

ESTIMATION OF FPS RELEASE FROM SODIUM POOL UNDER BDBA CONDITIONS IN A SODIUM FAST REACTOR

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Abstract

This work is focused on the prediction of the release of volatile fission products (NaI) from hot sodium pools. A critical review of a model based on the film theory and its estimates in comparison to data from the NALA experimental program has been conducted. Given the assumptions embedded in the approach, the fitting-to-data transport coefficients derived and some inconsistencies found between its formulation and the estimates reported, an alternate approach including other phenomena anticipated in the scenario is here proposed. Based on diffusive and convective mechanisms in the gas phase, the assumption of the analogy between heat and mass transport (HMT) and the Raoult's law to set the NaI concentration at the Na pool interface, a good agreement has been found with experimental data, which mean a substantial enhancement of qualitative and quantitative predictability and, no less important, at the same time do have a conservative nature.

1. INTRODUCTION

Fission products (FPs) release from hot sodium pools is a very important concern when considering the potential radiological consequences in the case of Beyond Design Basis Accident (BDBA) in Sodium-cooled Fast Reactors (SFRs). In the case of vessel failure, large amounts of fuel, FPs, and activated sodium (Na) can enter the inertized inner containment and the heated sodium by the decay heat would evaporate with the release of significant fractions of hazardous volatile FPs from the radiological point of view as iodine (as sodium iodide, NaI). In this scenario, the estimation of the Na-pool retention capability is of utmost importance.

Much work has been done in the past regarding the evaporation of volatile radionuclides from hot Na-pools in the reactor vessel to the inert cover gas region and analytical models for the prediction of this phenomena were derived. In these models, the release of the volatile FPs is governed by diffusive transport processes. Based on a mass transfer formulation, the retention factor (defined as the ratio between the relative transfer rates of FP with respect to their initial inventories in the pool) of the species of interest is calculated.

This paper synthesizes the results of the work developed by CIEMAT in the frame of the ESFR-SMART project (contract number 754501). A critical review of the earlier work done by [1]–[3] and others on FPs release from hot sodium pools has been done through the comparison of the model results against experimental data from the NALA program [4], [5]. In these works, the film theory was applied for the evaporative release of FPs calculation. With the assumption of a hypothetical stagnant film at the gas/liquid interface, transport through was assumed to be governed by diffusion. Given the weakness found in the formulation of these works, a more mechanistic approach including other phenomena anticipated in the scenario is proposed and the results are compared and discussed.

2. EXPERIMENTAL NALA PROGRAM

NALA is a German (KFK) experimental program that included the investigation of fuel and fission products release from a wide range of Na-pool temperatures, including sodium boiling and sodium aerosol behavior in a closed/open vessel for a variety of geometric and convective conditions and for a representative selection of FPs. All the tests were performed in an inert atmosphere (Ar or N₂). The program consisted of laboratory-scale experiments in a glove box (NALA I), technical-scale experiments (NALA II), and laboratory-

scale experiments in a forced convection loop (NALA III). In this work, the focus is on the results derived from the NALA II phase: a sodium aerosol system under natural convection [4], [5].

The main components of the NALA II facility are a 2.2 m³ heated vessel, a sodium pot of 26 cm inner diameter with external heating devices (acetylene burner) to be flanged to the vessel bottom, inert gas supply, survey instrumentation, and aerosol measurement instrumentation (Fig. 1).

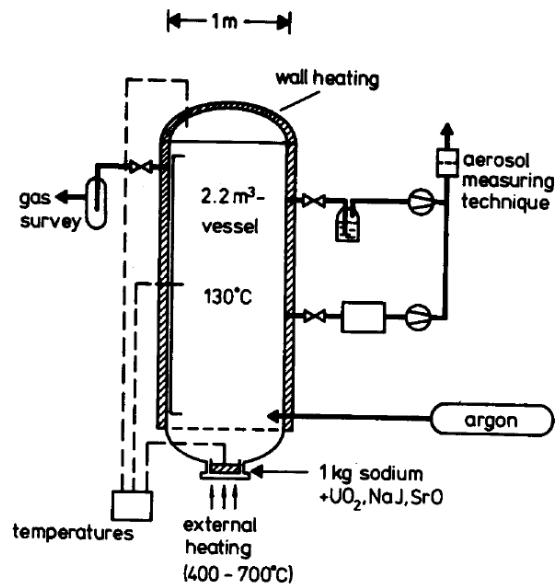


FIG. 1. DIAGRAM OF THE NALA-II FACILITY [4].

