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APPLICATION OF MELCOR FOR SIMULATING MOLTEN SALT REACTOR ACCIDENTS

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ABSTRACT

Molten salt reactor systems can be divided into two basic categories: liquid-fueled Molten Salt Reactors (MSRs) in which the fuel is dissolved in the salt, and solid-fueled systems such as the Fluoride-salt-cooled High-temperature Reactor (FHR). The molten salt provides a barrier to fission product release as actinides and many fission products are soluble in molten salt. Nonetheless, under accident conditions, some radionuclides may escape the salt by vaporization and aerosol formation. We present recent enhancements to MELCOR to represent the transport of radionuclides in the salt and releases from the salt. Some soluble but volatile radionuclides may vaporize and subsequently condense to aerosol. Insoluble fission products can deposit on structures. Thermochimica, a Gibbs Energy Minimization (GEM) code, has been integrated into MELCOR. With the appropriate thermochemical database, Thermochimica provides the solubility and vapor pressure of species as a function of temperature and composition, which are needed to characterize the state of the salt with fission products. Since thermochemical databases are still under active development for molten salt systems, thermodynamic data for fission product solubility and vapor pressure may be user-specified. This enables preliminary assessments of fission product transport in molten salt systems. In this paper, we discuss modeling of fission product releases in non-LWR systems with Thermochimica incorporated into MELCOR.

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KEYWORDS
Molten salt, fission product release, non-LWR MELCOR, MSR, FHR

1. INTRODUCTION

Molten salts have been proposed as a coolant because they have a broad temperature range over which they are liquid, have a high thermal conductivity, and are chemically inert [1]. For liquid-fueled MSRs, fuel and some fission products are soluble in the salts [2, 3]. Thus, a chemical processing unit outside of the reactor could be used to remove the fission products, and then return the salt for further heat production without requiring reactor shutdown. However, insoluble fission products could deposit on structural surfaces such as heat exchangers, which would reduce heat transfer and concentrate radionuclides on such surfaces. Removing deposited fission products may require the reactor to shut down, so soluble fission products are preferred. For solid-fueled MSRs, the fuel and fission products are isolated from the coolant, e.g., TRISO particles within pebble fuel elements. Thus, for these reactors, the radionuclides are isolated from the salt during normal operations. However, a severe accident may result in the rupture of TRISO particles, which could release radionuclides into the salt.

For both solid-fueled and liquid-fueled MSRs, a very attractive feature of molten salts is that the salt provides an inherent barrier to radiological releases in the event of an accident. The Generalized Radionuclide Transport and Retention (GRTR) model added to MELCOR applies to molten salt coolants in which radionuclides are released into or are already present in the salt, respectively.

The fission products have been classified into three forms: salt-seeking elements, noble-metal elements, and noble gases [1, 4]. Salt-seeking elements are soluble in the salt, whereas noble elements are not. A color-coding of some elements in the periodic table has been used to graphically display the solubility state of fission products in molten salt [5]. In this format, a fourth form was added of “sometimes soluble” elements [5]. Other workers have made a similar characterization of the fission products for the MSRE (Molten Salt Reactor Experiment) at ORNL (Oak Ridge National Laboratory) [3, 6]. More recently, a color-coding of some elements in the periodic table was presented by using three forms to indicate solubility [7].

1.1. Conceptual Framework of MELCOR Molten Salt Reactor Severe Accident Model

The GRTR model builds on the ideas of classifying radionuclides into different forms, but quantitatively tracks the masses of all species in a salt pool in five forms depending on both the solubility state and location of species in an MSR. This refined level of detail is necessary to capture the states and locations of radionuclides as observed in the MSRE [6]. However, the model does not require that a species be entirely soluble or insoluble, or in a single form. Therefore, a species can exist in more than one form and at different concentrations in each form. The amount of a species in each form is calculated by the GRTR model as a function of time, temperature, and salt composition.

A schematic of the GRTR model is given in Fig. 1. Shown in the top row of Fig. 1 are the five forms of the radionuclides. The inputs to the GRTR model include the control volume temperature, the control volume pressure, and the masses of all species in the five forms at the beginning of a timestep. Form 1 in the top-left corner of the first row is for soluble or salt-seeking radionuclides. If a radionuclide has reached its solubility limit, the insoluble mass in the salt may remain suspended in the salt, migrate to the top of the salt pool, or deposit on to fixed structural surfaces in contact with the salt. These three forms are shown in Fig. 1 as Forms 2, 3, and 4, respectively. The Form 5 is for gases such as Kr, Xe, and tritium. For the next step, as given in the second row, the GRTR model calls Thermochimica to determine the equilibrium masses of soluble and insoluble species. In addition, the vapor pressure of volatile species is determined by Thermochimica. The GRTR model then relocates the masses in Forms 1, 2, and 5 according to the equilibrium results. Only the forms pointed to by an arrowhead indicate mass transfer, with the form covered by the dashed part not to be included.

