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Thermochemical Modeling of Radionuclide Vapor-Liquid Equilibria in Sodium Pools for SFR Mechanistic Source Term Analysis

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ABSTRACT

Mechanistic source term (MST) analysis of sodium fast reactors (SFR) requires understanding of various radionuclide (RN) transport phenomena influencing potential releases from the fuel to the environment. One such phenomenon includes the retention or release of RNs from the sodium coolant pool, representing the step after possible fuel failures and influencing transport to the cover gas region. Thermodynamic vapor-liquid equilibria (VLE) calculations were performed on systems representing SFR sodium pools containing oxygen impurities and radionuclide (RN) inventories. First, an assessment and recreation of a previously developed thermodynamic database was completed, including updates to thermodynamic parameters. The RN inventories used in VLE calculations represented hypothetical source terms that might be released to the pool during previously analyzed fuel failure scenarios. The calculations were performed for all possible combinations of sodium pool size (i.e., total oxygen) and number of failed fuel pins (i.e., total RNs). In this way, multiple ratios of the RN relative to the oxygen impurity (RN:O) were compared for their impact on RN volatility, which is discussed in terms of the vapor fraction (VF), defined as the fraction of the RN that is calculated to exist in the vapor phase at equilibrium above condensed phases of that element. Similar trends in VLE behavior are seen in the equilibrium calculation results for elements of similar chemistry, and for some element types, it was found that the RN:O ratio can be important due to oxide formation, which typically exist in the condensed phase.

Keywords: sodium fast reactors, mechanistic source term, radionuclides, thermodynamics, vapor fraction

1. INTRODUCTION

To assist both the advanced reactor industry and U.S. Nuclear Regulatory Commission (NRC) in the pursuit of reactor design and licensing, the U.S. Department of Energy (DOE) Nuclear Energy Advanced Modeling and Simulation (NEAMS) program has established a mechanistic source term (MST) research project under the Multiphysics Applications technical area. MST analysis is central to the reactor licensing process and a focus of the NRC regarding their mission to provide reasonable assurance of adequate protection of public health and safety and environment. The objective of the MST project is to coordinate the development of comprehensive advanced reactor MST mod/sim capabilities to support risk-informed design and licensing decisions. As part of this effort, an MST mod/sim development pathway was developed [1], which outlines the high-level objectives and near-term tasks necessary to achieve the project objectives. This work is one such near-term task that involves the assessment of a previously developed thermodynamic database for modeling radionuclide vaporization from liquid sodium pools. Updated vapor-liquid equilibrium (VLE)

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calculations were performed to investigate the impact of oxygen content on RN volatility. Additionally, VLE calculations were performed containing the entire RN inventory to investigate the effect of interactions with other RNs as opposed to assuming the RNs only speciate with sodium and oxygen. The significance of relying on such approximations in MST analyses in the context of probabilistic risk assessments (PRA) is discussed. More details of this task, including the assessment of the sodium pool vaporization thermodynamic database and demonstration in VLE calculations relevant to SFR MST, can be found in a separate Argonne technical report that has been publicly released [2].

2. DATABASE DEVELOPMENT

The database includes thermodynamic data for all possible species for the considered elements that are available in the pure substance database of the FactSage commercial chemistry software [3]. The database is in the format of a FactSage Compound Database and activity coefficients are utilized in the FactSage Equilib module. The FactSage Compound Database can be easily converted to ChemSage format (.dat file) for use with the NEAMS Gibbs Energy Minimization (GEM) solver Thermochimica [4] which has been coupled with MOOSE-based codes as well as the integral severe accident analysis code MELCOR [5]. But because of the way in which activity coefficients are translated from FactSage to the ChemSage format in this workflow, the non-ideal behavior represented by these activity coefficients is not currently accounted for in Thermochimica GEM calculations, but this can be developed in the future if needed.

