

Comparison of LWR and SFR in-containment source term: Similarities and differences

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ABSTRACT

The fundamental differences between Light-Water Reactors (LWRs) and Sodium Fast Reactors (SFRs) (i.e., breeding ratio, neutron energy spectrum, power densities, coolant nature, reactor architecture and so on) entail major differences in safety aspects. In particular, the in-containment source terms in the event of a severe accident differ in major ways. Nevertheless, an in-depth analysis of similarities and differences based on the present available knowledge should allow assessment of the applicability of LWR safety-analysis tools to the SFR domain. This is the final goal of this paper. A thorough literature review indicates that the capabilities of present LWR safety-analysis codes to address in-containment SFR accident scenarios are uneven for different areas: while they appear acceptable for predicting aerosol evolution, they require significant expansion in other areas like aerosol generation and fission-product partitioning. Additionally, some areas in need of further experimental research are highlighted in this paper.

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1. Introduction

The characterization and behavior of in-containment radio-contaminants (aerosols and vapors of activation products, fission products and actinides) in the event of a postulated accident is of fundamental importance to assessment of the radiological consequences of worst-case scenarios and design of filtration systems and certain reactor components as well as to development of accident management strategies such as controlled filtered venting. Even though there are some commonalities regarding accident scenarios in different reactor types, there are major differences that would drastically influence source terms to the environment. This is particularly true when Light-Water Reactors (LWRs) and Liquid-Metal Fast Breeder Reactors (LMFBRs) are considered. A succinct review of the main safety advantages and drawbacks of the LMFBR technology compared to that of the LWR is proposed by Hennies (1990).

Nuclear safety studies of LWRs have traditionally given huge emphasis to Loss Of Coolant Accidents (LOCAs). The reason for this is the high pressure that needs to be maintained in the

primary-coolant system and the wide diversity of complex phenomena that occur in the event of system depressurization. Given the low pressure of the LMFBR coolant system and the specific reactor architecture, particularly in pool-type Sodium-cooled Fast Reactors (SFRs), attention has focused on very-low-probability scenarios leading to so-called Hypothetical Core Disruptive Accidents (HCDAs). In this type of accident, an event causing significant sodium voiding (boiling) occurs leading to high reactivity insertion in the core with substantial energy release and spreading voiding by further boiling. The total voiding of a large core can insert several dollars of reactivity. The power excursion only terminates with severe core damage and dispersal (subcriticality). In the worst case assumption, the primary system is breached, sodium is ejected and burns and sodium–concrete interaction occurs in the containment.

Beyond the specific accident scenario, there are some elements that make SFR safety threats rather different from those of LWRs; for instance, the significant activation of the primary sodium coolant and its high chemical reactivity with oxygen and water. On the other hand, key challenges in LWR technology may not be such a big concern in SFRs, e.g., if sodium–concrete interaction can be eliminated by a metallic liner in the containment, then hydrogen generation and combustion becomes much less of an issue for the SFRs.

A substantial body of experimental and analytical information on the accident source term has been generated to support the licensing and regulation of LWRs. Nearly all the currently operating

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LWRs in the USA and many more overseas, have been designed and licensed using the so-called “TID-14844 Source Term” (DiNunno et al., 1962). More recently, instead of source-term prescription, mechanistic models of the radionuclide release to containment in accident conditions were developed. There have been extensive efforts to validate the predictions of the source-term models by comparisons to small-scale, separate-effects tests and large-scale integral tests (see, e.g., Allelein et al., 2008). These efforts have been used to formulate an alternative to the TID-14844 source term for regulatory purposes, the “NUREG-1465 source term” (Soffer et al., 1995), a major change to the former specifications defining the in-containment source term. Therefore, NUREG-1465 provides a more realistic portrayal of the amount of fission products present in the containment in the event of a severe accident. It is worth noting, however, that this report was issued more than 15 years ago.

Given all the experience gained in the development of a mechanistic source term for LWRs, it might be foreseen that some of this experience could be applied to SFRs to some extent; however, a direct application of the NUREG-1465 source term to SFRs would be far from acceptable. This being said, it is no less true that NUREG-1465 could provide a useful framework for considering the development of a mechanistic source-term model for SFRs to be used both in licensing activities and risk analyses.

Issues of radionuclide release during accidents at SFRs have received research attention in the past but not to the same depth or breadth as for LWR accidents. Past work readily available in the literature addressed two major issues: retention in the primary sodium coolant of radionuclides released from reactor fuel and behavior of aerosols in the reactor containment. Investigations of radionuclide retention in sodium coolant were fairly integral in nature and exploratory in intent (Powers et al., 2010). Indeed, SFR safety analyses focused principally on partitioning of FPs between liquid sodium and a gas phase (i.e., evaporative release) while analysis of release in the form of aerosols (involving bubble scrubbing and sparging) received little attention. Experimental investigations of the behavior of aerosols in reactor containments were instrumental in the development of highly-sophisticated aerosol-behavior models such as MAEROS (Gelbard, 1982), implemented in the CONTAIN-LMR code (Murata et al., 1993). An extensive review of sodium-aerosol computer codes was reported by Dunbar and Fermandjian (1984). Presently, MAEROS models are implemented in major codes used for analysis of LWR accidents (Gauntt et al., 2005; Van Dorselaere et al., 2009). Despite there having been a good number of studies focused on sodium fires, few of them included speciation changes in sodium aerosols or, fewer still, radionuclide-transport aspects (Castleman, 1973; Clough and Fraser, 1973; Clough and Wade, 1971; Kawahara, 1976; Marimuthu et al., 1984; Subramanian et al., 2009). While it is true that literature sources dealing with radiocontaminant transfer in this context are rare, it is recognized that some significant experimental work performed in the 1970s and 80s is underexploited having remained proprietary or simply unpublished or, at best, mentioned at technical gatherings of which records have become rare (e.g., Jordan, 1976; Berlin et al., 1982; Sophy et al., 1982; Berlin and Geisse, 1989).