The next GRTR model calculational step, which is shown in the third and fourth row in Fig. 1, is to determine the transport among the five forms, and releases to the atmosphere, respectively. In this step, there are three transport processes within the forms. Insoluble fission products may transport between the interface layer and the pool. The second process is transport from suspended insoluble fission products to deposition on heat structures in contact with the salt. The third process is transport of insoluble material between the pool and deposition on surfaces in the core. Aerosols may be formed from the condensation or nucleation of volatile species that are released to the gas space. However, since this process occurs in the atmosphere above the pool, it is therefore modeled with MELCOR’s aerosol model. The state of the pool, deposited mass on surfaces, and the released masses are passed back to MELCOR for transport within a facility. The equations describing the transport between forms and to the atmosphere above the pool are given later in this work.

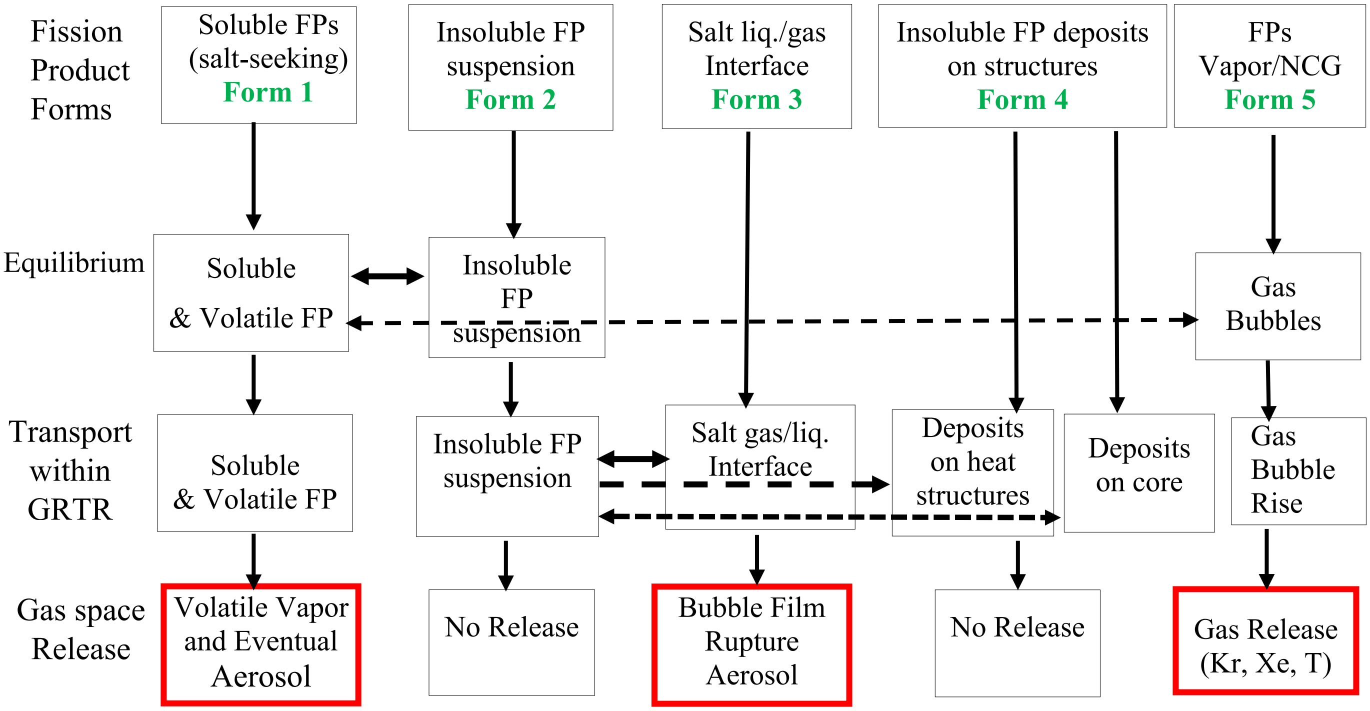

Figure 1. Schematic of the analysis steps of GRTR (Generalized Radionuclide Transport and Retention) model in MELCOR. The calculations performed are equilibrium, and then simultaneous transport within the pool and to the gas space.

1.2. GRTR (Generalized Radionuclide Transport and Retention) Model Data Requirements

To model the transport and release of radionuclides in the five forms, the three categories of properties needed are: (1) thermophysical, (2) thermochemical, and (3) mass transfer properties of the salt and the fission products. Thermophysical properties such as density, heat capacity, thermal conductivity, melting point, boiling point, viscosity and surface tension have been compiled for pure salts [8, 9, 10, 11, 12]. Thermochemical properties such as fission product solubility and vapor pressure as a function of temperature and salt composition have not been as extensively reported but some data are available. Thermochimica [13, 14, 15] is now included in MELCOR to determine solubility and vapor pressure. No transport property data has been found in the literature for the third category, such as diffusivity of the fuel and fission products in the salt and in the gas phase at high temperatures. Properties in this category are estimated using correlations. Accident analyses require these three property categories for pure salts with fission products and also for the effects on these properties from contaminants, such as water vapor, oxygen, abraded graphite particles, and corrosion products. Since severe accidents can extend to high temperatures significantly above normal operating temperatures, a wide temperature range is also needed.

Obtaining all the property data given above for an accurate radionuclide release model due to a severe accident is a daunting task. However, a system level code such as MELCOR is a viable tool for assessing sensitivities, uncertainties, and ranking the importance of different properties on releases. Thus, an important application of the GRTR model in MELCOR is for determining parameter sensitivities to help prioritize research efforts.