Details of the development methodology for the sodium pool thermodynamic database are reported separately [2] but it builds on and verifies the work that was reported previously [6]. The elements considered are listed in Table I along with the species that are considered to be in solution (ideal and non-ideal mixing) with the Na pool.

Table I. List of considered elements (left) and species assumed in solution together (right).

Elements considered			Species in ideal solution together*		
Am	La	Rh	Am	Mo	Sb
Ar	Mn	Ru	Ba*	Na	Se
Ba	Mo	Sb	NaBr*	Nb	Sm
Br	Na	Se	Ce	Nd	Sr*
Ce	Nb	Sm	Co	Ni	Tc
Co	Nd	Sr	Cr	Np	Te*
Cr	Ni	Tc	Cs*	Na_2O	Na_2Te
Cs	Np	Te	Eu	Pd	U
Eu	O	U	Fe	Pu	Y
Fe	Pd	Xe	NaI*	Rb*	Zr
I	Pu	Y	La	Rh	
Kr	Rb	Zr	Mn	Ru	

Assumptions are made regarding how to treat the mixing thermodynamics of various species. First, certain liquid phase elemental species (e.g., metallic species) were assumed in ideal solution with the Na pool based on operational experiences [7]. To verify this, multiple comparison runs were made with metal and semimetal species either included or excluded from ideal solution with Na, and no nonnegligible differences in vapor fractions were seen. Based on solubility measurements, the following sodium compounds were also assumed in solution with the Na pool: NaBr, NaI, Na_2O, Na_2Te. And due to a lack of solubility data of oxides in liquid sodium and based on past SFR operational experiences [7], all oxides and other compounds were not assumed in solution with the Na pool to treat their vaporization fractions to the cover gas as conservatively as possible. Finally, the species with asterisks (Cs, Rb, Sr, Ba, NaBr, NaI, Te) are modeled as exhibiting non-ideal behavior in the Na solution which is represented by temperature-dependent activity coefficient functions as shown in Table II.

Table II. List of species exhibiting non-ideal behavior in solution with activity coefficient data

	ln(gamma)=A/T+B		**log(gamma)=A/T+B**		
	A	**B**	**A**	**B**	**Reference**
Ba	254.14962	0	110.37578	0	[8]
NaBr	6893.9804	-3.01882	2994.01764	-1.31106	[8, 9]
Cs	1367.80094	-0.81805	594.0284	-0.35527	[10]
NaI	8028.78584	-3.46508	3486.85739	-1.50487	[10]
Rb	904.44404	0.00339	392.79506	0.00147	[8, 11]
Sr	1004.57061	0	436.27947	0	[8]
Te	-18712.974	12.59053	-8126.9412	5.468	[12]

3. EQUILIBRIUM CALCULATIONS

The database developed in Section 2 is demonstrated here with thermodynamic equilibrium calculations for RN vaporization from representative sodium pools in the context of an MST analysis for an SFR. The primary focus is the calculation of vapor fractions (VF) for various RNs from the sodium pool.

3.1. System Parameters and Elemental Composition

Several assumptions are made to qualify the applicability to SFR MST. First, it is assumed RNs mix homogeneously throughout the Na pool volume and concentration gradients are neglected. Second, the pressure is fixed at 1 atm in equilibrium calculations for temperatures below the boiling point of Na (400-882 ºC), and the resulting volume is calculated for each temperature. This is meant to physically represent the intentional depressurization of the cover gas region as the pool temperature increases. In scenarios where the reactor vessel is not depressurized intentionally, cover gas pressure will increase with T. Through comparison calculations, it was found that the VFs for this scenario would be lower than those calculated under the fixed pressure scenario. In this way, this method of solving for the phase equilibria is conservative in estimating the VFs. Third, the volume of sodium pool, and thus the Na content used in calculations, only includes the hot pool volume and not the cold pool volume, based on assumptions previously utilized in ref

[6]. As detailed in ref [6], for the severe protected loss of flow (PLOF+) and unprotected transient overpower (UTOP+) analyses, radionuclides are conservatively assumed to remain within the hot pool of the primary sodium circuit, as shown in Figure 1. This is considered a conservative assumption, especially for the UTOP+ transient, as primary system flow will likely disperse radionuclides throughout the system.

