VOLATILE FISSION PRODUCTS TRANSFER FROM HOT SODIUM POOLS

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ABSTRACT

This study focuses on the release of the most relevant volatile fission products from the hot sodium pools. From an exhaustive review of the open literature, 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. Due to certain assumptions embedded in the approach, the fitting-to-data transport coefficients derived and some inconsistencies found between its formulation and the reported estimates, 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 ideal-dilute solutions laws to set the fission products 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 while maintaining a conservative nature.

KEYWORDS: SFRs, Source Term, FPs Release, Mass Transfer Model, Heat-Mass Transfer Analogy.

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1. Introduction

Sodium-cooled Fast Reactor (SFR) technology was one of the most promising technologies in the 1960s-80s as an alternative to Light Water Reactors (LWRs). Although many of the programs for its development were abandoned in the 80's and 90's, there has been a renewed interest in this type of technology for some years now, being one of the technologies considered in the Generation IV International Forum (GIF) (https://www.gen-4.org/gif/jcms/c_9260/public) and one of the reactor technologies considered as a valuable option to be deployed as Small Modular Reactors (SMRs).

Fission products (FPs) release from hot sodium pools is a major concern when considering the potential radiological consequences in the case of Beyond Design Basis Accident (BDBA) in Sodium-cooled Fast Reactors (SFRs). In accidental conditions, the heated sodium by the decay heat would evaporate with the release of large amounts of fuel, activated sodium (Na) and significant fractions of hazardous volatile FPs from the radiological point of view into the inert cover gas region in the reactor vessel.

Sodium as a liquid metal has physico-chemical properties very different from those of water, which has a great impact on the behavior and transport mechanisms of FPs under accidental conditions, this being one of the main sources of uncertainty in the evaluation of the source term. This is why the estimation of the Na-pool retention capability is of utmost importance and the subject of numerous experimental programs and analyses.

Much work has been done in the past regarding the emission of volatile radionuclides in the context of evaporation from hot Na-pools in the reactor vessel to the inert cover gas region to determine the chemical and physical behavior of FPs under the wide range of conditions that can occur under accidental conditions. Castleman's studies (Castleman, 1970; Castleman et al., 1967; Castleman and Tang, 1968) about the chemical and physical aspects of FPs behavior in sodium was the mainstay of further research by developing theoretical description of these processes by thermodynamic principles. Their studies were focused on the essential data for predicting the FP behavior in sodium systems, namely their chemical potential in the liquid sodium, their volatility and extent to which they would be released during the evaporation of sodium and their chemical state upon release to the vapor phase. This work was continued by Clough (1971), Clough and Fraser (1973) and Clough and Wade (1971) with experimental and thermodynamic studies for the behavior of the main FPs with respect to both transport through liquid Na and vaporization from it into an inert atmosphere. In the 80's the German Kfk NALA program allowed the study of the delayed source term (Berthoud et al., 1988). On the basis of this program, the sodium retention capacity for fuel and FPs in terms of the retention factor, the release rate and releases mechanisms, sodium evaporation into an inert gas atmosphere, aerosol formation and properties and the aerosol behavior in the containment atmosphere were analyzed and compared with PARDISEKO code predictions (Bunz and Sauter, 1984; Sauter and Bunz, 1984; Sauter and Schütz, 1985, 1984, 1983, 1980; Schütz and Sauter, 1982).

Later on, Haga et al. (1992) experimentally determined the gas-liquid equilibrium partition coefficient of main volatile FPs (Cs, I, Te, among others) as a function of temperature between liquid sodium and the gas phase. Through this coefficient, the relative mass transfer rate of the fission products species compared to sodium was determined, being this the reciprocal of the retention factor (Lebel and Girault, 2018).

A mechanistic modeling of the evaporative release of volatile species from hot sodium pools into an inert atmosphere was developed by Brockmeier et al. (1994), Koch et al. (1993, 1991) and Starflinger et al. (1994) to be implemented in CONTAIN-LMR code (Murata et al., 1993) and validate it against the NALA experimental data. In this model, 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 Brockmeier et al. (1994), Koch et al. (1993; 1991), Kumada et al. (1976), Pradeep et al. (2016) and Starflinger et al. (1994) on FPs release from hot sodium pools has been conducted. Among the most important drawbacks of the models analyzed stand out: inconsistencies of the correlations derived for key properties, like NaI vapor pressure; and *as-hoc* fitting of the mass transport coefficients with the available data, which eventually impairs a true model validation. As an alternate, a model based on the film theory, the heat-mass transfer analogy and the Raoult law has been developed. To ease model-data comparisons, the results have been expressed in terms of mass fluxes and retention factors, as reported in the open literature. The data used for such comparisons come from the NALA program (Sauter and Schütz, 1983, 1980).