1.3. Paper Organization

Section 2 discusses the results of the thermochemical property literature survey. Much has been learned from the work at Oak Ridge National Laboratory (ORNL) with the Molten Salt Reactor Experiment (MSRE). Some general statements in the literature on fission product solubility have been reported. Data on the solubility limits for He, Kr, Xe, PuF_3, AmF_3, CeF_3 are available. For quantitative analysis that can be applied to a wide range of salt compositions and temperatures, species solubility limits and vapor pressures may be determined by the Thermochimica code [13, 14, 15] at the users' discretion if the appropriate database is available. By using equilibrium thermodynamics, we implicitly assume that the timescale for reactions to reach equilibrium is much shorter than the timescale for the accident events. Furthermore, the transport of reactants and products is assumed not to be limiting within the control volume. A similar approach using MELCOR but with a different GEM code external to MELCOR was reported recently for determining vaporization of fission products Cs and I from a fluoride salt [16].

Section 3 presents a discussion of research related to aerosol formation due to bubble bursting. As bubbles from gases that reach the pool surface, the busting of the liquid film at the pool gas interface provides a mechanism for aerosols from soluble species to be released directly into the gas space.

Section 4 discusses the integration of the GRTR model into MELCOR. As mentioned previously, there are five possible forms where radionuclides can exist in a molten salt pool, and MELCOR tracks the radionuclide masses in all these forms. For each control volume in the MELCOR model, the GRTR model allows for transport among these five forms. Radionuclides in Forms 1, 3, and 5 can lead to vapor or aerosol releases to the environment. In addition, radionuclides may deposit on surfaces in contact with the salt, and this may complicate operations. Once a radionuclide is transported out of the molten salt either by vaporization, aerosolization, or deposition on to surfaces, the existing MELCOR models are used to track these species within the reactor and containment. Finally, Section 5 presents a summary of the current capabilities and suggests some needed experiments. Example calculations are not included in this report but will be presented at the meeting.

2. RADIONUCLIDE SOLUBILITY AND VAPOR PRESSURE DATA IN MOLTEN SALT

2.1. Radionuclide Solubility

From a safety perspective, soluble fission products are not easily released as either vapor or aerosol. To maximize fission product solubility, an oxidizing solution is desirable in which the fission product atoms lose electrons by forming fission product fluorides or chlorides. However, there are structural metals such as steel and Hastelloy used for piping and vessels for which oxidation (which would be corrosion to form a fluoride or chloride), is very undesirable. For these metals, a reducing salt solution is desirable. Thus, the ideal molten salt is oxidizing for fission products but reducing for structural metals. This balance was achieved in the MSRE (Molten Salt Reactor Experiment) at ORNL (Oak Ridge National Laboratory) by controlling the UF_4/UF_3 molar ratio at ~100. This ratio increased as uranium fissions and thereby released fluorine into the salt, which formed UF_4. Beryllium was introduced and formed UF_3. Essentially, the beryllium may be viewed as a sacrificial metal to maintain a desired buffering effect. For the MSRE, if the salt is too reducing, then there was concern that uranium carbide would form.

Solubility is dependent on the redox potential of the salt and temperature [2, 4, 17, 18]. Solubility data of Pu, Ce, and Am in fluoride salts has been reported [19, 20, 21, 22, 23, 24]. An example of PuF_3 solubility as a function of temperature and composition is given in Fig. 2 as dots for the data from Barton et al. [19], and as a solid curve predicted with Thermochimica using the MSTDB [28]. The log of the solubility varies essentially linearly with the inverse absolute temperature. As can be seen from Fig. 2, the agreement between the Thermochimica predictions and the data is good. PuF_3 is soluble in FLiBe and PuF_3 solubility increases with increasing temperature. However, there is a solubility limit for a given temperature and salt composition. This limit is first checked in the GRTR model to determine whether new insoluble material is added as suspended particles or existing suspended material is dissolved.

