

Programm LWR-Sicherheit

Prediction of Fission Product and Aerosol Behaviour During a Postulated Severe Accident in a LWR

Methodology Developed in the
Program Light Water Reactor Safety (PSI)

S. Güntay, F. Aeby, M. Raguin, R. Passalacqua

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February 1990

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Abstract

Lack of appropriate energy removal causes fuel elements in a reactor core to overheat and may eventually cause core to degrade. Fission products will be emitted from a degraded reactor core. Aerosols are generated when the vapours of various fuel and structural materials reach a cold environment and nucleate. In addition to the fission products release and aerosol generation taking place in the reactor vessel, some more fission products release and aerosol generation will occur when the molten core debris leaves the pressure vessel bottom head and comes in contact with the pedestal concrete floor.

Fission products, if they are released to environment from the containment boundary, exert a great danger to public health. A source term is defined as the quantity, timing, and characteristics of the release of radionuclide material to the environment following a postulated severe accident.

At PSI a considerable effort has been spent in investigating and establishing a source term assessment methodology in order to predict the source term for a given Light Water Reactor (LWR) accident scenario.

This report introduces the computer programs and the methods associated with the release of the fission products, generation of the aerosols and behaviour of the aerosols in LWR compartments used for a source term assessment analysis at PSI.

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1. Introduction

Lack of appropriate energy removal causes fuel elements in a reactor core to overheat and may eventually cause core to degrade. This can happen after loss of coolant accident when the emergency core coolant systems are not available or after a transient with or without scram if the heat removal systems are not brought in operation. Fission products will be emitted from a degraded reactor core. Aerosols are generated when the vapours of various fuel and structural materials reach a cold environment and nucleate. In addition to the fission product release and aerosol generation taking place in the reactor vessel, some more fission products release and aerosol generation will occur when the molten core debris leaves the pressure vessel bottom head and comes in contact with the pedestal concrete floor.

It is expected that vapours of the control rods and structural or concrete materials will nucleate earlier than the vapours of the volatile fission products. The vapours of the fission products will condense on already formed aerosol particles. Therefore, inactive aerosols constitute an environment for the transport and removal of the fission products.

Fission products, if they are released to environment from the containment boundary, exert a great danger to public health. Source term is defined as the quantity, timing, and characteristics of the release of radionuclide material to the environment following a postulated severe accident. Estimation of the source term requires various strong interactions among various disciplines, such as primary system thermal-hydraulics, fission and aerosol release and transport in the primary coolant system, thermodynamics in the containment, core-concrete interaction, ex-vessel aerosol generation and aerosol retention in the containment and release to the environment. The thermal-hydraulic conditions existing in the primary coolant system and as well as in the containment strongly influence the timing and magnitude of the release of the gaseous fission products and fission product and structural material aerosols to the environment.

At PSI a considerable effort has been investigated to establish a source term assessment methodology in order to predict the source term for a given light water reactor (LWR) accident scenario. The methodology is composed of a series of coupled computer programs as shown in Figure 1. The codes are coupled in a sense that the

information from one code to the others is transferred in a forward manner and is used as the boundary condition in the next code. However, the information back feed to the code where it is originated is not considered. Integrated computer programs, such as MELCOR, where the information flow in both directions between the codes is considered, are also available at PSI which analyse the whole sequence of a postulated severe accident in one shot. However, it is believed that running mechanistic computer programs as shown in Figure 1, some of which have been developed at PSI, provides more detailed insights into various severe accident phenomena. The integrated or coupled source term computer programs permit a self-consistent treatment of individual accident sequences.

This report introduces the computer programs and the methods associated with the release of the fission products, generation of the aerosols, and aerosol behaviour in LWR compartments used for a source term assessment at PSI.

2. Fission Product Inventory Analysis

Fission products are generated in light water reactor fuel elements when they are exposed to irradiation in the core. Amount of them depends on the residence time of the fuel elements in the core and magnitude of the burn-up. Higher the burn-up and the longer the residence time cause more fission products being built up in the fuel pellets.

The ORIGEN-2 code (Ref. 1) is used to calculate the fission product inventory of a reactor core. ORIGEN is a widely used computer code for calculating the build up, decay and processing of radioactive materials. The cross sections, fission product yields, decay data, decay photon data are either hard wired in the program or are made available as data libraries during the execution of the code. The program has a very flexible input scheme that allows user to calculate the burn-up and the fission products fuel inventory for a given reactor power and history as well as the reactor decay power after the reactor scram.

The power history of various fuel batches, their burn-up and initial U^{238} and U^{235} contents are the only plant specific input data. In order to perform a relatively detailed fission products fuel inventory analysis, fission product build up in individual fuel batches must be calculated based on the in-core residence time and burn-up. The total fission products core inventory is then the summation of the individual batch inventories.

The fission product fuel-cladding gap inventory and released inventory at high fuel temperature may also be calculated using a set of user defined release rate constants. The program produces various types of outputs such as inventories of individual isotopes and elements in mass (gram), number of molecules (gram-atom), radioactivity (curie) and thermal power (watt).

The ORIGEN-2 code was obtained from ORNL (USA) and was installed (Ref. 2) on CDC and CRAY computers. Some minor modifications were made to correct programming errors and be able to execute larger problems than those foreseen by the code developers.

The ORIGEN-2 code does not require any user input parameter which tunes the internal calculation method. Therefore, uncertainties in the outcome of the code are stem from the uncertainties in the decay and cross section libraries and solution methods implemented in the code.

Reference 3 indicates that measured and predicted isotopic ratios agree generally within 5 percent, with a few exceptions on the order of 10 percent. However, the same reference also indicates that some uncertainty analyses performed for the source term purposes show 43 % underprediction to 76 % overprediction. This is observed when calculated inventories of nine isotope (H^3 , C^{14} , Kr^{85} , Ru^{106} , Sb^{125} , I^{129} , Cs^{134} , Cs^{137} , and Ce^{144}) are compared with the measured data from H. B. Robinson and Oconee fuel rod specimens experienced burnups around 30 GWd/MtU. With this comparison it is indicated that the ORIGEN-2 code estimates the individual nuclide inventories within a ± 30 % deviation.

3. In-vessel Release of Fission Products and Vapours of Structural Material

When a LWR core degrades, fission products are released from the fuel pins and transported away with the steam/non-condensable gas and aerosol flows. Determination of the release rates of fission products from uranium (UO_2) has been an important issue in the severe accident research programs. There have been several experimental programs carried out in various laboratories which provided certain data to enlighten the mechanisms of the release. Although the release data are put into Arrhenius diagrams to indicate fractional release at certain temperatures, the physics and chemistry of the fission

products are still partially understood. However, many detailed codes have been prepared to deal with the mechanisms of the fission product release from inside of the fuel pellets to the pellet surface and consequently into steam/non-condensable gas-aerosol environment.

Instead of using an existing code to calculate the release, a stand alone computer program, SASPROG, was prepared (Ref. 4) in order to use the necessary data from a core thermal response code available at PSI (e.g., SWR-BOIL (Ref.5)) and to provide the integral release of the whole range of fission products. The other aim is to calculate the release of vapours of various control rod and structural materials beside the calculation of the fission product release. The information pertaining to a) the predicted temperatures of the fuel, control rod and structural materials in the reactor vessel, b) fraction of initial fuel and other material inventories remained in their original locations (rest indicates the amount that has already relocated downward), c) fission product inventory in the fuel, d) decay power after the scram, and e) a set of fractional release rates for all the elements or element-groups are the main boundary conditions for the release calculation.