This paper analyzes the in-containment source term of both LWRs and SFRs and, based on the available open information, highlights the major differences. This comparison will provide a useful tool identifying for which issues application of LWR safety tools would be acceptable (even if some adaptations may be needed) and which non-generic issues would require ad-hoc model development and validation before implementation in SFR source-term analyses or before extending current LWR tools to the SFR domain.

2. Analyses

2.1. The major differences in core radionuclide inventory

At present LWR fuel enrichment is around 4–5% ^{235}U whereas typical SFRs (i.e., Phénix, Super-Phénix and others) used around 20% fuel enrichment in Pu isotopes. In LWRs about 70% of the thermal energy produced comes from ^{235}U fission, the balance being produced by ^{239}Pu fissions (neutron capture by ^{238}U during irradiation begins the creation of this plutonium isotope). This distribution is very different in a SFR, where Pu fissions supply nearly 85% of all the thermal energy produced and the remaining 15% comes from uranium fissions. Therefore, the irradiated-core inventory differences between SFRs and LWRs are due to:

- the energy spectrum of neutrons;
- the fission yields of ^{235}U and ^{239}Pu and their relative initial amounts;
- the fuel burn-up.

Fig. 1 shows the fission yields of ^{235}U and ^{239}Pu as a function of the isotope mass number (A) for thermal and fast neutrons. The asymmetric fission of both isotopes results in similar trends but with some marked differences. In those regions of high fission yields (and noting that the y-axis of Fig. 1 is logarithmic), the main differences between ^{235}U thermal fission and ^{239}Pu fast fission occur in the lower range of A (85–106), where significantly-different fission yields exist for fission products as radiologically important as Rb, Sr and Ru. Conversely, other major quantitative differences arise either in the low yield region of the curves or they do not affect radiologically-important isotopes, e.g., the noble metals (other than Ru). One can look at the following examples: for ^{110}Ag and ^{106}Ru the ^{239}Pu yield is nearly 2 orders of magnitude higher but silver remains refractory whatever the conditions while ruthenium, having very volatile oxides, can potentially become mobile (see Section 2.3.2 below); if one looks at, ^{131}I , ^{132}Te and ^{137}Cs , all are radiologically significant and have very similar fission yields. These SFR and LWR yield differences will be moderated in reality by the above-mentioned plutonium contribution to the LWR energy production. Another potential source of differences might be the fission-product neutron absorption which, generally speaking, will be a function of n_0 energy. However, as these represent a small mass fraction in the fuel, they should not have a major impact in the reactor neutron balance.

In short, the two most important discrepancies to be expected in terms of relative contributions of radiologically-significant radionuclides in the LWR and SFR inventories are ^{90}Sr (more in LWRs)

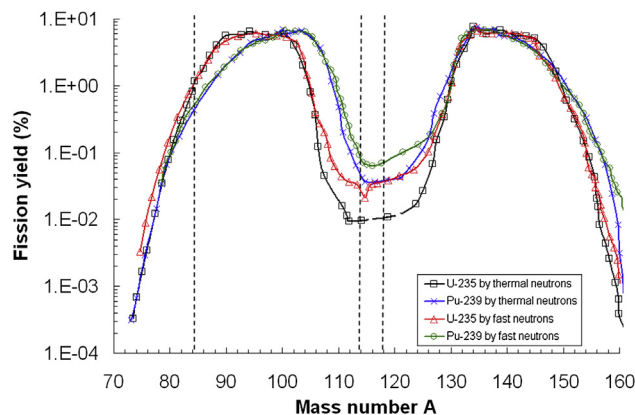


Fig. 1. Fission yield spectra of ^{235}U and ^{239}Pu for thermal and fast neutrons.

and ^{106}Ru (more in SFRs). Nevertheless, all absolute amounts of fission products could turn out to be larger in a SFR simply because SFR fuels are anticipated to be irradiated to higher burn-ups (100 GWd/t_{metal} or more).

2.2. The major differences in the source to the containment

In LWR accidents the main source of aerosols arises from nucleation of supersaturated vapors produced by vaporization of core materials. Broadly, radionuclide and structural vapor mixtures from the over-heating core materials are transported by a steam and hydrogen flow to cooler regions where they supersaturate and nucleate either homogeneously or heterogeneously (including condensing on structure surfaces). Once formed, the steam and hydrogen flow carries the aerosols toward the containment. According to Kissane (2008), an approximate average composition of a prototypical LWR particle entering the containment would consist of about one third metals, one third metal oxides and one third mixed fission-product compounds. Noble gases and a fraction of some volatile fission products, notably iodine, can reach the LWR containment in gaseous form.