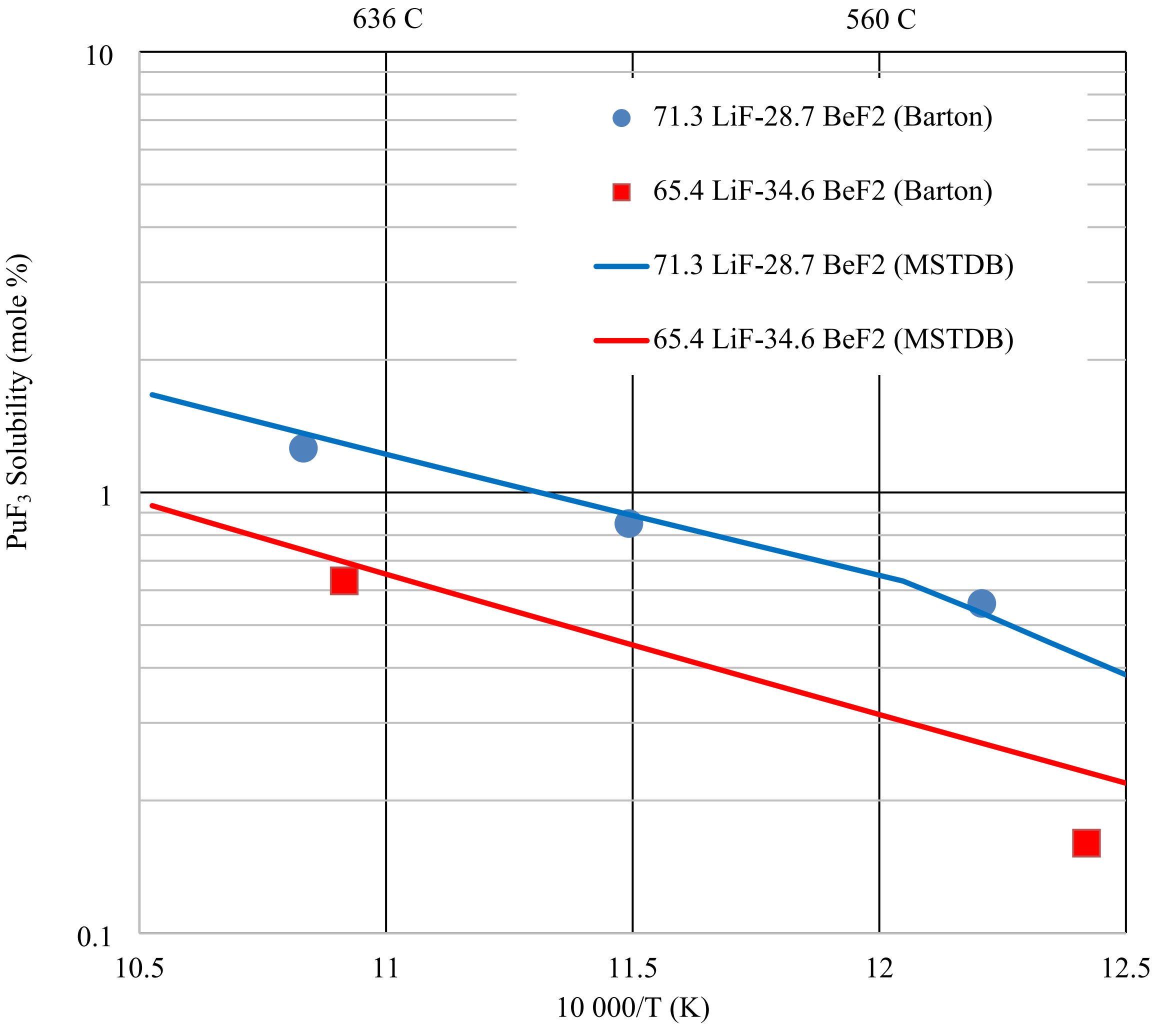

Figure 2. Solubility of PuF_3 in FLiBe at the mole percentage given in the legend for the data from Barton et al. [19] and calculated with Thermochimica using the MSTDB [28].

For solid-fueled reactors in which the fuel and fission products are isolated from the salt, fission product solubility may not be important during normal operations. There is still, however, radioactive tritium production in salts with lithium. A chemical processing step will be required to remove the tritium for long term storage as it decays or for ultimate disposal. For TRISO fuel at high temperatures, Ag-110m has been shown to migrate through intact TRISO particles [25] and should be insoluble in fluoride salts [4]. Other than these very small amounts of fission products, the salt chemistry should be maintained to minimize corrosion of structural metals for solid-fueled reactors.

There are also data for cesium and iodine fission products in molten salt [26, 27]. In the GRTR model, users may either specify a solubility limit or invoke Thermochimica to determine solubility limits with the ORNL MSTDB (Molten Salt Thermodynamic Data Base) [28]. Other databases may be utilized by MELCOR/GRTR in a separate file at the users' discretion. For soluble fission products that are volatile, the GRTR will pass the vaporized mass over a timestep to MELCOR. Upon encountering a cooler atmosphere, the volatile species may nucleate and form respirable aerosol particles.

2.2. Insoluble Fission Product Form

If the fission product is not soluble and is not a vapor, then such atoms may be isolated, or in the form of atomic clusters, or attached to existing particles in the molten salt to form a suspension. A suspension of small particles that are 1-1000 nm in diameter in a liquid is called a colloid [29]. The GRTR model tracks insoluble particles using Forms 2, 3, and 4, which are respectively, insoluble suspension, insoluble particles at the interface between the molten salt pool and the gas space, and deposited particles on fixed surfaces. The dynamics of these three forms shown in Figure 1 are discussed in Section 4 of this work.

3. RADIONUCLIDE RELEASE BY BURSTING BUBBLE FILMS

Whether the fission products are insoluble or soluble, the liquid salt retains these nonvolatile radionuclides, thus providing a barrier to release. However, a severe accident may involve considerable splashing and agitation of the salt. A liquid film or jet may be expelled by these processes with the film or jet breaking up to form droplets. These droplets are generally larger than 100 micrometers diameter and are not considered respirable. Respirable particles are most often defined as particles with an aerodynamic diameter of 10 micrometers or less [30].

