Coupled Thermal-Hydraulic Analysis and Species Mass Transport in a Versatile Experimental Salt Irradiation Loop (VESIL) using CTF

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ABSTRACT

With the resurgence of interest in Molten Salt Reactor (MSR) technology among commercial and governmental agencies, there is a need for new experiments and new modelling capabilities to characterize the unique phenomena present in this fluid fuel system. A Versatile Experimental Salt Irradiation Loop (VESIL) is currently under investigation at Idaho National Laboratory (INL). It is designed to be a dynamic fuel-bearing salt loop placed in the Advanced Test Reactor (ATR). One of the key phenomena this proposed experiment plans to elucidate is fission product speciation in the fuel-salt and the subsequent effects on fuel-salt properties, fission product transport and interactions, source term generation, corrosion and redox potential control. Specifically, noble gases (Xe & Kr) will bubble out to a plenum or off-gas system and noble metals (Mo, Tc, Te etc.) will precipitate and deposit in specific zones in the loop. This work extends the mass transfer and species interaction models in the general species transport capability of CTF and applies these models to give a preliminary estimation of fission product behavior in the proposed VESIL design. A noble metal (NM) – helium bubble sparging mass transfer model is coupled with the thermal-hydraulic results from CTF to determine the effectiveness of this insoluble fission product (IFP) extraction method for VESIL. Amounts of IFP species extracted to the off-gas system and species distributions in VESIL after a 60-day ATR cycle are reported. Additionally, redox control methods are suggested and their subsequent effect on IFP transport are discussed.

KEYWORDS: SPECIES TRANSPORT, MOLTEN SALT, NOBLE METAL HELIUM SPARGING

1. INTRODUCTION

The Versatile Experimental Salt Irradiation Loop (VESIL) shown below in Figure 1, is a proposed fuel-bearing integrated effects experiment currently under investigation at Idaho National Laboratory (INL) [1]. This natural circulation molten salt loop would contain a fuel-bearing salt and be irradiated in one of the I-positions of the Advanced Test Reactor (ATR). As the fuel-salt is irradiated, the process of insoluble
fission product (IFP) species migration, accumulation, and deposition will occur during the 60-day ATR cycle. Specifically, NM particle deposition and noble gas (NG) bubble formation may become significant and can be used as an online experimental indicator of the burnup process.

This work applies the species mass transport capabilities currently under development within the subchannel thermal-hydraulics code CTF [2], and acts as a feasibility study of the salt-loop in light of these complex transport phenomena. A previously developed NM deposition model [3] and helium sparging model [4] are coupled so the combined interaction of NM deposition and extraction via circulating helium bubble swarms shown in Figure 2 below can be modeled in determining what types of species will be extracted into an off-gas system.

![Figure 1. Preliminary VESIL Design [1].](image1)

![Figure 2. Noble Metal Flotation via Bubbles.](image2)

### 1.1. Importance of IFP Transport and Resulting Effects

Most liquid fueled MSR concepts are based off of the Molten Salt Reactor Experiment (MSRE) where the uranium is dissolved in the salt and is pumped through a loop or integral design. This key feature means that fission products will directly populate in the liquid fuel-salt during burnup and are free to migrate and mix in the reactor, rather than be held mostly stationary in a solid fuel-pellet. A majority of the fission products are soluble in the fuel-salt and form solid particulates and gaseous bubbles [5]. These insoluble species will have sink and source effects due to concentration gradient driven diffusion across material and phase boundaries that exist in the reactor. Therefore, the time and spatial dependence of these insoluble species must be tracked and predicted in determining mass accountability, homogenized cross sections in the reactor core, corrosion rates, and source term generation. Additionally, the cumulative decay heat from these IFPs could be significant and localized enough to cause structural damage in the reactor without active cooling [3].

