradient operator and the vector F represents the forcing function for the system in terms of body forces and volumetric heating.

These equations or their variants can be formulated with finite difference or finite element methods. The appropriate method for this work will be chosen based on a compromise between algorithmic requirements, existing code adaptability, economics, and schedule.

Additional numerical studies must be performed to determine what algorithmic simplifications are feasible. For example, if convective effects are small then a simple linearized equation may be employed in which the force vector is explicitly adjusted to accommodate the non-linear convection terms. If this simplification is permissible it may be more appropriate to use a

three dimensional heat conduction code as a base code for development of the melt flow model.

Another issue of concern is how to efficiently treat the moving melt boundary. For early ISV process times the melt zone will be small, requiring calculations only on a few computational cells. As the melt zone progresses an increasing number of computational cells will become active fluid cells. Because three dimensional Navier-Stokes calculations are expensive and time consuming a suitable scheme would perform calculations only on those cells that are active.

Because the fluid transients in the ISV process are long and slowly varying it will be necessary to solve the discrete flow equations with an implicit time integration procedure. An explicit method will require an excessive number of time steps and the resulting computation time will be prohibitively large. For an explicit method (typically implicit pressures and explicit velocities), the stable time step size has to satisfy the Courant condition and be less than the time step size associated with the viscous diffusion time scale, thus

$$\Delta t < \Delta t_{\text{courant}} \sim \left(\frac{h}{u_i} \right)_{\text{minimum}} \quad (3.25)$$

$$\Delta t < \Delta t_{\text{viscous}} \sim \frac{1}{4} \left(\frac{\rho h^2}{\mu} \right)_{\text{minimum}} \quad (3.26)$$

where h is the mesh size. For a melt velocity of 1 cm/s, a density of 2.5 gm/cc, a melt viscosity of 100 poise, and a cell size h of 1 m, the time step limits are $\Delta t_{\text{courant}} = 100\text{s}$ and $\Delta t_{\text{visc}} = 62\text{s}$. Therefore the maximum stable time step size is 62s. These time step sizes are clearly too small for a process that operates up to 300 hours. Moreover, since melt viscosity increases dramatically with decreasing temperature (viscosity is very large near the

melting temperature) the stable time step size will decrease dramatically as regions of the melt cool down.

The issues discussed above are being examined in more detail. Concurrently, existing implicit codes such as Commix 1B¹¹ and Sola (JR)⁽²³⁾ are being evaluated.

3.4 Process Chemistry

As discussed in Section 2.5, it will be necessary to minimize the system Gibbs free energy in order to calculate the equilibrium concentration of gaseous reaction products.¹² The total Gibbs free energy for a mixture and the species mass balance can be written as

$$\frac{F}{RT} = \sum_{i=1}^{N_o} m_i \left(\frac{F_i^o}{RT} + \ln \frac{f_i}{f_i^o} \right) \quad (3.27)$$

$$\sum_{i=1}^{N_o} a_{ji} m_i - b_j = 0 \quad \text{for } 1 \leq j \leq N_e \quad (3.28)$$

The problem is to find the set of m_i that minimize F/RT and also satisfy the species mass balance. Various approaches have been used to accomplish this minimization. When only a few reactions need be considered, a direct solution method can be used. When a large number of potential reactions must be considered, an optimization technique will be utilized to determine the distribution of the reaction compounds. The code SOLGASMIX-PV will be used as a chemistry submodel to perform this task.

Once the quantities of the reaction products are determined the volume of gas generated can be computed. Assuming the gas is ideal the volume of gas generated is given by

$$V_g = \frac{RT}{P} \sum_{i=1}^{N_g} m_i \quad (3.29)$$

where N_g is the number of gaseous compounds formed.

The volume of gas V_g produced will act as a source term for the gas diffusion model, the solution of which will determine the volume of gas entering the melt and the volume of gas flowing away from the melt.

If future analyses or experiments indicate the reaction kinetics must be considered, the rate equation (2.33) will be implemented in the following manner:

$$r_i = dm_i/dt = k'_i f(p, m_i) \quad (3.30)$$

where m_i is the number of moles of component i . In finite difference form, equation (3.30) becomes

$$\frac{m_i^{n+1} - m_i^n}{\Delta t} = k'_i f^{n+\theta}(p, m_i), \quad (3.31)$$

where the superscript indicates the time level, and θ is a numerical parameter that ranges from 0 to 1. For an explicit solution method $\theta=0$, and the approximate equation becomes

$$m_i^{n+1} = m_i^n + \Delta t k'_i f(p^n, m_i^n) \quad (3.32)$$

As part of this project $f(p, m)$ will be determined for each reaction of interest.

If ideal gas behavior is assumed, equation (3.29) may be discretized to obtain

$$\Delta V_g = \frac{RT}{P} \sum_{i=1}^{N_g} (m_i^{n+1} - m_i^n) \quad (3.33)$$

3.5 Gas Release from the Melt

Some features of the numerical approximations that belong in this section were introduced in Section 2.6 because these features determine the form of the governing equations discussed there. In particular, the concept of treating the continuous range of bubble sizes with discrete representative bubble sizes from a geometrically increasing series has already been introduced. The decision to integrate rate equations for bubble coalescence and movement relative to liquid melts but use equilibrium expressions for the rapid evaporation or condensation of vapors to or from bubbles was also discussed in conjunction with the governing equations.

