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Toward a Mechanistic Source Term in Advanced Reactors: Characterization of Radionuclide Transport and Retention in a Sodium Cooled Fast Reactor

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Toward a Mechanistic Source Term in Advanced Reactors: Characterization of Radionuclide Transport and Retention in a Sodium Cooled Fast Reactor

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A vital component of the U.S. reactor licensing process is an integrated safety analysis in which a source term representing the release of radionuclides during normal operation and accident sequences is analyzed. Historically, source term analyses have utilized bounding, deterministic assumptions regarding radionuclide release. However, advancements in technical capabilities and the knowledge state have enabled the development of more realistic and best-estimate retention and release models such that a mechanistic source term assessment can be expected to be a required component of future licensing of advanced reactors.

Recently, as part of a Regulatory Technology Development Plan effort for sodium cooled fast reactors (SFRs), Argonne National Laboratory has investigated the current state of knowledge of potential source terms in an SFR via an extensive review of previous domestic experiments, accidents, and operation. As part of this work, the significant sources and transport processes of radionuclides in an SFR have been identified and characterized. This effort examines all stages of release and source term evolution, beginning with release from the fuel pin and ending with retention in containment. Radionuclide sources considered in this effort include releases originating both in-vessel (e.g. in-core fuel, primary sodium, cover gas cleanup system, etc.) and ex-vessel (e.g. spent fuel storage, handling, and movement). Releases resulting from a primary sodium fire are also considered as a potential source. For each release group, dominant transport phenomena are identified and qualitatively discussed.

The key product of this effort was the development of concise, inclusive diagrams that illustrate the release and retention mechanisms at a high level, where unique schematics have been developed for in-vessel, ex-vessel and sodium fire releases. This review effort has also found that despite the substantial range of phenomena affecting radionuclide release, the current state of knowledge is extensive, and in most areas may be sufficient. Several knowledge gaps were identified, such as uncertainty in release from molten fuel and availability of thermodynamic data for lanthanides and actinides in liquid sodium. However, the overall findings suggest that high retention rates can be expected within the fuel and primary sodium for all radionuclides other than noble gases.

I. INTRODUCTION

To license and operate a commercial nuclear power plant, it is vital to ensure the safety of the public and environment by providing protections against the release of radionuclides. As part of the process to assess the safety of a nuclear plant design, the source term expected to occur during normal reactor operation and accident sequences is analyzed, where a source term is considered to be the types and amounts of radionuclides that could be released to the environment. Not only is this a fundamental part of the design process, but it is a requirement for regulatory licensing[1]. While past nuclear power plant source term analyses often used a deterministic, bounding, and conservative assessment of radionuclide release, over the past 25 years there has been an increased interest in the development of mechanistic source terms (MST) based on more realistic evaluations that also consider important uncertainties.

In general, an MST attempts to realistically model the release and transport of radionuclides from the source to the environment for a specific scenario, while accounting for retention or transmutation phenomena and any associated uncertainties. Determining an MST for radionuclide transport that involves complex phenomena requires significant knowledge and modeling capabilities. Assessing the processes that affect radionuclide transport in sophisticated systems can be difficult, as multiple chemical and physical interactions are occurring simultaneously.

To assist in the process of MST formulation, this paper qualitatively identifies and describes the transport processes of radioactive materials in a metal-fuel, pool-type SFR, which is characteristic of current industry designs, and assesses the current state of knowledge regarding these phenomena. This paper describes a small portion of the results of a U.S. Department of Energy Office of Nuclear Energy sponsored Advanced Reactor Technologies (ART) Regulatory Technology Development (RTDP) effort by Argonne National Laboratory[2] on the development of an MST that included:

- Extensive review of domestic SFR operating experience, accidents, and related experiments.
- Consideration of SFR source term efforts completed as part of early licensing interactions between the U.S. Nuclear Regulatory Commission (NRC) and the designers of the Clinch River Breeder Reactor Project[3,4], Rockwell International's Sodium Advanced Fast Reactor[5], and General Electric's Power Reactor Innovative Small Module[6].
- Identification of the sources and transport processes affecting radionuclide transport, including in-vessel and ex-vessel releases.
- Identification and assessment of the capabilities of existing codes methods that could potentially be used to complete an MST.

One of the key results of the Argonne effort was the creation of a concise and fully inclusive diagram describing the release and transport processes in an SFR. Unique diagrams for release from in-vessel fuel, pool fires during shutdown, spent fuel movement accidents (including sodium pots and gas cooled casks), cover gas cleanup systems, and sodium purification systems were developed. However, this paper only discusses in-vessel releases, and therefore only the diagram depicting release from fuel in-vessel is presented here.

As part of the MST development completed at Argonne, a thorough review of domestic SFR operating experience and accidents was completed to help characterize the source term. That review effort is summarized in ref. 7, which accompanies this paper.

