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The Development of a Realistic Source Term for Sodium-Cooled Fast Reactors: Assessment of Current Status and Future Needs

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Abstract

Sodium-cooled fast reactors (SFRs) continue to be proposed and designed throughout the United States and the world. Although the number of SFRs actually operating has declined substantially since the 1980s, a significant interest in advancing these types of reactor systems remains. Of the many issues associated with the development and deployment of SFRs, one of high regulatory importance is the source term to be used in the siting of the reactor. A substantial amount of modeling and experimental work has been performed over the past four decades on accident analysis, sodium coolant behavior, and radionuclide release for SFRs. The objective of this report is to aid in determining the gaps and issues related to the development of a realistic, mechanistically derived source term for SFRs. This report will allow the reader to become familiar with the severe accident source term concept and gain a broad understanding of the current status of the models and experimental work. Further, this report will allow insight into future work, in terms of both model development and experimental validation, which is necessary in order to develop a realistic source term for SFRs.

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Nomenclature

ABCOVE	Aerosol Behavior Code Validation and Evaluation
ABR	advanced burner reactor
ABTR	Advanced Burner Test Reactor
ACRR	Annular Core Research Reactor
AFCI	Advanced Fuel Cycle Initiative
ALMR/PRISM	Advanced Liquid Metal Reactor/Power Reactor Innovative Small Module
ANL	Argonne National Laboratory
AOO	anticipated operation occurrence
AST	alternative source term
ATR	advanced test reactor
ATWS	anticipated transients without scram
BDBE	beyond design basis event
BRISC	Burner Reactor Integrated Safety Code
BWR	boiling water reactor
CCDF	complementary cumulative distribution functions
CDA	core disruptive accident
CDF	core damage frequency
CFG	clad free gas
CRBR	Clinch River Breeder Reactor
CSTF	Containment Systems Test Facility
DBA	design basis accident
DBE	design basis event
DOE	Department of Energy
EBR	Experimental Breeder Reactor
ESF	engineered safety feature
FAST	Fuel Aerosol Simulant Test
F-C	frequency-consequence
FFTF	Fast Flux Test Facility
FP	fission product
GNEP	Global Nuclear Energy Partnership
HCDA	hypothetical core disruptive accident
HEDL	Hanford Engineering Development Laboratory
HEPA	high efficiency particulate air
IFR	integral fast reactor
INL	Idaho National Laboratory
LANL	Los Alamos National Laboratory
LBB	leak before break
LMFBR	liquid metal fast breeder reactor
LMR	liquid metal reactor
LOCA	loss of coolant accident
LOHS	loss of heat sink
LWR	light water reactor

MACCS	MELCOR Accident Consequence Code System
MSL	Mars Science Laboratory
NABRAUS	Natrium brand und Schwebstoffilter anlage
NRC	Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
PIRT	Phenomena Identification and Ranking Table
PNC	Power Reactor and Nuclear Fuel Development Corporation
PRA	probabilistic risk assessment
PWR	pressurized water reactor
QHO	quantitative health objectives
RF	retention factor
RSICC	Radiation and Safety Information Code Center
SBO	station blackout
SFR	sodium-cooled fast reactor
SGTR	steam generator tube rupture
SNL	Sandia National Laboratories
STCP	Source Term Code Package
TMI	Three Mile Island
TOP	transient overpower accidents
TREAT	Transient Reactor Test Facility
TUC	transient under cooling
ULOF	unprotected loss of flow
UTOP	unprotected transient overpower

1. Introduction and Background

1.1 Overview

Sodium-cooled fast reactors (SFRs) continue to be proposed and designed throughout the United States and the world. Although the number of SFRs actually operating has declined substantially since the 1980s, there remains a significant interest in advancing these types of reactor systems for several reasons. Some of these reasons include (1) closing the fuel cycle by fissioning actinides, allowing for a lower inventory of long-lived waste products to be stored in a repository, (2) the production of nuclear fuel by breeding, creating a virtually unlimited supply of nuclear fuel, and (3) the potential reduction of capital costs, by using modular construction and eliminating costly pressure vessels and piping and employing advanced power conversion systems, such as a supercritical CO_2 Brayton cycle.

Of the many issues associated with the development and deployment of SFRs, one of high regulatory importance is the source term to be used in the siting of the reactor. Source term is defined as the release of radionuclides from the fuel and coolant into the containment, and subsequently to the environment, following a severe reactor accident where a significant portion of the reactor core has melted. The radionuclides of interest include transmutation products, fission products, and actinides. Typically, the release of the fission product noble gases, halogens, volatiles, and semi-volatiles to the containment plays a major role in the downwind dose consequence from a proposed severe accident event. Mitigating the release of these fission products and actinides can have a significant effect on the downwind dose consequence. Therefore, the holdup of the radionuclides in the fuel, coolant, and containment following a severe accident will significantly affect the dose consequence to the public. The development of a realistic source term for SFRs based on mechanistic models that have been validated using benchmark experiments is needed.

A substantial amount of work has been performed over the past several decades to develop a realistic source term for light water reactors (LWRs). Both modeling and model validation using in-pile and out-of-pile experimentation have played a key role in developing models and computer codes that have gained the confidence of the United States Nuclear Regulatory Commission (NRC). These codes allow for improved modeling of severe accident phenomena for LWRs and the development of the NUREG-1465 (Ref. 1) source term.

Additionally, considerable modeling and experimental work has been conducted over the past four decades on accident analysis, sodium coolant behavior, and radionuclide release for SFRs. For the United States, this work resulted in support of the Clinch River Breeder Reactor (CRBR), which was authorized by Congress in 1970. Initial appropriations were provided in 1972. In 1977, however, President Jimmy Carter vetoed a bill that would have continued funding for the project. This lack of funding effectively halted the project for a short time. The project was temporarily revived under the Reagan Administration in 1981, but in 1983, partially because of ballooning costs to taxpayers, Congress terminated the project. Although some effort on SFR development has continued in the United States, most of the facilities used to perform fast reactor safety work and sodium coolant behavior have been shut down and decommissioned. Work has continued in France, Japan, and Russia.

The objective of this report is to aid in determining the gaps and issues related to the development of a realistic, mechanistically derived source term for SFRs. This report will allow the reader to become familiar with the severe accident source term concept and gain a broad understanding of the current status of the models and experimental work. Further, this report will allow insight into future work, in terms of both model development and experimental validation, which is required in order to develop a realistic source term for SFRs.

1.2 Background Information

The term sodium-cooled fast reactor is a subset of the more generic term liquid metal reactor (LMR) or liquid metal fast breeder reactor (LMFBR). A LMR could have a metal coolant of sodium, sodium-potassium, lead, or lead-bismuth. For this report, the thrust will be sodium coolant and, therefore, a SFR.

The majority of commercial nuclear power reactors around the world are thermal LWRs, and this is not likely to change in the near future. However, it is expected that at some time in the future, fast reactors will be necessary for breeding fuel and/or transmuting actinides. It is also hoped that fast reactors could be made cost competitive with LWRs by using advanced power conversion cycles and modular design and construction techniques. Recently, the Global Nuclear Energy Partnership (GNEP) program proposed the design and development of the Advanced Burner Reactor (ABR) (Ref. 2), which would be used to close the fuel cycle by fissioning actinides, thus allowing for a lower inventory of long-lived waste products to be stored in a repository.

Over the past five decades, many LMRs, in particular SFRs, have been proposed, designed, constructed, operated, and decommissioned around the world. The first liquid metal cooled fast reactor was CLEMENTINE, a mercury-cooled reactor built at Los Alamos, New Mexico in 1946 and operated at a power level of 25 kW. Other SFRs operated in the United States include Fermi, the Experimental Breeder Reactor I (EBR-I), the Experimental Breeder Reactor II (EBR-II), and the Fast Flux Test Facility (FFTF). SFRs that have been proposed in the United States include the Clinch River Breeder Reactor (CRBR), the Integral Fast Reactor (IFR), and the Advanced Liquid Metal Reactor/Power Reactor Innovative Small Module (ALMR/PRISM). A description of these and other LMRs proposed and operated around the world can be found in IAEA-1531 (Ref. 3) and IAEA-1569 (Ref. 4).

Many sodium test facilities of small-, intermediate-, and large-scale magnitude have existed in the United States and throughout the world to support the sodium coolant technology work required from the 1960s through the 1980s. Of these facilities, most located in the United States have been shut down and decommissioned. Some work continues at a modest level of effort in France, Japan, and Russia. A description of past and present sodium test facilities located in the United States and throughout the world, including their scale and the type of work performed, can be found in Sienicki and Grandy, 2007 (Ref. 5).