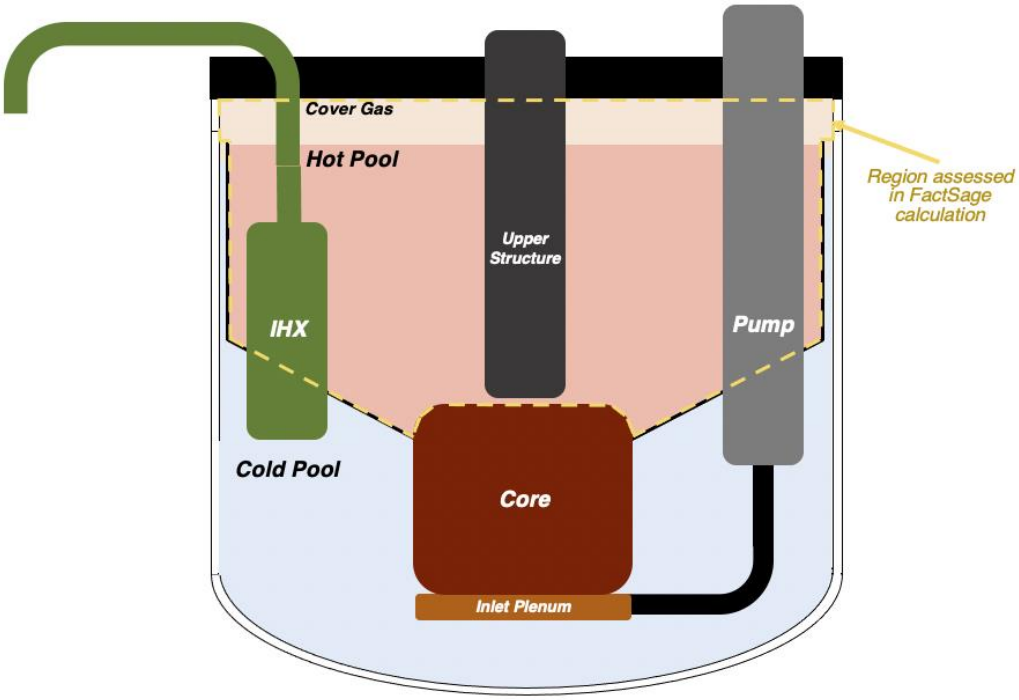

Figure 1. Region assessed by FactSage for Pool Vaporization Calculations.

The range of sodium hot pool and cover gas volumes is calculated based on a set of representative SFR designs including FFTF, AFR-100, ABTR, VTR, and available data from some commercial designs. From the set of representative designs, bounds of 1E3 and 1E6 kg were chosen for the sodium pool mass. A density value of 0.840 g/cm^3 and molar mass of 23 g/mol was used to convert to volumes and molar amounts, respectively. An oxygen content of five parts per million was assumed within the sodium pool, which is typically an upper-bound on reactor grade sodium oxygen content [13].

In fixed pressure calculations, the amount of Ar cover gas (in moles) in the system was estimated from the ideal gas law assuming P=1 atm and T=400 ºC. The volume of the cover gas region (i.e., Ar volume) is taken as a fraction of the sodium pool size used in each corresponding run, covering a range of sodium pool sizes. The cover gas region volume was taken to be 0.77 of the sodium pool size because this ratio represents the largest such value from a set of representative reactor designs, based on FFTF, AFR-100, ABTR, VTR, and some commercial designs. Through multiple comparison calculations, it was found that the VFs increase as the amount of Ar increases, therefore using the largest possible cover gas region corresponding to a specific sodium pool size is deemed conservative.