II. TRANSPORT PHENOMENA

This section provides a high level overview of the transport and retention phenomena affecting the release of radionuclides from in-vessel fuel. Phenomena related to the melting, relocation, freezing, and retention of fuel within the core are not discussed, but ref. 8 provides a detailed review of these issues. For a metal fuel SFR, there are five main barriers to the transport of radionuclides:

1) **Fuel Matrix**: Retention of radionuclides within the fuel matrix.
2) **Cladding**: Retention of radionuclides within the fuel pin.
3) **Primary Sodium**: Retention of radionuclides within the primary sodium.
4) **Primary Circuit Boundary**: Retention of radionuclides within the primary system (includes the primary sodium and cover gas region).
5) **Containment**: Retention of radionuclides within the containment building and guard vessel.

The following subsections step through each of these barriers and identify the phenomena that will likely be encountered by both radionuclide vapors/gases and particulates. A diagram depicting these phases can be found in Figure 1.

II.A. Retention within the Fuel Pin

As fuel burnup increases during normal operation, the creation of fission gases causes the fuel to swell and come into contact with the cladding. The fission gas pockets will interconnect and form passageways to the fission gas plenum, which in turn significantly slows further fuel swelling. Some fission products may migrate to the bond sodium, which is displaced to the fission gas plenum due to fuel swelling. Fission gases and vapors released from the fuel matrix must first travel through the bond sodium before reaching the fission gas plenum.

Elevated temperatures during an accident may cause eutectic penetration of the cladding to begin, with eutectic thinning rates of the cladding dependent on fuel temperatures and thermally-induced cumulative cladding stress. If fuel temperatures remain above the eutectic formation temperature for an extended period of time, cladding breach may occur. Creep in the cladding and excessive hoop stresses can also induce pin failure. However, to encounter fuel melting in areas of the fuel other than the eutectic regions, higher fuel temperatures would be required. If an accident causes extreme fuel pin temperatures with significant internal fuel melting, more rapid eutectic penetration and cladding failure is expected.

If a breach in cladding occurs, the radionuclide gases and vapors contained in the fission gas plenum will be released to the primary sodium, along with some fraction of bond sodium (and any fission products that have migrated to and dissolved in the bond sodium). If the fuel pin failed due to eutectic cladding penetration, only the eutectic regions of the fuel will be molten. Without significant additional fuel melting, there is likely to be minimal release from the fuel, due to the compatibility (*i.e.*, non-reactivity) between metal fuel and sodium.

Many radionuclides will be retained in the fuel matrix, as uranium acts as an excellent solvent. If additional fuel melting were to occur, due to a prolonged loss of cooling or a power spike that results in higher fuel temperatures, fuel material, including fission products retained in the fuel, may enter the coolant channel. This will result in a greater release of radionuclides into the primary sodium. However, once again, due to the compatibility of metal fuel and sodium, there is no chemical reaction between the two substances.