In order to solve for mass of the n th gas species in the L th bubble size (the variable $b_{n,L}$ defined in Section 2.6), the number concentration n_L of bubbles of size L must be known. This section will discuss the solution for $\{n_L\}$ first, then the solution for $\{b_{n,L}\}$. Assuming that the magnitude of the liquid velocity is small in comparison to the rise speed s_L , the governing equation for n_L is given by

$$\frac{Dn_L}{Dt} = -n_L \frac{\partial S_L}{\partial X} + Q_L \quad (3.34)$$

where D/Dt is the substantial time derivative and Q_L is the sum of the sources (coalescence) and sinks of bubbles of size L given by equations (2.48)-(2.51) and additional sources from chemical reactions and phase transformations. Expressing the substantial derivative in terms of local and convective derivatives, and assuming that bubbles rise only in the vertical direction, equation 3.34 becomes

$$\frac{\partial n_L}{\partial t} + S_L \frac{\partial n_L}{\partial x} = -n_L \frac{\partial S_L}{\partial x} + Q_L \quad (3.35)$$

where x is the vertical direction.

The initial condition for Equation 3.35 is given by

$$n_L = n_L^i \quad (3.36)$$

where $\{n_L^i\}$ is the initial bubble density. The boundary condition at the melting interface is given by

$$n_L = \frac{q_L}{\frac{4}{3}\pi r_L^3 \rho_g S_L} \quad (3.37)$$

where q_L is the mass flux of gas which goes into bubbles of size L . This mass flux will be the primary source of bubbles and will be caused chiefly by chemical reactions, evaporation of volatile wastes, or melting of porous solids. For large q_L , it may be necessary to check for frothing or gas volumes greater than the liquid volume with the relation

$$\sum_L n_L \frac{4}{3}\pi r_L^3 > 0.5 \quad (3.38)$$

and to simplify the coalescence terms of the governing equation to avoid unnecessary stiffness in the equations to be solved for this case.

Because Equation 3.35 involves derivatives of n_L in the vertical direction x and not in the y and z directions, it may be solved numerically by placing a grid in the x direction that extends through the liquid region. The liquid depth, d , is discretized into elements of size Δx so that

$$d = N\Delta x \quad (3.39)$$

where N is the number of elements. Also, the simulation time, T , is discretized into time steps of size Δt so that

$$T = M\Delta t \quad (3.40)$$

where M is the number of times steps. Varying the y and z coordinates yields a set of independent, one dimensional problems.

Let n_{Li}^m , S_{Li}^m and Q_{Li}^m denote and approximations to n_L , S_L and Q_L at grid point i and time level m , where

$$1 \leq i \leq N + 1 \quad \text{and} \quad 0 \leq m \leq M. \quad (3.41)$$

The approximate speed S_{Li}^m is obtained from equations (2.37) to (2.46) and the approximate source Q_{Li}^m is obtained from equations (2.48) - (2.51) of section 2.6. Using a backward difference approximation to the spatial derivatives, the approximation to equation (3.35) becomes

$$\frac{\partial n_{Li}}{\partial t} - \frac{S_{Li}}{\Delta x} n_{Li-1} - \left[\frac{2S_{Li} - S_{Li+1}}{\Delta x} \right] n_{Li} + Q_{Li} \quad (3.42)$$

for i from 1 to $N + 1$. Then we approximate the time derivative in equation 3.42 by a backward difference and obtain

$$n_i^m - -S_{Li}(n_{Li}^{m1} - n_{Li-1}^{m-1})\frac{\Delta t}{\Delta x} - n_{Li}^{m-1} (S_{Li}^{M-1} - S_{Li-1}^{m-1})\frac{\Delta t}{\Delta x} + Q_{Li}^{m-1}\Delta t + n_i^{m-1} \quad (3.43)$$

for m from 1 to M .

The approximation n_{Li}^m in equation 3.43 depends on the values n_{Li}^{m-1} and n_{Li-1}^{m-1} . Thus, the method is explicit and equation (3.43) can be solved by

"marching" along grid points at each time level, beginning with grid point 1 where a boundary value is specified. Starting with a set of initial values, $\{n_{L,i}^0\}$ for $i = 1$ to $N+1$, $n_{L,i}^m$ can be successively calculated for $i = 2$ to $N+1$ and $m = 1$ to M .

There are various numerical methods in addition to the particular one presented in this section. These methods will be examined and the one used for the In Situ Vittrification code will be selected on the basis of accuracy, stability, and efficiency.

Initial modeling will assume that the composition of all gas is the same for all time, bubble sizes, and position. When this initial modeling is completed and functioning well, the assumption that the composition can vary will be introduced. With this generalization, $\{n_{L,i}^m\}$ will be needed to determine the source term in governing equations for $\{b_{n,L}\}$ that are just like equation (3.35) with n_L replaced by $b_{n,L}$ and Q_L replaced by a source term, $Q_{n,L}$ for each species. Because the governing equation for $\{b_{n,L}\}$ is the same as the governing equation for $\{n_L\}$, the technique described to solve for $\{n_L\}$ will also work for $\{b_{n,L}\}$. The only change required is the additional step of using the method described in conjunction with equations (2.55)-(2.62) to account for evaporation or condensation of vapors into bubbles after each time increment, Δt .