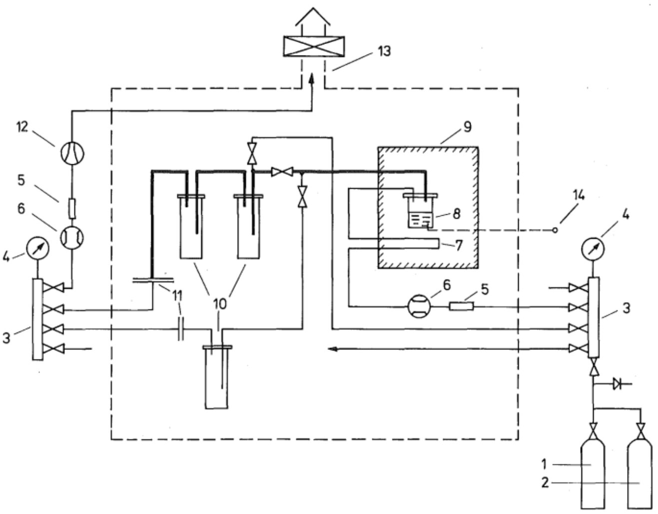
2. NALA program

The NALA program is a German experimental program (KFK) that aimed to study fuel and FPs release at various sodium pool temperatures, including boiling, and to observe sodium aerosol behavior in both closed and open vessels under different geometric and convective conditions. The tests were conducted in an inert atmosphere using either argon or nitrogen. The program included 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 I and NALA II phases and in those experiments involving the most important volatile FPs in terms of volatility from liquid hot sodium, it means, NaI and Cs as stated by Castleman et al. (1967) and Castleman and Tang (1968).

2.1. NALA I

Two experimental setups were used in the NALA I series: a closed test vessel in an oven and an open test vessel in a glovebox (Sauter and Schütz, 1983, 1980; Schütz, 1980; Schütz and Sauter, 1982). In this work only the experiments in a closed vessel in an oven will be analyzed. In these experiments, the closed SS vessel (7 cm diameter, 10 cm height) with the contaminated Na is placed in an oven at pool temperature between 700 and 1160 K at isothermal conditions into cold traps (Fig. 1). The sodium was contaminated with UO_2 powder of various mean particle sizes, UO_2 pellets, metallic Cs, NaI powder and SrO powder. The released aerosols were transported by a variable inert gas stream at isothermal conditions into cold traps and a filter. After washing and titration, the Na release rate and the fuel and FP traces were determined.



1. Argon storage bottle
2. Helium storage bottle
3. Valve block
4. Pressure gauges
5. Flow meters
6. Gas meter
7. Preheating arc
8. Test vessel
9. Oven
10. Cold trap
11. Filters
12. Pump
13. Exhaust system with filter
14. Thermocouple
15. Power supply unit for electric vessel heating
16. Nitrogen storage bottle
17. Suction funnel
18. Glovebox

Fig. 1. Structure of the experiments with closed test vessel in an oven (Schütz, 1980).

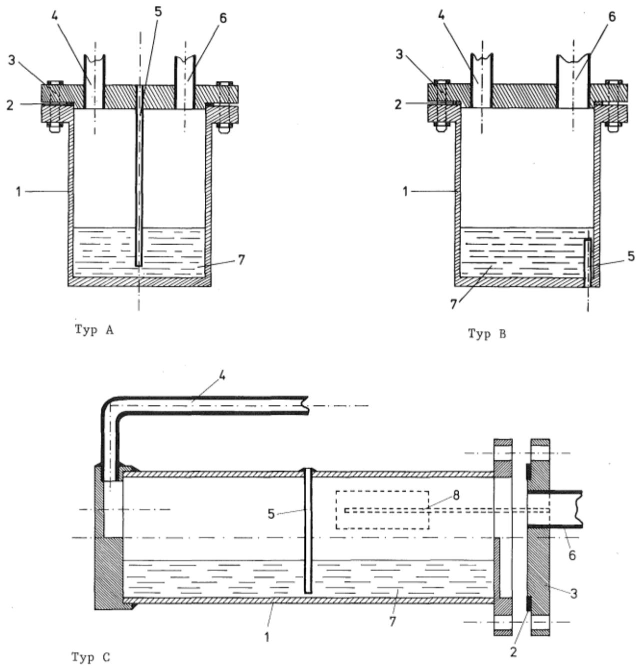
These experiments typically proceeded according to the following scheme: In a glove box, a certain amount of sodium (approximately 10 g) was weighed and filled into a test tube together with the substance to be examined. Thereafter, the vessel was installed in the experimental apparatus and heated in an oven to the desired temperature. Temperatures between 700 and 1160 K were achieved. After reaching the target temperature, the vaporizing or released by other mechanisms quantities were transported by an inert gas stream (forced convection). The inert gas (Ar) flowed through a preheating arc to the test vessel in such a way that sodium and carrier gas formed a nearly isothermal system. The released amounts were transported by the inert gas stream (forced convection) through heated pipelines into two cold traps (cooling with N_2 liquid) and a final filter (pore size 0.2 μm). Behind the filter was a feed pump with regulating valves. In order to avoid background effects, the inert gas flow during the heating and cooling phase was passed through a bypass line, which was also equipped with a cold trap and filter. Flowmeters and gas meters were used to measure the gas volume flow (at room temperature) and the amount of gas before the test vessel and after the filter. The vessel was completely closed, with carrier gas inlet and outlet perpendicular to the sodium surface and with direct contact between the gas and the pool surface. The net test duration was such that ± 10-20 % of the sodium inventory was released. By controlling the oven by hand, the set point temperature could be kept constant at ± 3%.

Three different test vessel types were used in these tests (Fig. 2):

- Type A: Cylindrical SS pot, 7 cm diameter, 10 cm height, tube for holding the thermocouple attached to the lid, inlet and outlet tubes of the same diameter, vertical arrangement. Most of the experiments were carried out with vessels of this type.