Following paragraphs describe the interactions with other computer programs required for a SASPROG analysis. Also described is the calculational method.

The thermal and mechanical behaviour of the fuel, control rods and structural materials are currently calculated using the SWR-BOIL code (Ref. 5) in the current LWR source term study at PSI. The SCDAP/RELAP5 (Ref. 6) can also be used for the same purpose. The SWR-BOIL or SCDAP/RELAP5 code represents the core with "nodes" and provides time dependent temperature and extent of material relocation of each node. The SASPROG code requires the time dependent temperatures of the nodes representing the fuel elements, control rods and structural materials.

Once the whole core slumps into the lower plenum, the SASPROG also requires the time dependent temperature of the core debris until the whole core debris is ejected out of the reactor pressure vessel.

Fission product fuel inventory can be taken from an ORIGEN-2 analysis. The control rod and structural material inventories are to be added to the inventory file. The SASPROG code requires the axial and radial power profiles in order to distribute the fission products as a function of nodes. The structural or control rod material is uniformly distributed among the nodes based on the node dimensions.

The SASPROG code offers three options associated with the selection of the release model. The first option allows user to define the temperature dependent fractional release for any given number of elements. The second option allows user to select the release model based on the SASCHA experimental program (Ref. 7). The third option provides the selection of the release model based on the NUREG-0772 data base (Ref. 8).

Release rates of various fission products and structural materials at 2400 °C determined during the SASCHA experimental program are used as the basis for the second option. The SASCHA fractional release rate constants at 2400 °C are shown in Table-1. The SASPROG calculates the release rate of an element at a temperature based on the following equation:

$$k(T) = k_{2400} * e^{(\ln 4 * (T/200 - 12))}$$

where k_{2400} is the fractional release rate (minute⁻¹) at 2400 °C, T is the temperature in °C and $k(T)$ is the fractional release rate (minute⁻¹) at temperature T.

The following equation correlates the temperature dependent release fractions based on the NUREG-0772 data base:

$$k(T) = A(T) * e^{B(T) * T}$$

Table-2 presents element and temperature dependent constants A and B adopted from Reference 3.

The details of complex phase interactions of various components within the melting core are, for most part, not known quantitatively. Hence these rates are valid only to the extent that the SASCHA experiments or NUREG-0772 data base adequately modelled a core meltdown situation. Table-3 compares the fractional release rates based on the NUREG-0772 data base and with those calculated using the SASCHA release rates at 2400 °C. Except for barium and ruthenium, fractional release rates of various elements based on the SASCHA experimental data are lower by a factor between 1 to 3 than those calculated based on the NUREG-0772 data base. Reference 7 indicates that barium, tellurium, antimony, and silver releases are depending very much on the extent of oxidation of the zirconium cladding. This factor is approximately 1000 for barium and 500 for ruthenium. Extent of zirconium cladding oxidation considered in SASCHA experiments and in the NUREG-0772 data base should probably be different and causes

the difference. Reference 3 indicates that the effect of zirconium oxidation on the release rates of these materials (also on tellurium) is not well quantified. This reference also states that it is not known with any certainty whether this phenomenon is exhibited in the release rates of other metals. For the difference observed between the fractional release rates of ruthenium, no explanation has been found in the open literature. However, due to small magnitude of the release of ruthenium, any inaccuracy in its fractional release rate is believed not to cause an extensive uncertainty in the ruthenium source term to the environment.

The SASPROG then calculates the time dependent release of an element using the following equation :

$$C(t)=C_0*e^{(-k(T)*t)}$$

where C_0 is the fuel or structural material inventory at the beginning of the period "t". C is the inventory after the release during the period "t". $k(T)$ is the fractional release rate at fuel or structural material temperature T during the period "t" depending on the user selected option. The amount of released mass during this time period is the difference between the initial and final inventories at the beginning and end of this period.

It is assumed that the release of the fission products or vapours of the structural material occurs only from the fuel or structural material that is residing in any node. The time dependent mass fraction of the initial node inventory is an user input data to the program. No release is considered from the portion of the node mass that has relocated. The code, however, makes accounting of the mass accumulated in the water. Once the water in the lower plenum boils off, the code continues in calculating the release of the fission products and vapours of the structural material from the accumulated core debris. The lower plenum boil off time is an user input to the code.

Major driving force for the transport of the fission products and vapours of the structural materials is the steam flow. Once the lower plenum of the pressure vessel is dried out, the steam flow stops. The SASPROG code, therefore, assumes that the transport of the fission products (except the noble gases) and vapours of the structural material does not occur. However, the code further assumes that the release continues and the vapours of the materials do not condense on the surfaces. Once the reactor bottom head fails, it is assumed that the mass already released from the core or core debris will be available for the aerosol generation outside of the reactor vessel just one second after

the reactor bottom head breaches. The time after which no more release is desired until the vessel breach time is an user input data. The program considers the time of the last data point entry in the temperature file as the vessel bottom head breach time.

The fission products and vapours of the structural materials released within the core can be used as an input to determine the chemical forms of the fission products, aerosol generation and in-vessel transport of the fission products and aerosols. The chemical forms of the fission products will be discussed in Section 4 and the transport of the fission product in Section 5. The current source term analysis at PSI assumes a complete aerosolization of the whole fission products and structural material vapours with a given chemical and physical forms of the aerosols as displayed in Table-4.

The SASPROG code also calculates the equivalent release in number of moles (gram-atom), and radioactivity (curie) and thermal power (watt). The change in the mass of elements due to the decay or parent to daughter chain is not considered in the program however, the decay is considered in the thermal power and radioactivity of the individual element or element groups. The decay data as a function of time and element or element groups shown in Table-1 is an user input data.

ORIGEN-2 provides inventories of 67 elements. This number will then be increased when the structural and control rod materials are included. In order to reduce the number of entries in the inventory file for further processing to a reasonable number, the SASPROG code performs a grouping by introducing the elements with similar characteristics, such as noble gases, in the same group. Each group is composed of one or up to 7 elements. 40 groups are established and are shown also in Table-1. The SASPROG then calculates the cumulative release for each group in four different units.

4. Chemical Forms of the Fission Products after the Release From the Core

After the release from the overheated or degraded reactor core, specific chemical reactions in the primary system may result in significant changes in the chemical and physical forms of the released fission products. The following processes are expected to occur:

- vapour with vapour,

- vapour with aerosol,
- vapour with clean or oxidized surfaces or surfaces with surface deposits,
- aerosol with aerosol,
- aerosol with surfaces,
- vapour with steam/hydrogen,
- aerosol with steam.

The extent of these reactions will primarily be depending on various parameters such as the chemical and physical forms of the volatilized material together with the timing of the release.

Determination of the chemical forms of the fission products can be done either using a thermodynamic code such as SOLGASMIX-PV (Ref. 25) or based on some engineering judgement using the information obtained from various code calculations or experimental data. The experience in using the SOLGASMIX-PV code at PSI has indicated that the code results are very much dependent on the accuracy of the Gibb's free energy constants supplied to the code. The other concern in using a thermodynamic code is the omission of the kinetics effects. The current source term analysis at PSI, therefore, employs the second approach. The assumed chemical forms of the fission products are shown in Table-4. This is accomplished in the ARES code and will be discussed in Section 8.