The RN inventory utilized is similar to that used in previous studies analyzing the RN release fractions into the hot pool due to the failure of 1 pin [6]. Through multiple preliminary runs of different Na pool sizes with varying, yet dilute, RN concentrations, the VFs were found to be independent of the relative proportion between RNs, except for Se, which is due to the thermodynamically favored species EuSe(g) and SrSe(g). Therefore, the VFs can be considered appropriate for nearly any dilute RN distribution in the hot pool considered here which do not involve considerable deviation from the relative amounts of Se, Eu, and Sr.

VFs vary as the relative concentration of RNs to Na/O become more dilute (i.e., low RN:Na ratios), therefore a set of test cases were completed covering the range of RN concentration levels involving the representative SFR designs with four fuel pin failure scenarios: 1 pin, 10 pin, 100 pin, 1000 pin. As such, the number of test cases (TC) is reduced from the 32 possible combinations of Na pool size and fuel pin failure amounts to just seven cases. This range of representative RN concentrations can be seen in Table III.

Table III. Equilibrium calculation test cases covering a range of ratios of failed fuel pins to Na, Ar, and O content representing various SFR designs

Test Case ID	Sodium (mol)	Oxygen (mol)	Argon (mol)	Failed fuel pins	~C_{Cs} (ppm)
TC1	4.35E+07	3.13E+02	1.66E+04	1	0.04
TC2	4.35E+06	3.13E+01	1.66E+03	1	0.4
TC3	4.35E+05	3.13E+00	1.66E+02	1	4
TC4	4.35E+04	3.13E-01	1.66E+01	1	40
TC5	4.35E+04	3.13E-01	1.66E+01	10	400
TC6	4.35E+04	3.13E-01	1.66E+01	100	4000
TC7	4.35E+04	3.13E-01	1.66E+01	1000	40000

For reference, the activities of several RNs were measured in the sodium coolant of EBR-II and FFTF [14] and are similar in value or up to a few orders of magnitude smaller than the lowest concentrations used here (i.e., TC1: 1 pin failure in the largest sodium pool volume). In this way, the majority of test cases utilized here correspond to SFR accident scenarios of varying severity in RN release. Finally, it is noted that structural material elements (i.e., corrosion products) are not included in the equilibrium calculations presented.

3.2. Equilibrium Calculation Results

Equilibrium calculations covering a range of ratios of RN concentrations to Na pool sizes (and thus oxygen contents available for reaction with the RNs) were performed with the objective of finding VF curves that can be used in generalizable MST modeling tools. The equilibrium calculations were conducted by keeping the RN amounts the same across all seven test cases (TC1 to TC7), only altering the amounts of Na, O, and Ar. As shown in Table III, TC1 represents the most dilute TC, and thus the lowest RN:Na or RN:O ratio, which is the parameter used often when interpreting the results. Only a subset of results are shown in this paper, while additional details and discussion can be found in the full report [2]. The RNs are divided into four groups based on similar trends in behavior, with examples provided for some groups. In all VF curve plots, the RN:Na ratios is represented by the concentration of Cs for each test case for convenience in visualizing RN concentrations in the Na pool, as the relative proportion of each RN to any other is constant.

3.2.1. Group 1: Cs, Na, Rb, Rh, Ru, Sb, Tc

All RNs in this group exhibit no concentration effects except at the highest temperatures for the highest concentration test cases (e.g., TC7). The dominant gas phase species of Na is always Na(g) with the dimer (Na_2(g)) at 1 order-of-magnitude (OOM) lower amounts. The VF curves for Na generally do not differ