### 2. VESIL DESIGN & PARAMETERS

The current VESIL design was chosen after several Thermal-Hydraulic analyses conducted at INL using STAR-CCM+ and SAM [7]. The selected design is a natural circulation driven loop shown below in Figure 3, where half of the loop (L_core) is exposed to the neutron flux, and the other half (L_upper) is outside of the core experiencing no flux. Shown in Figure 4 is the CTF VESIL geometry. The salt heats up and rises through the inner core in cells 1 – 4 and cools down flowing through the outer annulus in cells 5 – 8.
When using the helium sparging model to extract IFPs, the helium bubbles are introduced at the bottom of the loop in cell 1 and flow concurrently with the fuel-salt until they reach the liquid-gas interface in cell 5 where they are extracted to the off-gas system. The key parameters of the loop calculated previously by SAM/STAR-CCM+ are summarized in Table I below [7]. The CTF model is then forced to agree with the SAM/STAR-CCM+ results to ensure the correct natural circulation velocity and temperature.

The helium sparging parameters are listed in Table II where the amount of helium bubbles extracted at the upper plenum to the off-gas system is characterized by an extraction efficiency [4]. The proposed fuel-salt is UCl₃-NaCl₃ based off of the REBUS-3700 concept and the thermal-physical properties of the salt for the temperature range listed in Table I are summarized below in Table III where only the density is temperature dependent [8].

### Table I. VESIL Parameters [7]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value (units)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum Temperature</td>
<td>1011.87 (K)</td>
</tr>
<tr>
<td>Minimum Temperature</td>
<td>910.35 (K)</td>
</tr>
<tr>
<td>ΔT</td>
<td>101.52 (K)</td>
</tr>
<tr>
<td>Fission Power Density in Core</td>
<td>100.00 (W/cm³)</td>
</tr>
<tr>
<td>Salt Velocity</td>
<td>0.226 (m/s)</td>
</tr>
<tr>
<td>Total Height</td>
<td>78.00 (cm)</td>
</tr>
<tr>
<td>Height of Core</td>
<td>39.00 (cm)</td>
</tr>
<tr>
<td>Outer Salt Radius</td>
<td>2.778 (cm)</td>
</tr>
<tr>
<td>Inner Salt Radius</td>
<td>1.964 (cm)</td>
</tr>
<tr>
<td>Salt Mass</td>
<td>6.513 (kg)</td>
</tr>
</tbody>
</table>

### Table II. Helium Sparging Parameters [4]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value (units)</th>
</tr>
</thead>
<tbody>
<tr>
<td>He injection rate</td>
<td>2.0E-6 (moles/s)</td>
</tr>
<tr>
<td>Initial Bubble Diameter - D_{ref}</td>
<td>0.3175 (mm)</td>
</tr>
<tr>
<td>Removal Efficiency</td>
<td>80.0 (%)</td>
</tr>
</tbody>
</table>

### Table III. UCl₃-NaCl₃ Fuel-Salt Properties [8]

<table>
<thead>
<tr>
<th>Property</th>
<th>Value (units)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density (ρ)</td>
<td>3860.4 – 0.837×T (K) (kg/m³)</td>
</tr>
<tr>
<td>Dynamic Viscosity (µ)</td>
<td>0.00217 (kg/m-s)</td>
</tr>
<tr>
<td>Thermal Conductivity (k_T)</td>
<td>0.7 (W/m-K)</td>
</tr>
<tr>
<td>Specific Heat Capacity (C_p)</td>
<td>950 (J/kg-K)</td>
</tr>
<tr>
<td>Prandtl Number (Pr)</td>
<td>2.9450</td>
</tr>
</tbody>
</table>
3. SPECIES MASS TRANSFER MODELS IMPLEMENTED INTO CTF

3.1. Noble Metal Deposition Model Review & Assumptions

The NM deposition model is a thin film model where a mass transfer coefficient $K$ captures the combined effect of advection and diffusion, and the transfer is driven by a concentration gradient across the film [3]. Starting from the left-hand side in Eq. (1), the terms include source term change in mass density with respect to time, the mass transfer coefficient $K$ multiplied by the surface area available for deposition divided by the control volume multiplied by the difference in the mass density in the bulk control volume and the equilibrium mass density that exists in the liquid at the solid-liquid interface or deposition surface. As can be seen the surface area to volume ratio is an important geometrical factor in NM deposition.

$$S_{\text{deposition}} = \frac{K A_{\text{surf}}}{V_{\text{cell}}} \left( \rho_{\text{bulk}} - \rho_{\text{interface}} \right)$$

The first assumption made is the value of the $\rho_{\text{interface}}$ term, the equilibrium mass density of the species that exists in the liquid. Since NMs acted hydrophobically in FLiBe salt in the MSRE, all NMs in the fuel-salt can be assumed to be completely insoluble, and therefore the equilibrium mass density of the species that exists in the liquid is zero $-\rho_{\text{interface}} = 0$ [9]. This simplification is strongly dependent on the assumption that the redox of the fuel-salt (the fluorine/chlorine potential) is well controlled.