A significant amount of in-pile transient testing work was performed in the 1980s on both metal and oxide fuels (Refs. 6-13). The work was performed at both the Transient Reactor Test (TREAT) Facility at Idaho National Laboratories (INL) and the Annular Core Research Reactor

(ACRR) Facility at Sandia National Laboratories (SNL). Fuel failure testing and debris bed coolability were performed with and without sodium coolant in order to better understand the failure mechanics associated with hypothetical core disruptive accidents (HCDAs). No in-pile testing work has been performed using fast reactor fuel with the sole purpose of studying the source term for HCDAs in sodium coolant. LWR source term experiments were performed in the 1980s and 1990s in the ACRR (Ref. 14).

Models and computer codes were developed to perform accident analysis on SFRs, including anticipated transients without scram (ATWS) and HCDAs. Some of these codes included SAS4A/SASSYS-1 to analyze the accident sequences (Ref. 2; Refs 15-18) and SIMMER-II/III/IV to study the transitional phase of HCDAs (Ref. 19). Models and computer codes, including CONTAIN-LMR, were developed to perform release of fission products from the sodium pool to the containment, aerosol generation and plate-out, and sodium spray and pool fires (Ref. 20). The code requires the source term input for the calculation.

A significant amount of work was performed from the 1960s through the 1980s on liquid sodium chemistry, the release of fission products (in particular iodine) and uranium from liquid sodium, aerosol generation in sodium spray and pool fires, and the release of fission products under spray and pool fire conditions. Two documents summarized the extensive amount of work performed and the state of the technical knowledge base as of 1983. Morewitz (Ref. 21) and Randich (Ref. 22) both reference much of the same work and describe areas of future work to determine release fractions under more realistic conditions.

2. Objectives and Scope

2.1 General

The primary objective of this report is to determine the requirements for defining an accident source term for regulatory application for SFRs. The intent is to ascertain the current capabilities and deficiencies with respect to properly characterizing mechanistic source terms for SFRs. The source term should be expressed in terms of times and rates of appearance of radioactive fission products into the containment, the types and quantities of the species released, and other important attributes such as the chemical forms of iodine. For regulatory purposes, this mechanistic approach would, therefore, present a realistic portrayal of the amount of fission products present in the SFR containment from a postulated severe accident.

The mechanistic source term results should provide the same level of information as that generated for LWRs in NUREG-1465 (Ref. 1). However, there are some differences that should be considered in deriving a SFR mechanistic source term. For example, NUREG-1465 lists point estimates for the source term calculations. Due to the increase in computational capabilities in the years since NUREG-1465 was compiled, the analyses could include full distributions and quote quantile values of the resultant distributions for all calculations, thereby capturing all uncertainties in the development of the source term. This approach will dictate some of the suggestions for how to develop tools to aid in calculating the source term.

SFRs are significantly different from LWRs such that the same source term cannot be used for both reactor types. SFR cores are much smaller than LWRs. The active fuel length in a SFR core is typically one meter, with an extended gas plenum above the fuel pin region. SFRs have liquid sodium as the coolant and do not operate at significant pressures. A significant quantity of sodium extends above the core region. In the event of a leak or rupture of the reactor vessel or primary piping, the sodium does not vaporize since it is not at saturated conditions; however, it can ignite in the presence of air or concrete. Although similar fission products and actinides are generated in both reactor types, their chemistry with the sodium coolant and release to the containment are unique. Although it is expected that the noble gases would be immediately released to the containment in a SFR severe core disruptive accident, as for LWRs, the volatile halogens (iodine, bromine), alkali metals (cesium, rubidium), alkali earths (strontium, barium), and chalcogens (tellurium, selenium) are highly soluble in liquid sodium metal, some of which form soluble sodium compounds. These are released from the sodium coolant by vaporization from the surface of a pool or though aerosol production if the sodium is burning.

SFRs can vary by fuel form, fuel loading, and coolant flow paths. Many fuel forms have been proposed for SFRs, including oxide, metal, nitride, and carbide. The most typical forms are oxide and metal. Oxide forms, popular in Europe and Japan, may allow for the expertise with LWR fuel to be extended to SFRs. Metal fuels (where the metal is typically zirconium) could allow for more simple recycling of the used fuel.

Fuel loading depends on how the SFR will be used. As a breeder reactor, blanket regions will be included to generate Pu-239 from U-238, or U-233 from Th-232. As a transmuter/burner reactor, actinide fissioning will require the fuel to maintain significant additional inventories of recycled actinides. As a simple burner/converter reactor, a nominal fuel loading of U-235, U-233, or Pu-239 could be used with extended burnup as the goal.

The SFR reactor coolant system can be either loop type or pool type. Both have been considered and operated throughout the world. The impact on source term is that the HCDAs considered will be different and lead to different release scenarios, in much the same way that boiling water reactors (BWRs) and pressurized water reactors (PWRs) differ.

2.2 Accidents to Be Considered

In order to determine accident source terms for regulatory purposes, a range of severe accidents will need to be analyzed for SFR plants. Ideally, a report containing all available data on experiments related to SFRs should be compiled. Such a report would be comparable to NUREG/CR-5747 (Ref. 23), which documents the work done in support of NUREG-1150 (Ref. 24) and was used as the basis for NUREG-1465 (Ref. 1). NUREG-1465 defines an accident source term for LWRs. In order to develop a realistic source term for SFRs, a suite of codes is likely to be needed. The work for NUREG-1465 used the integrated Source Term Code Package (STCP) (Ref. 73) and MELCOR (Ref. 74).

A key decision to be made in defining an accident source term is the severity of the accident or group of accidents to be considered. Footnote 1 to 10 CFR Part 100 (Ref. 25), in referring to the postulated fission product release to be used for evaluating sites, notes that “Such accidents have generally been assumed to result in substantial meltdown of the core with subsequent release of appreciable quantities of fission products.” Possible choices for a realistic source term range from (1) slight fuel damage accidents involving releases into containment of a small fraction of the volatile nuclides such as the noble gases, (2) severe core damage accidents involving major fuel damage but without reactor vessel failure or core-concrete interactions, similar in severity to the Three Mile Island (TMI) accident, or (3) complete core-melt events with core-concrete interactions. These outcomes are not equally probable. Since many reactor systems must fail for core degradation with reactor vessel failure to occur and core-concrete interactions to occur, one or more systems may be returned to an operable status before core melt commences. Hence, past operational and accident experience, together with information on plant designs and a vigorous program aimed at developing accident management procedures, indicate that complete core-melt events resulting in reactor vessel failure are considerably less likely to occur than those involving major fuel damage without reactor vessel failure. These accidents, in turn, are less likely to occur than those involving slight fuel damage.

It is important to note that the purpose of this report is to define how a realistic source term should be developed for SFRs and identify gaps in current abilities for developing such a source term. Therefore, the approach suggested is to use data from experiments complemented by expert judgment and elicitation in order to develop distributions with a best estimate central value and all associated uncertainties for each phase of the modeling process. By combining the latest statistical techniques with validated phenomenological modeling, this process can produce the most informative results by which decisions can be made for the sake of safety calculations. The source term to be calculated will give information that can be used to make both best-estimate and conservative calculations for siting issues. Appropriate decisions can then be made by choosing the level of conservatism that is needed for the specific calculation by requiring a predetermined level of confidence in the results. Linking such a source term to consequence assessment software would be relatively straightforward. The output from these codes can then be compared to the quantitative health objectives (QHOs) that are required to be met. This type of integrated risk assessment is being utilized currently in the assessment of source terms for the Mars Science Laboratory (MSL) program at SNL (Ref. 26).

For LWRs, the NRC allows an applicant to propose changes in source term parameters from those specified in NUREG-1465 (Ref. 1), as long as these changes are based upon and justified by design-specific features (Ref. 1). This should be taken into account when developing a strategy for calculating a SFR source term.

NUREG-1465 (Ref. 1) also states that credit will be allowed for removal or reduction of fission products within containment via engineered safety features such as filters, as well as by natural processes such as aerosol deposition. The ability to model this should also be taken into account when developing capabilities to determine SFR source terms.

2.3 Limitations

Limiting factors for source term development could include burnup range, reactor coolant system type (loop or pool), fuel form (oxide or metal, Ref. 71), and loading (quantities of actinides). It is envisioned that the source term would be developed for one reactor system type initially and then extended to other systems using the same modeling process.

2.4 Margins and Uncertainties

NUREG-1465 (Ref. 1) lists five major categories of uncertainties. They are:

1. Accident severity and type
2. Onset of fission product release
3. Release phase durations
4. Composition and magnitude of releases
5. Iodine chemical form

These uncertainties are discussed in Chapter 4 of NUREG-1465 (Ref. 1). All five of these categories should also be assessed for SFRs. However, the uncertainty quantification should be an integral part of the analysis and should be presented in the form of frequency-consequence (F-C) curves and complementary cumulative distribution functions (CCDFs). In this manner, the central values (median, mean, etc.) for all calculations can be presented simultaneously with the uncertainty bands that accompany them. A well-informed decision can be made on the basis of the entire distribution.