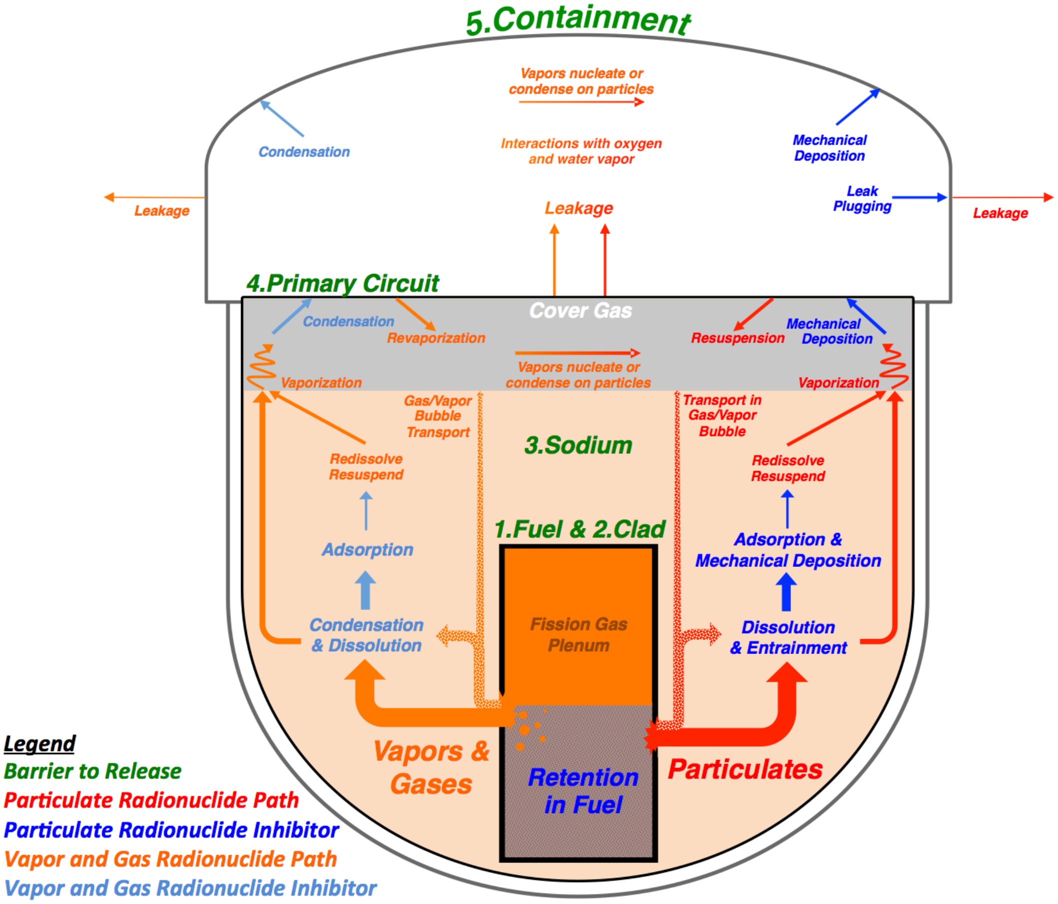

Fig. 1. Radionuclide transport phenomena for in-pin releases in primary sodium.

II.B. Retention within the Primary Sodium

Vapors and gases released from the fuel pin have differing fates depending on the vapor pressure and solubility of the particular element or compound in the sodium solution. Gases or vapors with high vapor pressure and low sodium solubility (e.g. noble gases) will be directly transported through the primary sodium and to the cover gas region. Some vapors may condense completely to the liquid phase once they come in contact with colder sodium and could dissolve, or they may nucleate within a bubble and be transported as aerosols. Other vapors will directly dissolve in the primary sodium from the gas phase due to high solubility of the element or compound in sodium. Dissolved vapors may also adsorb onto primary system structure, but this phenomenon is dependent on properties of the individual element or compound and the material of the primary system structures.

Particulates that are released from the fuel pin may become entrapped within a vapor or gas bubble. These particulates may settle on or migrate to the surface of the bubble and interact with the sodium; the same outcome will occur if the bubble collapses as the vapor condenses when colder sodium is encountered. The particulates may dissolve in the primary sodium or become entrained in the moving sodium stream. The particular phenomenon that will be encountered by the particulate depends on whether compounds are formed (*i.e.*, chemical reactions with the sodium) and the solubility of the element/compound in sodium. From there, adsorption on structure may occur, especially in lower temperature regions of the primary system where dissolved radionuclides may precipitate. Mechanical deposition within the primary system is also a possibility, especially for entrained particles.

For those radionuclides that have adsorbed onto structure, redissolution is possible if temperature changes occur in the primary sodium, as solubilities are typically highly dependent on temperature. If a radionuclide has mechanically deposited onto structure, resuspension may be possible is flow conditions adjacent to the structure change.

II.C. Behavior in Cover Gas

Those vapors and gases that were directly transported through the primary sodium in bubbles due to their high vapor pressure and low solubility (such as the noble gases) will be released to the cover gas region upon reaching the sodium surface. If any entrained particles are present within these bubbles, they may also be initially released to the cover gas region when the bubble bursts at the surface of the sodium pool.

The vapors and gases that dissolved in the primary sodium, along with dissolved particulates, must vaporize in order to escape the sodium. Barring any significant sodium boiling event, this can only occur through evaporation. The vapor pressure of a substance determines its ability to evaporate from a liquid; mixing in the sodium pool can greatly affect a substance's vapor pressure.

The cooler (relative to the sodium pool) thermal environment in the cover gas region can lead to the condensation of vapors on structural surfaces or onto particles. Mechanical deposition can again remove particulates from the cover gas region, as particles impact the various structures present, or settle back onto the sodium pool due to gravity. For highly volatile vapors or gases, such as the noble gases, condensation will not occur even in this cooler environment.

Lastly, resuspension/revaporization of some radionuclides is again possible if temperature changes occur in the cover gas region, as volatility typically increases with increasing temperature. Mechanically deposited particles could resuspend if a mechanical shock occurs on the structure where the particles are located, or if vapor flow increases adjacent to the structure.

II.D. Behavior in Containment

Airborne radionuclides may transport to the upper containment area through leakage paths in the reactor head. Typically, the reactor head has a design basis leak rate, which is very small (assumed to be on the order of 0.1% volume per day). Once in containment, some of the same phenomena associated with radionuclide transport in the cover gas region will occur, while additional phenomena are possible due to the presence of oxygen and water vapor.

First, much colder temperatures than the cover gas region will likely be found in containment, which will encourage additional condensation of vapors either onto the surface of structures, or onto aerosols (although the highly volatile noble gases will remain in the gas phase). Mechanical deposition will once again reduce any particles that managed to successfully transport from the cover gas region into containment.

Unlike the primary sodium or cover gas region, there is likely to be oxygen and water vapor in the containment, as it is typically not inerted to allow for maintenance activities. Leakage from the cover gas region means that sodium vapor in the cover gas region may also enter containment (the concentration of sodium vapor in the cover gas region is likely to be much higher than the concentration of radionuclides). The sodium vapor will react with the oxygen and water vapor to form aerosol particles. These particles will agglomerate and may remove vapors or particulates that have condensed or mechanically deposited on their surfaces. Other radionuclides may also react with the oxygen and water vapor and decompose to form new compounds.