3.6 Gas Migration in the Surrounding Soil

The finite element approximation to the gas flow problem is given by

$$G \hat{P}_g + H \frac{d\hat{P}_g}{dt} - F_g \quad \text{in } \hat{\Omega} \quad (3.44)$$

$$\hat{P}_{gj}(0) - P_a \quad (3.45)$$

$$\hat{P}_{gj}(t) = 0 \text{ on } \hat{\Gamma}_{12} \cup \hat{\Gamma}_{22} \cup \hat{\Gamma}_{13} \cup \hat{\Gamma}_{23} \quad (3.46)$$

where $\hat{P}_{gj}$ is the approximation of P_g at node j in the computational mesh and

$$G_{ij} = \int_{\hat{\Omega}} \rho_g \frac{K_{nm}^g}{\mu_g} \psi_{i,n} \psi_{j,m} dx + \int_{\hat{\Gamma}_{13} \cup \hat{\Gamma}_{23}} \rho_g \frac{K_{nm}^g}{\mu_g} \psi_i \psi_{j,m} ds \quad (3.47)$$

$$H = \int_{\hat{\Omega}} \rho_g \pi \beta_g \psi_i \psi_j dx \quad (3.48)$$

$$F_g = \int_{\hat{\Omega}} \left[Q_g - \rho_g \pi \beta_g \frac{\partial \hat{T}}{\partial t} \right] dx \quad (3.49)$$

$$\frac{d\hat{P}_{gj}}{dt} = \frac{\partial \hat{P}}{\partial t}(\mathbf{x}_j^i) \quad (3.50)$$

where Ψ_l is the finite element basis function for node l and $\hat{\Omega}$ is the approximation to domain $\Omega_1 \cup \Omega_2$.

The finite element approximation to the pressure is given by

$$\hat{P}_g(\mathbf{x}_l) = \sum_{k=1}^N P_{gk} \psi_k(\mathbf{x}_l) \quad (3.51)$$

where N is the number of nodes in the mesh.

To solve equations (3.44) - (3.46), an approximation to the time derivative is introduced

$$\frac{d\hat{P}_g}{dt} = \frac{1}{\Delta t} (\hat{P}_g^{n+1} - \hat{P}_g^n) \quad (3.52)$$

where the superscript denotes the time level (n is a non-negative integer) and Δt is the time step. Using a generalized trapezoidal method

$$\left(\frac{1}{\Delta t} H^{n+\theta} + \theta G^{n+\theta} \right) \hat{P}_g^{n+1} - \left(\frac{1}{\Delta t} H^{n+\theta} + (\theta-1) G^{n+\theta} \right) \hat{P}_g^n = F_g^{n+\theta} \quad (3.53)$$

$$\hat{P}_g^0 = P_a \quad (3.54)$$

where θ is a numerical parameter that varies from 0 (explicit method) to 1 (implicit method).

3.7 Flow and Combustion in the Confinement Hood

The governing transport equations representing conservation of mass, momentum, and energy used in the combustion module of the ISV code may be represented by the following discrete equations

$$A_p + B_p = 0 \quad (3.55)$$

$$A^u u + B^u u = f^u \quad (3.56)$$

$$A^U U + B^U U = f^U \quad (3.57)$$

for mass density ρ , mixture velocity u , and internal energy U , where A and B are coefficient matrices and F represents the various source terms.

A reacting flow code called COYOTE⁹ has been secured and will be used as the combustion module for the ISV code. This code was originally developed for representing flow and combustion in an internal combustion engine. The above equations, representing the phenomena that are desired to be modeled in

the combustion module, will initially be implemented as they are in the COYOTE code. COYOTE incorporates the following features in its implementation:

1. Cartesian and cylindrical coordinates in two dimensions with a swirl model for representing three-dimensional effects,
2. nonuniform, staggered mesh (velocity nodes off-set from pressure/scalar nodes),
3. variable hybrid upwind/centered differencing for the advection (first derivative) terms,
4. centered differencing for all other terms (second derivatives),
5. diffusion and convection limited time stepping,
6. pressure iteration at each time step,
7. mass leakage at user-determined grid cells,
8. flow visualization via path tracing of massless marker particles, and
9. turbulence modeling.

COYOTE has the capability to employ either the $k-\epsilon$ or sub-grid scale (sgs) turbulence models. In the case of the latter, turbulence is modeled below the size of the grid cells, but computed directly at larger scales. The $k-\epsilon$ model computes only the ensemble-averaged field. Both are eddy viscosity type models. Initially, no turbulence model will be employed in the combustion module, since it is not certain that turbulence effects will be present in the flow inside the ISV confinement hood.

3.8 Numerical Parametric Studies

The various component models described in the foregoing sections will be used to perform parametric studies. These analyses will indicate if the

models formulated cover all the important phenomena and will provide a benchmark for comparison with future versions of the models. Results of sensitivity studies of various input parameters will identify the most sensitive parameters and models and will be used to recommend specific measurements in laboratory and field tests. Also, specific solution techniques used in the calculations will be evaluated for accuracy and speed. Finally, these early calculations will provide a set of test problems to verify that the integrated ISV model is correct.