The main characteristics and boundary conditions in NALA II experiments were collected in Table 1: pool temperatures from 600 to 900 K, inert gas temperatures of 400 K, natural convection and admixtures of UO₂, NaI and SrO in the pool [4], [5]. The vessel is filled with argon at normal pressure (1 bar) and heated up to 400 K and maintained constant during the whole experiment. In the sodium pot, sodium amounts of the order of 1 kg are heated up by external heating (0.7 to 3 h) adjusted to the evaporation rate in such a way that ~10% of the Na evaporates. Temperatures are measured at six different locations in the vessel and at two locations in the sodium pot. The aerosol instrumentation consists of washing bottles, filters, and an automatic, continuously measuring device for sodium mass concentration measurements. An Andersen impactor is used for size distribution measurements. In the next table, the main characteristics of NALA II tests are shown (Table 1).

TABLE 1. EXPERIMENTAL BOUNDARY CONDITIONS IN NALA II [5]

Geometry	Values
Vessel volume (m ³)	2.2
Vessel height (m)	2.30
Tests conditions	
Na pool area (cm ²)	531
Na amount (kg)	1
Na temperature (K)	600-900
Atm. Gas temperature (K)	400
Contaminants	UO ₂ , NaI, SrO

The present analysis is focused on the experiments on Na and NaI vaporization. Table 2 compiles the experimental data for the tests of interest. As it can be seen T5 and T7 are exclusively devoted to the Na vaporization estimates. Note that being Na boiling point around 1156 K, the Na vaporization rate is controlled by increasing Na temperature and, in no case, Na pool reached boiling conditions.

TABLE 2. TESTS CONDITIONS [5]

Exp. Nr.	T (K)	Na amount (g)	NaI amount (g)
T1	835.15	258	1.00
T2	905.15	1100	1.06
T3	754.15	1315	1.00
T4	769.15	1322	10.00
T5	837.15	1146	-
T6	823.15	1321	0.20
T7	830.15	1007	-

3. THEORETICAL MODELS OVERVIEW

In this section, an overview of the earlier modelling work by other authors is given and the accuracy of their estimates is shown against NALA data. Then, the formulation of a more mechanistic model is thoroughly described. And, finally, additional phenomena are included in the formulation and their effect are assessed against the same database.

3.1. Earlier modelling critical overview

Fission products emission from hot sodium pools was analyzed and compared against experimental data from NALA II program by [1], [4], [6], and later by [2] among others. In all these works, the evaporative release is modeled as governed by diffusive and convective transport processes. Based on the film theory by Nerst (1904) a mass transfer coefficient approach is made: the mass flux of the volatiles is assumed to be controlled by a driving force, i.e. the partial density difference between the gas side of the liquid surface and the atmosphere, and the gas mass transfer coefficient represents the reciprocal resistance of the species against mass transfer [1].

In NALA program [4]–[6], the experimental release of a fission product solute (i) is given with respect to the release of the solvent (liquid sodium) by the Retention Factor (RF):

$$RF = \frac{\left[\frac{m_i}{m_{solvent}} \right]_{pool}}{\left[\frac{\phi_i}{\phi_{solvent}} \right]_{released}}, \quad (1)$$

Where m denotes the initial inventory of the FP (i) and solvent (liquid Na) and ϕ the release mass fluxes. In other words, the retention factor is a ratio between the relative transfer rates of Na and FP (relative with respect to their initial inventories in the pool).