5. In-vessel Fission Product Transport Analysis

Radionuclides released from the melting core will undergo chemical and physical changes and will deposit on various surfaces as they are transported through the Reactor Coolant System (RCS) to containment. What fraction of these released materials escapes the physical boundary of the RCS depends mainly on the accident scenario which defines the release path and thermal-hydraulic conditions in the RCS. The retention in the RCS is directly determining the source term to the environment for the accidents in which the containment is by-passed.

Two computer programs, namely, PULSE from EG&G (USA) (Ref. 9), and TRAP-MELT from BCL (USA) (Ref. 10) are available at PSI for the fission product transport calculation in the primary system. Main input to these codes are the source rates of fission products, such as iodine, cesium, strontium, tellurium, etc, and thermal-hydraulic conditions prevailing in various parts of the primary coolant system. A representation of the primary coolant physical system with control volumes is required. In each control volume a transported species can reside on at least two carriers either in particle or vapour form. Four states in which species may reside can be described: a) suspended-molecular (vapour), b) suspended particle, c) deposited-molecular (vapour condensation), and d) deposited-particle. A fifth state, surface-reacted, is used to describe the vapours which have reacted with reactor coolant surfaces and are not considered to be subject to revolatilization. The main cause for the revolatilization is the possible fission product heating and is not considered in both programs. Both codes consider the following mechanisms for deposition on surfaces:

- . Vapour reaction with surfaces (chemical reaction),
- . Submicron and supermicron particle depositions from fully developed turbulent flow when no temperature gradient exists between the wall surface and carrier gas,
- . Particle deposition from laminar flow when no temperature gradient exists between the wall surface and carrier gas,
- . Particle deposition due to the thermophoresis,
- . Particle deposition due to gravitational sedimentation.

The interaction processes considered are:

- . Brownian coagulation,
- . Turbulent coagulation,
- . Gravitational coagulation.

Both codes are complementary to each other in terms of modelling the various phenomena. The PULSE provides detailed simulation of the transport in pipings with various bends. The TRAP-MELT is limited by the number of control volumes by which the physical system is to be represented.

6. Analysis of Aerosol Generation During Core Debris Interaction with Concrete

Progression of a severe reactor accident will eventually lead to the point where molten core debris penetrates the primary vessel and falls into the reactor cavity. The interaction of molten core debris with structural concrete is known to produce vast amounts of aerosols and cause release of fission products. Experimental investigations of melts interacting with concrete have shown that the concrete is aggressively attacked. A most important aspect of this attack is that hydrates and carbonates within the concrete are thermally decomposed to produce steam, carbon monoxide and carbon dioxide. It has been observed during various core debris simulant-concrete experiments that these gaseous products of concrete decomposition sparge up freely through the melt. As the gases pass through the melt there is significant generation of aerosols. A code, VANESA (Ref. 11), was prepared by SNL (USA) to calculate the aerosol generation arising from such a core debris-concrete interaction. The code considers two main mechanisms of aerosol generation:

- i) **Vaporization:** Species with relatively high vapour pressures at given temperature evaporate into the gas stream. When the gas stream cools, these vapours condense to form aerosol. Vaporization of species from the melt can be enhanced because of the gases sparging through the melt are quite reactive. Chemical reaction taking place between some of the species and the water vapour may generate more volatile hydroxides. In addition to this enhancement, internal heat generation due to oxidation of metallic constituents of the melt increases the evaporation.
- ii) **Mechanical:** Violent agitation of the melt by sparging gases leads to formation of particulate melt in the gas stream. Mechanical generation of aerosols occurs by two means. The first develops when melt first contacts concrete and there is a period of exceptionally violent gas generation. During this period, droplets of melt are thrown into the gas. Inertial forces cause the particles to disintegrate until they become small enough. As a result, maximum size of the entrained droplets is naturally controlled.
The second mechanism is associated with the bursting of the gas bubbles rising in the melt at the melt surface. As the bubble breaks, the melt film over the bubble surface is thrown off in the form of fine particles and resulting particles are entrained in the gas stream.

The composition of the aerosol produced by vaporization depends on the vapour pressure and chemical reactivity of the constituents of the melts. Consequently, the aerosol composition is not necessarily similar to the melt composition. On the other hand, the composition of aerosols produced by the mechanical processes is very nearly the same as that of the melt.

The aerosol generation rate is mainly depending on the melt temperature, gas flow, and concrete ablation rate. These information can be calculated using the WECHSL (Ref. 26) or CORCON (Ref. 27) code.

Experience with the VANESA code application has shown that the impact of the calculated melt temperature on the aerosol generation and fission product release is large. Figure 2 adapted from Reference 14 indicates that the aerosol generation rate is exponentially dependent on the melt temperature.

When the core debris attacks pedestal concrete, hydrates and carbonates within the concrete are thermally decomposed to produce steam, carbon monoxide and carbon dioxide. As these gases flow through the melt, aerosols are generated as indicated earlier. Figure 3 adopted from Reference 14 indicates the effect of the flow rate on the aerosol production rate. Except for very large and very small flow rates, the aerosol production rates are essentially linearly dependent on gas flow rates.

Concrete erosion rate defines the rate of concrete and steel additions from the basement into the melt and consequently affects the aerosol generation rate.

The VANESA code from the US Source Term Code Package (STCP) (Ref. 11) is implemented on CYBER and VAX computers. The gas generation and concrete ablation and melt temperature information are calculated using the WECHSL code at PSI. A small modification in the WECHSL code is made in order to prepare a file containing required information. The WECHSL code prepares this information relatively often at the beginning of the core debris-concrete interaction and the time span is increasing between the time points later in the transient. On the other hand, the VANESA code is written to accept this information only at regularly spaced time points. Therefore, a modification is made in the VANESA code to process the WECHSL file and prepare an internal file which contains this information at equally spaced time points. The composition and inventory of the melt at the beginning of the core-concrete interaction are calculated in the code depending on the initial fission product and structural material inventory at the

scram time (ORIGEN-2 output) and the cumulative released fission product and structural material inventory (SASPROG output) at the end of the in-vessel release phase. The difference between the initial inventory and what has been emitted is the required melt inventory for the VANESA code. Other modification in the code is to predict the release of each SASPROG fission product and other material groups in addition to the release in original VANESA fission product groups. The release is calculated in mass (gram), number of molecules (gram-atom), radioactivity (curie) and thermal power (watt). The decay in thermal power and radioactivity is also considered based on the decay information received from the ORIGEN-2 analysis. The modified VANESA version also appends the ex-vessel release information to the in-vessel release information. The combined file is then an input to the Aerosol Retention and Environmental Safety Code (ARES) developed for the containment aerosol behaviour analysis at PSI (see Section 8). The description of the PSI version of the VANESA and the input requirement for this version is given in Reference 13.

7. Analysis of Aerosol Retention in Water Pools

The determination of "Source Term" for a given core damage accident requires that the accident progression, the plant thermal-hydraulic response and the associated radionuclide release and transport be analyzed for the identified escape pathways. Many of these pathways currently identified for what are believed to be risk significant sequences involve pathway segments through water pools. Specifically, transport through BWR pressure suppression pool and PWR quench tank or secondary side of a PWR steam generator represent examples of such segments. In order to properly analyse the fission product transport for these sequences, it is therefore necessary to describe the effect of these water pools on the steam/non-condensable gas mixtures and associated entrained radionuclides.