- Type B: Similar to A), but with thermocouple tube in the bottom and enlarged outflow tube. This vessel was used to additionally determine the fuel or FP concentration as a function of the height in the pool.

- Type C: SS tube, 6 cm diameter, 20 cm length. Thermocouples for the determination of the pool and gas temperature, horizontal arrangement. In this type of vessel, the flow conditions of the inert gas are better defined than in A) or B). The deflection of the gas flow at the surface is eliminated. The gas temperature can be measured.



1. Cylindrical SS pot
2. Cu sealing ring
3. Lids
4. Inlet pipe for inert gas
5. Tube for thermocouple (Na *T*)
6. Exhaust pipe for inert gas
7. Sodium
8. Tube for thermocouple (gas *T*)

Figure 2. Test vessel types for NALA I experiments on a laboratory scale (Schütz, 1980).

2.2. NALA II

The purpose of the NALA II phase was to demonstrate that the NALA I results could be scaled up so that representative results concerning sodium aerosol system under natural convection would be obtained and translated in terms of *RF*s (Sauter and Schütz, 1984).

The main components of the NALA II facility are a 2.2 m^3 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. 3).

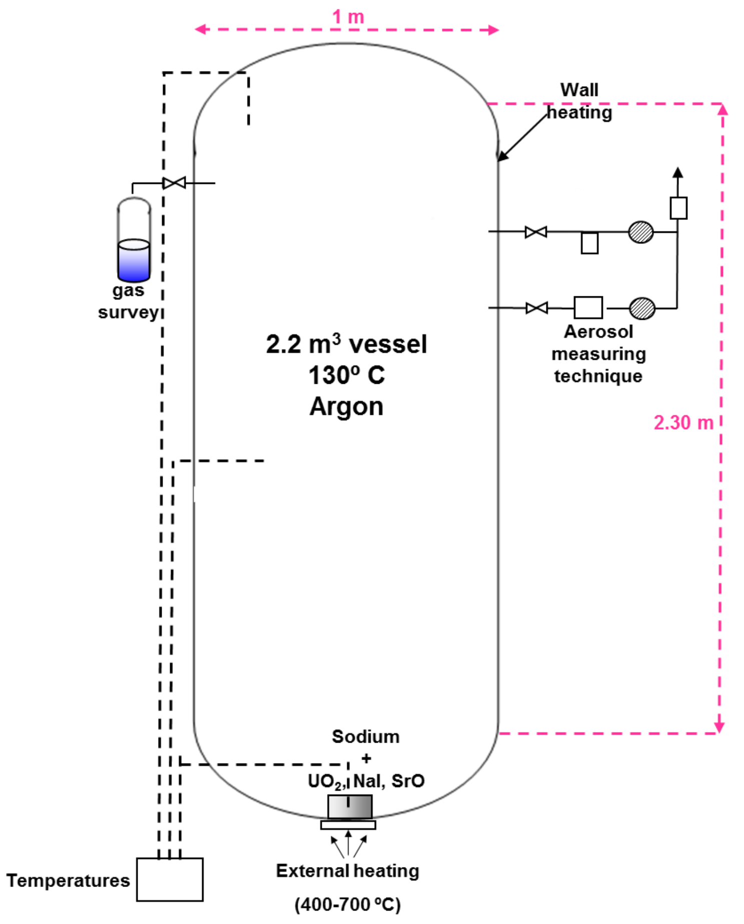


Fig. 3. Diagram of the NALA-II facility (Sauter and Schütz, 1980).

Boundary conditions in the experiments are defined as follow: pool temperatures from 600 to 900 K, inert gas temperatures of 400 K, natural convection and admixtures of UO_2, NaI and SrO in the pool (Sauter and Schütz, 1983, 1980). The vessel is filled with argon at normal pressure (1 bar), heated up to 400 K and kept constant during the whole experiment. In the sodium pot, sodium amounts of the order of 1 kg are heated up to the desired pool temperature. By flame regulation, a temperature plateau is approximately achieved during the tests. The duration of the external heating (0.7 to 3 h) is adjusted to the evaporation rate in such a way that ~10% of the sodium 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.

3. Models critical review

Fission products emission from hot sodium pools was analyzed and compared against experimental data from NALA II program by Brockmeier et al. (1994), Sauter and Schütz (1980), Starflinger et al. (1994) and later by Pradeep et al. (2016). All these studies are based on modelling the FPs transpot by evaporation as governed by diffusive and convective transport processes. Through a mass transfer coefficient approach based on the film theory by Nerst (1904), 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 (Starflinger et al., 1994).

To measure the fission product release, the main variable reported in NALA program (Brockmeier et al., 1994; Sauter and Schütz, 1983, 1980) is the Retention Factor (*RF*), defined as the ratio between the relative transfer rates of Na and FP (relative with respect to their initial inventories in the pool):

$$RF = \frac{\left[{m_i}/{m_{solvent}} \right]_{pool}}{\left[{\phi_i}/{\phi_{solvent}} \right]_{released}} \tag{1}$$

Where *m* denotes the initial inventory of the FP (i, fission product solute) and solvent (liquid Na) and ϕ the release mass fluxes.