The containment is expected to have a small design basis leak rate, similar to that of light water reactors (LWRs), which will permit some release of the radionuclides that have managed to remain airborne. The pressure within containment is unlikely to be elevated much beyond normal conditions (ambient atmospheric conditions), barring some other associated accident (such as sodium fire associated with the intermediate sodium piping). Unlike LWRs, the primary circuit of an SFR is near atmospheric pressure, meaning any failures in the primary system barrier do not result in the release of significant amounts of energy to containment. Leak plugging is another interesting phenomenon that may limit leakage from the containment. The aerosols formed in containment due to condensation and reactions with oxygen/water vapor may plug leakage pathways. As airflow through these leaks carries the aerosols to them, they can mechanically deposit around and in the leak causing a reduction in flow area.

III. CURRENT KNOWLEDGE STATE

The following section reviews the current state of knowledge regarding the phenomena of radionuclide transport for a core damage accident. The radionuclides are separated into representative categories, as is done in TID-14844 (ref. 9) and NUREG-1465 (ref. 10), the regulatory-recommended LWR deterministic source terms. However, the dominant radionuclide groups and isotopes may vary from what have historically been important for LWR source term considerations.

For each radionuclide group, the knowledge regarding the following factors is assessed:

- Behavior in the fuel pin.
- Behavior in sodium pool (solubility, mixing).
- Possible release from the sodium pool (vaporization).
- Aerosol behavior (in cover gas region and in containment).

Not every factor is reviewed for each radionuclide group, as some radionuclides will likely be retained by one of the earlier occurring mechanisms, and are unlikely to vaporize and reach the aerosol state. The current state of knowledge focuses on experimental results and theoretical models for the behavior of the radionuclide groups.

III.A. Noble Gases

The noble gases of interest, xenon and krypton, are formed from both the direct fission of uranium and through the decay of other fission products in the fuel matrix. During operation, fission gas bubbles containing xenon and krypton are created, causing the fuel to swell and internal pin pressure to rise as the burnup level increases. The fuel porosities begin to interconnect, forming pathways out of the fuel matrix, which allows the fission gas bubbles to travel through these new pathways and eventually reach the fission gas plenum[11].

Any breach of the cladding is likely to cause some fraction of the bond sodium to escape from the fuel pin and result in a pathway from the fission gas plenum to the primary sodium. As shown in Figure 2, fission gas release to the plenum increases substantially beginning with a fuel volume increase due to swelling of ~30%.

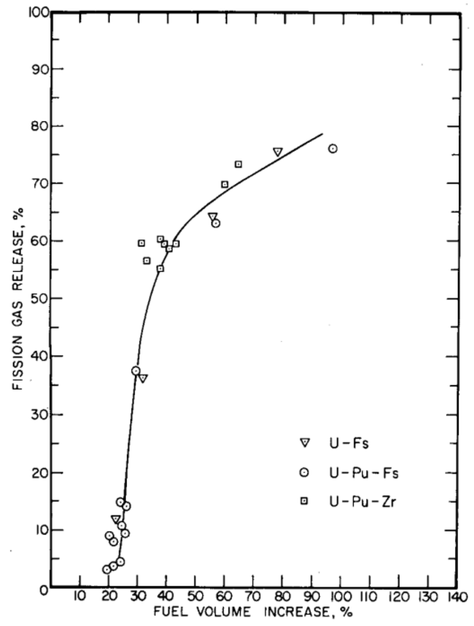

Fig. 2. Fission gas release to plenum versus fuel volume increase[12].

The magnitude of volume increase is correlated with the burnup level, but the exact composition and smear density of the fuel will also have an effect. However, as Figure 3 shows, it is likely that the majority of the xenon and krypton formed in the fuel pin will be released from the fuel pin during a cladding breach if burnup levels are beyond a few percent[12,13].

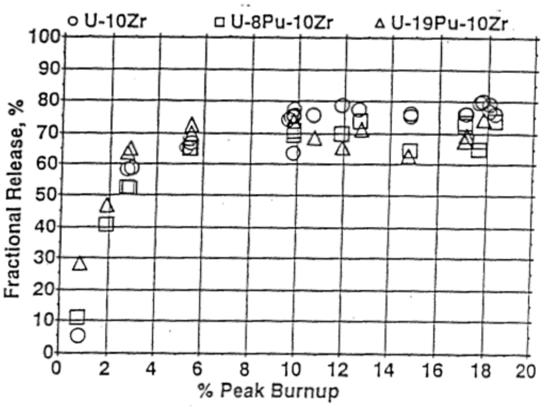

Fig. 3. Burnup dependence of fission gas fractional release[13].