3.8.1 Heat Transfer and Fluid Flow

During the remainder of FY-90 heat transfer and fluid flow processes will be simulated using in-house codes ABAQUS²⁴ and FIDAP²⁵. In these studies, since the electrostatic field module is not complete, the presence of electrodes will be approximated by prescribing a power source that is equal to i^2/k_e , where i is the current density and k_e is the electrical conductivity. It will be assumed that the current density is constant and a range of values up to the maximum current density that can be handled by the electrodes will be examined. Note that since electrical conductivity increases with temperature, the power source will decrease with increasing temperature. Also, the power will be set to zero in regions in which the temperature is less than the melting temperature so that power is "on" in the melt region only. Requirements for heat transfer numerical calculations are that the soil temperatures reach a maximum of about 2000°C and the melting rate be about 1-10 cm/hr. Simulation times will be on the order of 100 hours in order to model a representative ISV process.

The following characteristics of heat transfer and fluid flow will be assessed by the numerical studies:

1. the importance of melt flow and its influence on the shape of the liquid region and temperature field,
2. the location of high temperatures and the distance at which heat energy progresses into the solid region,

3. the heat flux from the soil and melt surfaces and the formation of a cold cap, and
4. the requirements on mesh and time step sizes to produce accurate simulations.

3.8.2 Process Chemistry

One of the purposes of the numerical calculations will be to determine the reaction products and their relative distribution as a function of temperature and pressure. The initial calculations will focus on those reactions anticipated to be of interest in the initial experimental program. The main reactions of interest are assumed to be the pyrolysis of wood and simple organics. Therefore, the parametric studies will focus on developing the product distribution for the pyrolysis of wood and simple organics as a function of temperature and pressure. Chemical equilibrium calculations will be performed at several temperatures between 300 and 2000 K. It is anticipated that the product distribution in these calculations will be insensitive to pressure. To verify this assumption, calculations will be made at pressures of one and three atmospheres at a few selected temperatures. Although the product distribution is anticipated to be insensitive to pressure, the volume of gases produced will be a linear function of pressure.

3.8.3 Gas Release From The Melt

Preliminary calculations will be performed for a one-dimensional melt geometry in which bubbles are injected at a mass rate q_i and are released to the atmosphere at a mass rate q_r . The objective of these calculations is to determine which physical parameters dominate the relationship between q_i and q_r . To this end, the following questions need to be answered.

1. How long does it take for the process to reach a "steady-state", at which $q_i = q_r$?
2. Where in the liquid is the highest concentration of bubbles?

3. What are the dominant bubble sizes after coalescence? Can we neglect bubble sizes that are outside this range?
4. How does the range of initial bubble sizes and coalescence model affect q_r ?
5. How does a vertically varying temperature field and melt viscosity influence the relationship between q_i and q_r ?

Some proposed calculations are described below. For the preliminary calculation, temperature is assumed constant at 1500 K and the melt is assumed to have a uniform composition typical of INEL soils (Table 3.1). Bubbles of an intermediate size classification, 10^{-3} m diameter, are injected at a constant mass flux of $2.8138 \cdot 10^{-5}$ kg/(m² s). Hand calculations indicate that the bubble rise speed for bubbles of this size is $1.279 \cdot 10^{-2}$ m/s so the flux proposed should produce a steady state bubble volume of 0.01 m³ for each m³ of liquid. After 78.186 seconds, the gas should begin to escape from the top of the one meter deep melt at a rate of $2.8138 \cdot 10^{-5}$ kg/(m² s). The bubble size should remain at 10^{-3} m diameter because all the bubbles rise at the same speed and do not coalesce.

Table 3.1. Physical properties of typical INEL soils near 1500 K

Property	Value	References
Viscosity	10^{-1} kg/(m s)	PNL-4800 ²⁶ page 100, 1000P
Liquid Density	$2.35 \cdot 10^3$ kg/m ³	PNL-4800 page 99, 2.2-2.5 g/cm ³
Gas Density	0.22 kg/m ³	Ideal gas law for nitrogen

The second proposed calculation introduces coalescence effects by distributing the $2.8138 \cdot 10^{-5}$ kg/(m² s) flux equally between bubbles of 10^{-3} m diameter and 10^{-4} m diameter, $1.4069 \cdot 10^{-5}$ kg/(m² s) of each size. In this case, release from the top of the liquid should begin a little before 78 seconds because of coalescence to form bubbles larger than 10^{-3} m. However, the steady-state release rate should not be attained until well after 78

seconds because of the slow rise speed of the smaller bubble size, $1.279 \cdot 10^{-4}$ m/s.

The third, and most realistic calculation is an attempt to duplicate parameters expected in ISV systems. Maximum release rates of 50 std m³/min are reported on page 44 of reference (PNL-4800) for the large scale test system. With a 25 m² surface area and rise speeds on the order of 1 m/s, this suggests a flux rate near 1 m² gas / m². In order to test the model, a constant source of nitrogen bubbles of 0.3 porosity times .1 m melt rate/hour * .22 kg/m³ gas = $1.8 \cdot 10^{-6}$ kg N₂ /m² is proposed. The rate of gas injection from this source is small, but the bubble size is also likely to be small so that the bubble rise speed is slow and gas may accumulate in the melt. The size of these bubbles should approximate the soil pore size, 10^{-4} m. After 50 hours, this source should be supplemented by an additional source of about $1.67 \cdot 10^{-2}$ kg/s steam for 600 s, representing boiling of 1 kg/m² water trapped under the melt per minute for ten minutes. The bubble size of this source would be 10^{-2} m diameter. This more rapid injection also tests the model for the boiling of volatile metals like cadmium and the rapid evolution of gases from pyrolysis and other chemical reactions. In addition, the model for transport of vapors of intermediate volatiles by bubbles is tested by assuming the melt contains sufficient silver to saturate the bubbles with silver vapor. The equilibrium vapor concentration of silver vapor at 1500 K is approximately 10^{-2} kg/m³.