The *RF*s of NaI in the sodium pool (black dots) by Starflinger et al. (1994) approach are shown in the next figure (Fig. 4). In this approach (red line), the gas mass transfer coefficient for sodium vapor has been fitted to the NALA database through an adjusted Sherwood number (Eq. 3.10 from Starflinger et al. (1994)). 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 also shown (green line). Even by using the same fitted Sherwood number and Na and NaI properties, these estimations are far from Starflinger et al. (1994) ones.

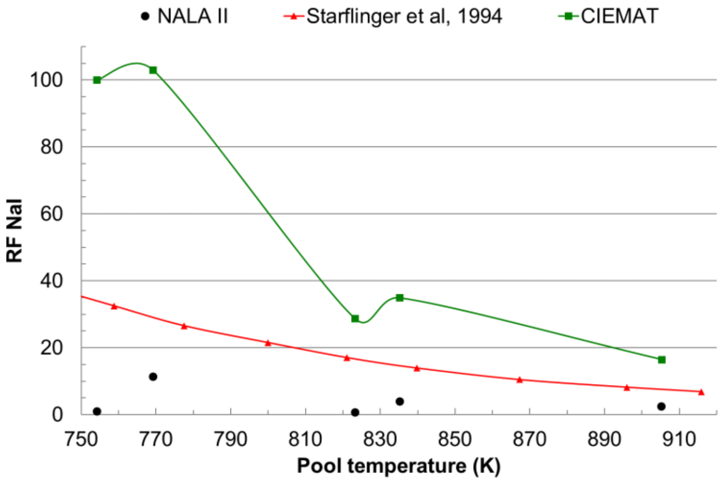


Fig. 4. *RF* of NaI in Na vs. pool temperature.

After analyzing the variables included in the model by Starflinger, an incorrect formulation of the NaI vapor pressure (p_v) correlation given by Starflinger et al. (1994) (Eq. 4.29) has been found to be the reason of the higher values predicted by CIEMAT´s reviewing. Therefore, a new correlation based on the experimental data reported by Cogin and Kimball (1948) is proposed (Eq. 3):

$$\left[p_{v,NaI}\right]_{Starflinger} = 10^5 \cdot 10^{\left[7.557 - \frac{10924}{T}\right]} \tag{2}$$

$$\left[p_{v,NaI}\right]_{CIEMAT}=133.322\cdot 10^{\left[11.681-\frac{11028}{T}\right]} \tag{3}$$

As can be seen in the next figure (Fig. 5), while the correlation by Starflinger gives vapor pressure values up to one order below the experimental data, good agreement can be found with CIEMAT's correlation.

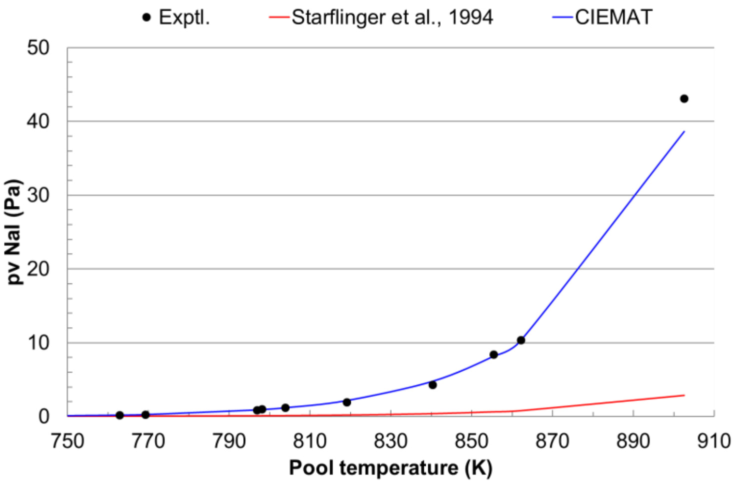


Fig. 5. NaI vapor pressure vs. pool temperature.

4. Extended Gas Mass Transfer (EGMT) model

In the analyzed scenario, FPs dissolved in the hot sodium pool diffuse away from the liquid surface at the same time as sodium vaporization takes place enhancing the FPs release. Based on this, the Extended Gas Mass Transfer (EGMT) model is based on the film theory (Green and Perry, 2008) and the dragging effect of the vaporized Na on the volatile species release is taken into account. Film theory is based on a simple conceptualization of the gas-liquid transfer processes. It describes a scenario where there is a hypothetical stagnant film near the interface, with no movement in the bulk liquid. Transport is primarily governed by molecular diffusion; at the interface, it is assumed thermodynamic equilibrium between the liquid and gas phases based on mass-transfer principles (the equilibrium is reach almost immediately when a gas and a liquid are brought into contact, Green and Perry (2008)).

The application of the film theory in the gas phase for the calculation of the release of mass flux of volatile FPs in the scenario analyzed is supported by estimations of the gas/liquid mass transfer coefficients (mass transfer coefficient in the gas phase is up to four orders of magnitude higher than the liquid mass transfer coefficient in the analyzed scenarios). Therefore, the release mass flux (ϕ) of volatile FPs (i) from the liquid phase is calculated through the following equation. In this equation, the release mass flux 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 (first term of equation). In the second term of the equation, the entrainment of FPs to the gas phase by the vaporized Na is taken into account (Herranz et al., 2009):

$$\phi_i = k_{\mathrm{i,g}} \cdot \Delta\rho_{\mathrm{i,g}} + \frac{\phi_{\mathrm{Na}}}{\rho_{\mathrm{sat,Na}}(T_{\mathrm{pool}})}\rho_{\mathrm{i,gif}} \tag{4}$$

The mass transfer coefficient ($k_{\mathrm{i,g}}$) (Eq. 5) 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:

$$k_{i,g} = Sh_{i,g} \cdot \frac{D_{i,g}}{L} \tag{5}$$

Where NaI binary diffusion coefficient in Argon ($D_{\mathrm{NaI,Ar}}$) has been taken from Tashiro and Sagawa (2001), and Cs binary diffusion coefficient in Argon ($D_{\mathrm{Cs,Ar}}$) from Aref'ev et al. (1982). L , the characteristic length of the system, is taken as a quarter of the Na pool diameter ($d/4$) as recommended by Incropera and Dewitt (2011).

