

## Review

## In-containment source term in accident conditions in sodium-cooled fast reactors: Data needs and model capabilities

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## ABSTRACT

Sodium-cooled fast reactors (SFRs) are one out of the six technologies considered in the so-called Generation IV initiative. A full-scope safety analysis of this reactor type would need to have computation tools developed and properly validated. The present paper focuses on the review of currently available data and modeling capabilities for in-containment source term analysis in accident conditions. Generally speaking, it has been found that improvements to characterizing particles (i.e., density and shape) and particle–particle interaction processes are required. However, beyond any doubt, two of the strongest modeling needs for SFR accidents are: aerosol generation which, in turn, means to properly model sodium vaporization, chemical reactions with the surrounding gas, nucleation of combustion products and primary particle agglomeration; and fission products partitioning. Current LWR integral codes, although not yet furnished with such models, look promising as a computational platform to be extended to the SFR domain. In any case, once they are adapted to anticipated SFR scenarios, extensive validation should be undertaken against a comprehensive and sound database.

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## 1. Introduction

In the framework of the Generation IV initiative (<http://www.gen-4.org/>), the sodium-cooled fast reactor (SFR) is one of six technologies that have been identified for development as a next-generation nuclear-energy system. At present, seven organizations - viz. CEA (France), DOE (USA), JAEA (Japan), JRC (Euratom), MEST (Korea), MOST (China) and ROSATOM (Russian Federation) - have signed the corresponding Gen IV System Arrangement. In addition, a further international project on SFR research and development started at the beginning of 2009 with partial funding from the EC 7th Framework Programme (Fiorini, 2009). This project encompasses a comprehensive range of actions including the subject of this paper.

Worldwide more than twenty experimental or power reactors have been built and operated where there exist two main concepts for SFRs (see, e.g., IAEA, 1999): pool-type designs such as PFR (UK), SuperPhénix (France) and BN-600 (Russia); and loop-type designs such as Rapsodie (France), FFTF (USA) and Monju (Japan).

During hypothetical, i.e., very low probability, SFR accidents leading to severe core damage, one can envisage highly-energetic

phenomena in the vessel leading to ejection of sodium mixed with fuel, fission products (FPs), steel and cover gas from the primary system into the containment. Indeed, despite their improbability, it is a requirement of level 2 probabilistic safety assessments that such events are analyzed and, ultimately, an associated source term evaluated (i.e., proving, above all, significant mitigation with respect to public health and safety). The present paper focuses on the review of currently available data and modeling capabilities of in-containment source term analysis in accident conditions, i.e., excluding any consideration of phenomena upstream in the vessel and downstream in the outside environment. In this way, the scope of such a review has a generic nature in the sense that it does not address any specific reactor configuration (i.e., pool/loop architecture) and/or specific accident scenarios such as unprotected loss of flow (ULOF), unprotected loss of heat sink (ULHS), loss of coolant (LOCA), etc.

Finally, it is worth noting that in the past the potential radiological risk arising from core degradation has always been the focus of safety evaluations. In fact, the consequences due solely to the primary sodium are significant. Firstly, the primary sodium is significantly activated during its successive passages through the core; the interested reader can find details in IAEA (1993) where, here, we cite the maximum <sup>24</sup>Na activity of 0.75 TBq per kg of primary sodium for the BN-600 reactor. This implies that a substantial release of primary sodium would constitute an

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elevated source of  $\beta^-$  and  $\gamma$  activity for many days. Secondly, the sodium represents a serious chemical hazard in all its forms in the containment (Na, Na<sub>2</sub>O, NaOH, etc. – see below) essentially until it forms - if it can - the carbonate or bicarbonate. Hence, prediction of the evolution of the physico-chemical forms of any released sodium is an important safety issue.

## 2. Major sources of in-containment aerosols

Under accident conditions, aerosols form essentially by two processes (Schikarski, 1976):

- The vaporization of core materials and subsequent re-condensation.
- The coolant vaporization (trace materials included) and subsequent re-condensation (Na aerosols) and/or chemical reaction (e.g., Na<sub>2</sub>O/NaOH/Na<sub>2</sub>CO<sub>3</sub> aerosols). It is worth noting the large amount of sodium inventory in an SFR primary system (about 2000 tonnes in a ~1500 MWe pool-type SFR, according to IAEA, 2006).

The significance of these aerosol sources will depend on the course of the accident, the materials involved and the lay-out of the containment system. Furthermore, these aerosol sources may combine and form a sort of “mixed aerosol source”, the composition of which can be variable depending on the prevailing conditions and, as a result, the in-containment particles might well be of heterogenous composition with different densities and sizes. In the case of Na aerosols, their importance lays in their abundance and nature as both a radioactive source and a radioactive species carrier, and the potential harm associated with some of the chemical species (i.e., the effects of exposure to sodium oxide and hydroxide aerosols, depending on the level of exposure, can vary from mild irritation to burns, pneumonitis and serious damage to the upper respiratory tract). In addition, the total amount of Na aerosols in the containment strongly affects the source term evolution within containment (Fermadjian, 1985).

### 2.1. Vaporization of core materials

As discussed by Clough and Fraser (1973), a key aspect for the accident evolution would be the potential segregation of fission products from sodium in the process of aerosol formation. If the size of fission product-bearing particles was significantly different from that of Na particles (either much bigger or much smaller), the aerosols populations would evolve in quite different ways since their governing mechanisms will be different. Contrarily, if fission products were uniformly associated with Na (and remained so), fission products and sodium would both evolve together and existing data on Na particles behavior would be applicable to the “mixed system”.

Several works were published in the 60s and 70s on the interaction of Na and fission products (e.g., Castleman, 1973). Their essence can be synthesized in a few points according to the different nature of important radionuclides:

- Noble gases, particularly krypton, have negligible solubility in Na; in terms of the Henry’s law, their constants are very low;
- Iodine remains in the form of NaI under anticipated conditions such as the presence of cesium and/or Na burning; in other words, NaI is so stable that it does not release any free iodine even in oxidizing conditions (i.e.,  $2\cdot\text{NaI} + 0.5\cdot\text{O}_2 \rightleftharpoons \text{Na}_2\text{O} + \text{I}_2$ ) if an excess of Na is available; studies of the NaI activity coefficient indicated that most of iodine would be retained in Na,

**Table 1**  
Characteristics of major radionuclides in Na solution.

	Element	Characteristics	State	Location
Noble Gases	Xe, Kr	High volatility, negligible solubility	Xe	Gas phase
Halogens	I	Volatile, soluble	NaI	Na (interface)/ Gas phase
Alkali Metals	Cs	High Volatility, soluble	Cs	Na (interface)/ Gas phase
Alkaline Earth Metals	Sr, Ba	Non-volatile	SrO, BaO	Na (suspended/ precipitated)
Soluble FPs	Te		Na <sub>2</sub> Te	Na (solved)
Solid FPs	Metals		MeO	Na (suspended/ precipitated)
	UO <sub>2</sub> , PuO <sub>2</sub>		MO <sub>2</sub>	
	MOX		Na <sub>3</sub> MO <sub>4</sub>	

with a large fraction concentrated near the gas–liquid interface;

- Cesium should be expected in its elemental state in liquid sodium rather than combined with other fission products and, although more homogeneously distributed than iodine, its concentration is somewhat higher at the gas–liquid interface too;
- Alkaline earth metals (Sr, Ba) seem to interact with dissolved oxygen to form relatively non-volatile species, i.e., remaining in the liquid coolant.

Most of the previous insights have been summarized in Table 1. These come from experimental observations though there have also been many thermodynamic analyses. However, given their importance and the fact that hardly any new studies have been published since the early 90s, further investigation would be welcome, particularly concerning cesium, strontium and tellurium.

The above statements give meaningful information for the Na-fission product interactions once these are well mixed. An additional study carried out by Clough and Fraser (1973) measured to what extent fission product vapors would be absorbed into condensing Na vapor. They found that, whereas iodine was mostly absorbed (99.5%), up to 40% of cesium could be still found in the gas phase. In other words, cesium under specific conditions could be responsible for a substantial contribution of radioactivity in the vapor phase.