Fig. 2 shows the main results obtained by [1] regarding RFs of NaI in the Na pool (red line). In this approach, the gas mass transfer coefficient for sodium vapor has been fitted to the NALA database. Despite this, there is an overestimation in the predictions that in some tests is as high as a factor of 30. An attempt made to reproduce these results is shown (green line). Even by using the same fitted Sherwood number (Eq. 3.10 from [1]) and Na and NaI properties, these estimations are far from [1] ones.

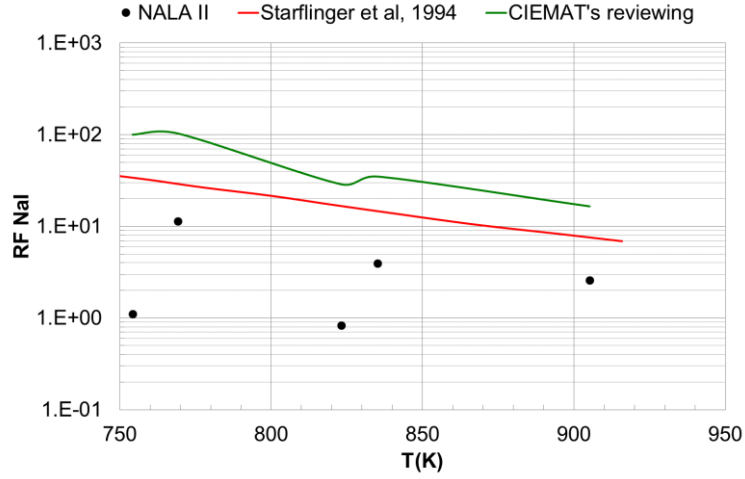


FIG. 2. RF OF NAI IN NA VS. POOL TEMPERATURE.

The correlation given by Eq. 4.29 by [1] in the temperature range of interest ($298\text{K} \leq T \leq 933\text{K}$) is as follows:

$$p_{v,NaI} = 10^5 \cdot 10^{\left[7.557 - \frac{10924}{T}\right]}, \quad (2)$$

However, results from this equation differ from the experimental data reported by [7]. In the next equation, a correlation of these experimental values [7] for the NaI vapor pressure is presented:

$$p_{v,NaI} = 133.322 \cdot 10^{\left[11.681 - \frac{11028}{T}\right]}, \quad (3)$$

By using this correlation, pressure vapors up to one order of magnitude higher than those predicted by Eq. 4.29 [1] are found which would account for the overestimations in RF calculations (Fig. 3):

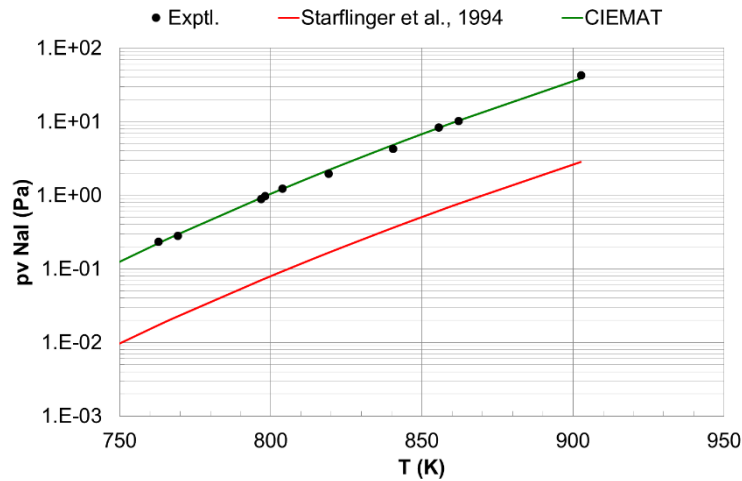


FIG. 3. NAI VAPOR PRESSURE VS. POOL TEMPERATURE.