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) (Incropera and Dewitt, 2011):

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

$$Sh = 0.332 \cdot \mathrm{Re}^{1/2} \cdot Sc^{1/3} \quad (forced\,convection, \mathrm{Pr} \ge 0.6) \tag{7}$$

The partial density difference in the gas phase between the gas interface (gif) and the gas bulk (gb) is calculated through:

$$\Delta\rho_{i,g} = \rho_{i,gif} - \rho_{i,gb} \tag{8}$$

Where $\rho_{\mathrm{i,gif}}$ is estimated from the saturation vapor pressure of the chemical species, which is just correlated with pool temperature (Green and Perry, 2008). This implicitly entails that the potential interaction between FPs (solute) and Na (solvent) is considered negligible. This is true on the limiting concentration region of ideal-dilute solution (10^{-5} mole fraction, Castleman et al. (1967)) which is the case of interest since the total quantity of FPs dissolved in the sodium coolant is in this order of magnitude (Castleman et al., 1967; Castleman and Tang, 1968). On these assumptions, $\rho_{\mathrm{i,gif}}$ is calculated through the Henry's law as:

$$\rho_{i,gif} = \frac{p_{v,i}(T_{if})}{(R/M_i)\cdot T_{if}} \cdot \gamma_i \cdot x_i \tag{9}$$

with $p_{v,\mathrm{NaI}}$ calculated from Eq. 3 and $p_{v,\mathrm{Cs}}$ from Starflinger et al. (1994) , x_i denoting the mole fraction of the species i in the liquid, and γ_i the activity coefficient of the species i.

The activity coefficients of NaI and Cs (γ_{NaI} and γ_{Cs} respectively) are calculated through the excess Gibbs free energy of mixing at an infinite solution (Castleman et al., 1967). When calculating the activity coeficients for NaI, significant differences have been observed between the values obtained using different methods found in the open literature. The activity coefficients calculated using Haga equation (Haga et al., 1992) are approximately twice as high as those using the Castleman method (Castleman et al., 1967). In EGMT model, the Castleman method was chosen for reasons of conservatism. The resulting values, larger than 100, indicate that NaI in sodium is more prone to scape than pure NaI (Haga et al., 1992). All gas phase properties involved in the above equations are evaluated at the pool temperature ($T_{\mathrm{if}} \approx T_{\mathrm{pool}}$). Note that $\rho_{\mathrm{i,gb}}$ grows along time due to the incoming NaI flow from the pool over time (time evolution is stopped when mass flow rate reaches 10% of its initial value to prevent mistakes in the total mass flow rate value calculated as an average during the time interval).

For the entrainment term, $\rho_{\mathrm{sat,\,Na}}$ is the density of sodium vapor above the saturated liquid, calculated from the thermodynamic relation (Fink and Leibowitz, 1995):

$$\rho_{\mathrm{sat,Na}} = \left(\frac{\Delta H_g}{T \cdot \beta_{sat}} + \frac{1}{\rho_l} \right)^{-1} \tag{10}$$

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.

The mass flux of vaporized sodium (ϕ_{Na}) is also calculated through the film theory:

$$\phi_{\mathrm{Na}} = k_{\mathrm{Na,g}} \cdot \Delta\rho_{Na,g} \cdot f_{cond,Na} \tag{11}$$

Where the gas mass transfer coefficient ($k_{Na,g}$) is calculated with equation 5 and the partial density difference in the gas phase between the gas interface and the gas bulk ($\Delta\rho_{Na,b}$) through equation 7. Na binary diffusion coefficient in Argon ($D_{Na,Ar}$) is taken from (Pradeep et al., 2012), $p_{v,Na}$ from (Fink and Leibowitz, 1995) and due to the small amount of FPs solved in the sodium pool, the Raoult´s law is applicable ($x_{Na} = \gamma_{Na} = 1$) (Kumada et al., 1976; Pradeep et al., 2016; Starflinger et al., 1994)).

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. 11. This correction factor, applicable when pool and gas temperatures differ (non-isothermal conditions in NALA II tests) is calculated by the following equation (Clement and Hawtin, 1977; Hills and Szekely, 1969; Rosner, 1967):

$$f_{\text{cond,Na}} = \frac{\beta_{Na}(T_{if})\cdot\left(\Delta T\cdot Le^{-n}+\left(\Delta h_{v,Na}\cdot\Delta p\right)/\left(Le\cdot c_{p,gb}\cdot P\right)\right)}{\Delta p\cdot\left(1+\left(\beta_{Na}(T_{if})\cdot\Delta h_{v,Na}\right)/\left(Le\cdot c_{p,gb}\cdot P\right)\right)} \quad (12)$$

Where β_{Na} corresponds to the first derivative of the vapor pressure $p_{\mathrm{v,Na}}$ 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, $\Delta h_{\mathrm{v,Na}}$ the latent heat of vaporization of sodium and *Le* is the dimensionless Lewis number (ratio between the Schmidt and Prandtl numbers).