As for the fission product release from a Na-metal solution, Castleman (1973) delivered an expression for the rate assuming that release is a gas diffusion-controlled process. The ratio of Fick’s law for each component of the solution (Na and fission product) leads to,

$$F_2 = 1 - (1 - F_1)^{A\phi} \quad (1)$$

Being  $F_1$  the fraction of sodium vaporized and  $F_2$  the fraction of fission product vaporized. The constant A can be calculated from the vapor pressures and the appropriate thermodynamic data for any particular fission product and  $\phi$  is the ratio of the gas phase diffusivities. By considering  $\phi$  equal to unity, the equation reduces to the Rayleigh equation for equilibrium vaporization. In fission product-sodium systems, the diffusivity ratio is always less than unity (since gas phase diffusivities are inversely proportional to the square root of the reduced mass of the system), so the Rayleigh equation may be seen as the upper limit of the fractional fission product release as a function of the fraction of sodium vaporized. By conducting simple hand calculations assuming dilute solutions, one can note the enormous difference between the alkaline metals (i.e., Cs and Rb) and the rest of fission products (Fig. 1). As noted, the tendency for alkaline metals to vaporize is outstanding; it is seen that at lower temperatures the volatility of these metals relative to

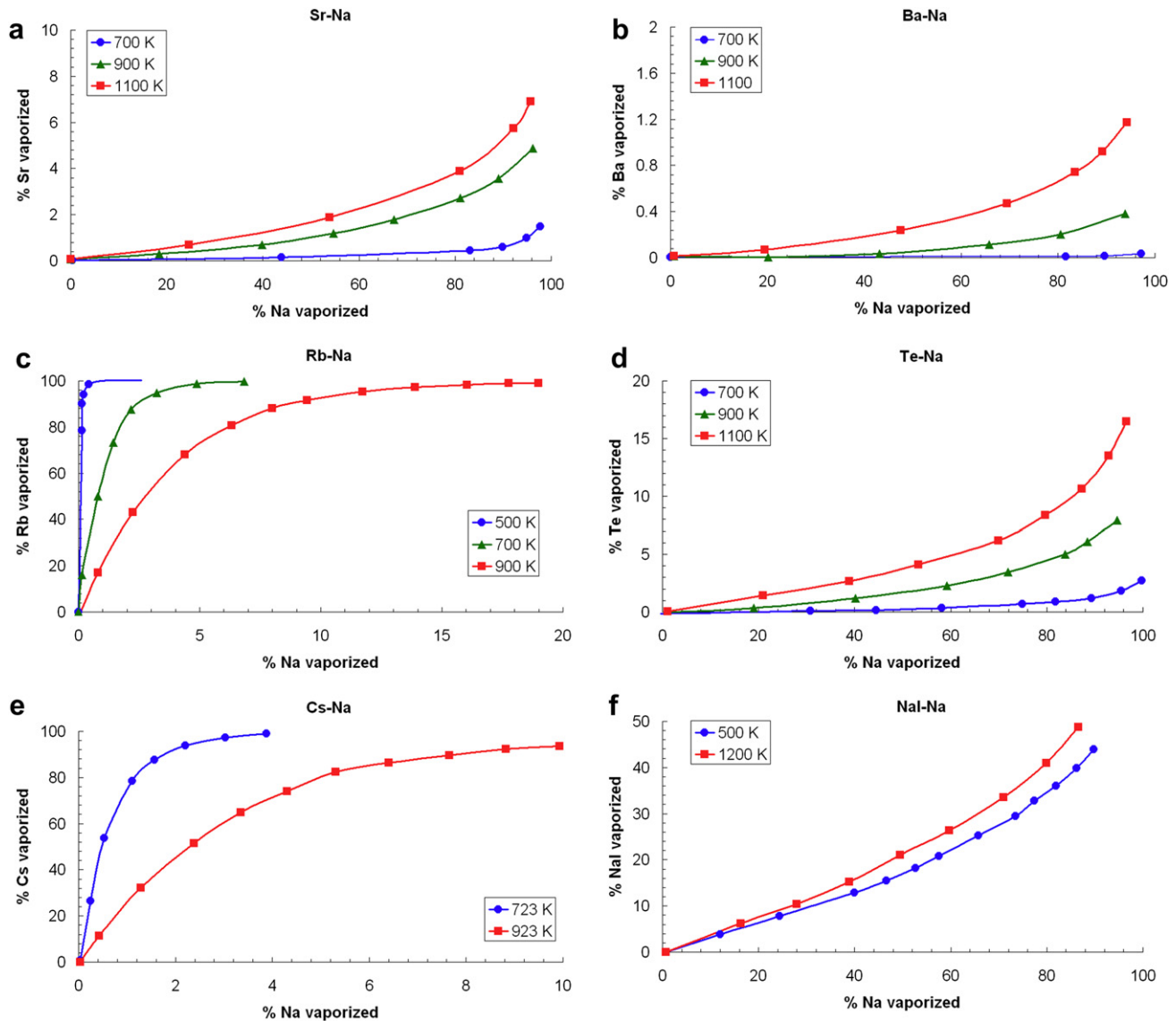


Fig. 1. Fission products–Sodium equilibrium vaporization.

that of sodium increases. Contrarily, alkaline earth elements (i.e., Sr, Ba) and Te hardly vaporize (less than 5%, even at high temperatures); in this case, the higher the temperature, the higher the vaporized fraction. This vaporization trend is more pronounced, but still moderate in the case of NaI.

Models for evaporation of sodium in conditions of forced or natural convection in the gas phase have been developed by various authors (Koch et al., 1991, 1992; Brockmeier et al., 1994) and validated against NALA experiments performed at Karlsruhe.

In addition to the tendency to release fission products during Na vaporization, sodium combustion has also been addressed experimentally as a potential release mechanism. Kawahara (1976) determined that whereas Na and I releases from a crucible were similar and of a moderate magnitude (from around 10–20% between 400 and 600 °C), that of cesium reached values over 60% at 600 °C (Fig. 2). The Sr release hardly reached 0.5%. In addition, Kawahara et al. also investigated how conditions other than temperature, viz. O<sub>2</sub> concentration and humidity, affected the release; in both cases, the higher the concentration of these compounds, the lower the fraction released where pollution of the sodium surface by combustion products is proposed as the

hindering mechanism. Including data from Lauben with their own, Kawahara et al. derive an isotope release fraction vs. Na release fraction where it is observed that there is a near 1 to 1 ratio in the case of iodine and a much higher ratio in the case of cesium (Fig. 3).

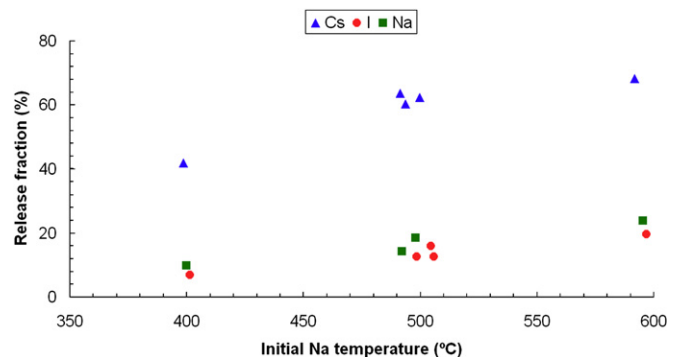


Fig. 2. Release fraction as a function of initial Na temperature.

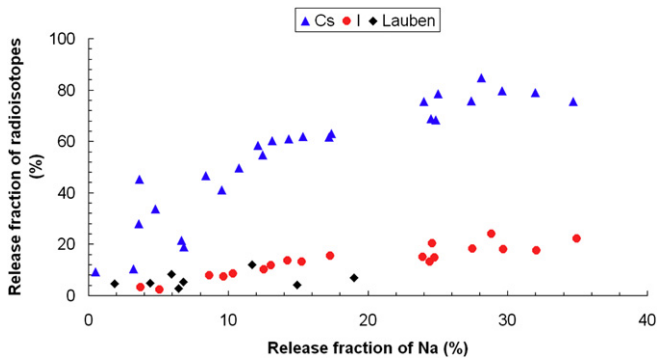


Fig. 3. Relation of release fractions between sodium and radioisotopes.

Adams et al. (1979), by conducting experiments with  $\text{Na}_2\text{O}$  and  $\text{U}_3\text{O}_8$  aerosols in the Nuclear Safety Pilot Plant (NSPP) facility, demonstrated that although individually they behaved differently due to their different properties, the two particle types co-agglomerated acting as a single aerosol of specific properties (see Section 4.2 below).

## 2.2. Sodium fires

The potential influence of sodium fires on source term analysis of SFRs takes place through its effects on:

- The release of fission products and aerosols;
- The production of particles carrying chemically-aggressive compounds (i.e.,  $\text{NaOH}$ );
- The change of in-containment boundary conditions (i.e., T, P, relative humidity, etc.).

