



Radionuclides emissions from sodium pool fires: Learning from the 1970s-era FANAL experiments

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ABSTRACT

The emission of radionuclides from sodium fires is a very important concern when considering the potential radiological consequences of an accident at a sodium fast reactor. Radionuclides can be released when contaminated sodium from, for example, the primary coolant system, catches fire. Not all radionuclide species will behave the same way, however, and some will have a higher tendency to become aerosolized than others. This paper uses historical experimental results from the 1970s-era FANAL program in order to come up with estimates for the combustion partition coefficients for radioiodine, radiocesium, and several other radionuclides from pool fires, and uses information from the program to characterize the chemical interaction between radioiodine vapor and sodium combustion product aerosols.

1. Introduction

Sodium fast reactors (SFR) were one of the major nuclear reactor designs that were being pursued in the 1960s, 1970s, and 1980s as alternatives to light water reactors. Although many of the programs to deploy these were abandoned in throughout the 1980s and 1990s, there is a lot of renewed interest in advanced SFR technology today, and it is one of the key technologies under consideration in the Generation IV International Forum (Kelly, 2014). There are a number of advantages to SFRs, in terms of safety, versus light water reactors, but also several disadvantages, and areas where not of a lot of information is known (Herranz et al., 2012).

Sodium as a liquid metal is very different from water in terms of its physio-chemical properties, and this would have a large impact on the behavior and transport mechanisms of fission products in the case of an accident. These major chemical differences mean that different radionuclides will dissolve and react with in a much different way than they would with water. Sodium also has a much higher boiling point than water, which means that it is more likely to stay in liquid form while being transported through the primary coolant system during an accident. Once in contact with air, however, sodium will also tend to ignite and catch fire. One of the major sources of uncertainty in evaluating the safety of some of the new SFR designs is the behavior of fission products in these fires, and their relative release to the air or retention in the liquid phase.

This is the primary topic that this paper aims to address, and is doing so by re-examining previously unpublished results from the

FANAL program. This program was carried out in the late 1970s at the Cadarache research center in France by IPSN (the *Institut de Protection et de Sûreté Nucléaire*, the predecessor of IRSN), and involved a set of 12 different experiments that studied the behavior of real radionuclides in sodium fires.

In the 1970s and 1980s, there was a considerable amount of research that was being done in SFR safety, mainly to support the safety cases for the commercial prototypes that were being put forward at the time. Much of this work was never published in open journals, however, and was only shared publicly in a very limited manner in conference proceedings. A major consequence of keeping the information closed for so many years (~40 years at the time of writing) is that it is very difficult for researchers today to access the data, analyses, and insights gained in the past when trying to assess safety in the new generation of SFR projects. With the renewed interest today in SFR technology, IRSN is conducting a project to retrieve and re-analyze information from its archives as part of in new studies (Lebel and Girault, 2017a). The present study, along with another study that uses the 1980s-era EMIS experiments on sodium pool fire aerosol emissions (Lebel and Girault, 2017b), are products of this effort.

2. Radionuclide release phenomena

Volatile radionuclides can be emitted from hot liquid sodium during a sodium fire, compelled by thermodynamic drive to establish a vapor-liquid equilibrium between the radionuclide species in the two phases. Not all of the radionuclide species that can become dissolved or

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suspended in the sodium coolant during an accident are very volatile (e.g., Sr, Te, Ce, Pu, etc.), but important ones like cesium and iodine (as NaI) are volatile enough to be released in significant fractions from the sodium liquid.

A lot of the previous work examining radionuclide emissions from liquid sodium has been done in the context of evaporation from the hot sodium pool in the reactor vessel to the inert cover gas region. In this case, radionuclides and sodium are each evaporating into an inert argon atmosphere, and the driving force for evaporation is the heat in the sodium pool. In their study, Haga et al. (1992) described how this process is controlled by the equilibrium partition coefficient, K_d , which relates the mole fractions of sodium and the radionuclide species in the gas phase, $y_{na} = p_{na}/P$ and $y_i = p_i/P$, to the mole fraction of these in the liquid phase, $x_{na} = n_{na}/n_{tot} \approx 1$ and $x_i = n_i/n_{tot}$.

$$K_d = \frac{y_i/y_{na}}{x_i/x_{na}} \quad (1)$$

An alternative is to express the phase partitioning of the radionuclide solute with the activity coefficient, γ_i :

$$\gamma_i = \frac{y_i}{y_i^o x_i} \quad (2)$$

This description is related to the equilibrium partition coefficient, but is defined in terms of the saturated composition of the pure element, y_i^o , rather than the saturated vapor composition of sodium. The value of the activity coefficient depends on the excess Gibbs free energy of mixing, such that $\Delta G_i^E = RT \ln \gamma_i$ (Castleman and Tang, 1968).