3.8.4 Flow and Combustion in the Confinement Hood

Preliminary calculations will be made with the COYOTE code to investigate the effects of various parameters and situations on the combustion and rate of exhaust for the off-gases inside the confinement hood. These calculations will have the objectives of determining which parameters are important to the combustion/flow processes in the confinement hood and for which configurations. Initially, the confinement hood will be assumed to be of rectangular cross-section since COYOTE can only handle Cartesian and polar coordinates. The flow situation outlined in Ref. 27 will be simulated first. This EDF assumes that only H₂ and CO are contained in the off-gases and that they do not combust.

Additional studies will investigate the effects of varying (1) the location and rate of supply air into the hood, (2) the rate and location of off-gas penetration into the hood, and (3) the location and flow rate of exhaust gases from the hood into the treatment system. After confidence is gained in the combustion module, flow scenarios with transient boundary conditions will be investigated to explore the possibilities for explosions in the event of combustion being extinguished and then reignition occurring. Other scenarios will be investigated as their importance becomes apparent as a result of the preliminary studies.

The first calculations that will be made using COYOTE for off-gas flow in the confinement hood will use 2D rectangular geometry and will be based on an Engineering Design File that was produced for the Intermediate-Scale ISV Test 2²⁷. In this EDF²⁷, it is assumed that only carbon monoxide (CO) and hydrogen (H₂) are present in the off-gas entering the confinement hood. The carbon, hydrogen, and oxygen in the off-gas are assumed to have the same concentration as pyrolyzed wood plus carbon from the electrodes. It is further assumed that all of the carbon is used up to form CO and that all of the hydrogen is generated in the form of H₂. The rate of pyrolyzed gas production is based on the amount of combustible material buried, the assumed melt front velocity, and the premise that all of the gas enters into the confinement hood.

The EDF²⁷ gives flow rates for the carbon monoxide, hydrogen, and supply air. It will be assumed that the exhaust will have the same volumetric flow rate as the total inflow. Initially, it will be assumed that no combustion takes place, as does the EDF²⁷. Later, combustion will be added in which case the boundary temperatures of the confinement hood will be assumed to be constant for a given calculation, but varied for various runs. The temperature of the entering off-gas will be assumed to be that of the melt.

Other configurations of supply air, exhaust, and off-gas incursion will also be explored in order to determine important operating parameters and find the safest modes for operation.

4. TOP LEVEL SYSTEM DESCRIPTION

The ISV simulation code is a software package which performs the numerical simulation of the In Situ Vitrification process. The governing equations of Section 2 are implemented in the software by the numerical approximations and algorithms identified in Section 3.2. This section describes the major components of the ISV simulation code in terms of the input and output requirements for each model and the data flow and controls for these components at a high level of preliminary design of the software architecture. The conceptual design for the ISV simulation code is subject to change as more detailed analysis is performed on the simulation models.

4.1 ISV Analysis Package

The major components of the ISV analysis package are the input processor, the computational processor which includes the various component models, and the output processor. The top level data flow diagram for the ISV code, shown in Figure 4.1, illustrates a typical process with user input to the code and error messages and a simulation report returned from the code to the user. The level "0" data flow diagram, shown in Figure 4.2, depicts the three major software components of the ISV code, providing the user interface, the ISV numerical models implementation, and the output generation.

4.1.1 Input Processing

The ISV code receives the user input and establishes all conditions necessary to perform a simulation with the selected models. It supplies default values for all input not specified by user input and computes parameters which are implicitly specified. The data is validated for consistency, values within allowable ranges, and correct specification of input for models selected. The input processor defines the computational meshes and initializes all arrays and variables for either an initial simulation or for a continuation (restart) of a previous simulation. Section 4.10 describes the input processing in more detail.

Figure 4.1. Top level flow diagram of the ISV analysis package.

Figure 4.2. Zero-level flow diagram of the ISV analysis package.

4.1.2 Computational Processing

The computational processor executes one step of the simulation, using data values at the beginning of the time step (from initial conditions or results of a previous time step). Each subordinate model is called in the correct order to compute values for that model for the time step requested. After all models have been invoked, the results are optionally output to the simulation results file, the graphics file, and the restart file. If the simulation has been completed to the desired time or if the computational time limit is reached, then the computation is terminated. Otherwise, the data values are updated to the new time and the computational processor begins the next time-step calculations.

4.1.3 Output Processing

After the computation is terminated, the results are summarized.

4.2 Simulation Control

The simulation control for the computational process is shown in Figure 4.3. Simulation input is passed from the input processor to the simulation control at 2.1. For each time step, the processor initiates problem execution and invokes the individual simulation models, selected by the user, and in the order starting at 2.2 and advancing sequentially to 2.8. Dependent variable data are communicated between the models. Each model typically relies on the results of one or more of the previous models in the sequence for some of its input data. After all specified models have been invoked, the simulation control may request intermediate results to be written to the simulation report, the simulation graphics, or the simulation restart. The process is terminated when either the simulation is complete or the computational time limit is reached. Each individual simulation model is described in more detail in the following sections.

Figure 4.3. Data flow diagram of the computational processor.