The last two points are addressed in the next sections while the fission product aspect has been discussed above. Thus, below attention focuses on Na aerosol generation.

The phenomenology specifically related to different forms of sodium fires is beyond the scope of the present study. Here we simply indicate that sodium fires are traditionally classified in three types: pool fires, spray fires and columnar fires. A good number of researchers have addressed the related fire phenomenology for each of these types and the reader is referred to the associated literature (e.g., Cherdron et al., 1983; Malet et al., 1981; Morewitz et al., 1977, Morewitz, 1979; Newman, 1972, 1983; Newman and Payne, 1976, 1978).

Malet et al. (1981) showed that the laws of aerosol formation and growth as well as their production in fires were not settled at that time. Later on, theoretical research work based on an instantaneous chemical-equilibrium model (Doda et al., 2003; Yamaguchi and Tajima, 2009) obtained reasonable agreement with data in terms of aerosol release fraction (i.e., fraction of sodium mass transferred upward to the atmosphere with respect to the total burning mass). As these studies showed, the aerosol release fraction seems to be highly dependent on temperature, oxygen molar fraction and convection conditions over the Na flame (Fig. 4a, b). We note, therefore, that convection depends on pool dimensions (diameter).

## 2.3. Sodium-concrete interactions

In the case of a sodium leak in an SFR, the liquid metal may come into contact with the structural concrete (floor and/or walls). The sodium-concrete interaction involves thermal and chemical processes that result in the release of water vapor from the

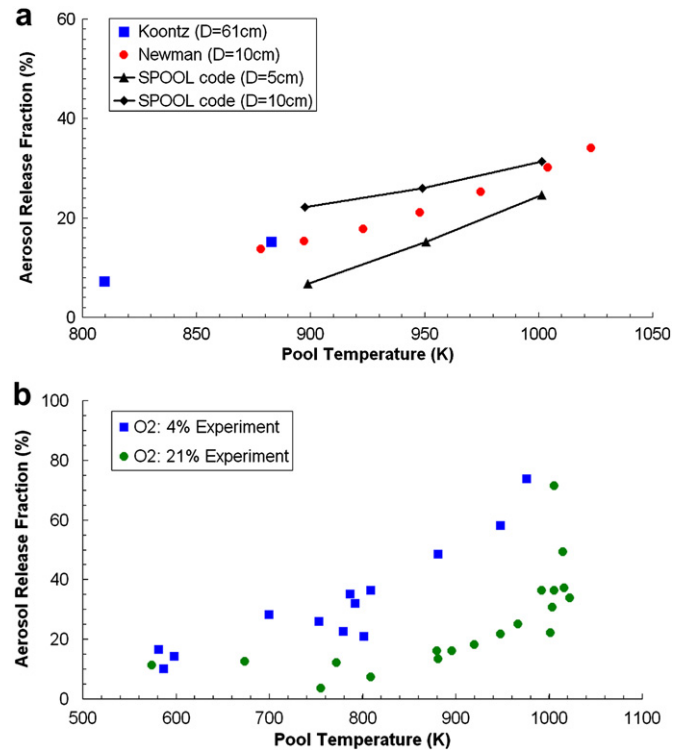


Fig. 4. Aerosol release fraction. a) Convection effect (Doda et al., 2003) and b) Oxygen effect (Yamaguchi and Tajima, 2009).

concrete, the generation of hydrogen from the sodium-water and/or sodium-concrete reactions, and, in the case of molten fuel in the sodium, the interaction of molten fuel with concrete. Four major effects of this phenomenon on the containment would be:

- The temperature and pressure build up including noncondensable gases ( $\text{CO}$ ,  $\text{CO}_2$  and  $\text{H}_2$ );
- The release of combustible gases ( $\text{CO}$  and  $\text{H}_2$ );
- The potential penetration of the concrete horizontal and vertical walls;
- The release of fission products and aerosols from the molten pool where this may include mechanical sparging.

A number of theoretical and experimental studies have addressed the scenario from the containment-loading perspective (Bae et al., 1998; Baker, 1977; Casselman, 1981; IAEA, 1996). Models of the ablation process and melt chemistry have been formulated and these can be used to provide boundary conditions for the calculation of radionuclide releases. Contrarily, no data on radionuclide release during such interactions have been found.

## 3. Aerosol characterization

The characterization and behavior of in-containment nuclear aerosols is of fundamental importance for assessing the radiological consequences of reactor accidents and for setting up filtering systems and even reactor components.

### 3.1. Chemical speciation

As said above, the main source of aerosols is sodium burning. A number of experimental studies have been conducted to physically and chemically characterize Na compounds in aerosols. Some of the



most outstanding facilities involved in this investigation are: CSTF in U.S. (Hilliard et al., 1985; Postma and Owen, 1980; Souto et al., 1994), FAUNA in Germany (Cherdrón and Jordan, 1980; Cherdrón and Charpenel, 1985; Cherdrón et al., 1985), ATF in India (Subramanian and Baskaran, 2007; Subramanian et al., 2009), NSPP in U.S. (Adams et al., 1979) and ESMEALDA in France (Jordan et al., 1988).

Sodium fires resulting from accidental releases of liquid sodium from an SFR would produce aerosols consisting of sodium oxides which would react with available water vapor and carbon dioxide in the air. The exposure of these sodium oxide aerosols ( $\text{Na}_2\text{O}$  and  $\text{Na}_2\text{O}_2$ ) to humid air results in the formation of sodium hydroxide ( $\text{NaOH}$ ) (forming solution droplets if water vapor is abundant). The  $\text{NaOH}$  particles thus formed would be particularly corrosive and hazardous (as are the oxides). Reaction of hydroxide with carbon dioxide to form the carbonate ( $\text{Na}_2\text{CO}_3$ ) would make the aerosol substantially less chemically hazardous (Cooper, 1980). Formation of the bicarbonate almost renders the aerosols completely chemically innocuous.

Tables 2 and 3 (Casselmann, 2009; Cooper, 1980) gather the relevant reactions of sodium with oxygen, water and carbon dioxide and some physical constants of sodium compounds, respectively.

Most of the experimental studies conducted have focused on the timing of Na-oxide aerosols transformation to hydroxide ones ( $\text{NaOH}$ ) and, subsequently, to carbonates ( $\text{Na}_2\text{CO}_3$ ) in a range of conditions varying, in particular, relative humidity (Cherdrón et al., 1985; Hofmann et al., 1979; Subramanian et al., 2009). Essentially, all studies agree that transformation to  $\text{NaOH}$  is a fast process of the order of 1 to tens of seconds, depending on the conditions (Hofmann et al., 1979). According to data available, the formation of carbonates is nearly complete after hundreds of seconds as long as the relative humidity is appreciable, e.g., over 260 s at RH above 20% (Cherdrón et al., 1985), and within 500 s at RH between 50 and 65% (Subramanian et al., 2009); at low RH such transformation has been measured to be much slower. Fig. 5 illustrates the carbonate formation kinetics according to Cherdrón et al. (1985). Cherdrón et al. explained the carbonate formation kinetics based on the concepts of saturation of solution and on particle condensation.

The above observations were supported by theoretical studies (Clough and Garland, 1971; Cooper, 1980). These are both based on gas phase transport theory and in-particle diffusion; in other words, they assumed that chemical reactions are much faster than