Once xenon and krypton enter the primary sodium, their solubility obeys Henry's law[14]. Multiple data sources have demonstrated that the retention of xenon and krypton in sodium under conditions of the primary system is negligible[14,15], due to a high vapor pressure and a low solubility that is about a factor of 10^4 smaller than in water. Therefore it can be assumed that any xenon and krypton released into the primary sodium will reach the cover gas region. The time delay between release from the fuel pin and entrance to the cover gas will vary with reactor design, but has been estimated to be on the order of several minutes for previously constructed reactors[16].

Condensation of xenon and krypton in the cover gas region will not occur due to their high vapor pressures. Some xenon and krypton will escape the cover gas region, assuming a design basis leak rate from the cover gas region into containment. Once in containment, the noble gases will mix with the existing gas in the containment and escape to the environment according to the containment design basis leak rate.

It is important to account for the time delays that occur during the release of noble gases from the fuel to the cover gas region and into containment. Many of the radioactive isotopes of xenon and krypton have relatively short half-lives. This delay provides time for a reduction in the xenon and krypton radionuclide inventory. While transport within the primary sodium may only take several minutes, the retention of xenon and krypton in the cover gas region and within containment may be much longer.

III.B. Halogens

Iodine and bromine are the halogens usually assessed during source term development. However, due to their low fission-yield and short half-lives, isotopes of bromine are not likely to be a major contributor to the radiological source term. Iodine, however, is of interest and its form and behavior in metal fuel differs greatly from what is found in oxide fuels, such as those used in LWRs.

There are four isotopes of iodine that are likely to be formed in significant quantities in the fuel: ^{127}I, ^{129}I, ^{131}I, and ^{133}I. For radiological hazard reasons, the ^{131}I isotope is of primary interest in the development of an MST; while all isotopes of iodine collect in the thyroid, the longer half-lives combined with a relatively high fractional contribution to the total core inventory make ^{133}I an important MST contributors from the iodine group as well. They are formed almost entirely from the decay of fission products within the fuel[17]. The presence of elemental I_2 in the fuel is very unlikely since iodine will bond with other elements[18], and gaseous I_2 has not been seen in past reactors. In order of thermodynamic preference, iodine will form the following iodides: CsI, BaI_2, SrI_2, LaI_3, CeI_3, NdI_3, and UI_3.

Even though UI_3 is not the thermodynamically preferred iodide, the ratio of uranium to the other possible iodide elements within the fuel is so high (several orders of magnitude greater) that UI_3 is still likely to form and be a major factor in the retention of iodine within the fuel[16]. CsI is also likely to be found since cesium is produced at a faster rate than iodine and has thermodynamic preference compared to other fission products[18].

CsI may migrate to and collect in the bond sodium of the fuel pin, and during a cladding breach, may be released into the primary sodium. In sodium, CsI will decompose to form Cs and NaI[14]. NaI is a very stable compound of iodine.

Uranium, and therefore UI_3, will not be released from the fuel in any appreciable quantity following a cladding breach. Only if fuel melting is encountered can this iodine be released into the primary sodium to form NaI (ref. 15 & 19). Experimentation with melting of an U-Fs alloy in a crucible for four hours at 1250°C resulted in the release of only ~1% of the iodine from the fuel[20]. Only when held at higher temperatures (≥1300°C) for multiple hours were significant fractions of the iodine released from the fuel[20].

Once in the primary sodium, iodine will remain as NaI and not recombine with cesium[19]. Past experiments have shown that NaI will collect near a gas-liquid boundary, rather than disperse as a homogenous mixture[15]. This observation may influence vaporization rates from the sodium pool surface. Precipitation is unlikely to occur since the iodine concentration will likely be very low, and NaI stays in the liquid phase at lower temperatures when highly diluted in sodium[21,22].

NaI has a very low vapor pressure[23], and multiple studies and experiments have examined the vaporization of NaI (ref. 24, 25, 26, & 27). Equilibrium vaporization results from ref. 24 are shown in Figure 4. At typical primary sodium temperatures (~800 K) NaI vaporization lags the vaporization of sodium as a whole (*i.e.*, more than 50% of sodium needs to vaporize for approximately 10% of NaI to vaporize). The comparison to sodium vaporization provides an intuitive measure to gauge the vaporization of a radionuclide. Since sodium vaporization from the pool is usually very small (the fraction released to the cover gas is usually on the order of 10^{-4} - 10^{-6}), and NaI vaporization lags sodium, it gives an indication of the very small release fraction of NaI. Barring a significant sodium vaporization event, such as very high pool temperatures or bulk pool boiling, it is unlikely that significant quantities of NaI will vaporize from the pool[18].