When the flow passes through the spargers or vents, gas bubbles are produced. As the bubbles rise in the water pool, a phenomenon called "Scrubbing" takes place and fission product and structural material aerosols and gaseous fission products leave the bubble boundaries. It is expected that these pools will retain a significant fraction of the gaseous fission products and aerosols. This attenuation is usually expressed quantitatively as the "Decontamination Factor (DF)" which is defined as the ratio of the mass injected to the mass which escapes from the surface of the pool. The Reactor Safety Study

(WASH-1400) (Ref. 15) assumed a decontamination factor of 100 for subcooled water pools and 1 for saturated pools. Since this early study, various experiments have been conducted, and based on the information gained several computer programs have been prepared. The predicted DF is based on the existing conditions that exist at the point at which steam and non-condensable gas mixtures with associated entrained gaseous fission products and aerosols are injected into the water pools.

At PSI three codes are available to calculate the aerosol removal from bubbles in water pools, namely, SUPRA (Ref. 16) from EPRI (USA), SPARC (Ref. 17) from PNL (USA), and BUSCA (Ref. 18). The last one is a result of a joint development effort of PSI, GRS (Köln/FRG) and SRD (UKAEA/UK). The three codes use somewhat similar methods of modelling of basic aerosol removal mechanism. However, the bubble hydrodynamics are treated differently. A lot of developmental effort has been and will be put into the development of BUSCA. The main advantages of developing this code further at PSI is to incorporate the models which are to be developed at PSI based on the data from the iodine and aerosol experimental programs (e.g, the POSEIDON program at PSI). The final version of this code is planned to replace the SUPRA or SPARC.

Following parameters are controlling the aerosol removal from the bubbles: a) particles size, b) bubble shape and size, c) fraction of steam in inlet gas, d) particle density, e) pool height and temperature, f) composition of the particles (soluble and insoluble), g) non-condensable gas composition, and h) pressure above water surface. The information associated with the aerosols (such as particle size, composition) are calculated using the Aerosol Retention and Environmental Code (ARES) (see Section 8) at PSI. The pool height and temperature and pressure above the pool surface are the information taken from the containment thermodynamic analysis. The parameters associated with the bubble hydrodynamics are calculated by the pool scrubbing codes themselves. These parameters are depending on the incoming gas flow rate, sparger type and dimension, etc..

The mathematical model describing fission product transport is based on fundamental mass and energy conservation laws. The way how they are formulated forms system of equations. The coupled set of first-order ordinary differential equations and various auxiliary equations are solved in a spatial domain using different differential equation solving methods in the codes. Table-5 presents the similarities and the differences between the three codes. Mechanisms causing particle capture during the bubble jet and consequent swarm formation include: i) excess steam (above the pool

vapour pressure) in the inlet gas causes condensation that moves particles to the bubble surface, ii) inlet gas temperatures higher than pool temperature cause thermophoretic deposition, and iii) high inlet velocities cause inertial impaction. A mechanism that hinders particle capture during swarm formation is diffusiophoresis if the steam content of the entering gas is lower than the pool vapour pressure. This mechanism is always active during bubble rise. Gravitational settling and Brownian diffusion are the additional effective deposition mechanism during the bubble rise.

The present pool scrubbing analysis at PSI employs the application of the three codes in addition to the engineering judgement based on the information gained from various experiments. This is intended to reduce the uncertainty in Decontamination Factor due to differences in modelling the bubble hydrodynamics and aerosol scrubbing.

8. Analysis of Aerosol Behaviour In Reactor Containment

The main issue from a postulated severe accident analysis is to predict the environmental release of the gaseous fission products and aerosols from a LWR containment. What could be released to the environment depends mainly on the aerosol retention in various compartments and transport from those compartments to the environment. Therefore, aerosol behaviour and its modelling under given thermodynamic conditions are important tasks to be accomplished within a source term analysis.

Radionuclides in aerosol form will be removed from a compartment atmosphere in a postulated severe accident by natural deposition processes in addition to the leaks and pool scrubbing. The rate of deposition by natural processes is in most circumstances very sensitive to aerosol particle sizes. Below particles sizes of $\sim 0.1 \mu\text{m}$, deposition is dominated by diffusion to the compartment walls under the influence of Brownian motion and increases with decreasing particles size. Above $\sim 1 \mu\text{m}$, gravitational sedimentation increases. Particles that have sizes in the range from ~ 0.1 to $\sim 1 \mu\text{m}$ may consequently persist to stay as airborne in a compartment atmosphere longer. In addition to these removal processes, two more mechanisms, namely thermophoresis and diffusiophoresis, play some role in aerosol removal due to deposition. Thermophoretic deposition takes place when particles suspended in a gas experience a force toward cooler temperature if the gas exhibits a temperature gradient. This force results from the interaction between the gas molecules and the particles. Diffusiophoretic deposition accounts for the force acting on particles in concentration gradients in a compartment

with dissimilar gas components (e.g. water vapour and air). Such situation is of special importance in compartments of a LWR where large amounts of steam condense on cold walls. Inter-compartmental transport due to steam/non-condensable gas flow and removal of aerosols due to pool scrubbing taking place in water pools make aerosol behaviour more complex and should be considered in addition to those natural removal processes.

In order to calculate aerosol behaviour and characterize the released aerosols, several informations are required, such as, a) thermal-hydraulic conditions in compartments, b) in- and ex-vessel source rates of gaseous fission products and fission product and structural aerosols, c) aerosol removal in water pools and filters, and c) geometry of compartments. The aerosol code at PSI, NAUA (Ref. 28), performs an aerosol analysis, however, with certain restrictions, such as requiring considerable user time for input preparation, and also with certain modelling inefficiencies. Therefore, a new code, Aerosol Retention and Environmental Safety Code (ARES) was developed at PSI. The objective of this new code development is to provide a best estimate treatment of aerosol behaviour in various compartments of a LWR and aerosol release to the environment. The specific objectives are to: a) determine characterization of the aerosols, b) improve modelling of aerosol removal to consider i) condensation of steam on aerosols, ii) hygroscopicity of aerosols and iii) thermophoresis, c) include ability to simulate filters and pools, d) calculate dose rates in each compartment in a LWR and total released activity to the environment, e) model any LWR or any experimental facility with a user friendly input, f) transfer directly information from the containment thermal-hydraulic (e.g. WAVCO (Ref. 23)), and fission product and aerosol release codes (e.g. SASFROG and VANESA) and to the iodine behaviour code, IMPAIR2/M (see Section 9). Figure 4 presents various modules in the ARES code. Also shown is the information required to run the code.

The first module in the ARES is a preprocessor code, PRETGEO, to handle the thermodynamic input and extract the information required for the aerosol analysis. It also reads the geometric data pertaining the physical system as it is modelled for the thermodynamic analysis. This code prepares some files to transmit the required information to the other modules.

The second module is composed of a driver program and various sub-programs. The driver program, SOURCE, reads and processes the fission products and aerosol source information. This information covers the in-vessel release from the degraded core (e.g., calculated by SASPROG code) and ex-vessel release due to core-concrete

interaction (e.g., calculated by VANESA code). The file contains the cumulative release of various elements or groups of elements (currently 40 elements or element groups are handled) in mass (gram), number of molecules (gram-atom), radioactivity (curie) and thermal power (watt). The information is further processed and the gaseous fission products and fission product, structural and concrete aerosols are classified into 10 groups. Each group is further classified generally into three modes, namely, gas, soluble and insoluble forms. Table-4 presents the 10 classes and the modes within each class. The first class represents fission gases. In order to trace the dose rates in compartments and also the released activity to environment, although they are not part of the aerosol behaviour calculation, they are specifically considered in the first class. The second class includes the halogens which appear as gas and soluble aerosol forms. The third class represents the alkali metals which appear only in soluble form. The rest of the classes include various insoluble aerosols. This table also indicates the chemical forms of the aerosols. The main objectives of specification of classes and modes are to a) consider implicitly the chemical forms of the fission products, b) better model the behaviour of different aerosol materials, c) determine the dose rates in various compartments, and d) distinguish sources of various iodine species .