4.3 Electrical Resistance Heating

4.3.1 Description

The data flow diagram in Figure 4.4 shows the Electrical Resistance Heating model. This model solves the electric potential field equation. The material composition, electrode configuration, and the applied electrode voltage conditions define the problem. A typical finite element solution scheme is to form the nonlinear system, apply the boundary conditions, and then solve the linearized system of equations. The solution provides the electrical heat source.

4.3.2 Input Requirements

The input data required for this model include the finite element mesh description and the material properties for each element. In particular, the electrical conductivity for the material in each element is required. Time dependent line source conditions are required to describe the electrode position and potential. This is external to the problem and must be supplied by user input.

4.3.3 Output Requirements

This model will produce the electric potential, electric current density, and heat generation rates for each element. The heat source will be used in the Thermal Energy Transport model.

4.4 Thermal Energy Transport with Melting

4.4.1 Description

The data flow diagram in Figure 4.5 shows the Thermal Energy Transport with Melting model. This model solves the thermal energy transport equation

Figure 4.4. Data flow diagram of the electrical resistance heating model.

Figure 4.5. Data flow diagram of the thermal energy transport model.

including convection and diffusion effects. The material composition, domain boundary conditions, and electrical heat source conditions define the problem. A typical finite element solution scheme is to form the nonlinear system, apply the boundary conditions, and then solve the linearized system of equations. The solution provides temperatures and melting rates within each finite element.

4.4.2 Input Requirements

The input data required for this model include the finite element mesh description, the melt velocity field, and the material property data for each element. In particular, density, heat capacity, heat of fusion and thermal conductivity are required for the material composition within each element. Initial temperature conditions are required at each node. The source term includes heat generation rates from electrical heating and chemical reactions. Radiation boundary conditions and convection boundary conditions are required at the soil-air interface. Temperature boundary conditions are required on the far-field boundary.

4.4.3 Output Requirements

This model produces the temperature distribution and melt geometry. Both of these are used in the Melt Flow model. The temperature distribution is used in the Process Chemistry model, and the Gas Migration and Gas Release models. The energy flux at the soil-air interface is used in the Combustion model. Since many of the natural properties are dependent on temperature, the temperature obtained from this model is also used to update material properties at the conclusion of each time step.

4.5 Melt Flow

4.5.1 Description

The data flow diagram in Figure 4.6 shows the Melt Flow model. This model determines the velocity and pressure fields in the melt.

Figure 4.6. Data flow diagram of the melt flow model.

4.5.2. Input Requirements

The input data required for this model include the finite element mesh description, melt boundary location, and the material property data. In particular, the viscosity and density are required for the material composition within each element. Required boundary conditions include prescribed velocities on the melt boundary and air pressure at the soil-air interface.

4.5.3 Output Requirements

This model produces the pressure and velocity fields in the melt. These quantities are used in the Process chemistry model and in the Gas Migration model. The velocity field is used in the Thermal Energy Transport model.

4.6 Process Chemistry

4.6.1 Description

The data flow diagram in Figure 4.7 shows the Process Chemistry model. This model solves the process chemistry for a limited number of chemical reactions.

4.6.2 Input Requirements

The input data required for the process chemistry model include the temperature and pressure distributions and material property data for each element in the mesh. In particular, the quantity and composition of the chemical species present are required along with Gibbs functions and fugacities for the chemical components.

4.6.3 Output Requirements

Quantities and chemical compositions of gases and products generated by chemical reactions will be produced by the process chemistry model. These

Figure 4.7. Data flow diagram of the Process Chemistry model.

values are used in both the gas Migration and the Gas Release models. Heat source from chemical reactions will be used in the Thermal Energy Transport model.

4.7 Gas Migration in the Surrounding Soil

4.7.1 Description

The data flow diagram in Figure 4.8 shows the model for transport of gases to the melt and through the surrounding soil. A typical finite element solution scheme is to form the nonlinear system, apply the boundary conditions, and then solve the linearized system of equations. This model computes the gas flow rate into the melt and through the porous regions surrounding the melt.

4.7.2 Input Requirements

The input data required for the gas transport model include the finite element mesh description, gas viscosity, density, compressibility and thermal expansion coefficients, and soil permeability and porosity. The required boundary conditions are atmospheric pressure at the ground surface and far-field boundaries. The rate of gas generation is supplied by the Process Chemistry model and the prevailing temperature field is supplied by the Thermal Energy Transport model.

4.7.3 Output Requirements

This model produces the gas pressure and flow fields in the melt and the surrounding porous region of the soil.

Figure 4.8. Data flow diagram of the gas transport model.

4.8 Gas Release from the Melt

4.8.1 Description

The data flow diagram in Figure 4.9 shows the Gas Release model. This model determines the rate and composition of gases released at the melt surface.

4.8.2 Input Requirements

Primary input data for the gas release model include melt viscosities and densities; gas densities; the flow rate and composition of the gases entering the melt; the chemical species that are destroyed in the gases; the initial distribution and sizes of bubbles, temperature, pressure, and velocity fields in the melt; and a coalescence frequency function.

4.8.3 Output Requirements

This model computes the quantities, sizes, rate, and composition of gas bubbles released at the melt surface. These values are used by the Combustion model.

4.9 Flow and Combustion in the Confinement Hood

4.9.1 Description

The data flow diagram in Figure 4.10 shows the Flow and Combustion model. This model solves the flow and combustion problem in the off-gas confinement hood.