across the range of RN:Na values except at the highest ratio, where the VF diverges at high temperatures approaching the boiling point (882 °C) due to the formation of less Na_2O(liq) formed. It is concluded that there are no inter-radionuclide (inter-RN) effects, and the VF of Na is not affected by the inclusion of other RNs in the calculation. For Cs and Rb, TC7 starts to diverge appreciably from the other six at around 700 °C, and especially so around 860 °C, where temperatures are above the boiling points of Rb (688 °C) and Cs (671 °C). The dominant liquid and gas phase species for both elements is always the elemental species, with species formed with sodium or iodine formed at OOM lower amounts. But because CsI and RbI formation increases with RN:Na ratio, concentration effects may not be negligible on the VF calculation for cases with very high RN:Na ratios and temperatures. Similarly, for the transition metals Rh, Ru, Tc, and metalloid Sb, a similar VF behavior is seen where TC7 starts to diverge around 800 °C and especially above 860 °C. The dominant liquid and gas phase species for all four elements is always the elemental species by multiple OOM. Therefore, concentration effects as well as inter-RN effects may be ignored in the calculation of VFs for these elements up to 800 °C.

3.2.2. Group 2: Am, Ba, Eu, Mo, Nd, Pu, Sm, Sr, U, Y, Zr

All the RNs in this group exhibit some concentration effects mostly due to the formation of oxygen-containing compounds dependent on the RN:O (i.e., RN:Na) ratio and temperature, where the impact of oxygen is most pronounced at the most dilute RN concentrations (lowest RN:O). The VFs for Am, Ba, Eu, and Sm generally do not exhibit concentration effects except when the oxygen content is similar in magnitude to the total RN content (i.e., dilute RN concentrations), which causes varying behavior in terms of the most thermodynamically favorable condensed phase (solid oxide vs liquid metal), which in turn affects the vaporized amount. The VFs are driven by the dominant gas species for Am, Ba, Eu, and Sm, which for all temperatures and all test cases is the metallic monomer by many OOM. As a result, aside from the extremely dilute cases where the VF for these elements may be lower, as well as a special case where Ba forms an inter-RN compound with uranium at lower temperatures and low RN:O, the VFs for these elements can be conservatively estimated using TC7 and ignoring inter-RN effects.

For most temperatures Mo exhibits similar VF behavior as the other transition metals in Section 3.2.1 but the favorability to form one inter-RN compound at lower temperatures slightly increases the VFs for the lowest three RN:O ratios, showing similarities to the VF curves of Ba. Therefore, it may be conservative to assume a VF from TC3 at lower temperatures and a VF from TC7 at all other temperatures.

For elements Nd, Pu, Sr, U, Y, and Zr, the formation of complex oxides, sometimes with other RNs (in the case of U, Zr, Sr), complicates the VF curves across the different test cases. Additionally, the formation of gaseous metal oxide monomers becomes more favorable at low RN:O ratios over the gaseous metal species for most of these elements. Especially for Nd, Pu, and Y, the VF behavior is driven by RN:O ratio influencing both gaseous and condensed phase oxide formation with negligible inter-RN effects. The VFs for these elements can be conservatively estimated from TC1 at high temperatures and TC7 at low temperatures. For Sr (Figure 2), inter-RN effects with U, Zr, and Se, and concentration effects (i.e., RN:O) need to be considered if a best estimate VF curve is desired given its potential dose consequence in MST analyses. In this case, the appropriate Sr:Na(O) ratio should be modeled and assumptions may need to be made about the appropriateness of allowing inter-RN effects based on if Sr co-transports with U, Zr, or Se, or if the Na pool can be assumed to be well-mixed. Otherwise, the VF curve can be conservatively assumed from TC7 at all temperatures, which is mostly driven by the distribution between the gas and liquid phases of the elemental metal species. Inter-RN effects for U and Zr also drive complex oxide formation behavior that is sensitive to the oxygen content. The VF for U is more driven by the speciation and vaporization of U(g), UO(g), or UO_2(g), while the VF for Zr may be sensitive to the iodine content in the system relative to oxygen, as ZrI_2(g) may form over Zr(g) and ZrO(g) for higher RN:O systems where it is also assumed interactions with iodine are possible.