**Table 2**  
Relevant reactions of sodium and their compounds.

Sodium combustion	
$2\text{Na} + 1/2\text{O}_2 \rightarrow 1/2\text{Na}_2\text{O}_2$	$\Delta H_{298}^0 = -100.7 \text{ kcal/mole}$
$2\text{Na} + \text{O}_2 \rightarrow \text{Na}_2\text{O}_2$	$\Delta H_{298}^0 = -120.6 \text{ kcal/mole}$
Chemical reactions in open atmosphere	
$\text{Na} + \text{H}_2\text{O}(\text{l}) \rightarrow \text{NaOH} + 1/2\text{H}_2$	$\Delta H_{298}^0 = -33.7 \text{ kcal/mole}$
$\text{Na} + \text{H}_2\text{O}(\text{g}) \rightarrow \text{NaOH} + 1/2\text{H}_2$	$\Delta H_{298}^0 = -45.7 \text{ kcal/mole}$
$\text{Na}_2\text{O} + \text{H}_2\text{O} \rightarrow 2\text{NaOH}$	$\Delta H_{298}^0 = -46.8 \text{ kcal/mole}$
$2\text{Na} + \text{H}_2\text{O} \rightarrow \text{Na}_2\text{O} + \text{H}_2$	
$\text{Na} + \text{NaOH} \rightarrow \text{Na}_2\text{O} + 1/2\text{H}_2$	$\Delta H_{298}^0 = 1.59 \text{ kcal/mole}$
$\text{Na} + 1/2\text{H}_2 \rightarrow \text{NaH}$	$\Delta H_{298}^0 = -13.7 \text{ kcal/mole}$
$\text{Na}_2\text{O}_2 + \text{H}_2\text{O} \rightarrow 2\text{NaOH} + 1/2\text{O}_2$	$\Delta H_{298}^0 = -124.0 \text{ kcal/mole}$
Sodium-concrete interaction	
$2\text{Na} + \text{CO}_2 + 1/2\text{O}_2 \rightarrow 2\text{Na}_2\text{CO}_3$	$\Delta H_{298}^0 = -175.3 \text{ kcal/mole}$
$2\text{NaOH} + \text{CO}_2 \rightarrow \text{Na}_2\text{CO}_3 + \text{H}_2\text{O}$	$\Delta H_{298}^0 = -40.0 \text{ kcal/mole}$
$\text{NaOH} + 1/2\text{CaCO}_3 \rightarrow 1/2\text{Na}_2\text{CO}_3$ $+ [1/2\text{Ca}(\text{OH})_2 \rightarrow 1/2\text{CaO} + 1/2\text{H}_2\text{O}]$	
$4\text{Na} + \text{CaCO}_3 \rightarrow 2\text{Na}_2\text{O} + \text{CaO} + \text{C}$	
$4\text{Na} + 3\text{SiO}_2 \rightarrow 2\text{Na}_2\text{SiO}_3 + \text{Si}$	
$2\text{NaOH} + \text{SiO}_2 \rightarrow \text{Na}_2\text{SiO}_3 + \text{H}_2\text{O}$	

**Table 3**  
Some physical constants for sodium compounds.

Name	Formula	Density (g/cm <sup>3</sup> )	Melting Point (°C)	Boiling Point (°C)
Sodium monoxide	$\text{Na}_2\text{O}$	2.27	1132	1950
Sodium hydroxide	$\text{NaOH}$	2.13	318	1388
Sodium peroxide	$\text{Na}_2\text{O}_2$	2.80	675	decomp.
Sodium carbonate	$\text{Na}_2\text{CO}_3$	2.53	851	1600
Sodium superoxide	$\text{NaO}_2$	2.20	551	decomp.

physical transport. By comparing with data from Hofmann et al. (1979), Cooper found a noticeable consistency of his predictions. Table 4 summarizes the characteristic time determined by Cooper as a function of the particle size.

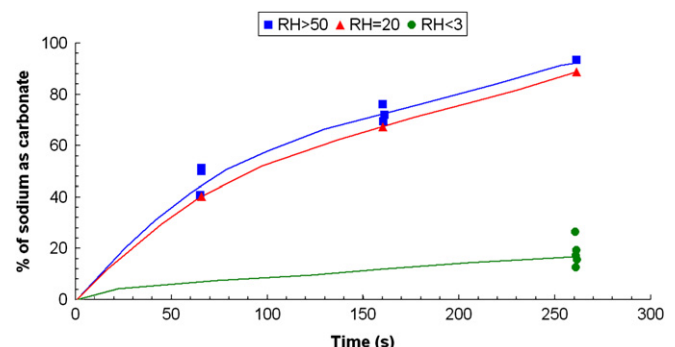
### 3.2. Size distribution

Most of the experimental data available till 1980 were gathered in NEA-CSNI (1979). As noted in Table 5 (NEA-CSNI, 1979, Appendix B), the primary particle size of nuclear fuel and coolant aerosols were reported in terms of geometrical diameter. At first sight, diameters of  $\text{UO}_2$  and mixed  $\text{UO}_2\text{-PuO}_2$  particles ranged from 0.003 to 0.1  $\mu\text{m}$ ; sodium oxide particles were larger, ranging from 0.1 to 0.5  $\mu\text{m}$  diameter; and mixed fuel and sodium produced particles of a size similar to sodium oxide alone (NEA-CSNI, 1979, Chapter II).

Several later studies indicated that the “initial particle size” of aerosols generated during a sodium fire in terms of Aerodynamic Mass Median Diameter (AMMD), was around 1  $\mu\text{m}$  (0.9  $\mu\text{m}$  according to Cherdrón et al., 1985 and 1.0  $\mu\text{m}$  according to Jordan et al., 1988). More recent investigations (Subramanian and Baskaran, 2007) measured AMMDs around 1.2  $\mu\text{m}$  in the temperature range 250–550 °C. In addition to temperature, RH is indicated to affect the primary particles size as well. By assuming a geometric standard deviation ( $\sigma_g$ ) of 2.0, and assuming a log-normal size distribution, the consistency of these values and those coming from earlier studies for sodium-based aerosols can be noted. Nevertheless, some authors (Sundarajan et al., 1982) found that the initial sodium fire aerosol distribution was bimodal.

## 4. In-containment aerosol phenomena

A thorough aerosol phenomenological description requires knowledge of some fundamental properties like size, shape, density and chemical composition, as well as particle dynamics. The former are dependent on particle–particle (agglomeration) and particle–gas/vapor interactions (vaporization/condensation); the latter is a result of the net effect of various driving forces that are usually estimated in terms of process rates.



**Fig. 5.** Carbonate formation in sodium fire aerosols.

**Table 4**  
Characteristic time (in seconds) of mass transport processes for different particle size.

Mechanism	0.1 μm	1.0 μm	10 μm
Diffusion within a reacting droplet	$1 \times 10^{-6}$	$1 \times 10^{-4}$	$1 \times 10^{-2}$
Gas phase transport of H <sub>2</sub> O	$6 \times 10^{-6}$	$3 \times 10^{-4}$	$3 \times 10^{-2}$
Diffusion of H <sub>2</sub> O within a Na <sub>2</sub> O <sub>2</sub> agglomerate	–	$1 \times 10^{-3}$	$1 \times 10^{-1}$
Gas phase transport of CO <sub>2</sub>	$2 \times 10^{-4}$	$1 \times 10^{-2}$	1
Diffusion of CO <sub>2</sub> within a NaOH agglomerate	–	$1 \times 10^{-1}$	$1 \times 10^{-1}$
Diffusion within a reacting solid particle	$1 \times 10^{-2}$	1	$1 \times 10^{-2}$

4.1. Thermal hydraulic boundary conditions

A set of studies found in the open literature allows identification of potential prevailing conditions in containment in the case of a CDA. Using CONTAIN-LMR/1b-MOD1 (Murata et al., 1993) and assuming a breach of the primary coolant boundary in the Advance Liquid Metal Reactor (ALMR), Chiao et al. (1994) demonstrated that consequences of pool and spray fires could be drastically different in terms of the thermal characterization of the containment-atmosphere depending on specific conditions such as the amount of steam in the atmosphere and the sodium inventory reaching the containment (sprays). Fig. 6 displays the containment response in the case of a pool fire. Pool fires lead to pressure increases of about 0.6 atm ( $P_{peak} = 1.7$  atm) and maximum containment-atmosphere temperatures around 315 °C; these spikes decrease over time and, in the long run (over 400 min), the temperature approaches values around 150 °C and the pressure spike practically disappears. These numbers contrast with those from the spray scenarios where pressure rise can be as much as six times higher ( $P_{peak} = 4.3$  atm) and atmosphere temperature can reach 1067 °C (assuming that 136 kg of Na entered the containment in 1 s).