3. Accident Source Terms

The first prescribed source term and the currently applied methodology present informative insights to aid in developing the framework for a deterministic SFR source term. Early analyses of "severe accidents" for reactor siting were performed using an instantaneous release to containment with prescribed core inventory percentage, 100% of noble gases, 50% of iodine, and 1% of solid fission products, presented in TID-14844 (Ref. 27). This early approach demonstrates little understanding of the evolving radionuclide states that transpire during a radiological accident. General improvements came during later analyses where the TID-14844 (Ref. 27) source was adapted with a probabilistic scope to include potential containment bypass and failure mechanics culminating in the WASH-1400 reactor safety study (Ref. 28). Current operating reactors were licensed with either of these two analyses or are receiving license renewals through the application of NUREG-1465, an applied methodology for source term analysis for reactor siting purposes which best describes the current regulatory approach to severe accident analysis.

NUREG-1465 (Ref. 1) was produced from research and code packages funded to investigate severe accident source terms after the TMI accident, partially replacing TID-14844 for severe accident analysis. NUREG-1465 (Ref. 1) produces a methodology to investigate postulated accidents by presenting release rates for materials of interest to radiological consequences for various release phases (coolant, gap, melt, ex-vessel, late in-vessel). These prescribed release rates result from mechanistic analyses of various postulated accidents from several reactor facilities, which can be applied to the chronology of accident events to produce an in-containment source term. Assuming a reactor is well represented by the facilities and accidents presented in the studies, the predetermined release rates may be used to aid in postulated accident considerations for equipment residing in containment, safety system mitigation performance, and the eventual off-site radiological and control room dose calculations.

An alternative source term (AST) to the proposed LWR severe accident source term presented in NUREG-1465 (Ref. 1) must meet the following requirements as described in Regulatory Guide 1.183 (Ref. 29):

- The AST must be based on major accidents, hypothesized for the purposes of design analyses or consideration of possible accidental events, which could result in hazards not exceeded by those from other accidents considered credible. The AST must address events that involve a substantial meltdown of the core with the subsequent release of appreciable quantities of fission products.
- The AST must be expressed in terms of times and rates of appearance of radioactive fission products released into containment, the types and quantities of the radioactive species released, and the chemical forms of iodine released.

- The AST must not be based upon a single accident scenario but instead must represent a spectrum of credible severe accident events. Risk insights may be used, not to select a single risk-significant accident, but rather to establish the range of events to be considered. Relevant insights from applicable severe accident research on the phenomenology of fission product release and transport behavior may be considered.
- The AST must have a defensible technical basis supported by sufficient experimental and empirical data, be verified and validated, and be documented in a scrutable form that facilitates public review and discourse.
- The AST must be peer-reviewed by appropriately qualified subject matter experts. The peer-review comments and their resolution should be part of the documentation supporting the AST.

The development of a SFR severe accident source term must meet the requirements above, which would require analyses of postulated accidents/bounding events to establish necessary release characteristics to satisfy the AST requirements.

The past analyses of the several postulated accidents were performed in NUREG-1150 (Ref. 24) to produce the above source term through the employment of lumped parameter codes. The AST requirements and tools applied in developing the current LWR source term suggest a similar code suite be developed to analyze the postulated events provided from probabilistic risk assessment (PRA) analyses.

3.1 PRA Selection of Bounding Events

A complete PRA aids in identifying design basis events (DBEs) and beyond design basis events (BDBEs), as well as assessing the frequencies and consequences associated with said events. A DBE, as defined in NUREG-0800 (Ref. 30), is a "condition of normal operation, including anticipated operational occurrences (AOOs), design-basis accidents, external events, and natural phenomena, for which the plant must be designed to ensure functions of safety-related electric equipment that ensures the integrity of the reactor coolant pressure boundary; the capability to shut down the reactor and maintain it in a safe shutdown condition; or the capability to prevent or mitigate the consequences of accidents that could result in potential offsite exposures." Since the condition of the operating reactor system can be common among various events, bounding events must be selected to minimize the necessary analyses of DBEs. Deterministic judgment employing risk-informed insights from PRA is prescribed in the selection of limiting events, which are typically categorized as either normal operation, AOOs, or postulated accidents dependent on frequency of occurrence as well as the consequence of the event. Since normal operations have relatively no consequence, no discussion is presented. AOOs must abide by dose guidelines and are discussed briefly. Postulated accidents represent the reactor states capable of producing severe consequences and are the primary focus of this report.

Initiating events whereby the frequency of occurrence is such that the event is anticipated to occur within the lifetime of the reactor are known as AOOs. In anticipation of such events, a reactor is designed to maintain fuel and reactor coolant system integrity to ensure that accidents do not advance to postulated accidents and that the site boundary dose is within a small percentage of the 10 CFR Part 100 (Ref. 25) guidelines. Source term determinations for these anticipated conditions are less dependent on melt events. Examples include rod loading accidents, coolant system activity releases, minor fuel failure events, etc. As such, DBEs of this nature, being less sensitive to fuel behavior, lend themselves to mechanistic analysis more readily than postulated accidents.

Postulated accidents, unlike AOOs, are not anticipated to occur within the lifetime of a reactor and, therefore, are often labeled as BDBEs; however, postulated accidents are still considered in safety-related equipment qualification, safety system performance analyses, and reactor siting. Postulated accidents, commonly referred to as severe accidents, may involve significant core disruption and be classified as HCDAs. The resulting consequences of HCDAs have a strong dependency on the fuel performance. Therefore, fuel performance must be well understood for both normal and off-normal conditions to support the use of a mechanistic analysis (Ref. 31). The additional fuel dependency (core inventory, gap inventory, fission product chemical and physical properties, etc.) increases the complication of determining a mechanistic source term for postulated accident considerations. Prior safety analysis reports have been submitted for LMR designs. The bounding events selected for the PRISM design by the NRC staff and vendor (General Electric Co.) have been placed in Table 1, the details of which may be found in SECY-93-092 (Ref. 31).

Table 1. Bounding Events.

Event		Description
UTOP	Unprotected Transient Overpower	The most severe insertion event (typically a control rod withdrawal accident) where rod drive control is unavailable
LOHS	Loss of Heat Sink	Heat removal through conventional means is lost (intermediate loop no longer removes heat)
ULOF	Unprotected Loss of Flow	Pump failure removes forced convection and rod drive control is unavailable
SBO	Station Blackout	Scram and natural circulation occur
SGTR	Steam Generator Tube Rupture	A justifiable number of steam generator tubes rupture
Large Intermediate Loop Break		Intermediate piping break
Flow Blockage		Blockage of flow near a fuel assembly
External Events		Consistent with those considered for LWRs

It should be noted that with the recent adoption of passive safety features, system thermal-hydraulic analyses require significant coupling between containment and the reactor coolant system. This has the potential to complicate the code development for system response analysis.

3.2 Fission Product Transport

Fission product release occurs when safety barriers fail. The physical safety barriers in the nuclear facility include the fuel clad, reactor coolant system boundary, and/or containment, which are necessary to provide redundant protection of public health. When determining how and when barriers are defeated, system dependencies become prevalent in determining the failure criteria, likelihood, and variability associated with different accident progressions. While there is no means to address system dependencies, general anticipations are discussed below. Ascertaining the accident that will result in the barrier defeat is not possible at this time, given that a reactor system has yet to be proposed for analysis; rather barrier defeat is simply assumed, allowing general discussion of the figures of merit. The system activities and co-interactions are discussed below for the following:

1. Coolant Radioactive Inventory Release
2. Clad Free Gas Volume Radioactive Inventory Release
3. Early In-Vessel Release
4. Ex-Vessel Release
5. Late In-Vessel Release

3.2.1 Coolant Activity

In design basis accident (DBA) analysis the coolant activity would not include the effects of core disruption; therefore, coolant activity levels are associated with normal operation coolant activity. The coolant activity release is presented solely as a release to containment. The reactor coolant activity is a product of the local fault of fuel pins causing fission products and fuel to be dissolved and the following relocation throughout the reactor coolant system, which occurs in addition to impurity as well as the sodium coolant activation during reactor operation.

Should the reactor coolant boundary be lost, a release of coolant and its associated radioactive inventory to the containment would result. The radioactive inventory in the coolant includes radioisotopes that are produced by activation of the stable isotope Na-23 by neutron absorption. Na-24, which decays by (β,γ) emission with a half-life of 14.96 hours, can be produced by the (n, γ) reaction. Na-22, which decays by (β^+/γ) emission with a half-life of 2.6 years, can be produced by the (n, 2n) reaction. Some fission products also can be expected to be in the coolant from leaking fuel pins. The coolant release into the containment could be in the form of a spray that could ignite in an air atmosphere, as discussed in Section 3.6. This type of failure would be more appropriate for loop-type systems as opposed to pool-type systems. For the primary coolant to escape a pool-type system, the reactor vessel would have to leak or rupture; however, the possibility of a loss of coolant is potentially impacted with the inclusion of guard vessels for both pool-type and loop-type configurations.