In SFRs the picture is drastically different. It is the sodium coolant, which is itself activated (^{24}Na , a gamma emitter, being the main concern), that comprises the main source of aerosols inside the containment building. Aerosols are produced when a severe accident ejects primary-system sodium into the containment and these burn in contact with the air creating sodium oxide aerosols from the homogenous nucleation of the combustion products. The initial sodium ejection into the containment occurs following core disruption and assuming energetic fuel–coolant interaction (i.e., sodium-vapor explosion): the primary sodium above the core is uplifted and impacts the vessel head and either the rotating-plug seal or the cover-gas purification circuit may fail. This sequence of events is important in that the core contamination may not reach the containment until shortly after the initial sodium-ejection event – the sodium in the upper part of the vessel will not have been contaminated before it is partly ejected. Contamination from the core can reach the containment either as vapors and aerosols transported through the sodium by bubbles or, if absorbed into the liquid sodium, as vapors by evaporation into the (breached) cover-gas space. The aerosols produced inside the primary system by the core disruption and vapor explosion may be largely retained by bubble-scrubbing mechanisms but evaluation of this phenomenology is complex. While vaporized sodium could entirely recondense before bubbles reach the cover-gas space (thus aiding retention), bubbles would persist due to an incondensable component comprising fuel-pin filler gas and the fission gases; furthermore, cesium is highly volatile in the expected conditions and so will contribute to the gas phase. So, although in principle core-material vaporization and coolant vaporization with subsequent re-condensation and chemical reactions could be considered separately (Schikarski, 1976), there are numerous experimental results (Castleman, 1973) indicating efficient uptake of radionuclides in the liquid sodium except for noble gases and cesium. In particular, most iodine will probably be absorbed into the sodium liquid becoming sodium iodide whereas (as already said) a large fraction of the cesium would remain in the gas phase – FP speciation is dealt with in more detail below in Section 2.3.2. In addition, while fission-product aerosols and coolant aerosols from combustion could be created largely independently and even behave differently, they would evolve together in the containment as a consequence of condensation and co-agglomeration (Adams et al., 1979; Clough and Fraser, 1973). This picture indicates that SFR aerosol particles will comprise a chemical mixture but in which Na oxides would be by far the most abundant substances.

In addition to the above differences, Na burning is known to produce high concentrations of aerosols that can exceed 10^2 g/m^3 (Jordan et al., 1988). This is at least one order of magnitude higher than that expected in LWR accidents ($1\text{--}10 \text{ g m}^{-3}$) (Allelein et al., 2008). Concerning the aerosol size distribution, several studies are fairly consistent indicating that the average initial (i.e., near-flame) particle size from representative sodium combustion is around $1 \text{ }\mu\text{m}$ or more AMMD (Cherdron et al., 1985; Barbe-le Borgne et al., 1986; Jordan et al., 1988; Subramanian and Baskaran, 2007); this is smaller than the initial size measured in representative LWR experiments such as PHEBUS-FP ($2\text{--}3 \text{ }\mu\text{m}$ entering the containment, Kissane, 2008). This trend for LWR aerosol initial size to be somewhat bigger than for a SFR accident is probably influenced by the aerosols being able to agglomerate during their transport in the primary system whereas the vast majority of SFR aerosols are produced directly in the containment (but nucleated sodium-oxide particles do agglomerate in the flame explaining the $\geq 1 \text{ }\mu\text{m}$ AMMD initial size). Furthermore, this difference would be short lived because the much greater aerosol concentration for the SFR would lead to faster aerosol growth by agglomeration (since it depends on particle concentration squared). On the other hand, the LWR and SFR initial size distributions appear to have similar geometric standard deviations of around 2 and in both contexts particles are initially compact, or spheroidal. Lastly, as the accidents progress, in the case of failure of the LWR pressure vessel a new source from the corium–concrete interaction would result in an aerosol population with a different composition (the additional contribution of concrete constituents would be considerable) and slightly larger size. Similarly in SFRs, it is foreseeable that Na–concrete interaction changes particle composition to some extent with accommodation of concrete constituents.

Turning to fission-product composition, there are two major differences regarding this at entry into a SFR containment: the nearly total presence of iodine as iodide in a Na-based solution and cesium largely in vapor form. The former, though, should be taken with caution since iodine is likely to become volatile when the Na fire happens since the iodide will decompose to produce molecular iodine. Furthermore, once volatilized, iodine could react with other chemical species and form compounds of high relevance to the source term, like organic iodides, or other less volatiles species such as sodium iodate (if residual humidity after reaction with sodium-oxide aerosols is sufficient). However, even in such a case, given the absence of an aqueous phase (see next Section), iodine chemistry for the SFR would not be as complex as it is in LWR scenarios since iodine chemistry in Na solution looks rather simpler; below, Fig. 2 outlines this statement.

2.3. In-containment chemical speciation

2.3.1. Sodium speciation

Sodium fires resulting from accidental releases of liquid sodium would produce aerosols consisting of sodium oxides where it is the oxide, Na_2O , that forms in the flame which then partially converts to the peroxide, Na_2O_2 , away from the flame at lower temperatures, i.e., the peroxide is unstable at flame temperatures.² These oxides form aerosols that would react with available water vapor (H_2O_v) and carbon dioxide (CO_2) in the air. The exposure of these sodium oxide aerosols to humid air in the free atmosphere can result initially in the formation of sodium hydroxide (NaOH) in aqueous solution, i.e., droplets. The NaOH particles thus formed would be

² It has been reported that some sodium superoxide, NaO_2 , can form but this is not a well-supported result.