Agitation may also entrain gas and thereby form bubbles in the molten salt. Bubble bursting releases two types of liquid droplets [31, 32]. When the depression in the liquid due to the bubble is filled in by the surrounding liquid, an upward liquid jet forms that disintegrates into droplets that are not respirable. However, the cap of the bubble is a thin liquid film that can be on the order of a micrometer thick or less [33, 34]. When the cap ruptures, respirable droplets may form. Consequently, soluble fission products and insoluble particles that are in the film can be aerosolized [35].

Evidence of molten salt aerosolization was reported the MSRE. Bubbles and mists were reported [36, 37]. Reference [37] states, "The origin of most of the mist in the MSRE pump bowl is undoubtedly the spattering and splashing of the streams from the spray ring." And later states, "the pump bowl difficulties were caused by an aerosol-type dispersion of salt particles." The basis for describing the particles as aerosol-type is unclear since no particle size distribution is given. Fig. 3 below (which is Fig. 4 in [37]), shows spheres of droplets collected from the gas space above the salt. Using the 1/2-inch width of the strip for scaling, the smallest particle is about 95 micrometers in diameter with most of the particles much larger. Such particle sizes are generally not considered to be aerosol particles. The particles in Fig. 3 may be due to the breakup of the liquid jet or crown from splashing. In addition, because the sample shown is time-integrated over 12 hours, possibly smaller droplets were deposited that coalesced over time into much larger droplets shown in Fig. 3. The report later states that "A filter about 15 ft downstream from the pump bowl trapped salt particles (all 10µ or smaller) at a rate of about a cubic inch per 100 hours of salt circulation." If the parenthetical remark, "all 10µ or smaller" as given in the report corresponds to 10-micrometers diameter or smaller, then this would be evidence of respirable aerosol release in tests for the MSRE.

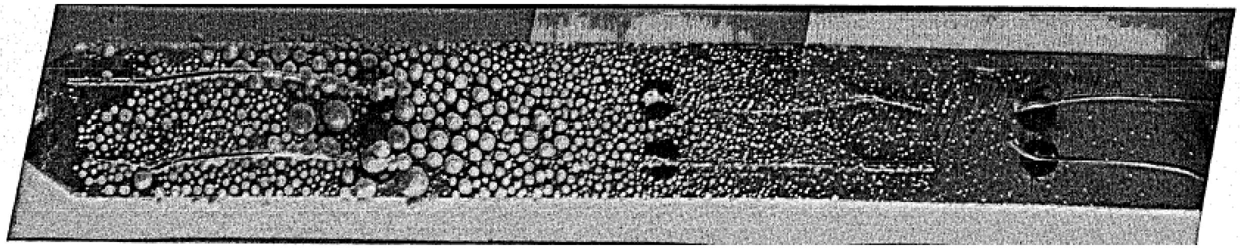

Figure 3. Photograph of droplets on a ½ inch wide metal strip exposed in the MSRE pump [37].

A more detailed description of respirable aerosol particle sizes produced as part of the MSRE is available. In reference [38] three sizes of particles were collected from the gas phase above a 50-gram salt sample. The smallest particles had diameters of 3.5-18 nm, the medium size had diameters of 100-200 nm, and there were particles that were described as more than ten times the medium diameter size. Two micrographs that provided the data for these small aerosol particles are shown below in Fig. 4. Such particles are respirable and believed to be of the salt.

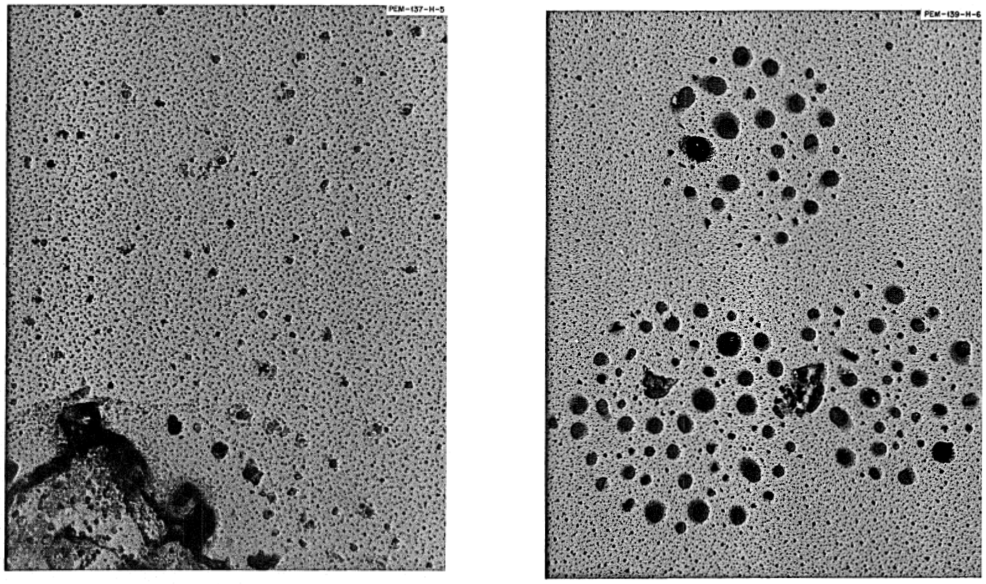

Fig. 10.4. Electron Micrograph of Particles in Gas Flowing over MSRE Salt. 88,000×.