4.9.2 Input Requirements

The input data required for this model include the gas in-flow rates, composition and thermodynamic state, thermal energy boundary conditions (boundary temperatures or energy fluxes), gas viscosities, thermodynamic

Figure 4.9. Data flow diagram of the gas release model.

Figure 4.10. Data flow diagram of the flow and combustion model.

properties, and reaction rate data. In addition, the geometry and computational mesh data and ignition information are required.

4.9.3 Output Requirements

This model produces the gas temperatures, pressures, velocity fields, densities and enthalpies.

4.10 User Interface

This section describes the user interface with the ISV simulation code.

4.10.1 Input Processor

The user input will be read and diagnostics generated. The simulation request will then be validated, checking for proper syntax of all input commands, values of all input data within appropriate range, and correct specification of all input for each model selected for execution. A problem description is created for use by each of the simulation models. Model specific data will be stored appropriately. After the simulation request is validated, problem specific data is created for the simulation properties and the finite element mesh description. The simulation controls are then set and passed to the computational processor. The input processing requirements are summarized from the draft ISV modeling requirements document:

1. Select initial/restart capability
2. Select initial/restart data files for input
3. Select final/restart/plot files for output
4. Select input units as SI or British
5. Provide (optional) echo of input data

6. Provide default values for all unspecified user parameters
7. Select models to be invoked by the computational processor. Provide default selection of models
8. Accept initial material composition conditions of waste site to be modeled
9. Accept material properties (physical and chemical) or provide default properties
10. Generate the computational meshes
11. Accept initial/boundary conditions for simulation models
12. Accept model control parameters, such as time-step limits, accuracy requirements, and convergence criteria
13. Validate input for consistency of type and value within range, as appropriate

4.10.2 Graphics Interface

The graphics interface is not depicted in the preliminary design of the software architecture. More information is required to define the software requirements. This will be completed in a software requirements specification. In concept, the graphics interface module manages all graphics input and output. The user-selected input from a graphics interface includes optional problem initialization and restart and dynamic selection of time dependent quantities for output and display. The graphics interface will operate on the Cray or on workstations communicating through TCP/IP protocol. The graphics requirements are summarized from the draft ISV modeling requirements document:

1. Input--Accept dynamic requests for time-dependent information summary.
2. Output--Provide time step summary. Display images of transient quantities, including:
 - a. Melt zone growth rates and geometries.
 - b. Power consumption and energy losses.
 - c. Composition and quantity of off-gasses.
 - d. Fluid flow, temperature, and concentration fields in the molten zone and surrounding soil.
 - e. Temperature and concentration fields in the containment hood.

4.10.3 Error Processor

An error processor will communicate all diagnostic messages and error conditions to the user.

4.10.4 Output Processor

The output processor provides the file interface for all generated output. A portion of the output processing is performed under the simulation control, since quantities must be output at the end of each time step or at some user-selected frequency. Another portion produces the final simulation summary report:

1. Generate report data files
2. Generate restart data files

3. Generate plot data files

4. Generate summary report.

5. ISV MODEL DEVELOPMENT PLAN

The ISV analysis package will provide analytical support for several tasks including safety analysis, test design and analysis, and equipment design. In order to support these objectives in a timely manner, it has been determined that the individual model development will proceed in parallel with the design of the overall code structure. This will allow separate effects studies to be made before the entire analysis package has been assembled and commissioned. Furthermore, development of the various models will proceed in stages of increasing complexity, such that parametric studies can be made earlier than otherwise. Such studies will, in fact, be helpful in determining how rigorous each model will need to be as well as in supporting the above ISV objectives. The parametric studies will be undertaken to simulate various burial conditions and will be expanded as experimental data becomes available. These studies will serve either to verify the models or indicate how they may need to be improved to provide more accurate simulations. Finally, it is planned to ensure the review and proper documentation of model and code design and that there will be code version traceability.

The immediate primary objective of the ISV Modeling and Testing Program is to provide a basic capability for assessing the safety of the large "hot" test that is scheduled to take place in the summer of FY-91. The following sections outline the plan to meet this objective. Primary emphasis is placed on testing and model development for the latter half of FY-90. Tests and modeling tasks are described and proposed activities in FY-91 are outlined.

5.1 ISV Tests and Modeling

Tests are necessary to identify and characterize the important phenomena which must be modeled. ISV Tests can be divided into four categories:

1. Laboratory tests to determine physical and chemical material property data,

2. Separate-effects tests which measure parameters described by individual models and inputs required by these models,
3. Bench and engineering size tests (electrode separations of 0.1 to 0.4 m) which permit detailed measurements of a limited number of phenomena and their interactions, and
4. Intermediate and large field tests (electrode separations of 1 to 5 meters) which provide data for an environment similar to actual production conditions.

The data gathered from the first three types of tests will be used to assess and define the functional requirements of the individual models. The fourth category of tests will provide data for integral assessment of the ISV models and evaluation of scale-up capabilities. Currently, three engineering tests (5, 7, and 8) and two intermediate tests (I^1 , I^2) are planned for the last quarter of FY-90. These tests will begin to provide data for model validation and future tests design and planning. The tests and variables to be measured for the different ISV models are illustrated in Figure 5.1.