Specific chemical reactions that may occur in the primary system may result, to a certain extent, in some changes in the chemical and physical forms of the fission products. In addition, revolatilization of fission products due to fission product heating may cause some late transient release of volatile fission products within the primary system. A transport code can be used to simulate the fission product and aerosol attenuation in the primary coolant system. Current source term analysis at PSI involves the application of Aerosol Code ARES to the RCS as well. It is believed that this will bring some underestimation in the calculated aerosol retention in the primary system because specific fission product and aerosol removal phenomena, such as turbulent deposition, chemisorption, etc., that occur in the RCS are not modelled in the code. However, this error should be small especially for the accident scenarios where the transport path in the RCS is short. For the accident scenarios where the containment is by-passed, specific phenomena governing radionuclide transport and retention in RCS should be considered.

The driver program, AERO, drives the sub-programs, FILTER, BUSCA, NAUA, and IMPIN.

The sub-program FILTER is prepared to calculate the total aerosol load in individual filters. It also checks if the design limit of a filter is reached. If it is the case, user provided decontamination factor is set to unity for that particular filter.

The sub-program BUSCA calculates the decontamination factors for the pools. The current version of the program in the ARES uses a constant-user input decontamination factor. However, it is planned that a multi-dimensional-surface model as a function of water height, flow rate, aerosol inlet conditions, etc will be used to determine the decontamination factor for the considered pool, inlet gas and aerosol flow conditions. The model will be developed using the pool scrubbing codes available at PSI.

These sub-programs, except NAUA, are run at time points found in the thermal-hydraulic data file. Information, such as decontamination factor, is then passed into the NAUA, and the NAUA code is executed for the next time interval starting at that particular time point. It is assumed that the information calculated by the sub-programs FILTER and BUSCA are assumed to remain constant for the current ARES interval.

The sub-program NAUA is the heart of the ARES. It is a modified version of the NAUA-5/M obtained from KfK Karlsruhe/FRG. The modifications have been made at PSI to extend the capabilities of the code to better model the aerosol behaviour in compartments. The following aerosol removal processes are considered in the program:

- Removal processes
 - . gravitational settling,
 - . diffusional plate out,
 - . thermophoretic deposition, (added at PSI)
 - . diffusiophoretic deposition.

- Interaction processes
 - . Brownian coagulation,
 - . gravitational coagulation,
 - . steam condensation on particles, (added at PSI)
 - . hygroscopicity of particles. (added at PSI)

- Transport phenomena
 - . aerosol sources,
 - . leakages.

The formulation of various removal mechanisms can be found in Reference 19. The program accepts only one source combination for various fission product and structural material groups shown in Table-1 in a compartment at any time. The user may move the source from one compartment to another at any time. Depending on the timing and constitution of the release, the constitution of aerosol particles may be dynamically changing. The initial particle characterization, such as particle size, density, shape factor, thermal conductivity and standard deviation, at the time of the release into compartments is an user input data. Currently at PSI, this data is prepared based on the experience for the in-vessel phase and is transferred (except the shape factor) from the VANESA calculation for the ex-vessel phase. It is assumed that particles are homogeneously distributed within a compartment except in boundary layers at walls. The rate of particle growth of the particles due to steam condensation on particles is considered in the sub-program NEWCON. Hygroscopicity allows particles to grow more rapidly and thus, increases the particle settling velocity. The rate of the growth due to steam condensation is described by the Mason equation. The ARES code specifically considers the cesium iodide and cesium hydroxide as the hygroscopic aerosols.

The sub-program IMPIN prepares the necessary information for the iodine behaviour program IMPAIR-2/M (see Section 9). The prepared files contain the source information pertaining iodine species, thermal-hydraulic conditions and geometry of the compartments.

Reference 19 presents the description of the code ARES and input requirement.

9. Analysis of Iodine Behaviour In Reactor Containment

Fission product iodine is a major potential contributor to accident consequences. Therefore, the iodine retention in containment is one of the prime concern in an accident analysis. Chemical state of iodine within the oxidic fuel matrix is not known with confidence (Ref. 20). The same reference indicates that the iodine is present within the fuel matrix predominantly as iodine atoms and molecular iodine with comparatively little formation of cesium iodide. Thermodynamic data and experiments to date suggest that, in the absence of other materials, iodine will be predominantly released into the primary coolant system of a LWR as cesium iodide under the reducing steam conditions of a degraded core conditions. Therefore, in formulating the aerosol classifications as shown in Table-4 the following assumptions are made: a) one percent of the iodine released at

any time will be in gaseous (elemental) form and b) the rest of the iodine is in cesium iodide form. The later is a salt. However, chemical reactions and mass transfer may cause this distribution being changed dynamically. Under the effect of strong chemical reactions taking place in the gas and water phases of compartments, iodine may appear in various forms, namely,

- in the gas phase,
Molecular iodine (I_2), Iodide (I^-), Hydroiodous acid (HOI), Iodate (IO_3^-), Organ_iodine (CH_3I), and Iodide (CsI), AgI and HOI in droplet form
- in the water phase,
Molecular iodine (I_2), Iodide (I^-), Hydroiodous acid (HOI), Iodate (IO_3^-), Methyl iodide (CH_3I) and Silver iodide (AgI).

A new computer program, IMPAIR-2 (Ref. 21), is written to analyse the iodine chemistry in a single compartment containing water and gas phases. Reference 21 describes the chemical reactions taking place between various iodine species. All the reactions are defined with a set of ordinary differential equations which are to be solved simultaneously using Gear's method to provide the concentrations of various iodine species. The reaction rates, mass transfer constants, droplet entrainment and fall back rates and dose rates are the main terms in these equations. The activation and rate constants to be used in calculation of the reaction rate constants are input to the program. The boundary conditions used in the calculation are the initial iodine species concentrations, dose rate, pH of the water and the leak flow rate out of the compartment.

Experimental investigations in various laboratories are underway to understand and create a data base (such as the LIRIC data base created at AECL-WNRE (Ref. 24)) for reaction rate constants governing the complex reactions taking place between the iodine species. Expected accurate information in determination of various rate constants will improve the prediction capability.

The IMPAIR-2 code is further extended for multi-compartment applications. The extended code is named IMPAIR-2/M (Ref. 22). The source rates of various iodine species are considered in addition to the initial concentrations. Each compartment may have its own source term. Compartments without water phase are also considered. Compartments may be in communication with each other through one or more connections. Therefore, a complete treatment of mass transfer of iodine species is realised. A link to an aerosol code for airborne iodine aerosol behaviour and also another link to a thermodynamic code for the intercompartment mass transport are established.

Description of the models and input requirements of the IMPAIR-2/M are given in Ref. 22.

9. Conclusions

Considerable effort has been spent in investigating and establishing a methodology to predict the source term to the environment. The methodology can be applied for a given severe accident scenario postulated to occur in a light water reactor. The methodology associated with the release of the fission products and behaviour of the aerosols in various LWR compartments is presented in this report. The codes calculating the release of the fission products and vapours of the structural materials (the SASPROG code) and aerosol behaviour in the containment (the ARES code) have been developed at PSI. The other codes have been obtained from foreign laboratories, but have been extensively modified.