In beyond design basis and severe accidents the coolant activity continuously changes with core disruption. The failure of the cladding barrier due to loss of heat removal or system control allows fuel pin discharge into the coolant, altering the coolant activity. Large-scale clad failures occur due to environmental stressors (radiation damage, high temperatures, pressure differentials etc.) which alter the physical properties of the clad or the strain experienced. Given a clad failure, the release results in fission product vapor and molten fuel injection (fuel type dependent), fuel-coolant interactions, dissolution of fuel, thermal stress on fuel particulates, mode of fuel transportation (melting, slumping, and sweeping), and eventual debris bed formation. The various fuel states and the local coolant conditions throughout the primary coolant system present rather difficult thermo-chemical problems when analyzing severe accidents given the large range of thermal conditions.

Tracking fission products throughout the accident raises the need to analyze two-phase sodium conditions. Various states will produce varying localized aerosol physics; therefore, the primary system phases and flow conditions impact the removal and re-suspension of aerosols throughout severe transients.

3.2.2 Clad Free Gas Volume

The clad free gas (CFG) volume release is discussed in the context of the release from the fuel into the reactor coolant. Various mechanisms affect the release of fission products from a fast reactor fuel (metal or oxide) to the CFG. The primary contribution to the relocation of fission gas from the fuel into the CFG is the diffusion of gases to the grain boundaries. Gases within the fuel form bubbles, both within the grains and the grain boundaries, although the energy restricting bubble growth within a grain is significantly larger. Thus, the ability for bubble interconnection in the grain is not quite as significant as for grain boundaries, but may still contribute through coalescing until contacting a grain boundary or free surface. The high porosity level expected for either fuel type, oxide or metal, results in interconnection of the bubbles, primarily at the grain boundary, to the surface of the fuel, thereby allowing virtually all fission gas to escape from the fuel to the CFG space. The fission product inventory and location must be determined to establish release rates from failed fuel pins.

The nature of a fuel pin failure will impact the fuel pin release mechanisms. Whether a cladding failure is due to molten fuel penetration or clad strain results in different fractional releases of fuel and gaseous species into the coolant, which is also dependent on the fuel. The percentage of released fuel and fission gas into coolant is established by the percent of fuel pins that have been ruptured and the pressure differential between the fuel pin and coolant. Transient analyses for the various accidents will permit pin and coolant conditions to be established and the initiation of the failure events and severity with respect to duration to be determined. The IFR program and others have initiated investigation into predicting failure times and fuel property evolution, providing good general agreement with experimental data during validation. The new fuel element composition suggests potential areas for further investigation to assure adequate fuel pin analysis is provided.

Another release identified here for completeness, which is not directly discussed in the NUREG-1465 (Ref. 1), is diffusion release. Diffusion release occurs between the time of clad failure and fuel melt via diffusion of fission products to the vapor volume and ultimately releases to the coolant stream through a clad failure site.

3.2.3 Early In-Vessel

The early in-vessel release phase encapsulates the release occurring during the melt progression and ends if/when the lower reactor vessel head ruptures. This phase would include significant fuel degradation with loss of fuel geometry, melting of the fuel and cladding, and slumping of the core materials to the bottom of the reactor vessel. The assumption is that all of the noble gases are now released from the fuel with a significant fraction of the volatiles and some of the semi-volatiles and other groups. The amounts released into the containment during the early in-vessel release are strongly influenced by the residence time within the reactor coolant system. Plateout of volatile and semi-volatile nuclides on walls and structures can add significantly to the residence time and retention factor.

Dissolution and vaporization of the chemical species produced from the fuel/stainless steel system dictate the relocation of the fission products and are dependent on the thermo-physical properties, flow rate, and time. Liquefaction processes, fuel clad chemical interactions, and coolant physical and chemical interactions yield a complicated subset of problems to physically represent for detailed fission product tracking. The various fuel states expected throughout the in-vessel phase, including fuel dispersion and crust formation, will also warrant consideration.

The inclusion of sodium coolant produces additional chemical forms which must be considered when analyzing fission product transport as well as dissolution and liberation of radionuclides from insoluble or nonvolatile compounds. The same considerations should be made for stainless-steel effects. The chemical form, physical form, and solubility of the fuel inventory in the sodium coolant play a major role in the release fraction. The transportation of fission products to the cover gas, if incorporated in the design, would need to be detailed for accidents involving releases of cover gas should they be relevant.

3.2.4 Ex-Vessel

Failure of the reactor vessel would allow for ex-vessel release of the coolant and inventory. A number of accident scenarios and phenomenological conditions can be envisioned for ex-vessel release of the sodium coolant, molten fuel, and debris bed. The lower portion of the reactor vessel could fail due to heating and melt through, causing a significant quantity of the coolant and fuel to be placed within the reactor vessel and the guard vessel or containment vessel. Burning of the sodium coolant could proceed if exposed to air or if contact with concrete results. The reactor vessel or upper head portion of the reactor vessel could be breached due to mechanical energy deposition. Mechanical energy could be produced by over-pressurization due to thermal considerations, positive void coefficient effects, or recriticality of the debris bed as the core is redistributed in the vessel. A breach of the upper head could allow for sodium pool burning of expelled sodium, or from that left in the vessel if exposed to air or concrete.

3.2.5 Late In-Vessel

Simultaneously, and generally with a longer duration, the late in-vessel release phase would release some of the volatile and semi-volatile nuclides that had deposited within the reactor coolant system into the containment.

3.3 Duration of Release Phases

NUREG-1465, “Accident Source Terms for Light-Water Nuclear Power Plants,” (Ref. 1) lists estimated durations for LWR release phases discussed in Section 3.2. The ability to provide this type of estimate for SFRs, both loop-type and pool-type, will be needed in order to provide a mechanistic source term. The capability to model either metal or oxide fuel employed in the final reactor design is also necessary.

Various mechanisms affect the release of fission products from a fast reactor fuel (metal or oxide) to the gap. The primary contribution to the relocation of fission gas from the fuel into the gap space is the diffusion of gases to the grain boundaries. Gases within the fuel form bubbles, both within the grains and the grain boundaries, although the energy restricting bubble growth within a grain is significantly larger. Thus, the ability for interconnection of the bubbles formed in the grain is not quite as significant as for grain boundaries, but may still contribute through coalescing until contacting a grain boundary or free surface. The high porosity level expected for either fuel type results in interconnection of the bubbles, primarily at the grain boundary, to the surface of the fuel, allowing virtually all fission gas to escape from the fuel to the gap space.

In order to determine whether a particular DBA represents a reasonable scenario upon which to base the timing of initial fission product release into the containment, PRAs for SFRs need to be reviewed to determine the contribution to core damage frequency (CDF) resulting from said DBA. Developers of any codes for calculation of SFR source terms need to be careful to consider all appropriate information relevant to the calculation. For example, both loss of coolant accidents (LOCA) and leak before break (LBB) scenarios need to be included to account for the fact that some plants may have LBB approval while other plants do not.

In order to provide a realistic estimate of the time for fuel rod failure, a code suite with the ability to perform calculations for the particular type of SFR will need to be developed. The code must be able to perform calculations for multiple types of accidents in order for the results to be considered realistic.

In the following sections, the different release phases of LWRs are described, including the associated phase durations. It is not known how these durations will compare to SFR phase durations. In fact, it is not known if the phases will even be the same.

3.3.1 Coolant Activity Release

The coolant activity release begins with a postulated pipe rupture and ends when the first fuel rod fails. During this phase, the activity released to the containment atmosphere is that associated with very small amounts of radioactivity dissolved in the coolant itself. For LWRs, this duration is less than one minute.

3.3.2 Gap Activity Release

The gap activity release phase begins when fuel cladding failure commences. This phase involves the release of radioactivity that has collected in the gap between the fuel pellet and cladding. This will be of concern mainly for ceramic fuels.

3.3.3 Early In-Vessel Release

During the early in-vessel release phase, the fuel and other structural materials in the core reach sufficiently high temperatures that the reactor core geometry is no longer maintained and fuel and other materials melt and relocate to the bottom of the reactor vessel. During this phase, significant quantities of the volatile nuclides in the core inventory as well as small fractions of the less volatile nuclides are estimated to be released into containment. This phase ends when the vessel fails, allowing molten core debris to fall onto liner or concrete below the reactor vessel. Release durations during this phase will vary depending upon accident sequence and reactor design. For LWRs this duration is on the order of one to 1.5 hours.

3.3.4 Ex-Vessel Release

The ex-vessel release phase begins when molten core debris exits the reactor vessel and ends when the debris has cooled sufficiently that significant quantities of fission products are no longer being released. During this phase, significant quantities of the volatile radionuclides not already released during the early in-vessel phase as well as lesser quantities of non-volatile radionuclides are released into containment. For LWRs this duration is on the order of two to three hours.