Lastly, the diffusion coefficient of these species in the salt is not entirely known, however an estimate value taken from the MSRE is applied to all of the NM species in the model [9]. This value was derived for species in FLiBe salt, and it is likely that diffusion coefficients for species in chloride salts will be different. However, since no measurements have been done in a chloride system, the FLiBe values are a practical first estimate to use in determining the mass transfer coefficients for VESIL. The diffusion coefficient $D$ is then used to determine the mass transfer coefficient $K$ from the mass transfer Dittus-Boelter correlation for turbulent flows shown in Eq. (2) below [9].

$$K = 0.023 \frac{D}{d} \left( \frac{\rho_{\text{avg}}}{\mu} \right)^{0.8} \left( \frac{\mu}{\rho \sigma} \right)^{0.4}$$

3.2. Helium Sparging Model Review & Assumptions

The helium gas sparging model is a two-phase flow model where the number of helium bubbles is determined by the gas inlet flow rate, the ideal gas law, and a set initial bubble diameter. The bubbles are then transported in CTF as a species around the loop by the fuel-salt velocity field [4]. It is important here to determine the interfacial area made by the helium bubble swarm so the correct surface area for species mass transfer to the bubbles can be calculated.

The ideal gas law is used to determine the changing diameter of the bubbles due to changes in pressure, mass, volume, and temperature traveling through the loop. Here the bubbles are assumed to be in thermal equilibrium with the fuel-salt, small and spherically shaped, and that the void fraction is sufficiently small to avoid bubble-bubble interactions.

$$P_b = P_f + \frac{4\sigma_l}{d}$$

The Young-Laplace equation is used to calculate the pressure inside the gas bubble due to the pressure outside the bubble and surface tension shown in Eq. (3) above, assuming the bubble is roughly static in the fuel-salt [4]. Here, $P_b$ is the pressure inside the bubble, $P_f$ is the pressure of the surrounding liquid, $\sigma_l$...
is the surface tension of the bubble, and \( d \) is the bubble diameter. Inserting Eq. (3) back into the ideal gas law, and assuming spherical bubbles, results in Eq. (4). This is then solved via Newtons Method to determine the average bubble diameter needed to calculate the helium bubble interfacial area available for mass transfer at each cell in the loop [4].

\[
[P_l + \frac{4\sigma_l}{d}] \frac{\pi d^3}{6} = nRT_l
\]

The coupling of the species mass transport/mass transfer model with the thermal-hydraulic results taken from CTF is clearly seen in Eq. (4) where the bubble interfacial area available for mass transfer directly depends upon the temperature and pressure calculated by CTF in each cell. Here \( n \) represents the number of moles in each bubble, which is the summation of all gas species in the bubble, including any NG fission products that have migrated into the bubble. The number of moles in each bubble is calculated by dividing the total moles of gas in the cell by the number of bubbles calculated from the CTF bubble species transport solve at the previous time step [4].

Lastly, the extraction boundary condition for the helium bubbles carrying any deposited species to the off-gas system needs to be determined. For simplicity, a bubble removal efficiency at the removal location in cell 5 of Figure 4 is defined. Here a percentage of the bubble mass flow rate is extracted as seen previously in Table II. Future work will incorporate buoyancy and diffusion forces to accurately determine the mass transfer of bubbles across the liquid gas interface in cell 5.

3.3. Noble Metal Helium Bubble Coupling & Off Gas Mass Accounting Methodology

It is important to note that many off-gas isotope species (which are primarily xenon) have NM precursors as seen in the decay chain \( \text{Sn} - \text{Sb} - \text{Te} - \text{I} - \text{Xe} \) where the color red indicates insoluble solid species, the color orange indicates a semi-soluble species, the color green indicates a soluble liquid species, and the color blue indicates an insoluble gas species.