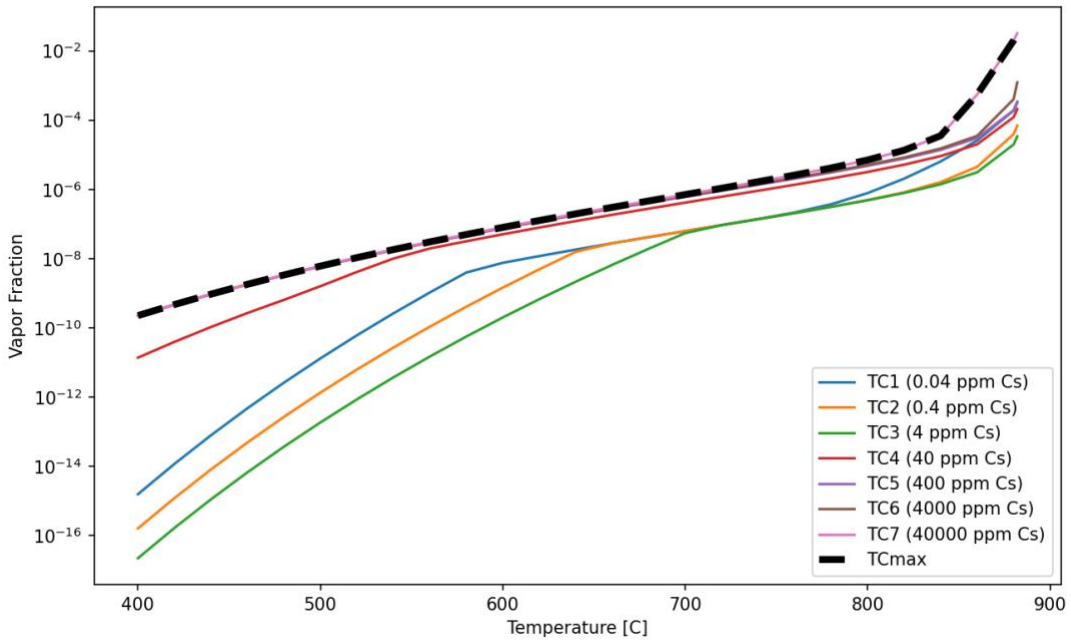

Figure 2. The vapor fraction curves for strontium for 7 test cases of varying RN:O ratio.

3.2.3. Group 3: Ce, La, Nb, Np, Pd

This group is generally characterized as oxide-forming elements, similar to those in Section 3.2.2, except the VFs of these elements tend to decrease for increasing RN:O, which is the opposite behavior of the previous group. For Ce, La, Nb, and Np, this behavior is likely because the dominant gas phase species for these elements is the monoxide (i.e., MO) while the dominant condensed phase species is usually the liquid metal. Therefore, the increasing RN:O ratios, which also represent decreasing oxygen content, will drive the thermodynamic favorability of the elemental species such as the liquid metal, and thus have the net effect of reducing the VFs for these elements in these systems. And for Pd, this behavior is seen because of inter-RN effects with U (UPd_3) in high RN:O test cases which will reduce the VF if this inter-RN compound forms. If it is deemed not likely that this compound forms, then Pd can be treated similarly as the other transition metals, driven solely by the favorability of its elemental metallic species. Nonetheless, all elements in this group can be conservatively estimated by their VFs from TC1.