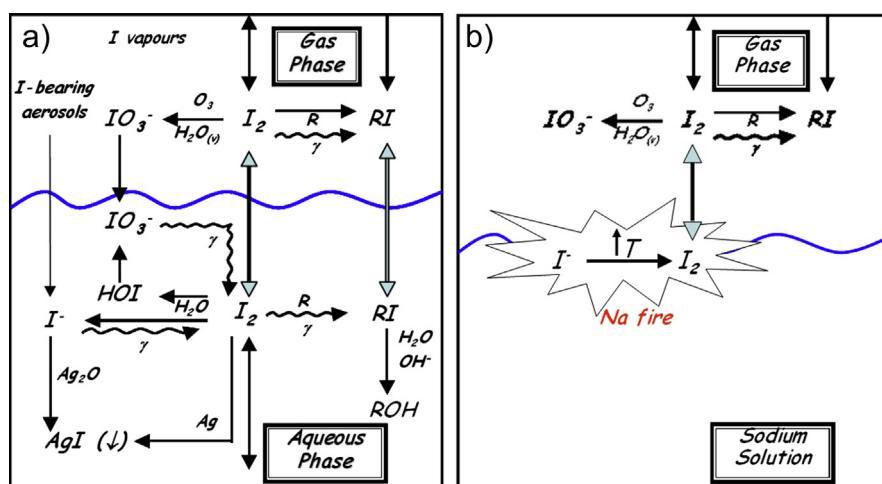


Fig. 2. Iodine chemistry under postulated accident conditions in-containment: a) LWR scenario; b) SFR scenario.

particularly corrosive and hazardous (as are the oxide aerosols). Reaction of hydroxide with carbon dioxide to form the carbonate (Na_2CO_3) would make this aerosol substantially less hazardous chemically (Cooper, 1980) where, ultimately, further reaction to form the bicarbonate (NaHCO_3) renders it almost harmless chemically. However, availability of humidity and carbon dioxide in the containment is limited. It is perfectly conceivable that the water-vapor and carbon dioxide content would not saturate the hydration and carbonation reactions. This would lead to aerosols of mixed composition, perhaps part oxide, part hydroxide and part carbonate. Furthermore, the hydroxide is strongly hygroscopic where seven hydrated forms of sodium hydroxide exist, i.e., crystalline varieties of $\text{NaOH} \cdot n\text{H}_2\text{O}$, $n = 1, 2, 3, 3\frac{1}{2}, 4, 5$ or 7 ; for the carbonate, three hydrated forms exist including a mono- and a decahydrate ($\text{Na}_2\text{CO}_3 \cdot 10\text{H}_2\text{O}$, or washing soda). All these hydrates require temperatures $\leq 100^\circ\text{C}$ and lead to mass increase for the particles concerned as well as size increase (the hydrates are less dense so particles expand). Table 1 gives a summary of key Na reactions (Casselmann, 2009; Cooper, 1980).

In summary, a given molar amount of sodium oxide aerosols forms then can consume many times more moles of water vapor in cooler, post-fire conditions changing the chemical and physical characteristics of the aerosols. A rough calculation for the EFR (European Fast Reactor) containment at 26°C and with only 25% RH

shows that all of a 1-tonne charge of sodium peroxide aerosols (i.e., 590 kg of sodium) could be converted to anhydrous hydroxide. On this same basis, however, even high ambient humidity in the containment atmosphere seems insufficient to create hydroxide droplets unless the sodium spillage is small. Using the same example, carbon dioxide, presently at a concentration of 394 ppm in the atmosphere, would be insufficient to convert fully the aerosol in the containment to carbonate except for irrelevantly-small sodium spills; in other words, a large spill would be only marginally converted to carbonate before exhaustion of the carbon dioxide.

Looking now at kinetics, given the corrosive nature of NaOH, most experimental studies conducted for SFR accidents focused on the timing of Na oxide aerosols transformation to hydroxide ones and, subsequently, to carbonate in a range of conditions varying, in particular, relative humidity (Cherdrón et al., 1985; Hofmann et al., 1979; Cherdrón and Jordan, 1988; Subramanian et al., 2009). Essentially, they all agree that transformation to NaOH is a fast process, of the order of 1 to tens of seconds, depending on the conditions and particle size.

Turning to carbonates, according to the available data, the formation of carbonates with constant concentration of carbon dioxide (free-atmosphere conditions) is nearly complete after hundreds of seconds as long as the relative humidity is appreciable (over 260 s at RH over 20% – Cherdrón et al., 1985 Cherdrón and Jordan, 1988 – 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. 3 illustrates the carbonate formation kinetics according to Cherdrón et al. (1985).

Table 1

Relevant reactions of sodium and their compounds.

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

2.3.2. Fission-product speciation

The available literature dealing with the specific case of fission-product and actinide partitioning between liquid, vapor/gas and aerosol phases during evaporation of contaminated liquid sodium into air (i.e., with combustion) is meager. While major studies have been undertaken for pool fires, it turns out that no studies for contaminated-sodium spray fires exist. Table 2 below summarizes what can be said about the locations and chemical forms of radionuclides in the containment.