The understanding of severe accident phenomena is believed to be far from complete. Very little experimental information is available to elucidate certain severe accident phenomena, such as core melting process. Therefore, various uncertainties associated with the code predictions must be recognized.

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Table-1 SASCHA Release Rate Constants for Elements or Element Groups

	Element	SASCHA (min ⁻¹)
	-----	-----
Group 1	: Kr,Xe	0.5
Group 2	: I	0.5
Group 3	: Br	0.5
Group 4	: Cs	0.5
Group 5	: Rb	0.5
Group 6	: Cd	0.5
Group 7	: Ag	0.14
Group 8	: In	0.2
Group 9	: Te	0.071
Group 10	: Se	0.071
Group 11	: Ge	0.071
Group 12	: Sb	0.043
Group 13	: As	0.043
Group 14	: Sr	1.*10 ⁻⁴
Group 15	: Ba	1.*10 ⁻⁴
Group 16	: Mo	1.*10 ⁻⁴
Group 17	: Tc	1.*10 ⁻⁴
Group 18	: Ru	1.*10 ⁻⁶
Group 19	: Rh	1.*10 ⁻⁶
Group 20	: Pd	1.*10 ⁻⁵
Group 21	: Nb	1.*10 ⁻⁵
Group 22	: Y	1.*10 ⁻⁵
Group 23	: La,Zr,Pr,Nd,Pm,Sm,Eu	1.*10 ⁻⁵
Group 24	: Gd,Tb,Dy,Ho,Er,Tm,Yb	1.*10 ⁻⁵
Group 25	: H,He	0.5
Group 26	: Li,Be,C,Sf,Co	1.*10 ⁻⁵
Group 27	: Ni,Cu,Zn,Ga	1.*10 ⁻⁵
Group 28	: Sn	0.014
Group 29	: Ac,Th,Pa,Np,Pu	1.*10 ⁻⁵
Group 30	: Am,Cm,Bk,Cf,Es	1.*10 ⁻⁵
Group 31	: Tl,Pb,Bi,Po	1.*10 ⁻⁵
Group 32	: At,Rn,Fr,Ra	1.*10 ⁻⁵
Fuel		
Group 33	: U	2.*10 ⁻⁴
Structural Materials		
Group 34	: Fe,Cr,Co	1.*10 ⁻³
Group 35	: Mn	1.*10 ⁻²
Group 36	: Sn	0.014
Group 37	: Zr	1.*10 ⁻⁵
PWR control rod materials		
Group 38	: In,Cd,Ag	1.*10 ⁻⁵
BWR control rod material		
Group 39	: B ₄ C ₃	1.*10 ⁻⁴
Concrete materials		
Group 40	: Concrete Materials	-

Table-2 Values Used for the Constants A and B in the Approximation of the Release Rate Coefficients, $k(t)=Ae^{BT}$, based on NUREG-0772 Data Base

Fission product group	800°C < T ≤ 1400°C		1400°C < T ≤ 2200°C		T > 2200°C	
	A	B	A	B	A	B
I, Xe, Kr	7.02E-09 ^a	0.00886	2.02E-07	0.00667	1.74E-05	0.00460
Ca	7.53E-12	0.0142	2.02E-07	0.00667	1.74E-05	0.00460
Te, Ag	3.88E-12	0.0135	9.39E-08	0.00630	1.18E-05	0.00411
Sb	1.90E-12	0.0128	5.88E-09	0.00708	2.56E-06	0.00426
Ba	7.50E-14	0.0144	8.26E-09	0.00631	1.38E-05	0.00290
Mo	5.01E-12	0.0115	5.93E-08	0.00523	3.70E-05	0.00200
Sr	2.74E-08	0.00360	2.78E-11	0.00853	9.00E-07	0.00370
Zr	6.64E-12	0.00631	6.64E-12	0.00631	1.48E-07	0.00177
Ru	1.36E-11	0.00768	1.36E-11	0.00768	1.40E-06	0.00248
Fuel	5.00E-13	0.00768	5.00E-13	0.00768	5.00E-13	0.00768
Cladding (Zr)	6.64E-12	0.00631	6.64E-12	0.00631	1.48E-07	0.00177
(Sn)	1.90E-12	0.0128	5.88E-09	0.00708	2.56E-06	0.00426
Structure	6.64E-10	0.00631	6.64E-10	0.00631	1.48E-05	0.00177

^a7.02E-09 denoted 7.02×10^{-9} .

Table-3 Comparison of US-NUREG-0772 and SASPROG Fractional Release Rates (min^{-1})

Temperature ($^{\circ}\text{C}$)	1200	1900	2100	2300	2400
NUREG I,Xe,Kr		.64441E-01	.24463E+00	.68452E+00	.10843E+01
SASCHA		.15625E-01	.62500E-01	.25000E+00	.50000E+00
NUREG Cs		.64441E-01	.24463E+00	.68452E+00	.10843E+01
SASCHA		.15625E-01	.62500E-01	.25000E+00	.50000E+00
NUREG Te,Ag		.14831E-01	.52286E-01	.15041E+00	.22686E+00
SASCHA		.22188E-02	.88750E-02	.35500E-01	.71000E-01
NUREG Sb		.40880E-02	.16845E-01	.46074E-01	.70545E-01
SASCHA		.13438E-02	.53750E-02	.21500E-01	.43000E-01
NUREG Ba		.13296E-02	.46970E-02	.10880E-01	.14540E-01
SASCHA		.31250E-05	.12500E-04	.50000E-04	.10000E-03
NUREG Mo		.12264E-02	.34907E-02	.30542E-02	.37304E-02
SASCHA		.31250E-05	.12500E-04	.50000E-04	.10000E-03
NUREG Sr		.30385E-03	.16733E-02	.44677E-02	.64681E-02
SASCHA		.31250E-05	.12500E-04	.50000E-04	.10000E-03
NUREG Zr		.10689E-05	.37758E-05	.86751E-05	.10355E-04
SASCHA		.31250E-06	.12500E-05	.50000E-05	.10000E-04
NUREG Ru		.29564E-04	.13735E-03	.42009E-03	.53833E-03
SASCHA		.31250E-07	.12500E-06	.50000E-06	.10000E-05
NUREG U		.10869E-05	.50498E-05	.23461E-04	.50569E-04
SASCHA		.62500E-05	.25000E-04	.10000E-03	.20000E-03
Cladding					
NUREG Zr	.12902E-07	.10689E-05	.37758E-05	.86751E-05	.10355E-04
SASCHA	.24414E-08	.31250E-06	.12500E-05	.50000E-05	.10000E-04
NUREG Sn	.89026E-05	.40880E-02	.16845E-01	.46074E-01	.70545E-01
SASCHA	.34180E-05	.43750E-03	.17500E-02	.70000E-02	.14000E-01
Structural Materials					
NUREG	.12902E-05	.10689E-03	.37758E-03	.86751E-03	.10355E-02
SASCHA	.24414E-06	.31250E-04	.12500E-03	.50000E-03	.10000E-02