5. Model validation

5.1. Experimental tests conditions

In NALA I tests, the closed SS vessel (7 cm diameter, 10 cm height) with the contaminated Na is placed in an oven at pool temperature between 700 and 1160 K. The released aerosols were transported by a variable inert gas stream (forced convection) at isothermal conditions into cold traps and a filter. A compilation of the main tests conditions are shown in Table 1 (Schütz, 1980).

In the case of NALA II tests, the boundary conditions were defined according to the SNR 300 core catcher design characteristics: pool temperatures from 600 to 900 K, inert gas (Ar) temperatures of 400 K, natural convection and admixtures of UO_2, NaI and SrO in the pool (Table 1) (Minges et al., 1992; Sauter and Schütz, 1983, 1980).

Table 1. Compilation of some of the available characteristics of NALA I and II facilities.

	NALA I	**NALA II**
Geometry		
Na pool area (m^2)	3.85×10^{-3}	5.31×10^{-2}
Vessel volume (m^3)	3.85×10^{-4}	2.2
Vessel height (m)	0.1	2.30 (cylindrical part)
Tests conditions		
Na amount (kg)	0.250-1.320	~ 1
Na *T* (K)	700-1160	673-973
Atm. gas (*T*, K)	Ar (Pool *T*)	Ar (403)
v_{flow} (l/min)*	1-11	-
Contaminants	UO_2, Cs, NaI, SrO	UO_2, NaI, SrO

* Gas volume flow of the transport gas through the test vessel, measured at room temperature.

Table 2 compiles the experimental data for the tests of interest in NALA I and NALA II phases. In this table, the test number and FP tested is indicated, *T*(K) is the pool temperature, t(min) is the net measuring time at set point temperature without heating and cooling, *v*(l/min) indicates the gas volume flow of the transport gas through the test vessel, measured at room temperature (applies to NALA I only). The last two columns show the quantities weighed of Na and FPs before the experiment and added to the test vessel. 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.

The experimental setup in the analyzed NALA I tests is one with vessel type A (see section 2.1). As it can be seen from the table, T5 and T7 in NALA II phase are exclusively devoted to the Na vaporization estimates.

Table 2. Tests conditions (Minges et al., 1992; Sauter and Schütz, 1983, 1980; Schütz, 1980).

	Exp. Nr.	**FP**	***T* (K)**	***t* (min)**	***v* (l/min)**	**Na amount (g)**	**FP amount (g)**
NALA I	T1	Cs	710	90	10.0	101	0.50
	T2	Cs	803	60	5.0	106	0.50

	T3	Cs	898	17	10.0	102	0.50
	T4	Cs	996	11	8.5	104	0.50
	T5	Cs	1087	8	4.0	99	0.50
	T1	NaI	795	60	10.0	98	1.67
	T2	NaI	876	20	9.0	52	1.04
	T3	NaI	998	12	10.0	98	1.43
	T4	NaI	1059	3	10.0	100	1.16
	T5	NaI	1111	5	1.0	101	1.57
NALA II	T1	NaI	835	45	-	258	1.00
	T2	NaI	905	43	-	1100	1.06
	T3	NaI	754	170	-	1315	1.00
	T4	NaI	769	175	-	1322	10.00
	T5	-	837	101	-	1146	-
	T6	NaI	823	170	-	1321	0.20
	T7	-	830	153	-	1007	-

5.2. Sodium evaporation

Sodium evaporation modeling is an essential element when modeling FPs transfer from hot pools to the gas phase over liquid surface. In the next figure (Fig. 6), a comparison between experimental data of free/forced convection controlled Na release from a liquid pool into argon gas atmosphere (NALA I and NALA II experiments) and the calculated predictions is presented: predictions by (Starflinger et al., 1994) (red line) for NALA II temperature range and EGMT model estimations (blue dots). At first glance quantitative and qualitative good agreement is obtained by estimations from Starflinger et al. (1994) 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. A subestimation of an order of magnitude can be observed when looking at the EGMT estimations. It is worth mentioning the low Na mass flux of evaporation predicted in the NaI test with pool temperature of 1111 K. The reason behind would be the low gas flow velocity through the test vessel although this is not reflected in the experimental value.