3.2. Model proposal

3.2.1. Gas Mass Transfer (GMT) model

Building on the previous works, the Gas Mass Transfer model (GMT) approach is based on the film theory [8]. This theory is a simple conceptualization of the gas-liquid transfer processes in which near the interface there exists a hypothetical stagnant film (bulk liquid is not moving) and transport is governed essentially by molecular diffusion; in the interface, equilibrium is assumed between liquid and gas phases. In this approach and based on estimations of the mass transfer coefficients, only the gas-phase mass transfer is calculated, neglecting the liquid one due to the fact that the resistance to mass transfer in the gas phase is up to four orders of magnitude higher than the liquid mass transfer coefficient [8]. Fig. 4 represents the decrease of the partial density from the gas side of the interface into the gas bulk.

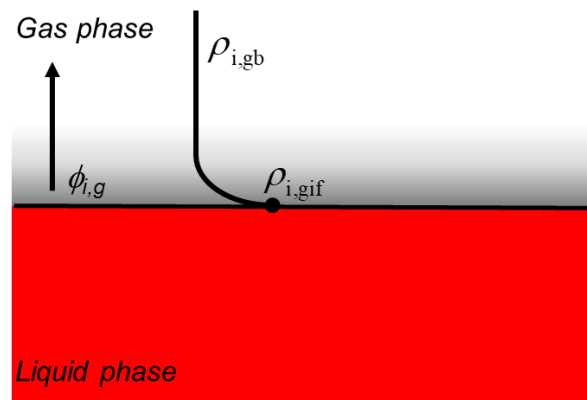


FIG. 4. GMT MODEL SKETCH.

In this approach, the release mass flux (ϕ) of any volatile species (i , FPs and Na) from the liquid phase is controlled by a driving force, i.e. the partial density difference between liquid surface and gas bulk and the reciprocal resistance of the species against mass transfer is taking into account through the gas mass transfer coefficient:

$$\phi_i = k_{i,g} \cdot \Delta\rho_{i,g}, \quad (4)$$

Where the mass transfer coefficient ($k_{i,g}$) is derived from the non-dimensional Sherwood number and the ratio between the binary diffusion coefficient of the considered species in the gas phase and the characteristic length of the system:

$$k_{i,g} = Sh_{i,g} \cdot \frac{D_{i,g}}{L}, \quad (5)$$

NaI binary diffusion coefficient in Argon ($D_{NaI,Ar}$) has been taken from [9], Na binary diffusion coefficient in Argon ($D_{Na,Ar}$) is taken from [10] and the characteristic length of the system (L) is taken as a quarter of the Na pool diameter ($d/4$) as recommended by [11]. By applying the HMT analogy ($Le \sim 1$ in the analyzed scenarios), the convective mass transfer Sherwood number is estimated from suitable heat transfer correlations, according to the prevailing flow regime (as determined through the non-dimensional Ra numbers) [11]:

$$Sh = 0.15 \cdot (Gr \cdot Sc)^{1/3} \quad (10^7 \leq Ra \leq 10^{11}, \text{ all Pr}), \quad (6)$$

The partial density difference in the gas phase between the gas interface (g_{if}) and the gas bulk (g_b) is calculated through:

$$\Delta\rho_{i,g} = \rho_{i,gif} - \rho_{i,gb}, \quad (7)$$

Where $\rho_{i,gif}$ is estimated from the saturation vapor pressure of the chemical species, which is just correlated with pool temperature [8]. This implicitly entails that the potential interaction between NaI (solute) and Na (solvent) is considered negligible. On these assumptions, $\rho_{i,gif}$ is calculated as

$$\rho_{i,gif} = \frac{p_{v,i}(T_{if})}{(R/M_i) \cdot T_{if}} \cdot \gamma_i \cdot x_i, \quad (8)$$

With $p_{v,Na}$ from [12] and $p_{v,NaI}$ calculated from Eq. 3, x_i denoting the mole fraction of the species i in the liquid, and γ_i the activity coefficient of the species i . Due to the small amount of fission products solved in the sodium pool, the approximation $x_{Na} = \gamma_{Na} = 1$ is made [1]–[3]. In the case of NaI, γ_{NaI} is calculated through the excess Gibbs free energy of mixing at an infinite solution [13]. The resulting values, larger than 100, indicate the strong tendency of NaI to escape from the Na pool [14]. All gas phase properties involved in the above equations are evaluated at the pool temperature ($T_{if} \approx T_{pool}$). Note that $\rho_{i,gb}$ grows along time due to the incoming NaI flow from the pool over time.