Table-4 Classification of Fission Products and Aerosol in ARES

1. class:	NOBLE GAS	100% of Kr, Xe, H and He are in gas form
2. class:	HALOGENS (volatile)	1% of I and Br are in gas form the rest are in CsI, CsBr and RbBr soluble form
3. class:	ALKALI METALS (volatile)	The rest of Cs and Rb are in CsOH and RbOH soluble form
4. class:	TELLURIAN GROUP (volatile)	100% of Te, Se and Ge are in elemental forms
5. class:	ALKALINE EARTHES (non-volatile)	100% of Sr is in oxidic and insoluble form 100% of Ba is in oxidic and insoluble form
6. class:	METALLIC (non-volatile)	100% of Cd, Ag and In are in elemental insoluble form 100% of Sb, As, Gd, Li, Cu, Sn, Am, Ti, At, U, are in oxidic forms
7. class:	NOBLE METALS (non-volatile)	Mo, Tc, Ru, Rh, Pd are in insoluble forms (Mo in oxidic)
8. class:	RARE EARTHES (non-volatile)	100% of Y, Nb, La, Ac are in insoluble oxidic form
9. class:	STRUCTURAL MATERIALS (non-volatile)	100 % of Fe, Mn, Sn are in oxidic form, Ag, In, Cd, or B ₄ C ₃ in insoluble forms
10. class:	CONCRETE MATERIALS (non-volatile)	100% of these elements are in oxidic insoluble forms (60 % is SiO ₂ , 30 % is CaO, 10 % is Al ₂ O ₃)

Table-5 SUPRA, SPARC and BUSCA, Similarities and Differences

	SPARC	BUSCA	SUPRA
Aerosol Removal Mechanism			
Sedimentation	yes	yes	yes
Inertial Impaction			
due to a)inertial circulation	yes	yes	yes
b)flow impaction			
against water-gas			
interface	yes	yes	yes
Brownian diffusion	yes	yes	yes
Thermophoresis	yes	yes	yes
Diffusiophoresis/stephan			
flow	no	yes	yes
Particle growth	yes	no	yes
Pool Temporal Analyses	no	no	yes
Bubble hydrodynamics			
Bubble shape	oblate-spheroid	all bubble shapes	6 mm dia. bubble
Bubble rise speed	"	Wallis 5-regime Corr.	Wallis 5-regime Corr.
Bubble break up	no	no	yes/final bubble size 6 mm dia.
Condensation of steam during bubble rise	no	yes	yes
Evaporation of steam into bubble	no	yes	yes
Non-condensable gas flux	no	no	yes