Two large tests, "cold" and "hot", are scheduled to be conducted in FY-90. The "cold" test will be a nonradioactive, nonhazardous test using the full size operational system. Test pit features will include waste containers, simulated waste materials and simulated plutonium tracers. The "hot" test will also use the full size system and will be conducted in pit #9 of the Subsurface Disposal Area (SDA). Efforts are being made to characterize pit #9 wastes to aid the selection of a suitable test site. Items such as large metal objects and wastes containing high levels of plutonium and uranium will be avoided.

5.2 Model Development

The goals for model development for FY 90 are 1) to determine the suitability of each model, including the increase in sophistication each may

Figure 5.1 ISV tests and variables to be measured for the ISV models.

require to meet model requirements, 2) to determine to what degree various models need to be coupled in order to adequately represent key integral phenomena, and 3) to make recommendations for modeling and experiment design for FY 91 based on insights gained from parametric studies combined with testing.

As shown in Figure 5.2, the FY-90 development plan consists of several tasks. These tasks and associated deliverables (reports) are described below.

5.2.1 Develop Electric Resistance Heating and Energy Transport Model (Task lines A and B)

Workscope: Develop and interface models describing the heating and melting processes. These models include:

1. A computer model describing the Electrostatic Potential and current density fields and the distributed heat source
2. A computer model describing the temperature field and melting process
Perform validation and parametric studies

Deliverables:

Model Design and Methods Verification: Heating and melting
Processes in ISV (09/27/90)

5.2.2 Develop a Fully Implicit Fluid Dynamics Code for Melt Flow (Task line C)

Workscope: Develop boundary condition logic for the moving melt boundary in two-dimensions and implement and test logic. Convert logic to indirect addressing and add three-dimensional capability.

Figure 5.2 The FY-90 ISV model development plan.

Deliverables:

Model Design and Methods Verification: Melt Flow in ISV (09/27/90)

5.2.3 Develop Process Chemistry Model (Task line D)

Workscope: Develop equilibrium chemistry computer model which will compute pyrolysis products generated during ISV and perform supporting analysis.

Deliverables:

1. Equilibrium Chemistry Model for ISV (08/24/90)
2. ISV Test Analysis and Equilibrium Chemistry Model Validation (09/27/90)

5.2.4 Develop Off-Gas Release Model (Task line E)

Workscope: Develop a computer model which will calculate the release of gases from the ISV melt and perform parametric studies and test analysis.

Deliverables:

1. OGRE/MOD1: A Computer Model For Predicting Off-Gas Release From In Situ Vitrification Melts (06/27/90)
2. Parametric Studies of Off-Gas Release During ISV (08/17/90)

5.2.5 Perform 1D Melting/Gas Release Calculations Using the code SCDAP (Task line F)

Workscope: Pre-and post-experiment calculations for the FY-90 engineering scale ISV experiments will be performed. Also, parametric

studies using coupled heatup, melting, and fission product behavior models in SCDAP/RELAP5 will be conducted. For these calculations SCDAP/RELAP5 will be modified to include: (a) the capability to specify electrical power and heat generation histories and distributions in the melt and adjacent regions and (b) replacement material property routines to be supplied by the ISV modeling group.

Deliverables:

1. Parametric Studies of Coupled Melting and Gas Release Processes in ISV (Results only)
2. Post test calculations (results) for FY-90 tests

Final Deliverable from the above two tasks 5.2.4 and 5.2.5

- ISV Test Analysis and Off-Gas Release Model Validation
(09/27/90)

5.2.6 Numerical Studies of Heat Transfer, Fluid Flow and Gas Diffusion (Task line G)

Workscope: Perform parametric studies using inhouse codes ABAQUS and leased code FIDAP to:

1. Examine melt flow and determine its effect on the temperature field and melting process,
2. Examine gas diffusion and determine its importance,
3. Establish requirements for mesh and time step size to produce accurate solutions,
4. Support Test Design and Analysis, and

5. Provide Benchmark Calculations for model testing and verification.

Deliverables:

1. Parametric Studies of Fluid Flow and Heat Transfer Processes in Relation to ISV (08/17/90)
2. Heat Transfer and Fluid Flow Processes in Engineering Scale Tests (FY-90)(09/27/90)

5.2.7 Develop Combustion and Flow Model (Task line H)

Workscope: Develop a computer model to calculate flow and combustion in the ISV Off-Gas confinement hood. Perform scoping calculations in support of ISV safety analysis.

Deliverables:

1. Model Design and Method Verification of flow and combustion in the ISV Off-Gas Confinement Hood (07/31/90)
2. Parametric Studies of ISV Off-Gas Flow and Combustion (09/27/90)

5.2.8 Define Software Requirements and Design Specifications (Task line I)

Workscope: Define software requirements and design specifications for the Input, Computational, and Output processors for the Electric Resistance Heating and Thermal Energy Transport Model.

Deliverables:

1. Draft ISV Modeling Software Requirements Document (06/29/90)

2. Draft ISV Modeling Software Design Specifications Document
(09/27/90)

The objectives for FY 91 are to validate the models using large "cold" test data and to provide the capability to assess the safety of the large "hot" test. As mentioned earlier, future model development will be guided by parametric studies and experimental results that will be obtained and/or interpreted during FY 90. It will become clear, after these results are obtained, which models will require improvement, and which models will be adequate to meet ISV objectives. Finally, the possibility of evolving ISV objectives, as new insights are gained, will certainly have an impact on future model development efforts.

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