The Haga et al. (1992) study experimentally determined values of K_d as a function of temperature for cesium, sodium iodide, and tellurium, and related them to some earlier theoretical models by Castleman and Tang (1968). To extend this to a non-equilibrium case, when the sodium pool and cover gas temperatures are not at the same temperature, they also presented a corrected non-equilibrium partition coefficient that includes differences in vapor phase diffusivity between the radionuclide species and sodium.

$$K_d^* = \frac{D_i}{D_{na}} K_d \quad (3)$$

This non-equilibrium partition coefficient also determines the relative mass transfer rate of the fission product species compared to sodium. The paper by Brockmeier et al. (1994) described the implementation of these concepts into a module of the CONTAIN-LMR code, and they described that the ratio of mass transfer rates would be related by:

$$K_d^* = \frac{\dot{m}_i/m_{i,pool}}{\dot{m}_{na}/m_{na,pool}} \quad (4)$$

Note that the original experimentalists actually defined Eq. (4) in terms of a retention factor (RF), which is the reciprocal of the non-equilibrium partition coefficient, $RF = (K_d^*)^{-1}$. The Brockmeier et al. (1994) and Haga et al. (1992) studies were informed by a few prior studies examining the relative evaporation rates of different fission product species and sodium. Castleman and Tang (1968) developed a lot of the early theoretical descriptions of these processes in terms of thermodynamic principles. This was followed up in experimental work by Clough (1971), Clough and Wade (1971), Clough and Fraser (1973), and Schütz (1979). These studies all evaluated the evaporation rates of different radionuclides into an inert atmosphere, and made a lot of progress describing that for a lot of the main radionuclides of importance. A few experiments studying radionuclide release from sodium fires had been carried out prior to the FANAL program, such as those by Chechetkin et al. (1973) and Kawahara et al. (1976). In a lot of cases, however, the observed release fractions during a sodium fire were somewhat different than what was experienced in inert atmosphere case. This was especially evident for radioiodine, whose release fractions were often higher than expected, and also often highly variable

depending on the mass of sodium used in the experiments.

The mechanics of fission product release in the fire case and inert evaporation case are related, but one has to reconcile these two cases by taking into account the different dynamics that exist in the boundary layer above the sodium pool. Sodium fires behave differently than hydrocarbon fires because the reaction products from a sodium fire (Na_2O and Na_2O_2) are solids. This profoundly affects the shape of the fire, and causes the flame to nearly attach itself to the surface of the pool, where the net outward flow of sodium vapor is balanced by the net inward flow of oxygen (Newman, 1983). The details of this reaction boundary layer from the point of view of mass transfer, heat transfer, and particle generation have been described by Sagae and Suzuoki (1985) and Lebel and Girault (2017a,b). One of the most important points is that sodium fires do not release all of their combustion products into the air. After the Na_2O particles are formed in the flame, a large majority often immediately re-deposits onto the surface of the pool to form a crust, and only the remaining fraction (usually only about 20%) actually escapes the fire.

In order to extend the partition coefficient concept to the sodium fire case, one must consider what happens to the radionuclides when it diffuses into the hot reaction zone. Certain species, like cesium, are also very reactive towards oxygen, and can burn to form solid oxides. In other cases, the species could be non-reactive, and would continue to diffuse past the flame front until condensing out on the other side. If the latter occurs, then radionuclide releases could be enhanced relative to sodium combustion product aerosols because they would avoid the re-deposition process that would bring them back to the pool surface. In addition, sodium oxide is present in the crust layer on the surface of the sodium pool, and makes up the particulates that are generated in the sodium fire. Radionuclides can adsorb and desorb from the surface of this material as well, and use the particulates leaving the fire as a vector for its release into the air. Taken together, these factors mean that partition coefficients for radionuclide releases in an inert evaporation case may or may not be the same when applied to the releases from sodium fires. It is convenient, therefore, to define a “combustion” partition coefficient, K_c , that relates the radionuclide releases to the sodium combustion product releases from a fire:

$$K_c = \frac{\dot{m}_i/m_{i,pool}}{\dot{m}_{na}/m_{na,pool}} \quad (5)$$

In this definition, note that \dot{m}_{na} refers to the sodium aerosol release rate, per mass of sodium, and $m_{na,pool}$ refers to the total amount of sodium remaining in the pool, in both Na metal form, as well as in the burned Na_2O form.

3. Experimental

3.1. FANAL experiments

In total, 12 tests were carried out between 1976 and 1979 as part of the FANAL program, with the goal of assessing the release of radionuclides from burning sodium. This would be a safety concern in the event of the ejection of primary sodium coolant into containment. In addition, the FANAL program was intended to be carried out at much larger scales than experiments that came before it, involving several kilograms of sodium at a time, rather than tens of grams. The program even employed real primary sodium coolant taken from the RAPSODIE reactor for some of the tests.

Information that is being presented in this paper was collected from archival material from IRSN at the Cadarache research center in France, including several unpublished internal reports, lab notes, and other material. Some results from the program were shared externally in an international conference on fast reactor safety in 1982 (Berlin et al., 1982), but the large majority of results remain in a series of unpublished files and internal reports. The main experimental reports are

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