These analyses agree with previous ones that also pointed to a spray fire posing more challenges to containment integrity than a pool fire (Bae et al., 1994; Malet et al., 1981; Morewitz, 1979). Figs. 7 and 8 show some of the results obtained in the Bae et al. (1994) studies, where the effect of the initial sodium temperature and the initial oxygen concentration were parametrically

**Table 5**  
Primary particle size measured by various investigations.

Reference	Material	Primary particle diameter, μm
Allent & Briant	Mixed UO <sub>2</sub> , PuO <sub>2</sub>	Log-normal, bimodal, ( $\sigma_g = 1.35$ ) 0.003–0.01
Chatfield	Pu, Na	0.02 maximum
Castleman	Mixed UO <sub>2</sub> , PuO <sub>2</sub>	Log-normal, 0.004–0.004, $\sigma_g = 2.0$
	PuO <sub>2</sub>	0.1, ( $\sigma_g = 1.9$ )
	UO <sub>2</sub> , Na	0.2–0.6
Morrison et al.	Clad & bare UO <sub>2</sub>	0.002–0.1
Kelly et al.	UO <sub>2</sub>	<0.005–0.02
Jordan et al.	UO <sub>2</sub>	$d_g = 0.073$ ( $\sigma_g = 1.85$ )
Kres	UO <sub>2</sub> , U <sub>3</sub> O <sub>8</sub>	0.014–0.034
Kitani	U <sub>3</sub> O <sub>8</sub>	$d_g = 0.037$ ( $\sigma_g = 1.6$ –2.2)
	U <sub>3</sub> O <sub>8</sub>	$d_g = 0.07$ ( $\sigma_g = 1.93$ )
Schikarski	Clad mixed	<0.08
	Oxide fuel	0.1–0.4
	Na <sub>2</sub> O	
Hilliard et al.	Na <sub>2</sub> O <sub>2</sub> , NaOH	0.1–0.5
Parker et al.	UO <sub>2</sub> , U <sub>3</sub> O <sub>8</sub>	$d_g = 0.034$

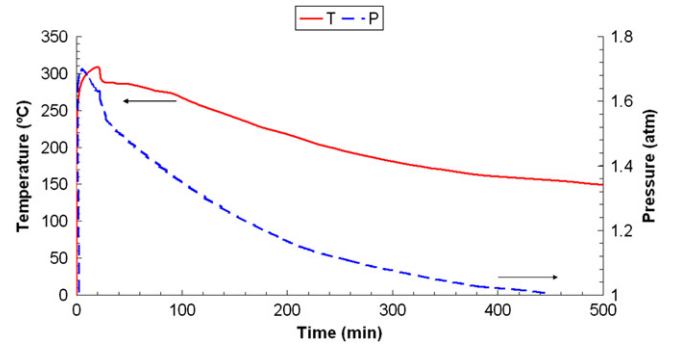


Fig. 6. ALMR containment response to a sodium pool fire.

investigated. The main observations were that pressure and temperature increases were higher and faster in the case of spray fires, even if less Na was injected. It was observed that the maximum pressure rise was far below the theoretical maximum (estimated on thermodynamic grounds) and they only approach each other when Na is sprayed in ultra-fine drops (Morewitz, 1979).

Analyses of large-scale containment response to postulated CDA scenarios were carried out for the Clinch River Breeder Reactor Plant (CRBRP) (Heisler and Morewitz, 1979). They investigated the effect of the extension of Na combustion in the thermal hydraulic response of the building by calculations with the SOMIX code. They found that the maximum pressure increase was in all the cases far from a threshold (6 atm) identified as the maximum practical pressure. In fact, in no case did they get pressure increases over 2.1 atm. Some interesting outcomes from their study were: maximum temperatures (1800 °C) and pressures (2.1 atm) were reached both for 27% and 84% of the oxygen reacted; and, no less important, substantial temperature gradients could be created in the containment, the profiles being a function of the amount of O<sub>2</sub> reacted. Fig. 9 illustrates this last observation.

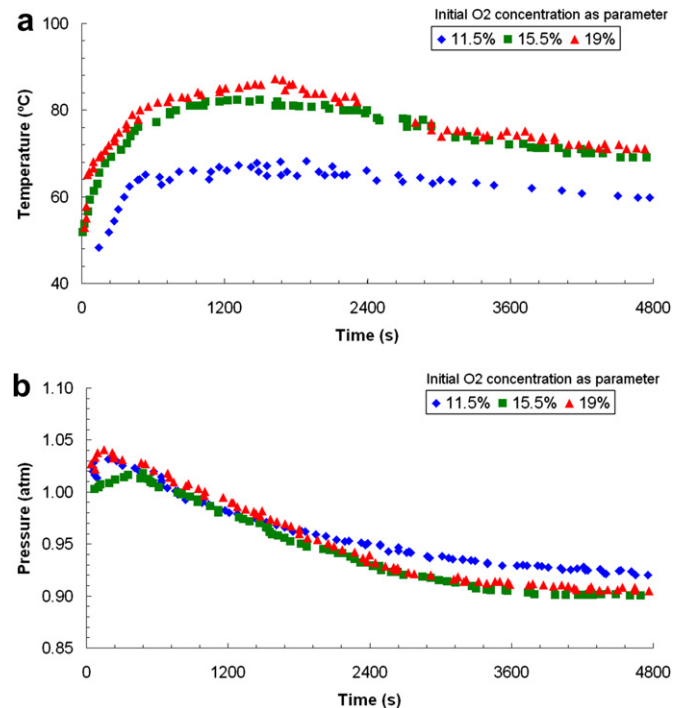


Fig. 7. Thermohydraulic variables of pool fires. a) Temperature and b) Pressure.

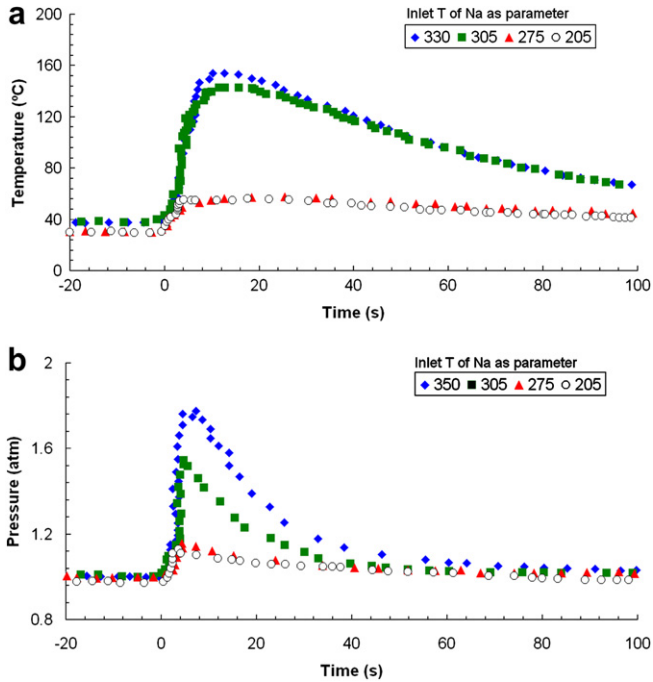


Fig. 8. Thermohydraulic variables of spray fires. a) Temperature and b) Pressure.

4.2. Aerosol evolution

4.2.1. Available data

A number of experimental programs were carried out in the past to understand the in-containment evolution of aerosols in the hypothetical case of an accident in an SFR. Many of those test campaigns were linked to code development and/or validation. Below a synthesis of their major observations is proposed.

ORNL (Adams et al., 1979, 1980) conducted a set of tests within the NSPP (Nuclear Safety Pilot Plant) facility (38.3 m<sup>3</sup>). In addition to the study of aerosol behavior in enclosures, they addressed specifically the potential interaction between two different aerosol populations. To do so, sodium oxide aerosols and uranium oxide aerosols were generated. Additionally, they reported on the different features of Na<sub>2</sub>O aerosols from pool and spray fires. As generic findings confirming some of the statements made in previous sections, they noted a much bigger impact on thermal hydraulics from Na-spray oxidation than from that of a pool. This difference extends also to the airborne aerosol concentration, which reached levels between 40 and 60 g/m<sup>3</sup> in the case of spray fires, whereas it was notably lower (6–25 g/m<sup>3</sup>) for pool fires (using

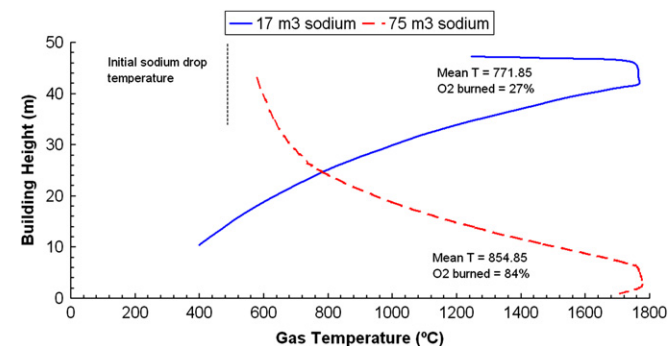


Fig. 9. Temperature axial profiles as a function of initial Na amount.