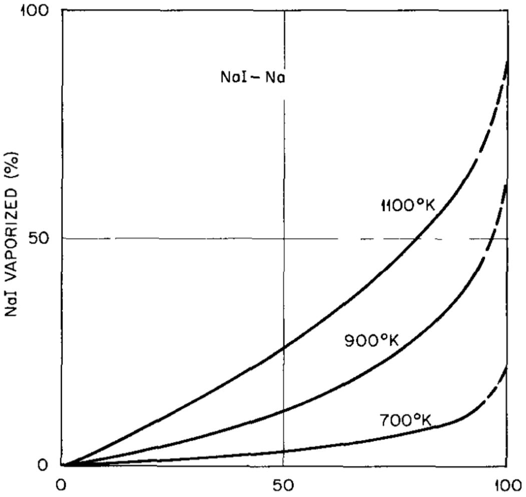

Fig. 4. Vaporization of NaI (ref. 24).
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Even if NaI were to vaporize, its very low vapor pressure would likely result in its condensation on cooler surfaces in the cover gas region[23]. If any vaporized NaI were able to reach the containment atmosphere before condensing, it could react with water vapor in the air to form molecular iodine[28].

III.C. Alkali Metals

Cesium and rubidium (Rb) are the alkali metals meriting strongest consideration for source term development, with the focus on the release and transport of cesium. The cesium isotopes formed in significant quantities include ^{133}Cs, ^{134}Cs, ^{135}Cs, ^{136}Cs, and ^{137}Cs. Due to its long half-life and high fission yield, ^{137}Cs is of particular importance for offsite release; ^{134}Cs is also an important MST contributor for short-term exposures, but is generally not considered to be a long term problem from the viewpoint of land contamination[15]. Both cesium and rubidium are highly reactive, like the fellow alkali metal sodium.

As described in the previous section, some cesium will form CsI within the fuel pin. However, since there is more cesium than iodine in the pin, not all cesium will form this bond and instead is likely to remain as elemental cesium and may be retained by the uranium[23]. Cesium that does escape the fuel matrix will collect in the sodium bond of the fuel pin, where it will decompose from CsI to form elemental cesium. Cesium is very miscible with the alkali metal of sodium. Due to this property, if the pin cladding is breached, the retention of cesium within the primary sodium is likely to be very high[16]. There has been some evidence that within sodium, cesium will collect at somewhat greater concentration near a gas-liquid boundary[15,16].

Both elemental cesium and rubidium have a relatively high vapor pressure when compared to sodium or NaI (ref. 29 & 30) and therefore are more likely to vaporize from the sodium pool. Figure 5 and Figure 6 show the equilibrium vaporization curves for both cesium and rubidium, with experimental results for cesium. As can be seen, the vaporization of both elements is greater than that of sodium.

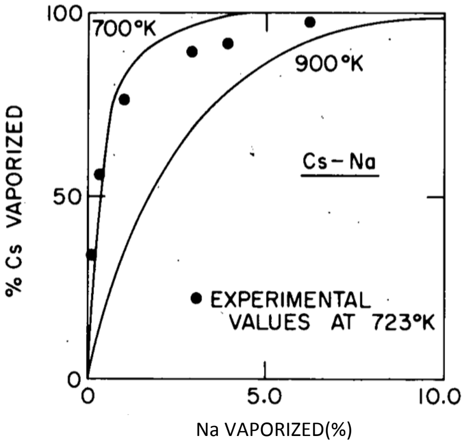

Fig. 5. Vaporization of cesium (ref. 24).
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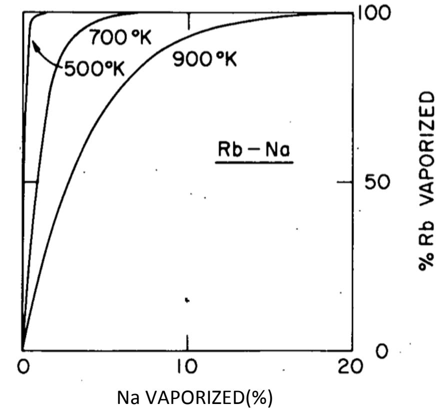

Fig. 6. Vaporization of cesium (ref. 24)
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Experimentation has shown a very small release fraction of cesium from a hot sodium pool, on the order of 10^{-4} - 10^{-5} (ref. 32). More recent analysis and experimentation on the vaporization of cesium has shown similar results[27,33,34], where the retention factor for cesium in a sodium pool increases as the temperature of the liquid surface increases. This phenomenon is a result of an increase in the vaporization of sodium with higher temperature, which means the ratio of vaporized cesium to sodium decreases[34]. Even though cesium is more volatile than sodium, very little vaporization of either element is likely to occur at expected accident sodium pool temperatures. The data appear to show cesium vaporization fractions of about one to two orders of magnitude greater than sodium vaporization, where sodium vaporization fractions are usually on the order of $10^{-4} – 10^{-6}$.

It is not expected that significant adsorption of cesium will occur in the primary system due to its high solubility in sodium[35]. If cesium were to vaporize to the cover gas region, past experimentation has shown rapid condensation on colder surfaces[32].