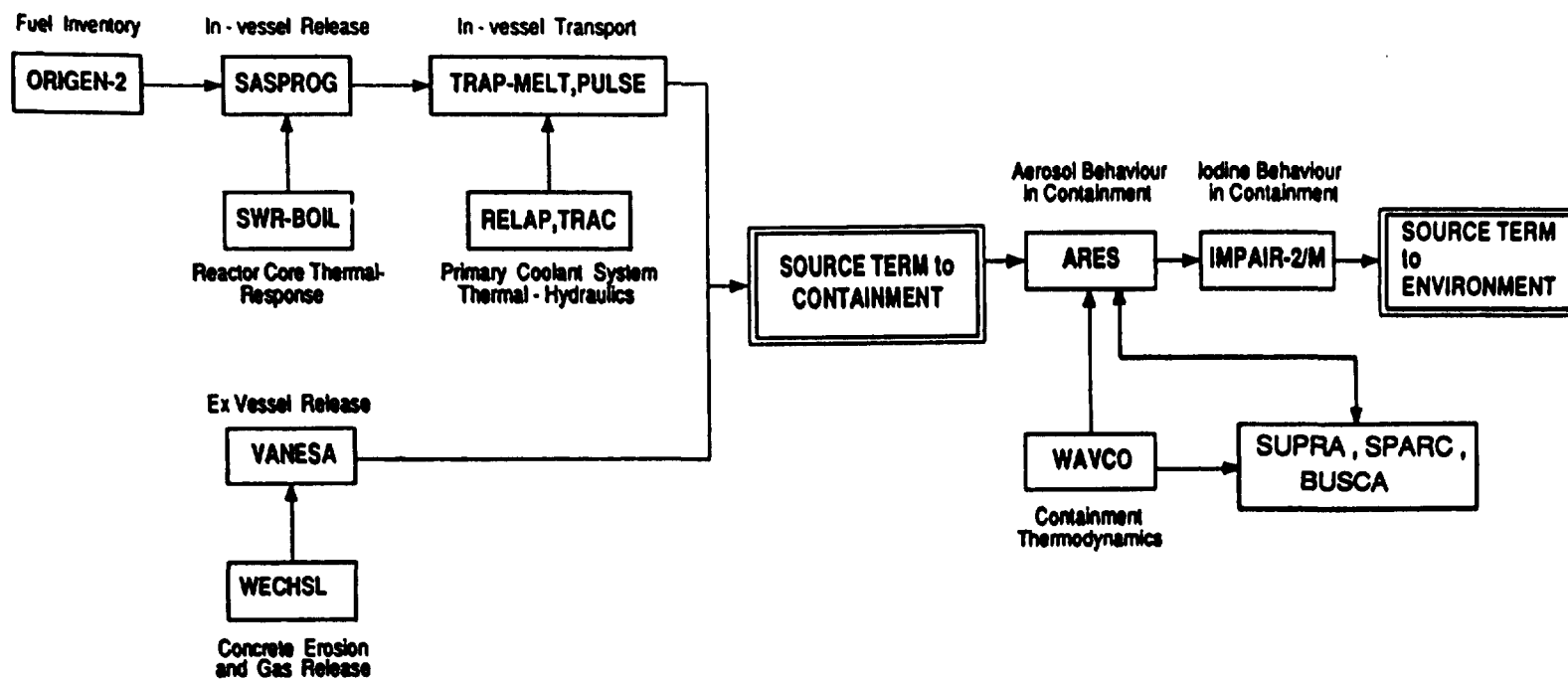


Figure 1 Analytical Tools used in the Reassessment of Source Term at PSI

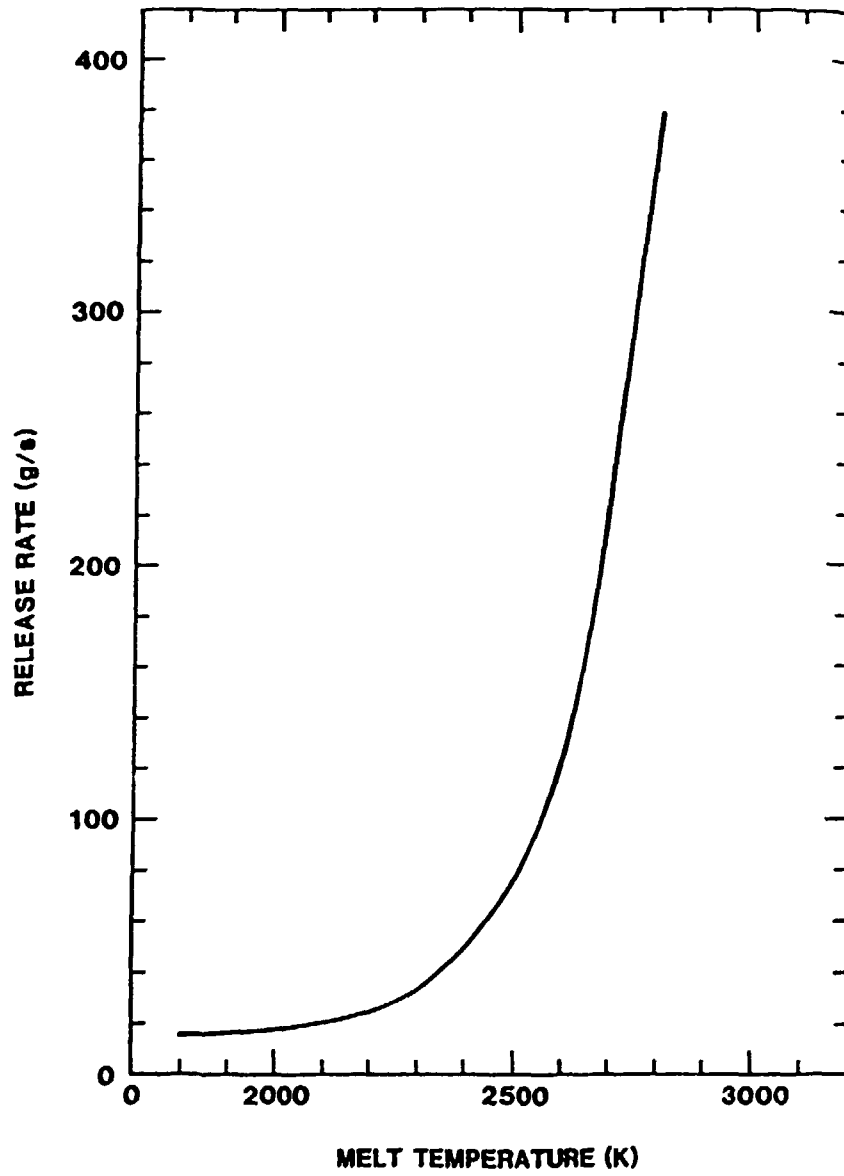


Figure 2 Effect of melt temperature on the aerosol generation rate

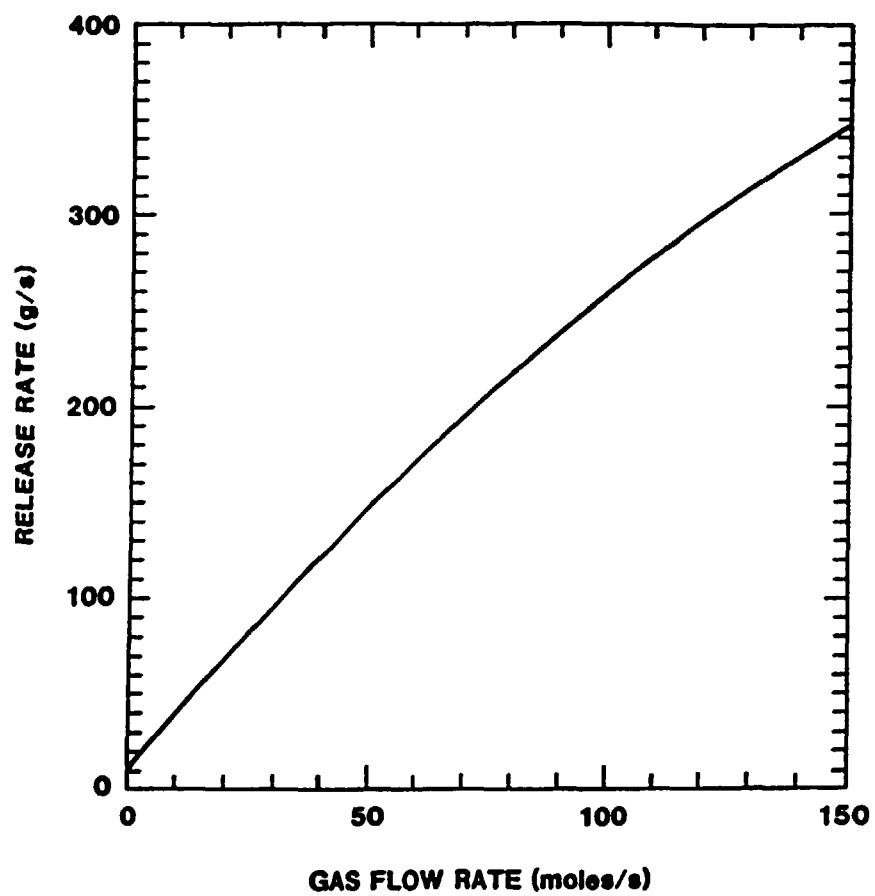


Figure 3 Effect of gas flow rate on the aerosol generation rate

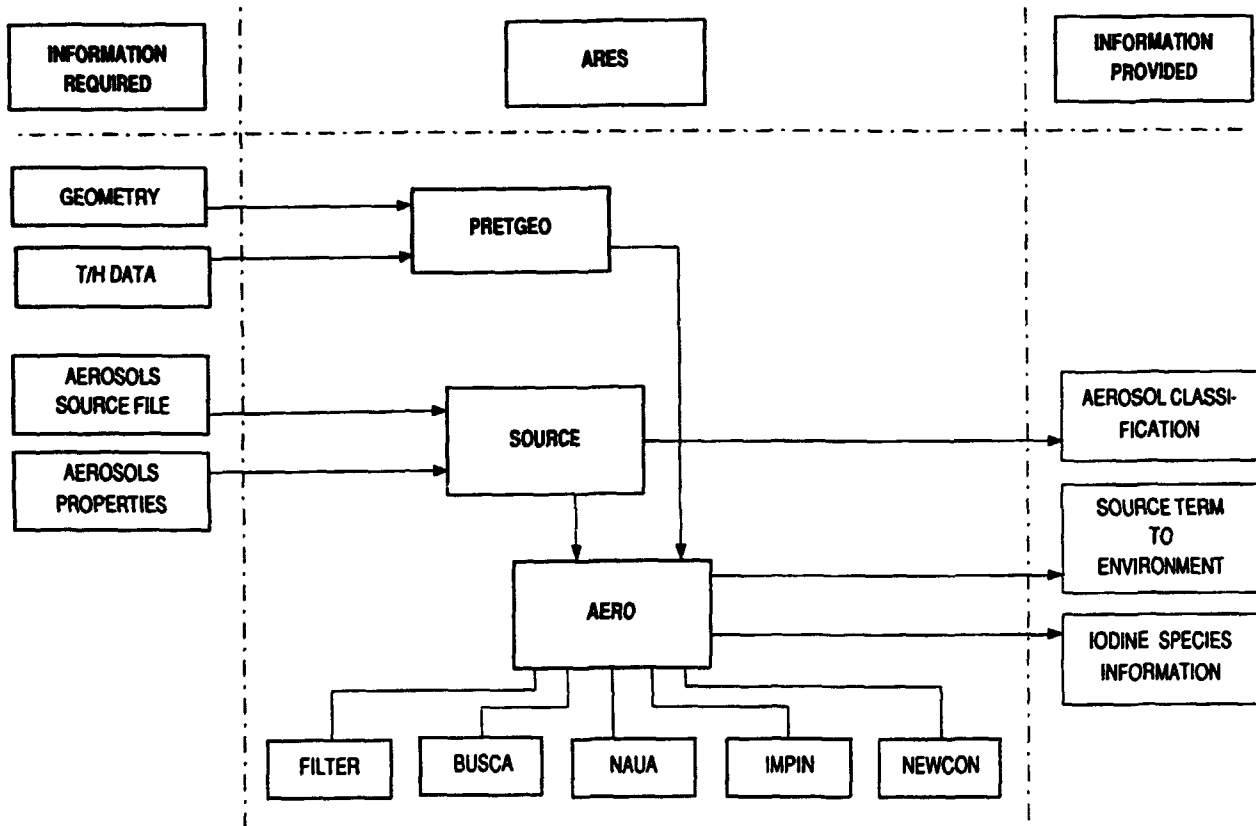


Figure 4 Aerosol Retention and Environmental Safety Code, ARES