When Na vapor moves away from the pool surface enters in a cooler region where molecules tend to nucleate in the form of tiny primary particles and, depending on conditions, grow by heterogeneous condensation or even coalescence. Beyond the local heat-up this entails in a region close to the pool surface, in terms of mass this means that the growth of gas bulk Na density, $\rho_{Na,gb}$, should be more moderate than initially considered; in other words, Na density difference between interface and gas bulk will be higher and so evaporation rates. To take this effect into account, a correction factor (f_{cond}) is introduced in Eq. 4:

$$\phi_{Na} = k_{Na,g} \cdot \Delta\rho_{Na,g} \cdot f_{cond,Na}, \quad (9)$$

This correction factor is calculated by the following equation [4], [9] and [13]:

$$f_{cond,Na} = \frac{\beta_{Na}(T_{if}) \cdot (\Delta T \cdot Le^{-n} + (\Delta h_{v,Na} \cdot \Delta p) / (Le \cdot c_{p,gb} \cdot P))}{\Delta p \cdot (1 + (\beta_{Na}(T_{if}) \cdot \Delta h_{v,Na}) / (Le \cdot c_{p,gb} \cdot P))}, \quad (10)$$

Where β_{Na} corresponds to the first derivative of the vapor pressure $p_{v,Na}$ to the temperature T at the interface, ΔT and ΔP the temperature and pressure differences in the gas phase, $\Delta T = T_{if} - T_{gb}$ and $\Delta p = p_{Na,gif} - p_{Na,gb}$ respectively, n is the coefficient corresponding to the heat transfer correlation used, $c_{p,gb}$ the specific heat capacity of the gas phase, $\Delta h_{v,Na}$ the latent heat of vaporization of sodium and Le is the dimensionless Lewis number (ratio between the Schmidt and Prandtl numbers).

3.2.2. Extended gas mass transfer (EGMT) model

Despite the good agreement obtained with GMT approach, the search for a more mechanistic model to include the dragging effect of the Na on the volatile species release leads us to propose an extension of the GMT by adding a term in Eq. 4 to take into account this phenomenon:

$$\phi_i = k_{i,g} \cdot \Delta\rho_{i,g} + \frac{\phi_{Na}}{\rho_{sat,Na}(T_{pool})} \rho_{i,gif}, \quad (11)$$

Where ϕ_{Na} is given by Eq. 9 and $\rho_{sat,Na}$ is the density of sodium vapor above the saturated liquid, calculated from the thermodynamic relation [12]:

$$\rho_{sat,Na} = \left(\frac{\Delta h_g}{T \cdot \beta_{sat}} + \frac{1}{\rho_l} \right)^{-1}, \quad (12)$$

Where Δh_g the enthalpy of vaporization, β_{sat} the temperature derivative of the pressure along the saturation curve and ρ_l the liquid sodium density.

4. MAJOR RESULTS

In the next figure (Fig. 5), a comparison between NALA II experimental data of free convection controlled Na release from a liquid pool into Argon gas atmosphere and the calculated predictions is presented: predictions by [1] (red line) and GMT model estimations (blue line). At first glance quantitative and qualitative good agreement is obtained by both approaches, specially estimations from [1] where a fitted to data mass transfer coefficient for sodium is used. This quantitative difference points out the sensitivity of the model to the gas transfer coefficient calculation.