III.D. Tellurium Group

Tellurium (Te) and antimony (Sb), which have very similar chemical properties, are the usual elements of interest from the tellurium group. In oxide fuels, these elements migrate to the cold part of the fuel matrix (the cladding interface), where high release rates (>50%) are possible when oxide fuel melts. This is not the case for metal fuel. While little information is available on the behavior of tellurium and antimony in metal fuels, past accident data along with solubility information can provide insight into their behavior during accidents.

Of all the tellurium isotopes, ^{132}Te and ^{129m}Te are of most importance. This is especially true for ^{132}Te, which decays to ^{132}I, a strong gamma emitter[36]. In past experiments, melting U-Fs alloy in a crucible for three hours at 1400°C, which is well above the melting point for metal fuel, 98.1% of the tellurium was retained in the fuel[37]. This high retention is in agreement with past SFR metal fuel accidents, where no tellurium has been found released from the fuel, even with significant melting.

High solubility of both tellurium and antimony in sodium has been seen in past work[22]. Tellurium will form $NaTe_2$, while antimony will form $NaSb_2$ in the sodium[22]. High rates of tellurium plateout (adsorption) have been observed in past sodium reactors[22], as it has reacted with stainless steel surfaces, even at low temperatures (such as those seen in the primary sodium purification cold trap).

Tellurium has a low vapor pressure, and is expected to vaporize at a rate far below sodium or even NaI. As seen by the equilibrium vaporization curve in Figure 7, even with sodium vaporization nearing 100%, tellurium vaporization is expected to be well below 20% at expected sodium pool temperatures. Due to its low vapor pressure, even if some tellurium were to vaporize, it would be expected to quickly condense on colder surfaces. The vaporization of antimony is considered negligible, since the concentration will be orders of magnitude less than tellurium[24].

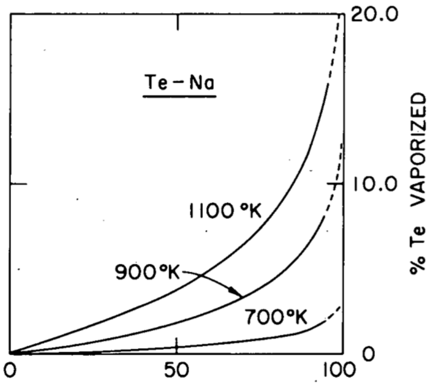

Fig. 7. Vaporization of tellurium (ref. 24).
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III.E. Alkaline Earths

Strontium (Sr) and barium (Ba), like the tellurium group, have noticeable differences in their behavior between oxide and metal fuel. In oxide fuel, both strontium and barium will form oxides that, when released, are not soluble in sodium[18]. However, the elemental states of both strontium and barium, which are more likely to occur in metal fuel due to the lack of oxygen, are soluble in sodium[18].

Experimental data on the release of strontium and barium from metal fuel is sparse but in past vented metal fuel concepts (which can be taken to represent fuel performance with cladding breach), strontium and barium have not been found released from the fuel pins during normal operation[15].

Even if strontium and barium are released to the sodium in their elemental forms, in low concentrations, both may react with oxygen impurities in the sodium and form oxides[15]. However, with larger releases, both strontium and barium will likely stay in their elemental form in sodium. Extensive plating and adsorption of strontium has been observed in experimentation[19] and in SFR accidents. The vapor pressures of strontium and barium are very low, which should result in very little vaporization from the sodium pool, as seen in the equilibrium vaporization curves in Figure 8 and Figure 9. Experimentation has seen vaporization values even lower than theoretically predicted (likely due to the formation of oxides with sodium impurities[15]).

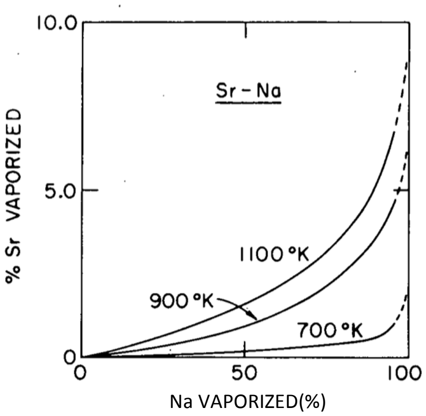

Fig. 8. Vaporization of strontium (ref 24).
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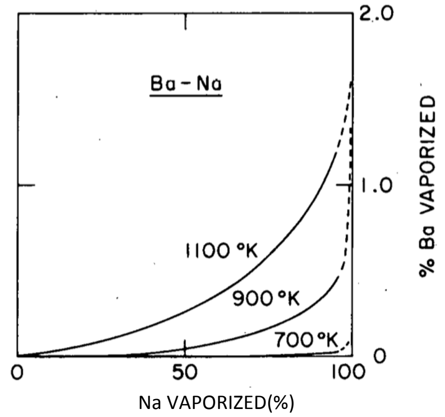

Fig. 9. Vaporization of barium (ref. 24).
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III.F. Noble Metals

Information on the behavior of the noble metals (ruthenium – Ru, technetium- Tc, molybdenum – Mo, palladium – Pd, rhodium – Rh, platinum – Pt, and silver – Ag) in sodium is less refined than some of the previous radionuclide groups. However, some general observations can be made, in conjunction with limited experimental evidence.