Some caveats must be added to Table 1. Firstly, concerning iodine, it is probable that the fraction of the NaI that evaporates with the sodium would decompose in the flame (e.g., Berlin et al., 1982) producing, at least initially, molecular iodine vapor. Furthermore, isolated NaI is neither stable in normal air (i.e., non-zero humidity) nor when exposed to light (photodecomposition)

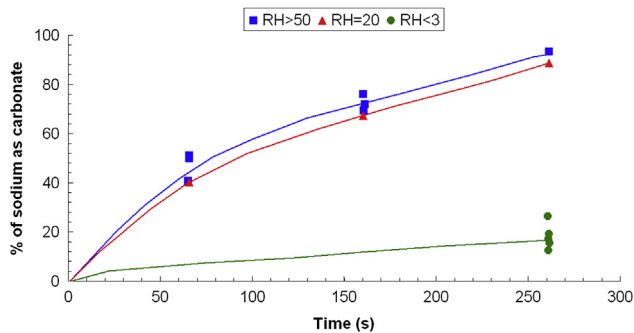


Fig. 3. Carbonate formation in sodium fire aerosols.

though in both cases the decomposition is slow at normal ambient temperatures. However, iodine as NaI remains fairly stable in liquid sodium (i.e., pool or droplets) and sodium peroxide (i.e., the fire-produced aerosols) systems up to moderately high temperatures (Baumash et al., 1970) and, hence, seems unlikely to change to a more volatile form (such as I_2) without prior transfer to the gas phase. Furthermore, similarly to severe accidents in LWRs, the fraction of the ejected iodine that forms molecular iodine can, in contact with organic substances (e.g., paints), lead to formation of volatile organic iodides. However, while methyl iodide would seem to be stable in a containment atmosphere with a persistent suspension of sodium oxide aerosols (Clough and Fraser, 1973; Kitani et al., 1975), dissimilarly to LWR accidents this may change as oxide aerosols are converted to solid hydroxide (for which no studies have been found). There is also the potential for products of air radiolysis to transform iodine vapor species into iodine oxides (c.f., LWR accidents). Hence, while molecular iodine may well probably be a significant product of the burning of contaminated sodium, once in the containment competing fates await this iodine, including reabsorption into the oxide aerosols which seems to be a stable retention. This is a significant difference from LWR accidents: SFR aerosols may well constitute an efficient trap for any volatile forms of iodine created in the containment.

Secondly, regarding ruthenium and technetium, while Table 1 indicates that these fission products would be suspended in solid form in the liquid sodium following core disruption, it is recalled that they have highly-volatile oxides, i.e., the trioxide, RuO_3 , the tetroxide, RuO_4 , and the heptoxide Tc_2O_7 . RuO_4 is the most volatile of these oxides exhibiting a vapor pressure of 10^5 Pa at 40°C (at 311°C for Tc_2O_7). Technetium, as ^{99}Tc ($t_{1/2} = 211,000$ years), a weak β^- emitter, constitutes a lesser threat than two isotopes of ruthenium that have very high specific activities, viz. ^{103}Ru ($t_{1/2} = 39$ days) and ^{106}Ru ($t_{1/2} = 1$ year). The radiological toxicity of ruthenium has been recognized for a long time with respect to water-cooled reactor accidents, e.g., Powers et al. (1994) pointed out that, if released in equal fractions, Ru can have early consequences comparable to those of iodine and longer-term consequences comparable to those of cesium. Mobilization of ruthenium has, therefore, received a lot of attention in the LWR domain, e.g., Auvinen et al., 2005. Furthermore, at the same burn-up, compared to UO_2 fuel the Ru inventory per kilogram of irradiated MOX fuel is far higher due to the larger contribution of ^{239}Pu fissions (see Fig. 1 above).³ Thus, a problem arises if oxide vapors can be produced from solid particles of ruthenium or technetium dioxide in burning liquid sodium. This could occur if such particles, dispersed in the

liquid sodium, were to reach a zone of sodium combustion and encounter favorable conditions of oxygen availability and temperature; flame temperature can reach about 1350°C , though the range 1000 – 1200°C is optimal for forming ruthenium tetroxide. A small-scale experimental program burning sodium in a small pool in which ruthenium particles were mixed showed that significant mobilization of the ruthenium is, indeed, possible (Berlin and Geisse, 1989). If, subsequently, ruthenium vapors converted to the far-less-volatile sodium ruthenate or ruthenium dioxide (many materials catalyze the non-congruent condensation $RuO_4(g) \rightarrow RuO_2(s) + O_2(g)$ which would otherwise involve a large endothermic intermediate step), this would induce mainly condensation of these species onto suspended aerosols since these, at the time the volatile oxides are produced, would present by far the largest solid surface. Hence, even with reduction of ruthenium volatility, a problem remains with significantly-increased radiological toxicity associated with the aerosols.

Finally, there is a possibility that the tellurium is in the liquid sodium as $CrTe$ since the tellurium is known to migrate in SFR fuel pins and react with the chromium in the stainless-steel cladding. If so, the reaction between the sodium and the tellurium would proceed very slowly, a sort of long-term leaching of the chromium from steel core debris. The probable alternative is that the $CrTe$ decomposes in the extreme conditions of core disruption and/or recriticality and, when ejected into the containment, is in the liquid sodium as telluride.

2.4. Thermal-hydraulic boundary conditions

Generally speaking, in-containment thermal-hydraulics affects aerosols in multiple regards: concentration, size distribution, driving forces, etc., and so have a determining impact on source terms.