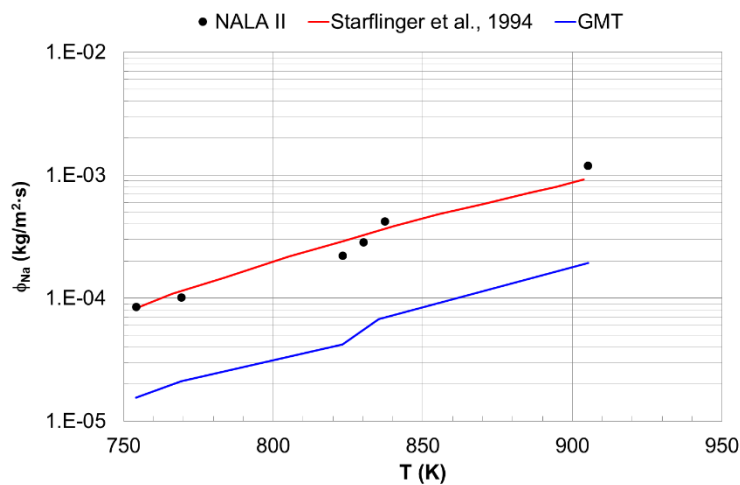


FIG. 5. NA MASS FLUX OF EVAPORATION VS. POOL TEMPERATURE.

Fig. 6 shows the models-to-data comparison in terms of the RF. In spite of the good estimations for the Na mass flux by [1], a huge overprediction is observed with the lower pool temperatures and as pool temperature increases, the overprediction is reduced. GMT good agreement with experimental data highlights the unnecessary of fitted mass transfer coefficients and the great sensitivity of the model to variables with empirical nature as pressure vapor or binary diffusion coefficients.

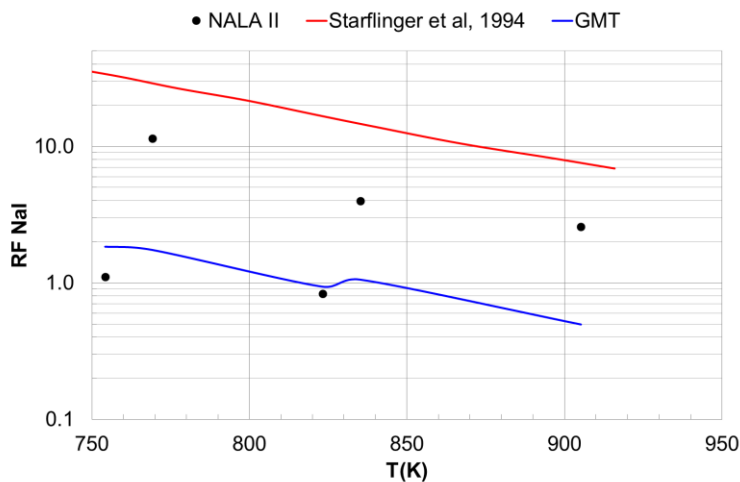


FIG. 6. RF OF NAI IN NA VS. POOL TEMPERATURE.

Fig. 7 illustrates the NaI vaporization rates given by GMT and EGMT approaches. In this case, no experimental data are available. From the figure, the effect of Na vapor dragging results in an enhancement of the NaI mass flux by a factor between 4 to 3, the lower the temperature, the higher the dragging impact on the mass flux. Regarding the absence of a clear trend of mass flux with temperature, it can be explained by the scattered initial inventories (Table II) that shield the increasing mass flux trends with pool temperature predicted by the models. As expected, an increasing trend with temperature of NaI mass flux estimations can be observed when looking at the gas mass transfer coefficient (embedded table in Fig. 7).

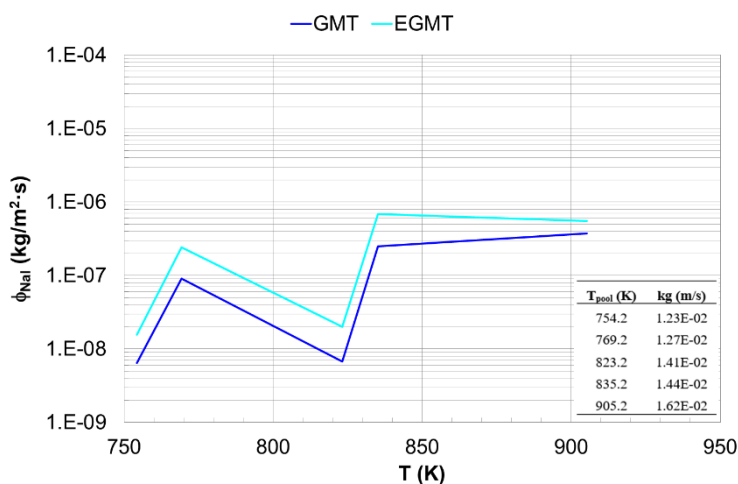


FIG. 7. NAI MASS FLUX VS. POOL TEMPERATURE.