First, the melting points of the noble metals are much higher than the expected fuel temperatures, even during most accidents. Table I summarizes the melting points for the noble metals of interest. Due to the high thermal conductivity of metal fuel and the sodium coolant, it is unlikely that fuel temperatures will reach the melting point of the noble metals (with a possible exception for silver). However, these melting points may be lower when the elements are mixed with uranium, as is the case for ruthenium[38]. Even so, experimentation has not observed significant releases, as melting of U-Fs alloys in a crucible to temperatures of 1200-1400°C, which is well above the melting point of metal fuel, for five hours showed almost complete retention of Ru, Mo, Rh, and Pd within the fuel[37].

TABLE I. Noble Metal Melting Point

Element	Melting Point (°C)
Ru	2334
Tc	2204
Mo	2623
Pd	1555
Rh	1963
Pt	1768
Ag	961

The noble metals are not very soluble in sodium, with the exception of silver[22], and adsorption is likely to occur. Very high adsorption rates for ruthenium have been observed in experimentation (>90%) (ref. 16) and past reactor operation[22].

III.G. Rare Earths

Historically, cerium (Ce) and yttrium (Y) are the only rare earths of interest for the source term. Lanthanum is also present, but tends to accompany its parent element barium. In oxide fuels, the rare earths form soluble oxides in the fuel matrix[30]. In metal fuel, at high temperatures the rare earths, along with uranium and plutonium, will form a eutectic with the cladding[39].

While past experiments have investigated the release of rare earths from U-Fs alloy melt in a crucible[37], the presence of oxygen in the crucible caused the formation of rare earth oxides and is not indicative of metal fuel behavior during an accident. Cerium has a relatively low melting point (795 °C) but it increases when in solution with uranium[40]. The same is true for yttrium, which has a melting point of 1522 °C, when at very low concentrations in uranium[41].

The rare earths are not soluble in sodium in their elemental forms[18]. Experiments have shown high retention and adsorption (>80%) of cerium in sodium[15]. The elemental forms of the rare earths also have low vapor pressures[42], which would likely limit any possible vaporization from the sodium pool. In past SFR metal fuel accidents, cerium has been found in the primary system. However, very high retention rates have been observed due to adsorption on primary system structure.

III.H. Actinides

One of the main motivations behind the use of metallic fuel is its compatibility with the sodium coolant, as sodium is even used as the bond material within the pin. The compatibility between metal fuel and sodium also permits the use of vented fuel designs. This is in contrast to oxide fuel (UO_2, PuO_2), which due to the presence of oxygen, can react with sodium[22].

Data on the solubility of elemental uranium and plutonium in sodium are sparse, as most experimentation has focused on the release of UO_2 and PuO_2 from oxide fuels, which would occur more readily during cladding failure in oxide fuels. However, even elemental uranium and plutonium could form oxides in the sodium if oxygen impurities exist above 1/100 of a ppm in the sodium[43]. What data that does exist appear to show that the solubility of uranium and plutonium in sodium is very low[43,44].

Experimentation investigating the solubility of plutonium in sodium over a prolonged period[45] shows that when using metal fuel, it may be possible for some very small fraction of the plutonium in the fuel matrix to dissolve in the sodium bond during normal operation. If this were to occur, then any dissolved plutonium could be released to the primary sodium during a cladding breach.

IV. SUMMARY

For SFR core damage accidents, there are typically five barriers to release, fuel, clad, sodium, primary system boundary, and containment. While a review of source term phenomena has revealed that numerous complex mechanisms affect radionuclide transport in a core damage accident, literature indicates that there exists the potential for significant retention of all radionuclide groups other than noble gases. Past experiments demonstrate that for the most likely core damage accidents, only the noble gases will be released from the sodium pool in significant quantities. All other radionuclides appear to have very low release fractions ($<10^{-3}$), due to high retention rates in the fuel and primary sodium.

Subsequent vaporization to the cover gas region is very small and requires an extended period of time to occur. These phenomena, along with radionuclide retention within the fuel matrix, are important as they can significantly reduce the radionuclide inventory that is available for release from subsequent barriers in the defense-in-depth philosophy. These retention mechanisms may result in the magnitude of release from a core damage accident falling below the release levels for SFR accidents where radionuclides originate from releases outside the active core.

Any MST calculation seeking an accurate assessment of radionuclide release to the environment must attempt to understand and credit these transport and retention phenomena. Significant experimental experience has provided sufficient insight for most phenomena, where most of the remaining gaps can be resolved with accident and test histories[2,7]. Leveraging this information, an estimate of radionuclide release for most accidents that accounts for the various retention and transport phenomena may be developed, where bounding release fraction values may be utilized if necessary. This effort is continuing into FY16 with additional quantification of release fractions from fuel, followed by a trial MST calculation intended to identify any gaps in the codes and methods that are used in support of an MST assessment.

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