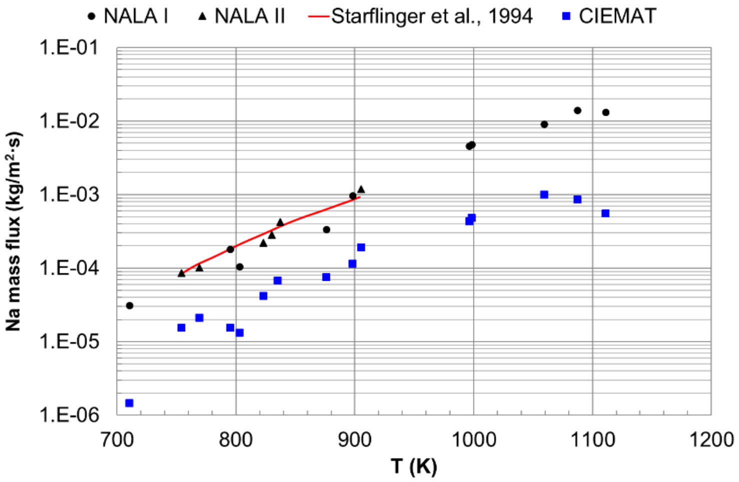
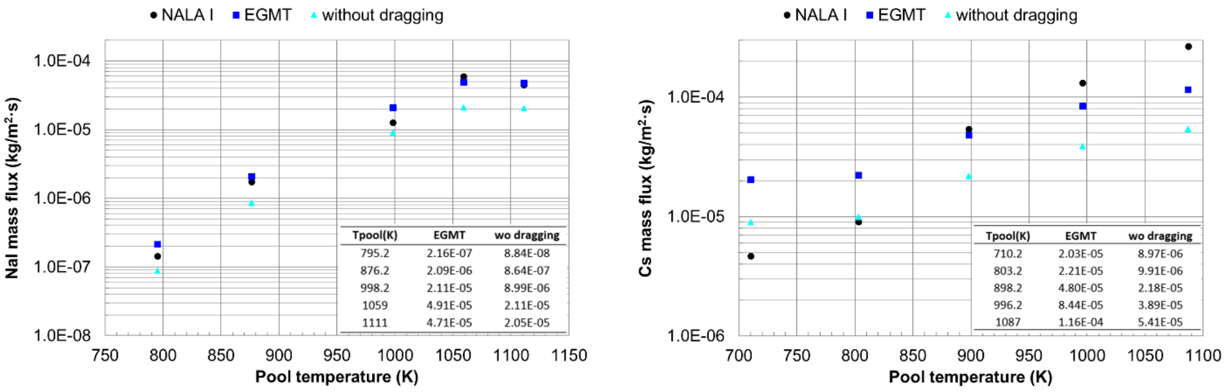


Fig. 6. Na mass flux of evaporation vs. pool temperature.

5.3. FPs release

Fig. 7 illustrates the NaI and Cs vaporization rates given by EGMT approach against experimental data from NALA I. A sensitivity calculation of the Na vapor dragging effect on the NaI release indicates that as expected, this phenomena results in an enhancement of the FPs mass flux by a factor of 2, i.e., the weight of each phenomena is similar. Quantitative and qualitative good predictions are obtained with EGMT model for both FPs; in both cases, EGMT model predictions correctly capture the exponential growth of the experimental mass flux with pool temperature. The release increase due to the change of the species thermodynamic state from the boiling temperature (933 K and 950 K, for NaI and Cs respectively) on is taken in both cases also. Nevertheless, more investigations concerning the partial pressures and activity coefficients of FPs of interest would be advisable. Finally, it is worth noting the higher release rate of Cs compared to that of NaI, up to 2 orders of magnitude at the lower temperatures, which points out the high volatility of Cs in liquid sodium.



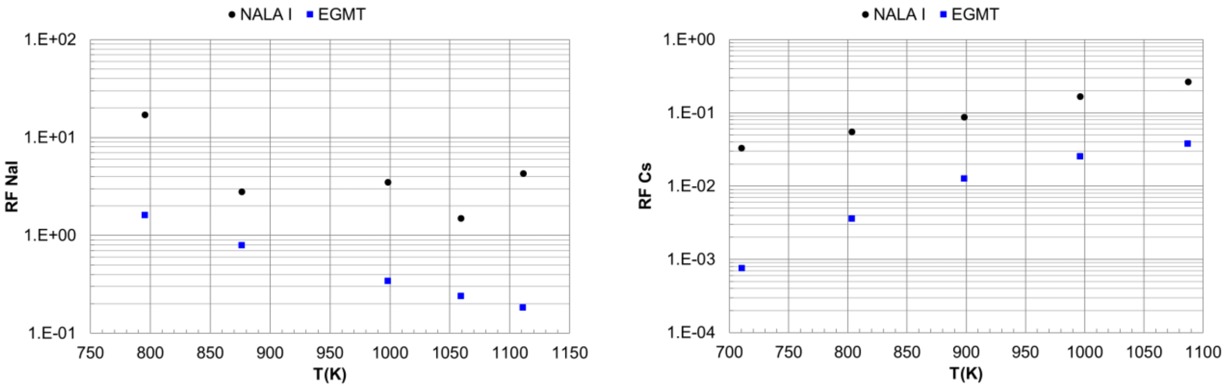
Tpool(K)	EGMT	wo dragging
795.2	2.16E-07	8.84E-08
876.2	2.09E-06	8.64E-07
998.2	2.11E-05	8.99E-06
1059	4.91E-05	2.11E-05
1111	4.71E-05	2.05E-05

Tpool(K)	EGMT	wo dragging
710.2	2.03E-05	8.97E-06
803.2	2.21E-05	9.91E-06
898.2	4.80E-05	2.18E-05
996.2	8.44E-05	3.89E-05
1087	1.16E-04	5.41E-05

a) NaI. b) Cs.

Fig. 7. FPs mass flux vs. pool temperature.