In LWRs containment thermal conditions are rather different from those resulting from SFR accident scenarios. In the short term, LWR coolant vaporization increases pressure rapidly to values that, although scenario dependent, could approach the containment design pressure. Then steam condensation (which can be greatly enhanced by containment sprays) reduces the pressure unless some energetic phenomena occurs in the containment, like hydrogen (H_2) combustion, creating pressure spikes that may again provoke pressure levels that could exceed the design pressure. In the long term, water vaporization and (more importantly) incondensable-gas production (H_2 , CO , CO_2) during the thermal decomposition of concrete by corium (reactor pressure vessel melt-through assumed) could again drive containment pressures higher than the design tolerance. In short, some thermal-hydraulic phenomena, directly or indirectly associated with the coolant vaporization, could make containment pressure attain values higher than the design tolerance. Average temperatures are usually around (100 – 120°C), although localized phenomena could result in higher values.

For SFR accidents, analyses of large-scale containment response of postulated HCDA scenarios (Heisler and Morewitz, 1979) investigated the effect of Na combustion in the thermal-hydraulic response of the building and found that the maximum pressure increase was in all the cases far from a threshold (6 atm) thermodynamically identified as the maximum possible pressure. As a matter of fact, in no case did they get pressure increases over 2.1 atm. Some interesting outcomes from the Heidler and Morewitz study were: maximum temperatures (1800°C at a specific elevation) and pressures (2.1 atm) were reached both for 27% and 84% of the oxygen reacted; and, no less important, substantial temperature gradients could occur in the containment, the profiles being a function of the amount of O_2 reacted. Fig. 4 illustrates

³ Some other FPs are also more abundant in irradiated MOX but these do not have a high volatility in either element or oxide forms (e.g., Rh, Pd, Ag).

Table 2

Characteristics, status and main location of radiocontaminants.

Element	Characteristics in sodium	State in containment	Main location(s)
Noble gases Xe, Kr	Inert, virtually insoluble	Element	Gas phase
Halogens I, Br	React with Na, moderate volatility	NaI, I ₂	Na (liq.) Aerosol
Alkali metals Cs, Rb	Soluble, high volatility	Cs ₂ O, CsOH	Gas phase Aerosol
Alkali-earth metals Sr, Ba	Very low volatility	SrO(s), BaO(s) in liquid suspension	Na (liq.)
Noble metals Ru, Pd, Rh, Ag	Very low solubility (except Ag), very low volatility	Metallic, in suspension	Na (liq.)
Other metals Mo, Tc	Very low volatility	MeO(s), MeO ₂ (s), Na ₂ MeO ₄ (s) in liquid suspension	Na (liq.)
Actinides, U, Pu	Refractory	MeO ₂ (s) in liquid suspension	Na (liq.)
Others Te	Reacts with Na, moderate volatility, decomposes in contact with air	Na ₂ Te Te	Na (liq.) Aerosol

this latter observation and includes the band of temperatures expected in the case of a LWR containment; as can be noted, the in-containment thermal load is much lower in the LWR case and even if gradients existed, they could never reach the magnitudes foreseen in the Heisler and Morewitz study (though the realism of such extreme gradients must be questioned given natural convection – their result would seem to require a static atmosphere, i.e., no mixing from natural convection). Nevertheless, the higher thermal challenge to SFR containments is real. Furthermore, concerning the pressure loading, and similar to the LWR containment scenario involving core–concrete interaction, the longer-term pressure would be increased if the primary-system decay-heat-removal (DHR) system(s) were inoperational. This is because, after the initial breach of the primary system, it is necessary to prevent core-debris decay heat from causing boiling of the sodium remaining in the reactor vessel. In a study by Scholtyssek and Maschek (1994) for EFR (the design of which included a passively-operating DRH dual system each of 45 MW capacity), the importance of this was highlighted: without the DHR system, containment pressure tolerance is challenged 28 h after the initial core excursion.

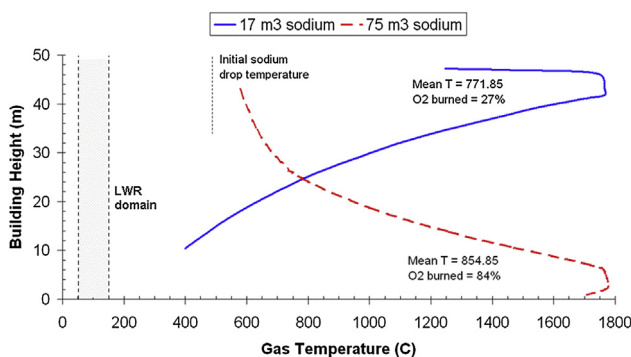
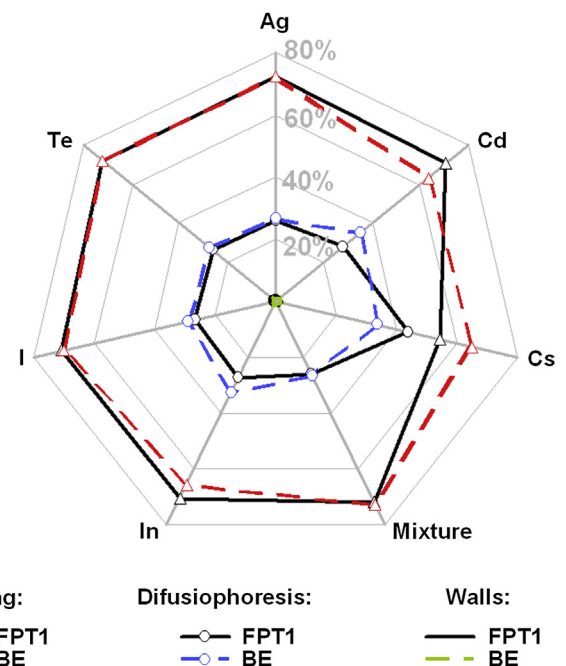
2.5. In-containment aerosol phenomena