3.2.4. Group 4: Br, I, Se, Te

The elements in this group are characterized by their anionic behavior in the Na pool, often forming compounds with Na or other RNs. In addition to these inter-RN effects, concentration effects may contribute as well evidenced by the importance of the proportion between RN, Na, and O in the VF curves of each of these RNs. Therefore, care must be taken for these system parameters if a best estimate is desired such as in the case of iodine which often has high importance in MST analyses. For conservative VF estimates, all elements in this group except for Se exhibit their highest VFs for the test cases with the higher RN:Na(O) ratios (i.e., TC7), mostly driven by favorable interactions of the anion with other RNs (e.g., CsI, CsBr, EuSe(g), EuTe(g)) over the Na solution at these extreme RN concentrations. Although it is worth noting that Se is the only element of this group that is not modeled with activity coefficient functions representing

nonideal behavior due to a lack of such experimental data. Therefore, it is likely that Se would experience reduced vapor pressures over the Na pool due to increased favorability for Na_2Se formation (similar to Te), therefore the VFs calculated here for Se, which assume ideality, are likely conservatively high and may be too sensitive to inter-RN effects. And if a best estimate is desired for Te, accurate understanding of RN:Na may result in a lower VF, especially if co-transport of Eu and Te are not deemed realistic.

The unusual VF behavior for iodine seen in Figure 3 is driven by inter-RN effects with Cs due to the thermodynamically favorable formation of the monomer and dimer of CsI(g) at lower temperatures, as well as the liquid or solid CsI depending on the temperature. As such, the dominant condensed phase species for iodine is NaI(liq) for all test cases and temperatures, except for lower temperatures of TC6 and TC7, where CsI(s,liq) formation is favorable due to the larger Cs content relative to Na. Similarly, the dominant gas phase species for iodine is NaI(g) by multiple OOM over CsI(g) for TC1-4. For TC5-7, the relatively high Cs content relative to Na starts to allow formation of CsI(g) and Cs_2I_2(g) in similar amounts as NaI(g) varying with the temperature and RN:Na ratio. These test cases with the largest VF values are caused by the formation of cesium iodide species due to the significantly higher Cs:Na ratios, therefore these may not be realistic for SFR MST scenarios but are worth noting for scenarios where homogeneous mixing does not occur and co-released cesium and iodine may preferentially form stable compounds during the event. Given the radiological importance of iodine, it may be advisable to investigate mixing assumptions and generate best estimate VF functions for iodine as a function of the ratio between Na, Cs, and I. And although Br is less important to MST, its VF behavior is similar, driven primarily by the formation of NaBr(g), or if the Na content is low enough, CsBr(g).