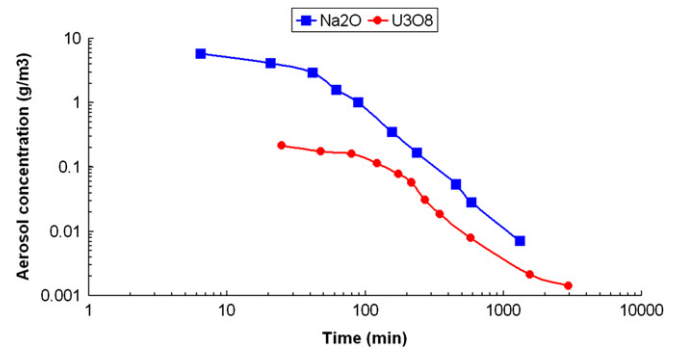


Fig. 10. Aerosol concentration vs. time.

the same amount of Na). They also reported on the Na<sub>2</sub>O particle size from these two sources, but it is not clear if this comparison can be made since they used different diameters to characterize the distribution (no GSD is even given). Anyway, it was seen that in all the individual aerosol tests the U<sub>3</sub>O<sub>8</sub> concentration was notably lower (0.05–0.22 g/m<sup>3</sup>) and the associated particle sizes were smaller than that measured in the Na<sub>2</sub>O tests.

The Na<sub>2</sub>O pool fire experiments showed a clear exponential decay in aerosol concentration. When comparing Na<sub>2</sub>O and U<sub>3</sub>O<sub>8</sub> depletion kinetics, it is noted that Na<sub>2</sub>O underwent faster removal (Fig. 10). In addition to the depletion kinetics of the single-component aerosol tests, the dual-component aerosol tests showed that the aerosol species co-agglomerated and, as a consequence, affected each other. Fig. 11 shows how the depletion kinetics was increased when injecting the second source of aerosols. By estimating the depletion rates of single and dual-species tests in both figures, it is realized that adding a new aerosol in the vessel meant an increase of the depletion rate by a factor between 2 and 3. The final mass distribution of these multiple-source tests confirmed the two compounds' co-agglomeration.

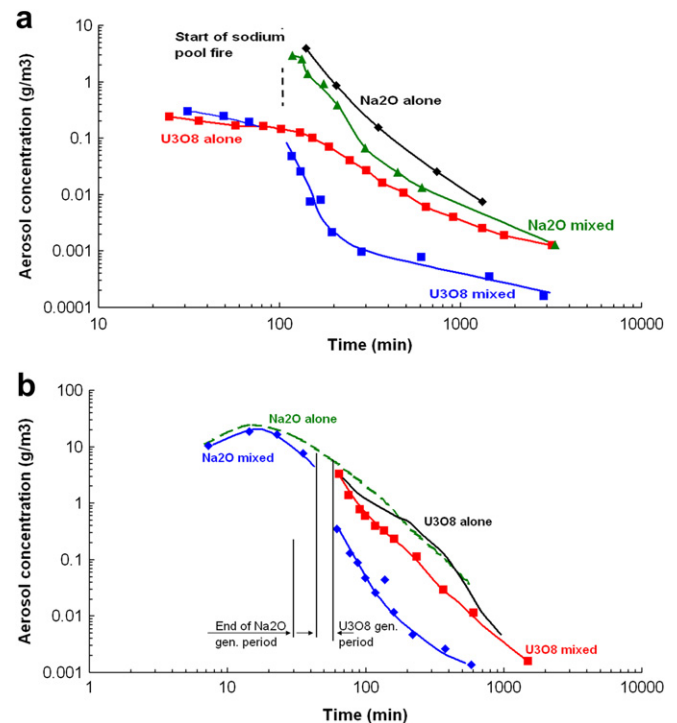


Fig. 11. Concentration evolution: single vs. multiple species aerosols.

The above observations regarding differences between aerosols from pool and spray fires were also highlighted by Cherdrón et al. (1985) in the FAUNA facility (220 m<sup>3</sup>). As shown in Fig. 12, where spray aerosols were generated in a very short period of time (around tens of seconds), the maximum aerosol concentration was much higher than that of pool fire tests. Deriving the time for the maximum aerosol concentration to reduce by a factor of 100, it would take around 100 h for pool fire aerosols and between 0.5 and 1 h for that of spray fires; in other words, in the conditions of the FAUNA tests, spray aerosols were removed nearly 100 times faster (but which attain, as stated above, substantially larger peak concentrations).

#### 4.2.2. Deposition mechanisms

Most relevant processes for “sodium-based” aerosols mentioned in the next sub-sections can be generally described by expressions found in the common aerosol literature (Davies, 1966; Loyalka, 1976; Pertmer and Loyalka, 1977, 1978). As a consequence, sodium aerosol evolution can be well described by the General Dynamic Equation (GDE) of aerosols, as proposed by Pertmer and Loyalka (1977),

$$\begin{aligned} \frac{\partial}{\partial t} n(r, t) = & \frac{1}{2} \int_0^r \varphi(s, r-s) \cdot n(s, t) \cdot n(r-s, t) \cdot ds \\ & - n(r, t) \int_0^\infty \varphi(r, s) \cdot n(s, t) \cdot ds - \frac{\partial}{\partial r} n(r, t) \cdot \frac{\partial r}{\partial t} \\ & - n(r, t) \cdot R(r, t) + S(r, t) \end{aligned} \quad (2)$$

which describes the evolution over time,  $t$ , of the number concentration,  $n$ , of particles with size  $r$ . On the right-hand side of the GDE the different terms are: the first two arise from particle agglomeration (addition and removal of particles with size  $r$ ), the third one corresponds to vapor condensation onto particles, the fourth one encapsulates all the particle removal mechanisms and the last one is the source term.

Next, only those aspects specific to sodium aerosols are discussed. Numerous studies (Adams et al., 1979, 1980; Jordan et al., 1988; Postma and Owen, 1980) have shown that in hypothetical accident conditions of SFRs, in-containment aerosols will undergo depletion processes that will carry a fraction of the airborne particles to containment floor and walls. Given the anticipated conditions in SFR containments, the major mechanisms expected to play a role would be: thermophoresis, which would take particles onto colder surfaces; gravitational settling, which would be effective for larger particles; turbulent deposition, which would be effective in those containment areas with highly turbulent flows (e.g., during spray burning). Moreover, the efficiency of these removal mechanisms would be highly dependent on particle size, density and shape; all three of these features are governed by

particle formation, agglomeration and vapor condensation (if any), especially water vapor given the hygroscopicity of the aerosols. Agglomeration occurs by different mechanisms, i.e., turbulence, gravity, Brownian diffusion, etc.

From all the above processes, in-containment thermophoresis is quite distinctive for Na aerosols. Specific studies were carried out to understand the effect of thermophoresis on Na<sub>2</sub>O particles in a 2 m-high chamber (Nishio et al., 1977). In particular, Nishio et al. looked at the combined effect of thermophoresis and “soaring”<sup>1</sup> of aerosols by thermal convection which they considered to hamper settling. They concluded that thermal convection had a noticeably impact on both thermophoresis and sedimentation; thermophoretic velocity could be reduced by half, whereas the “soaring” effect was highlighted as an essential phenomenon in the early stages of a postulated SFR accident.

One uncertain variable affecting thermophoretic removal is the thermal boundary layer thickness ( $\delta_T$ ). It has been attributed different values (NEA-CSNI, 1979, Chapter III), but usually these ranged between 100 and 1000  $\mu\text{m}$ .

Experiments conducted in large-scale facilities (Jordan et al., 1988; Postma and Owen, 1980) demonstrated that suspended aerosols were preferentially removed by settling, although plated mass (turbulence, diffusion and thermophoresis) was in no case negligible: in the FAUNA facility (220 m<sup>3</sup>) around 60% deposited onto the floor; in the ESMERALDA facility (3600 m<sup>3</sup>) only 10% of the mass deposited onto the walls (Jordan et al., 1988).

#### 4.2.3. Interaction processes

Agglomeration plays a key role in aerosol growth, particularly just above a pool fire. In the flame zone a high particle number concentration of small particles exists and, as a consequence, particle Brownian motion can result in substantial agglomeration rates. Above that region, the resulting larger particles are carried by thermal convection away from the burning zone and other agglomeration mechanisms (i.e., turbulent, gravitational, inertial ...) may dominate. From that location upward it is expected that agglomeration becomes less relevant (NEA-CSNI, 1979, Chapter III); nevertheless, while its intensity may decrease, the high aerosol concentrations mean that agglomeration continues and it is a key factor in accelerating deposition by sedimentation.