3.3.5 Late In-Vessel Release

The late in-vessel release phase commences at vessel breach and proceeds simultaneously with the occurrence of the ex-vessel phase. However, the duration is not necessarily the same for both phases. During this release phase, some of the volatile nuclides deposited within the reactor coolant system earlier during core degradation and melting may re-volatilize and be released into containment. For LWRs this duration is on the order of ten hours.

3.4 Fission Product Composition and Magnitude

In analyzing severe accidents WASH-1400 (Ref. 28) examined the spectrum of fission products and grouped fifty-four radionuclides into seven major groups on the basis of similarity in chemical behavior. The work done to support NUREG-1465 (Ref. 1) revised this study and produced thirty-one radionuclides that are divided into eight groups. This work will need to be revisited for SFRs. Of particular interest will be fission product and actinide chemical interaction with the sodium coolant, and the sodium coolant chemical interactions with air and concrete.

The eight groups from NUREG-1465 (Ref. 1) are shown in Table 2. Table 3 presents the radionuclide groups as suggested by Powers (Ref. 32). The bold elements in both tables represent the most important elements found in Appendix A. The importance rankings are based on the elements' downwind dose effects for an unmitigated release.

Table 4 shows the relative importance of the fission product and actinide elements that contribute to downwind dose compared to the noble gases. For each chemical group and the actinides, the relative ratio of the group's downwind dose contribution is compared to the noble release. The calculation used for comparative purposes is the same as that described in Appendix A. The parenthetical values following each element name is the percentage of that element contributing to the dose. The fourth column in Table 4 shows the comparison to the noble gases for an unmitigated total release. The sixth column shows the same comparison, but using the total release fractions for a BWR given in NUREG-1465 (Ref. 1). The results show that in order to model the release fractions accurately, the chemical forms and the release modes for iodine, cesium, tellurium, antimony, strontium, barium, ruthenium, molybdenum, yttrium, zirconium, lanthanum, niobiom, promethium, praseodymium, neodymium, cerium, plutonium, neptunium, americium, and curium must be well understood.

After applying the release fractions from NUREG-1465, iodine, tellurium, cesium, and strontium are still found to be the dominant contributors to the downwind dose consequence. Therefore, understanding the factors and details relating to the release of these elements will be important for the SFR source term development.

For elements and compounds that are soluble in sodium, their release mechanism from the coolant to the containment will be primarily by vaporization from the pool surface, especially at sodium coolant temperatures less than the boiling point of sodium. Different models can be used to determine the release of fission-product from the pool through vaporization, including the Rayleigh distillation method, Raoult's Law, or other semi-empirical methods (Ref. 22).

Table 2. Chemical Grouping from NUREG-1465 (Ref. 1).

Group Number	Group Title	Elements in Group
1	Noble Gases	**Kr**, **Xe**
2	Halogens	Br, **I**
3	Alkali Metals	**Cs**, Rb
4	Tellurium Group	**Te**, **Sb**, Se
5	Alkaline Earths	**Ba**, **Sr**
6	Noble Metals	**Ru**, Rh, Pd, **Mo**, Tc, Co
7	Lanthanides	**La**, **Zr**, **Nd**, Eu, **Nb**, **Pm**, **Pr**, Sm, **Y**, Cm, Am
8	Cerium Group	**Ce**, **Pu**, **Np**

Table 3. Chemical Grouping from Powers (Ref. 32).

Group Number	Group Title	Elements in Group
1	Noble Gases	**Kr**, **Xe**
2	Alkali Metals	**Cs**, Rb, Na, K
3	Alkaline Earths	**Ba**, **Sr**
4	Halogens	Br, **I**
5	Chalcogens	**Te**, Se
6	Platinoids	Pd, Rh, **Ru**
7	Early Transition Metals	**Mo**, Tc, **Nb**
8	Tetravalents	**Ce**, **Zr**, Th, **Np**
9	Trivalents	**La**, **Pm**, Sm, **Y**, Eu, Gd, **Pr**, **Nd**, **Pu**, Cm, Am
10	Uranium	U
11	Main Group Metals	Cd, In, Sn, **Sb**, Ag, Ge, Bi, Pb
12	Boron	B, Si, P

Table 4. Elements Contributing to Downwind Dose Compared to Nobles using NUREG-1465 Fractions.

Group Number	Group Title	Elements in Group	Group Importance Relative to Nobles – Unmitigated	NUREG-1465 Total Release Fraction	Group Importance Applying NUREG-1465 Release Fractions
1	Noble Gases	**Kr(60)**, **Xe(40)**	1.0	1.0	1.0
2	Halogens	Br, **I(99.5)**	7.1	0.6	4.3
3	Alkali Metals	**Cs(87)**, Rb(13)	1.0	0.6	0.60
4	Tellurium Group	**Te(91)**, **Sb(9)**, Se	5.0	0.3	1.5
5	Alkaline Earths	**Sr(70), Ba(30)**	5.8	0.12	0.70
6	Noble Metals	**Ru(90), Mo(8)**, Rh, Pd, Tc, Co	13.7	0.005	0.07
7	Lanthanides	**Y(30), Zr(28)**, **La(19)**, **Nb(9)**, **Pm(6)**, **Pr(6)**, **Nd(2)**, Sm, Eu	24.2	0.0055	0.13
8	Cerium	**Ce(100)**	43.4	0.0052	0.23
	Actinides	**Pu(87), Np(13),** Am, Cm	49.8	0.0052	0.26

Table 5 gives the vapor pressure for the volatile and semi-volatile fission-product elements that are important to the downwind dose in order from higher to lower volatility. Some of these elements can form other compounds with sodium or oxygen. Elements that form compounds with sodium, NaI for example, will have lower vapor pressures than the elemental form. Both the vapor pressure and the solubility in sodium at different temperatures play significant roles in the vaporization of fission products from non-burning sodium pools. Chemical modeling of vaporization of soluble elements in sodium pools can be found in Schram (Ref. 33).

Table 5. Vapor Pressures for Important Fission Product Elements.

Element	Vapor Pressure at 600°C (atm)	Vapor Pressure at 900°C (atm)
I	2.2E+2	1.4E+4
Cs	4.4E-1	4.3E+0
Na	3.1E-2	1.0E+0 (boiling point)
Te	8.2E-3	3.6E-1
Sr	8.0E-5	1.7E-2
CsI	5.2E-5	1.9E-2
NaI	2.5E-5	1.3E-2
Sb	7.4E-5	9.0E-3
Ba	7.5E-6	2.3e-3
others	<1E-14	<1E-9

3.5 Chemical Form

This section discusses the chemical form of the major fission product and actinide elements as they exist in the fuel and gap and their chemical form once transported into the sodium coolant. For oxide fuels, the chemical and transport behavior is well documented in Olander (Ref. 34). In oxide fuels, niobium, zirconium, barium, yttrium, strontium, cerium, praseodymium, neodymium, samarium, and lanthanum will all exist in oxide form. Molybdenum, cesium, and rubidium will be elemental in the high temperature regions of the fuel pellet and will exist in oxide form in the cool regions. Palladium, rhodium, ruthenium, tellurium, and technetium will be elemental. The metals will agglomerate to form inclusions in the fuel matrix.

3.5.1 Noble Gases – Xenon and Krypton

Since noble gases are chemically inert, they are easily transported from the fuel into the gap. For oxide LWR fuel, the typical value for the gap fraction of nobles is 3-5% (Ref. 1). For breached cladding of fuel pins, the gases are immediately transported into the coolant, but a significant quantity will remain trapped in the fuel matrix. If the fuel continues to be heated through melt, the gases are immediately released to the coolant. Berlin (Ref. 35) and Schram (Ref. 33) report that the noble gases xenon and krypton are only slightly soluble in liquid sodium. It is, therefore, expected that xenon and krypton will be transported rapidly to the cover gas in the reactor vessel for both gap releases and fuel melting releases.

3.5.2 Halogens – Iodine

Although some fraction of elemental iodine may exist in the fuel matrix in the vapor state, most will form a stable salt compound with the alkali metal cesium. The free energy of formation for CsI is quite negative, allowing for this reaction to proceed to completion. With the elemental yield for cesium from fission being about six times that of iodine, essentially all of the iodine should be removed from the gas phase in the fuel matrix (Ref. 34). This applies to both oxide and metal fuels.

Once the CsI is released to the sodium coolant, it will reformulate to NaI + Cs. If the NaI concentration in the sodium is low, this reaction will go to completion. The NaI is highly soluble in liquid sodium and has a melting point of 660°C
(Ref. 35).

3.5.3 Alkali Metals – Cesium and Rubidium

Both alkali metals cesium and rubidium behave with similar properties in the fuel matrix. The vapor pressure of cesium at 1 atm is 671°C. The vapor pressure of rubidium at 1 atm is 688°C.