Figure 8 shows the models-to-data comparison in terms of the *RF* (NaI and Cs tests). For both species, an underestimation of the *RF* is observed as a consequence of the smaller sodium evaporation rates predicted by the model. According to the *RF* definition, *RF* is a measure of the fission product retention capability within the pool. In other words, the higher the *RF*, the less the amount of FPs leaving the Na pool and entering the gas phase. Namely, the inventory of fission products available for a postulated release to the environment from the gas atmosphere would be larger. This means, underprediction of *RF* has a conservative nature. In light of the release mass flux results, a higher pool retention capability is predicted for the NaI case.



a) NaI. b) Cs.

Fig. 8. *RF* vs. pool temperature.

The model-to-data comparison in terms of the *RF* for the NALA II tests is shown in the next figure (Fig. 9). As indicated by Starflinger et al. (1994), experimental data of the NaI release under varying conditions of pool temperature and initial inventories show a considerable scattering of the retention factor, possible due to the temperature history during the experiments.

EGMT 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.

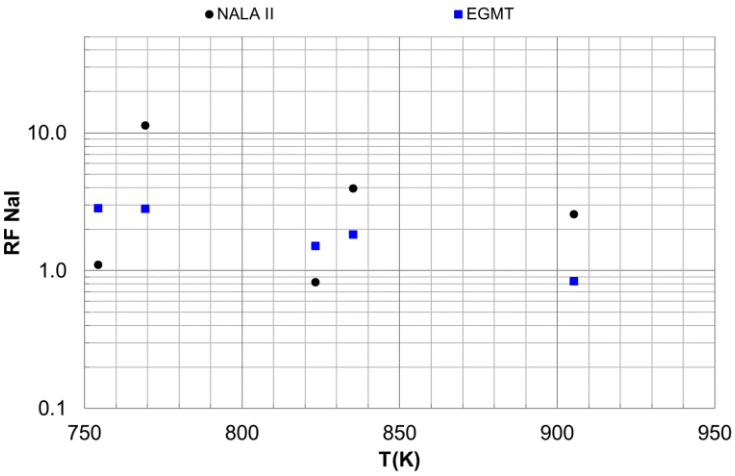


Fig. 9. *RF* vs. pool temperature.

6. Conclusions

This paper investigates the release of volatile fission products from hot sodium pools. A critical review of previous modeling works has led to the identification of their major shortcomings, one of the most important ones being their highly parametric nature. In order to avoid such a strong dependence on the ranges explored experimentally, a phenomenological approach has been adopted to develop a model that comprehensively accounts for the most significant phenomena governing the transfer of fission products (FPs) to the gas phase from hot sodium pools in a postulated accident scenario in a Sodium-cooled Fast Reactor. The results obtained and their comparison to the data available has been proved to enhance the predictive accuracy and, consequently, to provide insights into the FP transfer. The main conclusions can be summarized as follows:

- Some pioneering models based on the film theory included inconsistencies between their formulation and the estimates reported for some experimental scenarios addressed in this work.

- In some approaches authors used “fit-to-data” mass transport coefficients derived from the NALA experimental database and, consequently, got an excellent agreement in terms of Na vapor pressure. However, *RF* estimates,as reported in other researchers in previous investigations, largely overpredicted RF derived from measurements. No matter the goodness of the model, the use of an adjusted mass transfer correlation would potentially hinder the validation and further use of this approach.

- In a hot Na pool, fission products dissolved diffusion away from the pool liquid surface is enhanced by Na vaporization itself. The EGMT model is based on diffusive and convective mechanisms in the gas phase, the assumption of the analogy between HMT and the use of the Raoult’s law to set the NaI concentration at the Na pool interface. 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 release of volatile species is included through a term on the general mass flux equation. As a result, the EGMT model calculates the release in terms of mass fluxes and *RFs* in a wide range of conditions that can occur under accidental conditions in a SFR.

- Validation of the model by comparison with experimental data reveals that the gas mass transport coefficient is the most influencing factor. The formulation introduced in previous sections highlight that fitting it to data a vailable, as done in previous works, is not necessary, whenever the Sherwood non-dimensional number is properly calculated. The good agreement of the results with experimental data means a substantial enhancement of both qualitative and quantitative predictability, while maintaining a conservative approach. However, a more extensive experimental database would be needed both to support some properties used within the model and to perform a broader and sounder validation.

- Further progression in the modeling of FP transfer to the gas phase from hot Na pools would require fundamental data related to the physic-chemical interaction of FPs and liquid Na, which are barely found in the open literature, and a more extensive and better characterized experimental database to build a sound validation of enhanced models to come would be advisable.

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NOMENCLATURE

c_p	Specific heat capacity
D	Binary diffusion coefficient
d	Diameter
f_{cond}	Correction factor by condensation
H	Enthalpy of vaporization
h	Latent heat of vaporization
k	Gas mass transfer coefficient
L	Characteristic lenght
M	Molecular weight
m	Mass
n	Power coefficient at Sherwood number
P	Pressure
p_v	Vapor pressure
R	Ideal gas constant
T	Temperature
x	Mole fraction

Adimensional numbers

Gr	Grashof number
Le	Lewis number
Re	Reynolds number
Sc	Schmidt number
Sh	Sherwood number

Greek symbol

γ	Activity coefficient
ρ	Density
ϕ	Mass flux

Subscripts and superscripts

b	Bulk gas
g	Gas phase
gb	Gas bulk
i	Volatile FP species
if	Interface
l	Liquid phase
sat	Saturation condition

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