The effect of agglomeration on aerosol depletion was studied experimentally by Nishio et al. (1977). Through a battery of experiments, he observed how airborne aerosol concentrations decayed following different rates depending on initial particle concentration. At low initial concentration a net exponential decrease is observed whereas at high concentrations (5000 mg/m<sup>3</sup>) agglomeration accelerates the aerosol depletion at early times. As these experiments were part of a more ambitious program mainly focused on thermophoresis, another variable investigated (not relevant for this discussion, though) was the facility heating. Some of their results are given in Fig. 13.

Vapor condensation (heterogeneous nucleation) onto sodium aerosols plays also an important role in the region just above the vaporizing pool. The combined effect of condensational growth and stirred settling governs the aerosol behavior in the space above a hot sodium pool. After the formation of oxide aerosol fog, most of the aerosol is seen to be removed at an increasing rate until the

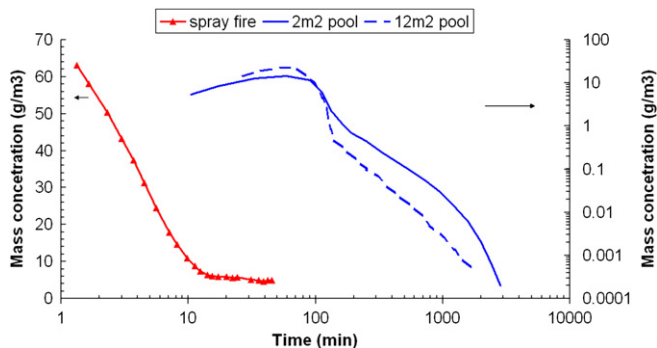


Fig. 12. Concentration evolution in the FAUNA facility.

<sup>1</sup> The authors use the term “soaring” for entrainment of particles by natural upward convection. However, in one experiment an apparatus has a chimney-type arrangement and in the other there is a heated plate at floor level: it seems likely that reverse-thermophoresis would be the main mechanism reducing deposition in this latter case since the general convection in both cases simply provides mixing. The notion of “soaring” may well be misleading.



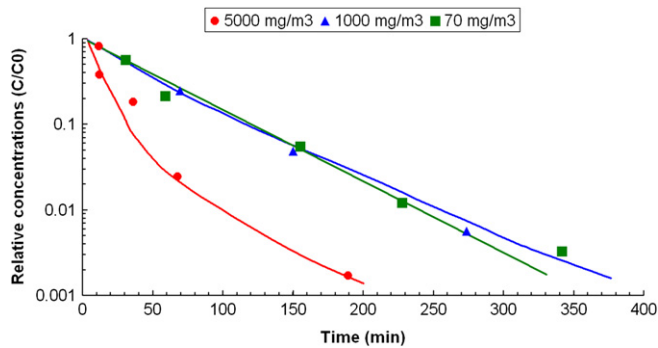


Fig. 13. Concentration decrease of sodium oxide aerosol (2000 W/m<sup>2</sup> heating).

cloud suddenly disappears (Van de Vate et al., 1976); this was presumably for a more-or-less open atmosphere. The aerosol settled on the sodium pool will, if sufficient, hinder the reaction of the pool surface with the atmosphere.

Further than Na condensation onto particle nuclei over the pool surface, steam condensation can also affect Na aerosol behavior. Although SFR containments will presumably contain low levels of humidity, accidental ingress of water cannot be ruled out. Concrete also contains much water in the form of hydrated species. A number of studies have been carried out on the effect of humidity on sodium aerosol growth. All of them agreed that Na aerosols grew even at relative humidity lower than 100%, no matter the Na species (NaOH, NaCl or Na<sub>2</sub>O). The most recent tests (Sundararajan et al., 1982) supplied growth rate data for Na<sub>2</sub>O concentration ranging between 80 and 500 mg/m<sup>3</sup> and relative humidity levels between 50 and 98%. Growth ratios around 3 at 95% relative humidity were reported.

Adsorption of vapors onto aerosol particle surfaces, though less important to aerosol growth, is very important in determining the eventual attenuation in the containment of fission products in the vapor phase (as indicated in Section 2.1). Information is contradictory: experiments have generally shown that sodium oxides are very active in adsorbing fission product vapors (NEA-CSNI, 1979, Chapter III) while there is evidence that oxide on a pool surface has little retaining effect on fission products in the Na beyond mass transfer effects (i.e., hindering evaporation from the liquid surface).

#### 4.2.4. Shape and density

Equations describing aerosol mechanisms are developed based on fully-dense spherical particles. However, reality may be far from such an ideal situation; in other words, particles might be fluffy and irregular. In order to overcome the resulting shortcomings from the “ideal approximation”, equations are corrected by shape factors and/or effective densities. Two shape factors are usually considered: the dynamics shape factor ( $\chi$ ) and the agglomeration shape factor ( $\gamma$ ) where the nearer the values of these factors are to unity the nearer the particle is to sphericity. The dynamic shape factor accounts for the effect of non-spherical forms on the drag force that a moving particle experiences. The coagulation shape factor corrects the actual target size that a particle exhibits when interacting with other particle. For sodium oxide aerosols formed in humid air containing CO<sub>2</sub>, shape factors for agglomerates were measured to be about 1.3 (NEA-CSNI, 1979, Chapter III). Smaller values very close to unity were found for sodium peroxide aerosols in experiments by Barbe-le-Borgne et al. (1986). The presence of humidity has the effect of rendering particles more spherical where, in excess, it will convert oxide aerosols into droplets of hydroxide solution; we note, also, that sodium hydroxide is very hygroscopic and will convert to droplets at quite low relative humidity.

An alternate approach combining particle mobility and collision correction factors was proposed for sodium particles by Gieseke et al. (1977): sodium oxide agglomerates are assumed to be spherical but porous; in other words, they are characterized by a reduced effective density ( $\rho_a$ ) and an agglomerate radius ( $R$ ). Then particle dynamic equations are corrected through,

$$r_e = \alpha^{1/3} \cdot R \quad (3)$$

where  $\alpha$  is the ratio of the actual and the theoretical densities and  $r_e$  is the particle equivalent radius.

Extensive experimental studies of sodium oxide agglomerates characteristics at BCL indicate a value for  $\alpha$  of 0.18 for agglomerates containing about 200 primary particles formed in dry, CO<sub>2</sub>-free air (Gieseke et al., 1977). Measurements of sodium oxide particles in moist air containing CO<sub>2</sub> indicated values for  $\alpha$  averaging about 0.61 (NEA-CSNI, 1979, Chapter III).

## 5. Overall assessment of modeling capabilities

Given the practical absence of activity during the last 20 years in the field of CDA modeling beyond core-disruption aspects, the status of the mid-1980s computer codes may be considered applicable today to a good extent. The main insights into aerosol-modeling drawbacks at the time were reported by Dunbar and Fermandjian (1984) - see also Dunbar (1985) and Fermandjian (1985). In the late 80s a benchmark against experimental data in a medium-scale facility (400 m<sup>3</sup>) for a 90 min pool fire was conducted using a number of these aerosol codes - see Lhiaubet et al., 1990. While most of the codes then reviewed and tested are no longer available, much (if not all) of the aerosol modeling was subsumed in codes such as ASTEC (Van Dorselaere et al., 2009), MELCOR (Gauntt et al., 2005) and CONTAIN (Murata et al., 1993) and conclusions from earlier work can be considered relevant to these codes. It is interesting to note that the Lhiaubet et al. benchmark concluded that aerosol codes were adequate for pool fire conditions in the sense that they tended to be conservative by overestimating (but not massively) the suspended aerosol concentration. This overestimation was in part due to significant underestimation of deposits on walls; this may mean that thermophoresis was underestimated or that account must be taken of mechanisms additional to those usually expected in the containment, e.g., turbulent deposition.

- **Agglomeration.** Several aspects of agglomeration were considered not to be properly taken into account in the models: particle shape, collision efficiency and turbulence (Dunbar and Fermandjian, 1984).

Concerning shape, aerosol equations are developed for spherical particles but allow deviations from this by introducing two coefficients (shape factors); these intend to approximate (usually empirically) the effect of particle shape on their motion (dynamic shape factor,  $\chi$ ) and on their agglomeration rate (agglomeration shape factor,  $\gamma$ ). However, computer codes assumed no variation of  $\chi$  with particle size and, occasionally,  $\gamma$  was adopted as a correction factor of the spherical particle surface instead of its diameter.