Therefore, as xenon isotopes are extracted via mass transfer into the helium bubbles, there are additional NM particulates that will also attach to the helium bubble and are hypothesized to be extracted into the off-gas system or at least the liquid gas interface at the top of the loop via flotation as seen in Figure 2 [9]. This NM deposition on the circulating helium bubbles acts to increase the overall off-gas extraction.

Each isotope species is fragmented into several different “sub” species in order to track the concentration of a given species in different locations (eg. \( ^{132}\text{Sn}(l) \) – species in the bulk liquid, \( ^{132}\text{Sn}(s) \) – species on the VESIL wall, \( ^{132}\text{Sn}(g) \) – species attached to helium bubbles, \( ^{132}\text{Sn}(\text{Og}) \) - species in the off-gas system) [3]. All species are born from fission in the bulk liquid location denoted by \( (l) \), and all subsequent species grow from the loss of the bulk liquid species through mass transfer mechanisms discussed earlier. Only growth from fission, radioactive decay, and mass transfer mechanisms are currently incorporated, but neutron absorption and other effects will be incorporated in the future.

4. VESIL SPECIES TRANSPORT RESULTS & ANALYSIS

The following five decay chains \( ^{128-132}\text{Sn} \rightarrow ^{128-132}\text{Sb} \rightarrow ^{128-132}\text{Te} \rightarrow ^{128-132}\text{I} \rightarrow ^{128-132}\text{Xe} \) are explicitly tracked in order to determine the total stable xenon concentration distribution that accumulates in VESIL during the 60-day ATR cycle irradiation. The helium gas sparging model is active with the parameters shown previously in Table II. After 60 days of irradiation, the steady state amounts of Sn and Sb in VESIL are insignificant due to their short half-lives ranging from seconds to minutes. However, Te, and I species are significant and approach their steady state distributions in VESIL with half-lives in the order days.
Selective results are shown to help elucidate the overall species transport behavior in VESIL. The helium sparging model is shown in Figures 5 and 6 where the bubble inlet is in cell 1 at the center lower plenum, and bubble extraction is at cell 5 at the top of the upper plenum where the species are extracted across the liquid gas interface to the off-gas system. The bubble interfacial area increases slightly in cells 2, 3, and 4 in Figure 6 as the bubbles rise upwards due to expanding bubble diameters from decreasing pressure and increasing temperature at the top of VESIL.

The effect of helium bubble sparging on NM deposition is shown in Figures 7 and 8. Examining the $^{130}$Te(s) surface concentration, there is heavy NM deposition occurring in cells 6 and 7 due to the high surface area of this annular region seen in Figure 4. Additionally, there is deposition occurring on the walls in cells 2 and 3, but it is suppressed due to the additional bubble interfacial area in this region available for deposition as seen in Figure 6. This extraction effect can be seen more clearly with the significant amount of $^{130}$Te(g) extracted to the off-gas system seen in Figure 8. After converting the surface concentrations in Figure 7 and comparing the total amount of $^{130}$Te in VESIL, it is found that 11.38% of all $^{130}$Te has been extracted to the off-gas system via helium sparging.

It is also worth mentioning that not only does the extraction of NMs enhance off-gas extraction through preemptive capture before decay – it can also lead to significant steady state amounts of isotopes in the off-gas system that may be unexpected. One example is $^{129}$I(\(l\)) which arrived in the off-gas system from the extraction of $^{129}$Sn(g), $^{129}$Sb(g), and $^{129}$Te(g), but also has a very long half-life \(t_{1/2} = 1.57 \times 10^7\) years. $^{130}$Te and $^{128}$Te are two other examples of long lived species extracted into the off-gas system \(t_{1/2} = 7.9 \times 10^{20}\) years and \(t_{1/2} = 2.2 \times 10^{24}\) years respectively. With large amounts of unexpected isotopes (other than NGs xenon and krypton) building up, it’s important to understand the
overall chemistry, and atomic/molecular behavior of the species that will accumulate in the off-gas system.