In the LWR containment thermal-hydraulic conditions presented above, it is generally accepted (Allelein et al., 2008) that particle sedimentation is the dominant aerosol depletion process followed by diffusiophoresis (AP1000 and AP600 reactor containments excepted). A relatively minor contribution to deposition in terms of mass could be also attributed to Brownian diffusion of small particles. As an example, Fig. 5 shows data and predictions in the case of the PHEBUS-FPT1 experiment (Herranz et al., 2007).

In-containment aerosol dynamics is expected to be somewhat more complex in SFRs than in LWRs. Aerosol dynamics heavily

depend on the generation process and in SFRs two major particle sources are pool and spray fires. Fig. 6 shows the normalized concentration in three different experiments as a function of time. As noted, data from the SFR-experiment aerosol database (Cherdron et al., 1985) and the LWR one (McCormack et al., 1987) have been plotted together. The main difference observed, apart from the particle nature, is associated with the maximum concentration reached in the experiments: the higher the agglomeration rate, the faster particles grow and can reach bigger sizes and, as a consequence, the faster the sedimentation. Given that aerosols concentrations are experiment-dependent, Fig. 6 is merely an illustration of the impact of high initial aerosol concentration in the case of a sodium spray injection. Other factors, such as the convective conditions including turbulent intensity over a burning Na pool can also have a significant influence on aerosol generation and evolution.

The concentration effect was studied by Nishio et al. (1977). They showed that particle formation in Na reactors is closely related to Na oxidation rate and subsequent formation of tiny, high mobility particles that, once agglomerated, result in the typical 1- μ m particles mentioned above (NEA-CSNI, 1979). Based on their data, two correlations were derived with correlation coefficients (R^2) higher than 0.94:

**Fig. 4.** Axial temperature profiles as a function of initial Na amount.**Fig. 5.** Mass distribution (% of the total mass injected).

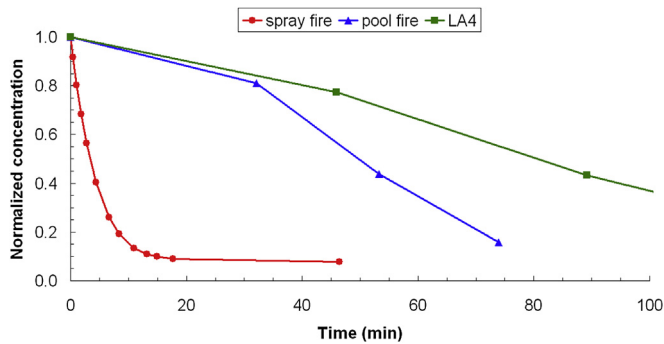


Fig. 6. Concentration evolution.

$$y = 125.3 \cdot x^{-2.06} \quad (1)$$

$$R^2 = 0.94$$

$$\text{for } C = 5000 \text{ mg/m}^3$$

and

$$y = 0.84 \cdot e^{-0.0185x} \quad (2)$$

$$R^2 = 0.99$$

$$\text{for } C = 1000 \text{ mg/m}^3$$

Fig. 7 shows the ratio of these two expressions. It indicates that concentration depletion is much faster at high concentration, so that most particles are swept out of the atmosphere and the airborne concentration is so sharply reduced that after a while the depletion rate is less than in the 1000 mg/m³ case.

In short, even though once generated the particle evolution would evolve quite alike in SFR and LWR scenarios, the generation process seems to largely impact aerosol dynamics through the total aerosol concentration produced.

As for the major mechanisms expected to play a role in SFRs, they would be: thermophoresis, which would take particles onto cold surfaces and, given SFR accident conditions, be more significant than in LWR accidents; settling, which would be effective for large enough particles; and turbulent deposition which, unlike in a LWR accident, could become significant in those containment zones where highly turbulent flows arise from strong natural convection. Broadly, turbulence effects (both deposition and agglomeration) and thermophoresis can be expected to play a more significant role in SFR conditions than in those of LWRs. Experiments conducted in large-scale facilities for SFRs (e.g., Jordan et al., 1988; Postma and Owen, 1980; Sophy et al., 1982) demonstrated that sodium airborne aerosols were preferentially removed by settling, although plated mass was in no case negligible: in the

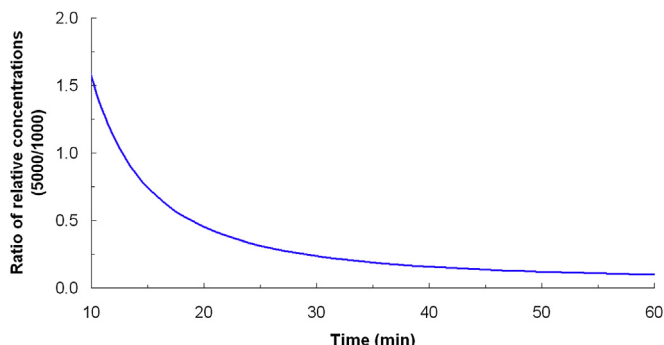


Fig. 7. Concentration decrease of sodium oxide aerosols.