Fig. 10.5. Circular Patterns Frequently Found on Screens in Gas Above MSRE Salt. 32,000×.

Figure 4. Electron microscope images of particles collected from the gaseous headspace above a 50-gram molten salt sample [38].

Aerosol instrumentation has improved considerably since the 1960s and a more recent study of the particles produced from the cap of a bursting bubble is available. In reference [34], there are impressive photographs of aerosol particle formation from a bursting bubble of an aqueous solution. Some of the data is given below in Fig. 5 for measurements made with a Condensation Particle Counter (CPC) and an Aerodynamic Particle Sizer (APS). The measured particle sizes are after the particles were dried and were therefore biased to smaller particle sizes. However, if 99.9% of the particle volume of an originally respirable 10-micrometer-diameter droplet was vaporized by drying, it would result in a 1-micrometer-diameter dry particle which is the upper detection limit of the CPC (Condensation Particle Counter). Therefore, the CPC data may be used to determine the number of respirable particles produced from a single bursting bubble since aqueous droplets that are 10-micrometers diameter and smaller are respirable.

From the left side of Fig. 5, the number of respirable particles from a single bubble is on the order of thousands and increases with increasing bubble diameter. From the right side of Fig. 5, the number of particles produced decreases with increasing surface tension. Water at ambient conditions has a surface tension of about 73 mN/m, for which there may not be respirable aerosol particles produced by a bursting bubble. A surfactant was added in the experiments to reduce the surface tension to 50 mN/m or less [34]. Pure molten salts, such as FLiBe and FLiNaK, have surface tensions over 150 mN/m at 800 °C, and the surface tension of pure KCl-$MgCl_2$ is over 70 mN/m at 800 °C [8]. For the fluoride salts, if the data on the right side of Fig. 5 are extrapolated to a surface tension of 150 mN/m, bubble rupture may not result in respirable particle release. Extrapolating may not be valid and does not support the assumption that the data in Fig. 4 were due to bubble cap rupture. Another consideration is that the salt may be contaminated with particles of noble metals, abraded graphite, and corrosion products. Colloidal suspensions act as a surfactant that can lower the surface tension [7, 39]. Other than for MSRE, we have not found data of the respirable aerosol produced from molten salt. Experiments are needed to resolve the discrepancy between the data in Fig. 4 and the extrapolation of the data on the right side of Fig. 5. Furthermore, as shown on the left side of Fig. 5, the bubble diameter is also important, and that needs to be determined as a function of the agitation level and contaminants in the salt.

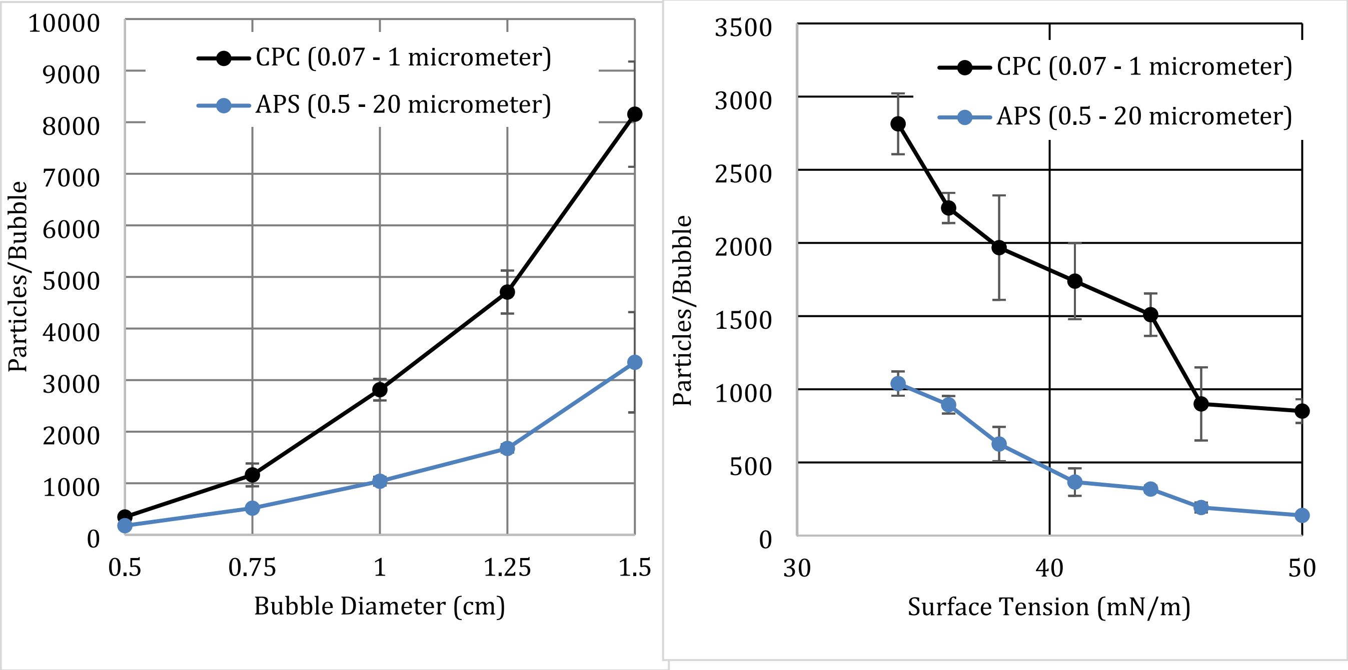

Figure 5. Aerosol particles generated by a bursting bubble of an aqueous solution [34].