Lastly, the amount of \(^{132}\text{Xe} (g)\) leaving the liquid stream and depositing onto the helium bubbles is plotted in Figure 9. A drastic growth of the amount of \(^{132}\text{Xe} (g)\) in the helium bubbles occurs as expected in cells 1 – 4, where the helium bubble interfacial area is the largest. This is followed by a drastic decrease in the density as the helium bubbles and the \(^{132}\text{Xe} (g)\) in the helium bubbles are removed to the off-gas system in cell 5 of the upper plenum. On the other hand, the spatial distribution of soluble species like \(^{131}\text{I} (l)\) shown in Figure 10, is mostly dependent on the changing density of the fuel-salt due to temperature. It is important to note this effect is small, as seen in the relative difference in the soluble \(^{131}\text{I} (l)\) concentration around the loop, and a homogenous well-mixed hypothesis for such species is a reasonable assumption.

The total amounts of stable xenon gas and the amounts of stable xenon collected in the off-gas system after 60 days of irradiation are shown below in Table IV. Since VESIL would operate at a power of 94.56 kW [7], these numbers are a reasonable estimate for the amount of stable xenon buildup.

<table>
<thead>
<tr>
<th></th>
<th>Total Stable Xenon</th>
<th>Stable Xenon in Off-Gas</th>
<th>Percentage (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{132}\text{Xe})</td>
<td>5.56 mg</td>
<td>5.54 mg</td>
<td>99.68%</td>
</tr>
<tr>
<td>(^{131}\text{I})</td>
<td>1.63 mg</td>
<td>1.62 mg</td>
<td>99.43%</td>
</tr>
<tr>
<td>(^{130}\text{Xe})</td>
<td>8.90e-2 mg</td>
<td>8.87e-2 mg</td>
<td>99.67%</td>
</tr>
<tr>
<td>(^{129}\text{Xe})</td>
<td>1.80e-13 mg</td>
<td>1.79e-13 mg</td>
<td>99.88%</td>
</tr>
<tr>
<td>(^{128}\text{Xe})</td>
<td>6.15e-4 mg</td>
<td>6.13e-4 mg</td>
<td>99.68%</td>
</tr>
<tr>
<td>Total Xe</td>
<td>7.28 mg</td>
<td>7.25 mg</td>
<td>99.63%</td>
</tr>
</tbody>
</table>

5. DISCUSSION & FUTURE WORK

As can be seen above, the helium sparging model with its current parameters and assumptions is more than capable of extracting nearly all of the stable xenon in VESIL. This conclusion will need to be revisited when a more realistic helium bubble extraction model is used. There will likely be significant amount of bubble recirculation depending on the bubble size and bubble concentration which will reduce the amount of NGs extracted.

Ultimately, with such minimal amounts of stable xenon building up, there is no need to actively sparge the fuel-salt with helium to extract the xenon. It is very unlikely that the pressure build-up will become prohibitive from an experiment design standpoint. However, the ability to actively extract IFPs

Proceedings of the PHYSOR 2020, Cambridge, United Kingdom
throughout the experiment to measure burnup, validate mass transfer correlations, measure corrosion rates, and determine chemical equilibriums will likely be invaluable.

Future work will investigate rudimentary redox control models to analyze the various options possible to implement within VESIL. These include gas sparging with hydrogen, dissolving a reducing metal into the salt, and controlling the U(IV)/U(III) ratio through U additions [10]. Through the coupling of CTF with Thermochimica, which is currently underway, the redox potential of the fuel-salt can be calculated, and corrosion rates can be estimated.

6. CONCLUSIONS

Species transport inside an in-pile salt loop were investigated and quantified. The analysis was able to provide a first order prediction of the species transport inside an in-pile experiment. The initial findings are very encouraging. The helium gas sparging model, while simple to implement, was shown to be very successful in extracting the majority of xenon and significant amount of NMs in the VESIL system. Additional analysis and sensitivity tests on the full 60-day ATR runs are currently being processed. The noble metal helium sparging model in CTF is behaving as expected and is able to account for the primary mass transfer mechanisms in MSR systems. Future work will further develop this coupling and incorporate more robust physical models to accurately determine the underlying phenomena behind NM and NG nano-micro-particle agglomeration and their behavior in molten salts.

ACKNOWLEDGMENTS

This material is based upon work supported under an Integrated University Program Graduate Fellowship. The work was partially supported through the INL Laboratory Directed Research & Development (LDRD) Program under DOE Idaho Operations Office Contract DE-AC07-05ID14517.

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