Gravitational collision efficiency had been approximated by either Fuchs or Pruppacher and Klett equations (Dunbar and Fermandjian, 1984), the former being 3 times greater than the latter. The difference arises from the Fuchs model not taking account of the motion of the smaller particle with which the larger particle collides. Presently, the Pruppacher and Klett formulation has been adopted in the majority of nuclear safety computer codes:

$$\varepsilon(r_1, r_2) = \frac{1}{2} \left( \frac{r_2}{r_1 + r_2} \right)^2 \quad (4)$$

The effect of turbulent agglomeration was and is approximated by the Saffman and Turner equations (Saffman and Turner, 1956), which accounts for both shear effects and in-eddy acceleration. These equations rely also on parameters like collision efficiencies, shape factors and turbulent energy dissipation rate. In some present codes, e.g., CONTAIN (Murata et al., 1997), the shear collision efficiency is set to 1.0 whereas that of inertia is estimated through Pruppacher and Klett's model. Even though some settings have been adopted and become "standardized", these parameters are still largely uncertain.

- **Numerics.** At the time the models were reviewed, different numerical methods were being used in aerosol codes: the moments method and different versions of the sectional method (i.e., piecewise constant and piecewise linear). The former was observed to have severe drawbacks when dealing with high agglomeration systems and it was abandoned. As a result, most of present aerosol codes rely on discretizing the size domain to turn the integro-differential aerosol equation into a set of algebraic ordinary differential equations.

According to Dunbar and Femandjian (1984), agglomeration issues should be investigated through analytical experiments. Likewise, integral experiments were recognized to be needed to assess the effect of aerosol injection (rates and total amount) from pool/spray Na fires on aerosol evolution within containment. This was investigated in the ABCOVE program (Aerosol Behavior Code Validation and Evaluation) carried out in the Containment System Test Facility (CSTF) at HEDL (Hilliard and Muhlestein, 1985; Hilliard et al., 1985). Although a number of experiments were conducted in the CSTF facility, only three of them belonged to the ABCOVE program. Nonetheless, these explored different aspects of aerosol generation: pool/spray sources, co-agglomeration, atmosphere composition, etc. A set of codes (i.e., HAA-3, HAARM-3, MAEROS, etc.) were compared to data and apparently they all showed reasonable behavior estimating airborne aerosol concentration and the total leaked mass was predicted within a 1.3–2.0 range from measurements, depending on the Na species (1.3 for NaI and 2.0 for NaO<sub>x</sub>). However, it should be underlined that the consistency of calculated results with data required a set of parametric calculations to determine a correction factor ( $\alpha \cdot \varepsilon$ ) of particle density and collision efficiency; and, even doing so, the codes were noted to behave better once the aerosol injection period was over.

According to the above discussion and the principal developments in the nuclear aerosol field (see the major review by Allelein et al., 2009), it seems very probable that the main model limitations identified in the mid-80s persist. This means that to achieve reliable computer code studies that do not need preliminary parametric calculations (i.e., tuning), improvements to characterizing particles and particle–particle interaction processes are required. Concerning particle characterization, the major variables governing a more accurate description are density and shape; improvement would mean coming up with models and/or applicable correlations for effective density and dynamic and coagulation shape factors. Regarding particle–particle interaction processes, validation of available collision efficiency expressions under anticipated conditions within SFR containments as well as determination of those parameters involved in some agglomeration mechanisms, such as the turbulent dissipation rate, would be needed.

In other words, aerosol transport mechanisms are accurate enough as long as particles and particle–particle interactions are well described. This can be concluded from the Lhiaubet et al.

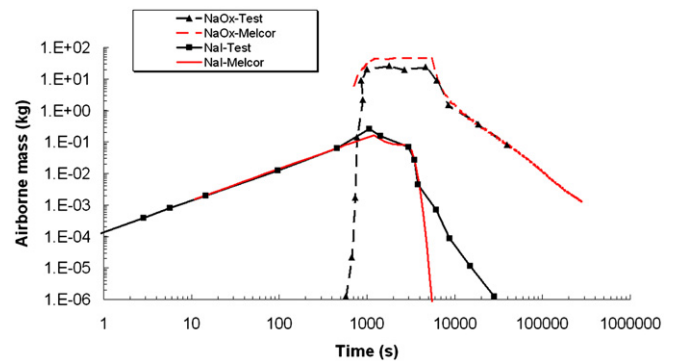


Fig. 14. NaO<sub>x</sub> and NaI airborne mass.

(1990) study and was demonstrated by Souto et al. (1994) who modeled the CSTF experiments with the MELCOR1.8.2 code obtaining reasonable agreement with data. By modeling the same tests with the MELCOR 1.8.6 (Gauntt et al., 2005) code, one may extend this observation on calculation-and-data consistency to current available codes in the arena of LWRs, whenever the initial processes or aerosol generation are not considered in the scenario (Fig. 14).

As discussed in previous sections, aerosol generation in SFR containments strongly depends on three major phenomena: sodium vaporization and nucleation, chemical reactions with the surrounding gas and primary particle agglomeration. Their consideration would require coupling thermal hydraulics, chemical reactivity of airborne and dissolved species, aerosol particle transfer and radiative heat transfer from sodium pool and spray fires (Yamaguchi and Tajima, 2009). A demonstration of this was the CONTAIN-LMR/1B-Mod.1 code (Murata et al., 1993) which included models for sodium pool and spray fires by adapting models coming from the SOFIRE II (Beiriger et al., 1973) and the NACOM (Tsai, 1989) codes, respectively. The result of integrating these phenomena should be the prediction of proper particle composition, concentration and size. Given the mixing conditions of the atmospheric layer where chemical reactions and nucleation are occurring, as well as the potential high concentration of the resulting primary particles, it is of utmost importance to achieve a good description of the appropriate agglomeration mechanisms in this region.

Another missing area in codes is fission products partitioning. This is of utmost interest for source term predictions and, although some data are available, both equilibrium and kinetic studies should confirm and extend the available information. A separate study has also identified the high uncertainty associated with iodine chemistry in an SFR containment, Powers et al. (2010). In turn, this points to the need of having a chemistry and nucleation model for those fission products capable of significant escape from Na solutions, such as cesium. Present LWR codes do different things: e.g., some assume that particles nucleate as soon as vapor become saturated and allow no supersaturation; others allow supersaturation and model kinetically the process of formation of aerosols by nucleation.

Therefore, current LWR integral codes would be capable of capturing in-containment source term evolution if they are improved with SFR aerosol generation and fission product chemistry and partitioning models: this is a *sine-qua-non* condition to obtaining a credible characterization of particles and airborne fission product inventory. Nevertheless, even if current LWR codes appear enabled with the major anticipated aerosol deposition mechanisms in SFR containments, it seems advisable to undertake

a thorough-as-feasible validation campaign. For that purpose a sound and comprehensive database against which to compare code estimates would be instrumental; this database should take into account previous and valuable experimental campaigns such as the ABCOVE and FAUNA programs.

## 6. Conclusions

The in-containment phenomenology governing evolution of sodium aerosols and radio contaminants in a postulated accident scenario in an SFR has been reviewed. It has been shown that many complex phenomena are involved that have a strong influence on radiological risk where, for the public, this arises from the amount and form of airborne contamination and the challenge it presents to filtration. It has been shown that a large body of experimental information exists but there are still some weak areas, e.g., aerosol characterization, radionuclide release during sodium-concrete interactions and mechanical, chemical and thermal re-volatilization.

Furthermore, important aspects of the existing knowledge have not found their way into major computer codes where, in particular, partitioning - or hosting - of radio contaminants by different phases (liquid sodium, vapor/gas phase, oxide aerosols, hydroxide droplets, etc.) is absent. It is clear that in the modern context of, e.g., expectations regarding analyses being conducted as far as a level 2 PSA (as is now usual for LWRs), investigation of safety and potential source terms will require development of additional modeling. In terms of the SFR source term itself, we can anticipate that it may be quite different from that of an LWR in, perhaps, giving greater emphasis (at least short term) to cesium isotopes: while iodine appears to be retained to a great extent by liquid sodium, cesium behaves in a very volatile manner.

In short, a full-scope SFR safety analyses would require furnishing the available codes with specific phenomena not presently considered and to undertake a sound validation of the resulting computational tools. As a consequence, it appears necessary to review all the valuable existing data, to assess their usefulness for validation and to identify gaps where further data is needed.

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