4. MELCOR INCORPORATION OF THE GRTR MODEL

4.1 Model Assumptions

A molten salt control volume consists of a salt pool of uniform composition, temperature, and pressure, and may also include heat structures. The atmosphere above the salt, if present, includes non-condensible gases, aerosols, condensible vapors, and possibly heat structures. MELCOR includes models for heat and mass transfer, and aerosol processes for the atmosphere in a control volume. Condensible vapors, non-condensible gases, and aerosol particles released from or to the molten salt transfers mass between the atmosphere and the salt pool. Radionuclides that are deposited on structures in contact with the salt transfer heat to the salt and structure via radioactive decay. In addition, the GRTR model allows for a top layer of foaming salt with bubbles that contain insoluble fission product at a different concentration than in the bulk salt solution. Such a layer was observed in the MSRE [6, 40].

The mass transfer process of insoluble particles has been modeled for the MSRE [7, 40]. Reference [40] states, "It is generally agreed that most of the fission products from niobium through tellurium are reduced to metals in the fuel salt, that they migrate to metal and graphite surfaces and to salt-gas interfaces, and that they adhere to the surfaces with varying degrees of tenacity." Determining the tenacity of the fission products onto surfaces in molten salt is a recognized research area [41]. For now, these particles are modeled as adhering to the metallic structures until data are available to indicate the level of disruption (i.e., either mechanic or thermal) needed to remove these particles. Therefore, the dashed arrow in Fig. 1 only points one way from form 2 to deposits on heat structures. However, deposits may move between form 2 and core surfaces which is shown as a double-headed dashed arrow.

In summary, radionuclides can be released from the salt as noble gases and by vaporization of volatile fission products. The vaporization process can result in respirable aerosol particles once the volatile species encounters a colder atmosphere, which will condense these vapors. There is evidence from MSRE that aerosol particles may also be released by agitation that engulfs gas and thereby forms bubbles. The cap of bubbles at the surface of a liquid has been shown to produce respirable aerosol particles for aqueous solution with a surface tension less than 50 mN/m. The surface tension is much higher for pure salts being considered for an MSR, and therefore bubble bursting in molten salt may not produce respirable aerosol. Because respirable particles were observed at the MSRE, bubble bursting is therefore included in GRTR model until data are available to exclude this phenomenon.

4.2 Model Transport Equations

The transport to surfaces in contact with the salt and releases are calculated simultaneously but shown as two steps in Fig. 1 for clarity. Transport of mass and heat to and from surfaces makes the problem time dependent. Mass is also exchanged between the top surface of the pool and the atmosphere above the salt pool. Volatile species and aerosol particles from bubble bursting may be transported from the pool to the atmosphere above the pool. Aerosol deposition from the atmosphere to the pool provides a source term for the pool. If there are soluble species in the deposited aerosol, they will be dissolved in the next timestep when an equilibrium calculation is performed with Thermochimica.

The conservation equations expressed in term of the masses and concentrations of species i in form j = 1, 2, 3, 4, 5 is given by $M_{i,j}$ and $C_{i,j}$, respectively. For simplifying these expressions, the core surfaces are included in the equations below, but deposits in the core are tracked separately in the code. The equations for $M_{i,j}$ are given next with the following notation,

A_{st} = salt top surface area,
A_{hs} = heat structure surface area in contact with the salt,
$C_{k,vap}$ = vapor concentration of species k in the atmosphere above the salt,
$H_{p\to q}$ = mass transfer coefficient of form p to form q,
$H_{p\to aer}$ = mass transfer coefficient of form p to aerosol by bubble bursting,
$H_{p\to vap}$ = mass transfer coefficient of form p to vapor in the atmosphere,
V = permanent gas release rate, and
t = time.

The values of $H_{p\to q}$ will depend on the geometry and flow conditions and are user input control functions based on correlations discussed in the next section.

Soluble mass in Form 1 changes due to vaporization and bubble bursting and is given by,

$$\frac{d(M_{i,1})}{dt} = -A_{st}H_{1\rightarrow vap}(C_{k,1} - C_{k,vap}) - A_{st}H_{1\rightarrow aer}C_{i,1} \qquad (k = \text{volatile species only}). \tag{1}$$

The first term on the right is nonzero only for volatile species.

Insoluble mass in Form 2 changes due to deposition on structures and transport to the gas-liquid interface. This mass change is given by,

$$\frac{d(M_{i,2})}{dt} = -A_{hs}H_{2\rightarrow 4}(C_{i,2}) - A_{st}H_{2\rightarrow 3}(C_{i,2} - C_{i,3}) \qquad (i = \text{insoluble species only}). \tag{2}$$

We assume that the deposit on structures is tenacious and remains attached to the structure, so there is no concentration difference in the first term on the right side of Eq. (2). However, deposit in the core may be removable at the users' discretion.