RF experimental values are compared against models predictions in the next figure (Fig. 8). As indicated by [1], experimental data of the NaI release under varying conditions of pool temperature and initial inventories (Table II) show a considerable scattering of the retention factor [1]. Differences between GMT and EGMT results indicate an overprediction of the NaI mass flux with the EGMT approach: the dragging effect of Na vapor on the total NaI mass flux seems to be overestimated.

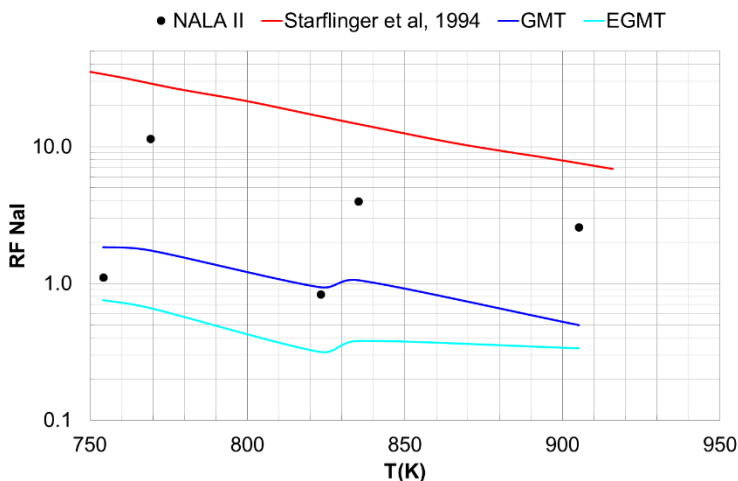


FIG. 8. NAI RF VS. POOL TEMPERATURE.

5. CONCLUSIONS

This work studies the release of volatile fission products from hot sodium pools. A critical review of previous works has been done and the main weakness of these approaches identified. A more mechanistic

approach including other phenomena anticipated in the scenario is proposed and the results are compared and discussed.

An exhaustive review of previous works based on film theory, besides the assumptions embedded in this approach and some inconsistencies found between its formulation and the estimates reported, reveals the use of fitting-to-data transport coefficients derived from the experimental NALA database, achieving in this manner excellent agreement of Na vapor estimations. However, this is not translated to the RF predictions with a large overprediction of the RF ones that is overcome with a different correlation for the NaI vapor pressure. Despite the overall good results, the use of an adjusted mass transfer correlation could impair somewhat the validation of these works.

After checking the suitability of the film model for the analyzed scenario through an estimation of the gas/liquid mass transfer coefficients, a mass transfer model based on diffusive and convective mechanisms in the gas phase, the assumption of the analogy between HMT and the Raoult's law to set the NaI concentration at the Na pool interface is build up. Besides, other phenomena anticipated in the scenario are included.

In this approach, the convective mass transfer Sherwood number is estimated from classical heat transfer correlations, according to the prevailing flow regime and the dragging of the Na vapor on the volatile species release is included through a term on the general mass flux equation. From the results, the gas transport coefficient is proved to be the most influencing factor and the results point out the unnecessary of fitted values by means of an adequate Sherwood number choice. Moreover, their formulation has an empirical nature that makes their use restricted to their development range. A good agreement has been found with experimental data, which means a substantial enhancement of qualitative and quantitative predictability and, no less important, at the same time do have a conservative nature. Besides this, there is still room for enhancement in the sodium evaporation modelling. Moreover, modelling of fission products transfer has been scarcely reported and a much more extensive comparison to data would be indispensable to consolidate a sound assessment of the model.

As future work, the validation of the proposed approach is planned to be extended to NaI and Cs in NALA I conditions, i.e., thermal equilibrium between the liquid pool and atmosphere and forced convection.

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