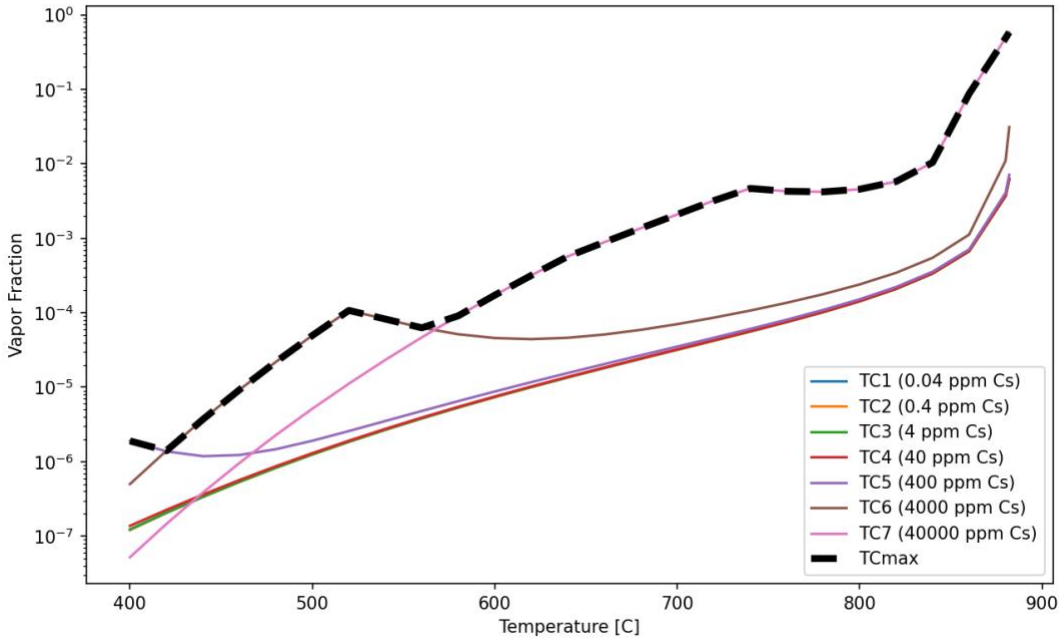

Figure 3. The vapor fraction curves for iodine for 7 test cases of RN:Na ratio.

4. CONCLUSIONS

In support of SFR MST modeling and simulation tool development, it was recommended to perform an updated assessment of a previously reported sodium pool thermodynamic database. Originally developed in the HSC Chemistry software, the database was recreated here (Section 2) in the FactSage commercial software to access updated and/or expanded sources of thermodynamic data and to facilitate conversion to the ChemSage format in the future, if necessary. The ChemSage database format is used by the NEAMS code Thermochimica, which is a Gibbs energy minimization solver that can be coupled to MOOSE-based tools or MELCOR. The database serves to model radionuclide vaporization from liquid sodium pools specifically in the context of SFR MST analyses, and therefore several assumptions were taken and documented.

The database is demonstrated through representative chemical equilibrium calculations (Section 3). The elemental vaporization fraction (i.e., the fraction of the element in the vapor phase above the condensed phases) is used as the figure-of-merit relevant to MST analyses. Several combinations of input compositions were modeled over the temperature range between the melting and boiling point of sodium. First, several RN inventories were used, assuming that either 1, 10, 100, or 1000 metallic fuel pin failures occurred (i.e., the corresponding radionuclide inventory is released directly from the failed fuel to the pool). Second, various sodium pool sizes were considered, covering a range representing available design data, which controls the total oxygen impurity content in the system. Major findings of the analysis include considerations for both potential concentration (i.e., RN:Na or RN:O) effects and potential inter-RN (i.e., RN1:RN2) effects:

- The sodium pool size is important due to the assumption that bulk sodium contains a constant concentration of oxygen impurities. In this way, larger sodium pool sizes may contain sufficient oxygen content to drive the favorability of oxide-forming reactions with some RNs. Therefore, it was found that the ratios of RNs to oxygen (and thus sodium) is an important parameter in understanding the thermodynamic behavior of RNs in sodium pools. As such, trends in behavior across the different chemical groupings of the RNs were discussed in the context of this ratio. For some RNs and MST analysis scenarios, it may be sufficient to assume the conservative value of VF (across all possible RN:O ratios). Otherwise, it is recommended to accurately characterize the oxygen content in the sodium pool and cover gas region.
- It was also found that the VFs of many RNs are independent of the existence of other RNs in the system, i.e., there are no inter-RN effects. Therefore, excluding other RNs as part of the calculation input for one specific RN in the pool may be sufficiently descriptive of that specific RN's VLE behavior. Many test case VFs were bounded by the VF that represents a species solely containing the RN, Na, and/or O. The lack of such interelement effects was found to exist for all tested RNs except for those listed below, most of which can behave as anions and are thus expected to be sensitive to the cationic RN concentration relative to Na.
    - The VF of Zr is dominated by $ZrI_2(g)$, therefore knowing the iodine concentration coexisting with zirconium in the pool is recommended.
    - The VFs of I and Br are driven by the cesium halide only at high concentrations of Cs relative to the Na concentration, otherwise the sodium halide is favorable.
    - The VF of Se is driven by SrSe(g) or EuSe(g) only at high RN concentrations relative to the Na concentration, otherwise the pure species Se(g) drives the VF.
    - The VF of Te is driven by EuTe(g) only at high RN concentrations relative to the Na concentration, otherwise the pure species Te(g) drives the VF.

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