Because of the high yield of cesium, substantial partial pressures (~1 atm) can develop in the fuel matrix, allowing for cesium vapors to condense in cooler regions of the fuel element. The process by which cesium migrates is simple distillation. For oxide fuels, at the interface between the fuel and the cladding, the temperatures are low enough that cesium can react with oxygen to form Cs_2O, which is much less volatile (Ref. 34). As described for iodine, CsI will also exist but in smaller quantities due to the roughly six times larger yield of cesium. Cesium can also react with steel to form CsOCr or uranium to form Cs_2UO_4, which is a solid (Ref. 21).

Elemental cesium and rubidium are very soluble in sodium. Their oxides are reduced by the sodium to form their elemental composition. Cesium and rubidium are expected to vaporize along with sodium (Ref. 21).

3.5.4 Tellurium Group – Tellurium and Antimony

Tellurium and antimony have very similar chemical properties. Tellurium and antimony are found in their elemental state and migrate easily from the hot regions of the fuel to the cold regions. They are found in the cold parts of the fuel pin and have been observed as amorphous deposits on the inside of cladding of high burnup LWR fuel. Cs_2Te has been observed in these deposits. In LMFBR systems, tellurium vapor is expected to react with cesium and sodium diffused into the stainless steel cladding and to become fixed to the surface (Ref. 21).

When a LMFBR fuel pin melts, roughly 50% of the tellurium and antimony are released from the fuel matrix to the sodium, where they are highly soluble.

3.5.5 Alkaline Earths – Strontium and Barium

In oxide fuel, strontium and barium are found as precipitates of fine particle oxides that are not soluble in the fuel matrix. At the melting point of the fuel, 80% of the strontium and 40% of the barium remain in the oxide fuel. These oxides are not reduced by sodium, and the oxides are not soluble in liquid sodium. However, elemental strontium and barium are soluble in liquid sodium (Ref. 35).

3.5.6 Noble Metals – Palladium, Rhodium, Platinum, and Silver

The noble metals palladium, rhodium, platinum, and silver remain elemental in oxide fuel. They accumulate in the hot regions of the fuel and form insoluble small inclusions. At the fuel melting point, 5-20% of these fission products remain in the oxide fuel. Except for silver, these noble metals are not very soluble in liquid sodium (Ref. 35).

3.5.7 Noble Metals – Ruthenium and Technetium

The metals ruthenium and technetium behave like noble metals but also can form volatile oxides at temperatures lower than 1200°C. In oxide fuel, ruthenium and technetium are uniformly distributed in the fuel matrix as small inclusions. At the fuel melting point, about 20% of these fission products remain in the fuel matrix. The oxides can chemically react with the sodium to form an oxide compound. Neither the oxides nor the elemental forms of ruthenium and technetium are very soluble in sodium (Ref. 35).

3.5.8 Rare Earths/Lanthanides – Lanthanum, Yttrium, Zirconium, Niobium, Molybdenum, and Cerium

In oxide fuel, these elements will be in a soluble oxide form in the fuel matrix. The rare earths are not very soluble in liquid sodium in either elemental or oxide form (Ref. 35). No data on the solubility of the lanthanides in liquid sodium was found by Schram (Ref. 33).

3.5.9 Uranium and Plutonium

Uranium and plutonium oxide are not very soluble in liquid sodium. However, there are almost no solubility data available of sodium-actinide systems (Ref. 33). Caputi and Adamson (Ref. 36) measured the solubility of uranium and plutonium in sodium. No data was found for the solubility of elemental forms of uranium and plutonium in sodium.

3.6 Sodium Releases

3.6.1 Release from Sodium Pools within the Reactor Vessel

In the event of a core disruptive accident (CDA), the most likely outcome is for the sodium to quench the fuel as the cladding ruptures and the fuel is exposed directly to the sodium coolant. In the extreme case, fuel may be grossly fragmented and form a coolable debris bed. With liquid sodium maintained in the reactor vessel, its function remains to extract heat from the fuel and dissipate it to heat sinks in the reactor coolant system so that the fuel does not melt. The sodium coolant temperature will continue to rise if sufficient secondary cooling paths do not exist. The coolant will be heated by the decay heat generated in the fuel and, to some extent, the fission products dispersed and dissolved in the coolant. While the fuel remains in contact with sodium, there will be a continuing dissolution of radionuclides from the fuel to the coolant.

The primary mode of radionuclide release from the sodium pool will be simple vaporization to the gas space above the fuel. The thermodynamic driving force for radionuclide release by vaporization is dependent upon the chemical activities of the radionuclides in the sodium. Typically, concentrations of the dissolved radionuclides will be quite low, but the chemical activities will not be directly proportional to the concentrations. A sub-regular solution model should be of adequate generality to describe the chemical activities of radionuclides dissolved in the sodium. Because of the low concentrations of the dissolved radionuclides, it should be possible to treat the sodium-radionuclide system as a set of independent binary systems rather than treating the pool as a multi-component liquid alloy. Then the chemical activity coefficients of each dissolved radionuclide can be found from:

$$RT \ln \gamma_R = (1-x)^2 \left(2\, h(T)\, x + w(T)\, (1-2x)\right)$$

where:

R = gas constant = 8.314472 J/mole-K

T = absolute temperature (K)

γ_R = activity coefficient for the radionuclide dissolved in sodium

x = atom fraction of radionuclide dissolved in sodium = moles radionuclide/(moles sodium + moles radionuclide)

h(T) and W(T) = temperature-dependent parameters associated with the radionuclide-sodium liquid system

Some care must be taken in the definition of the reference state for the activity coefficient. Two conventions are common in the literature. In one, the reference state is the liquid form of the radionuclide even if this is not the stable state of the radionuclide for the conditions of temperature and pressure of interest. The other takes the stable form of the pure radioactive element at the temperature and pressure conditions of interest to be the reference state.

It is essential in modeling vaporization releases to account for the chemical activities since these activities can deviate substantially from the ideal ($\gamma = 1$). Evidence for this is shown by the activity of NaI in sodium at the solubility limit shown in Figure 1.

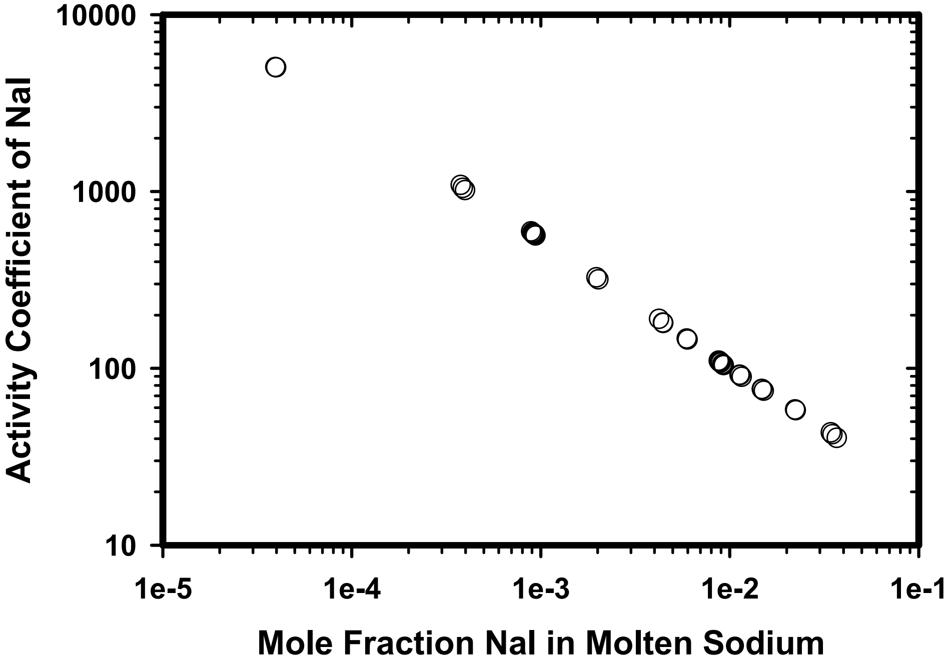

Figure 1. Chemical Activity Coefficient for NaI in Liquid Sodium.

The necessary parameters for the binary subsystems can be estimated from solubility data or from phase diagrams of the binary subsystems. For most elements of interest, these data seem to be available.

Kinetic limitations on the release of radionuclides will be primarily from mass transport. As long as the sodium pool does not boil, the mass transport limitations on the rates of radionuclide release will consist of a set of serially related steps:

- convective mass transport from the bulk pool to the sodium surface that can be treated using a surface renewal model
- gas phase mass transport that will depend on forced and natural circulation in the gas phase above the pool

These mass transport limitations can be coupled by assuming the gas phase and the molten sodium solution are at equilibrium in the immediate vicinity of the surface. From this assumption, the so-called "two-fluid" mass transport model can be used.