Mass in the gas-liquid interface can increase by aerosol deposition from the atmosphere, decrease by bubble rupture, and increase by insoluble species transported to the interface. These three processes for only insoluble species i are given by,

$$\frac{d(M_{i,3})}{dt} = Aerosol\ Deposition - A_{st}H_{3\rightarrow aer}(C_{i,3}) + A_{st}H_{2\rightarrow 3}(C_{i,2} - C_{i,3}). \tag{3}$$

Insoluble mass adhering to structures in contact with the salt accumulates according to,

$$\frac{d(M_{i,4})}{dt} = A_{hs}H_{2\rightarrow 4}(C_{i,2}) \qquad (i = \text{insoluble species only}). \tag{4}$$

Any non-condensible gases, such as Xe and Kr in the molten salt are tracked separately as Form 5 in the GRTR model. A simple conservative model is to release all such gases above the solubility limit to the atmosphere, and neglect delays for the time the permanent gases need to reach the top of the pool. However, if the release rate is known or assumed, the rate of change of permanent gas mass in the pool is given by,

$$\frac{d(M_{i,5})}{dt} = Source\ - A_{st}V(C_{i,5}) \qquad (i = \text{permanent gas}). \tag{5}$$

If the solubility and time to release from the pool is neglected, then the mass of non-condensible gases in the pool is zero, and any non-condensible gas released into the pool at the beginning of a timestep is always released into the atmosphere at the end of the timestep. If there is no vapor space in the control volume, then the Form 5 gases remain in the molten salt.

4.3 Model Parameters

The molten salt model developed for MELCOR as given by Eqs. (1-5) is very general and includes known and possible phenomena. This is intentional so that the user may investigate the relative importance of phenomena and decide what is worth further analysis or experimentation. A model with all the phenomena discussed in this work will include parameters that are unknown. Therefore, the code was written to allow users to specify model parameters in the input via user-defined control functions. This also enables the user to turn on and off selected phenomena in the model.

If an adequate database is not available for Thermochimica, then the GRTR model in MELCOR is designed to accept user-specified control functions for the solubilities and vapor pressures. One may choose to use the color-coded periodic table [5] as a guide for determining fission product solubility. For vapor pressures of soluble fission products, the data for a specific salt is a useful guide [26].

A potential process for direct aerosol release is for bubble cap bursting that may release particles with a composition the same as the salt pool and particles at the interface layer. The settling of aerosol particles into the pool is a source term for the third form and MELCOR provides the rate, composition, and sizes of aerosol particle settling.

All the mass transfer coefficients ($H_{p\to q}, H_{p\to aer}, H_{p\to vap}$) and areas ($A_{st}, A_{hs}$) can be provided as user input with user-defined control functions. For a well-mixed pool of molten salt, the mass transfer of a species to a surface can be modeled by analogy with heat transfer [42]. The correlations for heat transfer are applied for mass transfer with the Schmidt number replacing the Prandtl number, and the Sherwood number replacing the Nusselt number. This approach was used previously with MELCOR [16]. For the Sherwood number, the diffusivity of a species is needed and as shown in [16], the diffusivity can be estimated for fission products in the gas phase. The diffusivity of a colloidal particle is given by [29]

$$\mathcal{D} = \frac{kT}{3\pi D\eta} \tag{6}$$

where k is Boltzmann's constant, T is the absolute temperature, D is the particle diameter, and η is the solution viscosity.

The observed respirable aerosol particles at the MSRE may be due to bubble bursting, but the mass transfer coefficient $H_{p\to aer}$, is difficult to determine. Work is needed to determine the number and sizes of bubbles formed for a given level of agitation in molten salt.

5. SUMMARY AND CONCLUSIONS

The GRTR model has been incorporated into MELCOR. The GRTR model tracks the radionuclides released into the salt at the beginning of a timestep, and calculates the masses of each radionuclide; (1) uniformly dissolved in the salt, (2) insoluble particles uniformly suspended in the salt, (3) residing as insoluble particles at the gas-liquid interface, (4) particles adhering to structures in contact with the salt, and (5) gases released to the atmosphere above the pool.

To achieve the tracking of radionuclides in the five forms, the GRTR model first establishes the equilibrium masses dissolved in the salt and as insoluble species in the salt. This is accomplished using the GEM code Thermochimica, which is now part of MELCOR. In addition, Thermochimica provides the vapor pressure of volatile species needed for determining the release rate into the atmosphere above the pool. Transport among the five forms is next computed in the GRTR model before returning the end of timestep masses in these forms to MELCOR. Existing MELCOR models can then be used to determine releases out of a facility.

We recognize that there are parameters in the model that are not known. Therefore, the model is designed for sensitivity and importance exploration such that the unknown parameters can be varied using user-specified control functions. Some phenomena are poorly understood for molten salts, such as bubble bursting to form aerosol particles. Bounding experiments of a highly agitated molten salt, or a large salt release from a significant height are needed in which engulfed gas forms bubbles. The rate of bubble formation and bubble size for these conditions are needed. Furthermore, aerosol measurements of molten salt bursting bubbles are needed, similar to the measurements made for aqueous solutions [32, 34].

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