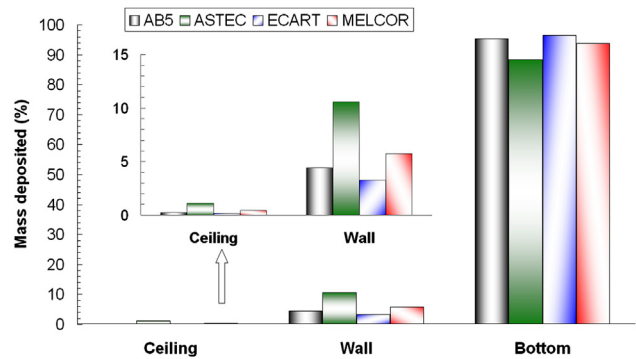


Fig. 8. Aerosol mass distribution.

FAUNA facility (220 m³) around 60% got onto the floor whereas this percentage was about 90% in the ESMERALDA facility (3600 m³). Fig. 8 shows the measured final mass distribution of the ABCOVE-AB5 experiment along with assessments conducted with LWR codes (Herranz et al., 2012); as noted, settling was the major driver mechanism of Na-particles removal from the containment atmosphere. It may be that in large-scale SFR conditions, the faster agglomeration of particles leads to faster deposition by sedimentation which tends to counter any effect of greater plate out due to faster thermophoresis.

In short, major differences should be expected concerning in-containment aerosols in two regards: relevance of agglomeration and different working phoretic phenomena (diffusiophoresis in LWRs and thermophoresis in SFRs). However, all the evidences seem to indicate that the final mass distribution would be similar, most of the aerosol mass being depleted onto horizontal structures by gravitational settling.

3. Conclusions

This study is an assessment of in-containment source terms for LWRs and SFRs based on the available literature highlighting some aspects and uncertainties of the less-well-known SFR source. By exploring sources and phenomena that fission products undergo within the containment, it is realized that sodium (Na) is the key SFR feature responsible for such differences. In a LWR accident iodine is the major concern; in a SFR sodium readily reacts with iodine to form NaI, so that I-131 and the other isotopes are partly but significantly trapped within liquid sodium (overall iodine volatility is drastically reduced); cesium, on the other hand, will behave with high volatility in both the LWR and SFR case and be a major radiological hazard associated with aerosols in the containments. This means that the potential radiological impact outside the SFR containment would be dominated by Cs-137 from the start of the release, instead of I-131 as in the case of LWRs. In addition, in a SFR a major release of Na would be a massive source of aerosols given the combustion of sodium in the oxidizing conditions of the containment (i.e., spray and/or pool fires); the formation of sodium-oxide particles would dominate the in-containment aerosol behavior whereas in LWR accidents the coolant (i.e., the water) is not a major contributor to aerosol mass; in a LWR accident, the aerosol mass is dominated by emissions from the degraded core (and core–concrete interaction if this were to occur). Once formed, the main aerosol depletion process will be, in both cases, gravitational settling. Nevertheless, whereas in LWRs diffusiophoresis will have a minor but significant impact, similarly thermophoresis will in case of SFRs; otherwise, turbulence effects in the case of the SFR accident will be more pronounced perhaps producing localized

zones of high deposition on walls or the roof and accelerating particle growth through agglomeration. In the end, the final deposited-mass distributions in the two containments could well be quite similar though the time evolution of the two airborne radiological hazards may be quite different.

It has been seen that initial aerosol generation in SFR containments strongly depends on the following major sequential stages: sodium vaporization, vapor mixing and combustion, nucleation of the combustion-product vapors (oxides), and rapid in- and near-flame agglomeration of primary particles. It has also been seen that some iodine, much cesium and perhaps just very small fractions of other fission products (except, potentially and very importantly, ruthenium) will transport with the sodium in the gas and aerosol phases. The proper consideration of aerosols in SFR containments would therefore require coupling thermal-hydraulics (including radiative heat transfer), chemical reactions of evaporating and airborne species and aerosol-particle dynamics. The result of integrating these phenomena should be the prediction of proper particle composition, concentration and size. Given the high mixing conditions of the atmospheric layer where chemical reactions and nucleation occur, as well as the potential high concentration of the resulting primary particles, it is of utmost importance to achieve a good description of the relevant agglomeration mechanisms in that region given the already rather large size (1 μm) of particles leaving the flame zone.

Another major phenomenological aspect missing in codes is fission-product partitioning. This is of utmost interest in source term predictions and, although some data are available, both equilibrium and kinetic studies should confirm and extend the available information. In turn, this points to the need to have a nucleation model for those fission products capable of escaping Na solutions in a significant fraction, like cesium. Present LWR codes assume that particles nucleate as soon as vapor get saturated; however, differences in boundary conditions and major uncertainties involved in such an approach, recommend not to apply such an approximation without supporting evidences.

Therefore, current existing LWR integral codes would be capable of capturing in-containment source term evolution for SFRs if they were enabled with SFR aerosol generation and fission-product partitioning models: this is a sine-qua-non condition for producing proper assessment of particles and airborne fission-product inventory.

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