A third mass transport limitation, again serially related to the two listed above, will involve radionuclide dissolution from the fuel. This third limitation will need to be considered only for radionuclides readily vaporized at the pool surface so that dissolution from the fuel significantly affects the average bulk sodium concentration of the radionuclides. Some data on the kinetics of radionuclide dissolution from irradiated oxide fuel are available. Rather little data on the dissolution of radionuclides from metal fuel into sodium have been found.

Experimental investigations have revealed some release of particulate suspended in molten sodium even when the sodium does not boil and is not sparged with gas (Ref. 37). These results have been replicated in other laboratories (Ref. 77). Some attempts have been made to account for what amounts to a mechanical rather than a thermochemical release process, but a definitive mechanism for the release under quiescent conditions has not been found.

Boiling of the sodium pool complicates the release of radionuclides. The mass transport of vapors can be enhanced by the vaporization of volatile radionuclides into the sodium bubbles rising to the pool surface. There is a large body of data on the "stripping" of volatile species from liquids by bubbling. Most of these data have been obtained for water or other conventional liquids. It is assumed here that the correlations derived from these efforts can be applied to the stripping of radionuclides from molten sodium by bubbles of sodium vapor.

A more important effect of boiling or any gas sparging of the sodium-radionuclide pool within the reactor vessel is the entrainment of liquid droplets at the surface where bubbles burst. The bursting of the bubble subjects the liquid to extremely high accelerations (~10,000 g). This is sufficient acceleration to comminute some liquid into particles of aerosol size (<~ 10 µm). The liquid droplets will be contaminated by dissolved radionuclides and can contribute to the source term. Entrainment of liquid droplets by bubble bursting is a mechanism by which non-volatile radionuclides dissolved in the sodium can contribute to the source term since vaporization is not involved. Again, the entrainment phenomenon has received quite a lot of study for conventional liquid systems. Both mechanistic models and dimensionless correlations of the entrainment are available. These models and correlations can be used to estimate radionuclide release by entrainment from sodium pools. Use is easiest if it can be assumed that concentrations at the pool surface are equal to those in the bulk pool. Corrections to the concentrations for surface tension effects of the solute are possible, but it is likely that the concentrations of radionuclides are sufficiently low that corrections will be small in comparison with other uncertainties.

Some experimental investigations for UO_2 and fission product (iodine, cesium, strontium) release from quiescent (non-boiling) sodium pools are summarized in Table 6. More comprehensive and additional experimental data is presented in Appendix B.

The extent of release of contaminants from pools in these tests are summarily described in terms of the retention factor (RF), which is the amount of contaminant present at the end of a test divided by the amount released. Larger RFs are indicative of smaller releases. RF will be used as the standard measure of release throughout this report. In order to convert RF to release fraction (the quantity released divided by the initial quantity present), add one to the RF and invert.

The results show that for non-boiling pool conditions, RFs of 10^3 are typical for UO_2 at different conditions in sodium. The results are independent of temperature. No values were found for other actinides or for metal fuel conditions. The fission products have RFs of ~3-4 for NaI, less than one for cesium, and ~20 for SrO.

Table 6. Investigations of Release from Sodium Pools without Boiling.

Investigator	Sodium Mass	Contaminant Mass	Reported RF	Notes
Jordan and Ozawa (Ref. 72)	Up to 100 g	Up to 10 g UO_2	10^3	Ar cover gas at various flow rates
Jordan and Ozawa (Ref. 72)	Up to 2000 g	20 g UO_2	10^3	Varied O concentration in Na
Berlin *et al.* (Ref. 35)	4-5 kg	UO_2 powder	10^3	T = 1073 K
Schütz (Ref. 37)	50-100 g	1-10 g UO_2	4×10^3 for 20μm UO_2; 3×10^4 for coarse UO_2	Inert cover gas; release independent of Na temperature
Schütz (Ref. 37)	100 g	0.5 g Cs	<1	Release dependent on Na temperature
Schütz (Ref. 37)	100 g	0.1 to 1 g SrO	20	Release independent of Na temperature
Schütz (Ref. 37)	100 g	1 g NaI	3	Release independent of Na temperature
Berlin *et al.* (Ref. 35)	4.4 kg	NaI	4.3	T = 973 K

Some experimental investigations for UO_2 and fission product (iodine, strontium) release from boiling sodium pools (greater than 900°C) are summarized in Table 7. More comprehensive additional experimental data are presented in Appendix B.

One would expect that if the United States were to have a robust SFR program, at least some of the experimental work would need to be repeated under very controlled conditions, especially for metal fuels where no data exists.

A significant amount of experimental work has been performed for releases of UO_2 and fission products from non-boiling and boiling sodium pools. Although this report documents some of the experimental work, it is by no means comprehensive of all of the work performed in the past. Further, attaining original references is difficult since much of the work was performed decades ago. A much more comprehensive effort is needed to collect and interpret the experimental data to determine its pedigree, validity, and usefulness in forming benchmarks for code validation. Some effort can then be made as to the uncertainty in the results and extrapolation to other conditions. Until this is done it is difficult to determine gaps and recommend experimental needs for future work.

Table 7. Investigations of Release from Boiling Sodium Pools.

Investigator	Sodium Mass	Contaminant Mass	Reported RF	Notes
Jordan and Ozawa (Ref. 72)	Up to 100 g	Up to 10 g UO_2	10^3	Ar cover gas at various flow rates
Sauter and Schütz (Ref. 50)	100 g	0.2 to 5 g UO_2	10^3	Particle sizes of 20 to 200 μm
Berlin *et al*. (Ref. 35)	4-5 kg	UO_2 powder	100	T = 1155 K particle size of 200 μm
Sauter and Schütz (Ref. 50)	1000 g	0.2-5 g SrO	500	No forced convection resulted in higher RF than non-boiling Na
Sauter and Schütz (Ref. 50)	1000 g	0.2-5 g NaI	1-11	Order of magnitude agreement with non-boiling Na

3.6.2 Release from Sodium Sprays

Liquid sodium contaminated with radionuclides can be expelled through holes or cracks in the reactor coolant system. If the sodium is pressurized, there will be a spray of sodium droplets. The kinetics of radionuclide release can be enhanced in the case of sodium sprays because of improved mass transport in both the liquid phase and the gas phase. It is useful to distinguish two cases. The first and simplest case involves spray into an inert atmosphere. The second case involves formation of sodium sprays in reactive atmospheres such as air or water vapor.

In the first case the enhanced radionuclide release occurs because:

- surface area per unit volume available for the vaporization process is made much higher by the formation of droplets;
- mass transport in the gas phase is enhanced by convection around a falling droplet;
- mass transport of radionuclides in the sodium to a free surface for vaporization is enhanced by circulation within the droplet.

The thermodynamic driving force for vaporization is the same as for the sodium pool discussed in the previous section. The temperature of the droplet is affected by the vaporization of sodium, which tends to cool the falling droplet. There seems to be general agreement that heat transfer and mass transfer must be coupled in the calculation of radionuclide vaporization and transport from burning sodium droplets.

In the case of sprays into a reactive atmosphere, the surface area effects of droplet formation and the effects of circulation within the droplet remain the same. The thermodynamic driving force for vaporization can be affected as the sodium becomes saturated in oxygen to the point that a crust can form on droplets during their flights through the atmosphere. The oxide crust is akin to that formed on sodium in a sodium fire, as discussed in the next section. Radionuclides that would ordinarily escape the liquid sodium can be trapped in this reactive crust material. Crust material forms by reactions that are usually exothermic, so the droplet temperatures can rise to the point where vapor bubbles can nucleate and rupture the droplets into much finer droplets. Some of these droplets may be of aerosol dimensions (< 10 μm). Reaction of atmospheric gases also affects gas phase mass transport of vapors around the falling droplet. It is not appropriate to treat the transport of vaporized radionuclides away from the droplet in terms of binary diffusion. Instead, the effects of reactive gases moving in toward the droplet must be recognized, and vapor transport needs to be treated in terms of the Stefan-Maxwell equations, even though the volume fraction of radionuclide vapors can be quite small.

There are good mechanistic bases for treating the vaporization of radionuclides from sodium spray droplets in an inert atmosphere. Aside from changes in mass transport modeling and freedom to neglect sodium dissolution of radionuclides from exposed fuel, the modeling is rather similar to that of radionuclide vaporization from a pool. The thermodynamic driving forces for releases of the radionuclides are the same.

Reactive atmospheres will affect the driving forces for release. The radionuclides will be dissolved in a solution of sodium and oxygen. Though the oxygen concentration in the sodium can be small, it can profoundly affect the chemical activities of the dissolved radionuclides. The reactive atmosphere can also affect vapor pressures because other vapor species become available. Perhaps the best example involves the vaporization of molybdenum and ruthenium. In a reactive atmosphere, vapor species such as $MoO(g)$, $MoO_2(g)$, and $MoO_3(g)$ dominate the vapor pressure. In the absence of a reactive gas, none of these vapors contribute to the vapor pressure.

Most of the aerosol produced in a sodium spray fire is from the sodium. For fine sprays, the aerosol production rate can approach the sodium combustion rate. The aerosols produced from sodium during combustion are initially mixtures of oxide, peroxide, and suboxide. The aerosols rapidly react with atmospheric gases to form NaOH. NaOH aerosols are deliquescent and will become NaOH solution droplets if the ambient humidity is sufficiently high. The droplets will grow by water adsorption until the water activities in the solution and the atmosphere are equilibrated. There is a good data base for predicting aerosol particle growth by this process (Ref. 78). A complication is that the NaOH will also react with CO_2 in the atmosphere to form Na_2CO_3, which will precipitate within the solution droplets.

Radionuclides released from sodium will condense to form aerosol particles in the sharp temperature gradients about the sodium fires. These radionuclide aerosols will coagulate with the sodium aerosols. For typical situations, the sodium aerosol mass is so large relative to the radionuclide aerosol mass that the properties of the aerosol are almost entirely determined by the sodium aerosols.

Reactions of aerosols produced from sodium with atmospheric gases greatly simplify prediction of aerosol shape factors. The particles rapidly become spheres as they absorb water, so both the dynamic and the collision shape factors are unity. Spheroidalization of the aerosol particles will also occur when reactive gases are not present if the aerosol particles are at temperatures above their melting points. When temperatures are well below the melting point, prediction of aerosol shape factors becomes more difficult. Typically, particles will have fractal geometries that vary with time as a result of sintering of the particle constituents. Chain agglomerates are expected to form initially. As these chains branch, they form high porosity agglomerates most closely reminiscent of dust bunnies, although substantially smaller.

3.6.3 Release During Sodium Interactions with Concrete

A significant number of experimental investigations of the interactions of molten sodium with structural concrete have been performed. The issues of radionuclide release during these interactions have not received a significant amount of consideration. The release process will be at least as complicated as that which arises during sodium pool fires. Molten sodium in contact with structural concrete will cause the evaporation of gel water in the concrete and even the decomposition of the $Ca(OH)_2$ and $CaAl(OH)_3$ that constitute the bonding of the concrete. The water vapor produced by these thermal reactions will react with sodium to form hydrogen. Rather little ablation of the concrete will occur until the oxygen concentration of the sodium is sufficient that NaOH precipitates to the interface between the sodium and the concrete. The molten sodium will dissolve the concrete, leading to ablation and the decomposition of calcareous aggregates such as limestone and dolomite in the concrete. The CO_2 released by the decomposition of these aggregates also will react with sodium to form CO. Condensed products of concrete decomposition can accumulate both at the interface with the concrete and on the surface of the sodium pool.

From the perspective of radionuclide release, the high temperatures and sparging of the molten pool by both hydrogen and CO are conducive to the release of volatile radionuclides. The vigor of the interaction can lead to entrainment of liquid containing radionuclides. Sparging by gases produced in the decomposition of concrete can be far more vigorous than that of simple boiling. On the other hand, slags of NaOH and the condensed products of concrete decomposition will have a high solubility for radionuclides that escape solution in sodium metal. At the surface of the molten sodium interacting with concrete, these slag layers can grow to substantial thicknesses. A significant thermal gradient can exist through the slag beds so that near the top the beds are quite cool. One can well imagine that the slag beds amount to porous media through which aerosols and vapors must pass before they can contribute to the source term.

Though numerous studies of sodium interactions with concrete have been found, definitive data on radionuclide release during such interactions have not been found in this research effort. Mechanistic models of the ablation process and melt chemistry have been formulated, and these can be used to provide boundary conditions for the mechanistic calculation of radionuclide release. Vaporization from the sodium-oxygen melt into sparging gases can be calculated much as described above. Interaction of the vapors and trapping of particles in the slag formed over the surface of the sodium will present a more formidable challenge in fully completing this model.

3.6.4 Release During Sodium Fires

Radionuclide release during prolonged sodium pool fires is challenging to predict. The problem of prediction is that a crust forms between the molten sodium and the ambient atmosphere. The crust is chemically complicated. Metastable species can form near the crust-molten sodium interface. These metastable species are converted to more stable species along the path from the sodium surface to the interface of the crust with the ambient atmosphere. At the interface with the atmosphere, crustal materials can react to form both NaOH and Na_2CO_3. Radionuclides released from the sodium-oxygen solution can condense and dissolve in the crustal material. There is not a rich data base on the chemical activities of important radionuclide species dissolved in crustal materials. Definitive models of the chemical state of the crustal materials have not been found.

There have been some large-scale experimental studies of sodium fires, involving a substantial amount of material in some cases. Rather less has been done regarding the radionuclide release associated with such sodium fires. Some experimental investigations of soluble fission product release during sodium fires are given in Table 8.

A detailed understanding of radionuclide release from a sodium pool fire includes knowledge of both the radionuclide chemistry and sodium pool fire physics. For the sodium pool fire physics, results from a recent Phenomena Identification and Ranking Table (PIRT) exercise performed at SNL provide insight into this topic (Ref. 38). The Na_2O aerosol, crust, and solutions are difficult phenomena to predict and understand. The PIRT results presented many phenomena associated with sodium pool fires. The most important to radionuclide release is the surface oxide crust to the progression of the amount aerosolized. Predicting the aerosol fraction of the Na_2O is important in predicting the amount that is removed from the oxide crust and the capability of predicting the consequences of this aerosolized oxide (Ref. 38).

For sodium spray fires, much of the phenomenology associated with sodium pool fires is the same. The difference is the capability of predicting the spray dynamics. Sodium spray fires tend to have high heat fluxes and a larger pressure gradient associated with them.

For consequence analysis of cascading events, it is also necessary to investigate the thermal damage from a sodium fire (radiant heat to nearby components and equipment). Understanding how the sodium aerosol might affect important electrical equipment is also a research area that is poorly understood.

Sodium fire research has included fundamental studies on droplet combustion, pool burning, suppression, and large-scale sodium fire experiments. However, there are gaps in the understanding of the basic combustion behavior and combustion mechanics due to the complexities involved. These gaps have led to little progress in understanding the basic combustion behaviors for sodium (Ref. 39). Many of these same concerns were noted as early as 1972 (Ref. 40).

New technologies have substantially improved fire computer modeling capabilities. Applying these tools to a sodium fire will require some additional model development and validation work.

Unfortunately, most of the experiments performed in the past cannot be used to support model development today. Clear definition of the experimental boundaries and initial conditions are necessary to create the modeled conditions, and most of the experimental results lack this information. Reports of precise conditions in experiments are rare in the literature, so the heat transfer evaluations have been almost impossible to determine (Ref. 39).

Table 8. Radionuclide Release During Sodium Fires.

Investigator	Sodium Mass	Contaminant Mass	Reported RF	Notes
Jordan (Ref. 49)	2-3 kg	100 ppm Cs		RF decreased with increasing O concentration
Jordan (Ref. 49)	5 kg	100 ppm St		No release detected
Berlin *et al*. (Ref. 35)	2-10 kg	Cs I	0.1	Air cover gas

3.6.5 Oxide versus Metal Fuel

It is as yet undetermined if new SFRs built in the United States would use metal, oxide, or some other fuel form. Metal fuels would allow for a potentially simpler reprocessing technique using pyro-processing. Oxide fuels have been used extensively throughout the world for SFRs. The experimental work performed in the past with sodium has focused on the use of oxide fuels (UO_2) and, as such, some contaminant fission products studied have been oxide form. For fission products, experimental tests using NaI and cesium will remain valid, since these forms will be present for all fuel types. Hence the main uncertainty is the RF for metal fuel forms and actinides in liquid sodium pools.

4. In-Containment Removal Mechanisms

The principal mechanism by which radioactive fission product gases and aerosols reach the environment is leakage from the containment atmosphere (Ref. 1). Fission product removal mechanisms fall into two categories:

1. Engineered Safety Features (ESFs)
2. Natural processes

General Design Criterion 41 in Appendix A of 10 CFR Part 50 currently requires LWRs to have ESFs in place to remove or reduce fission products within the containment. These ESFs include containment atmosphere sprays, BWR suppression pools, and filtration systems utilizing both particulate filters and charcoal adsorption beds for the removal of iodine, particularly in elemental form (Ref. 1). Filtrations systems are the subject of Section 4.1 of this report while aspects of aerosol behavior are covered in Section 4.2.

4.1 Filtration Systems

ESF filtration systems are discussed in-depth in Regulatory Guide 1.52 (Ref. 41). These systems are in place chiefly to reduce aerosols and iodine in the containment. Typically, ESF filtration systems consist of demisters to remove steam and water from the air, heaters to decrease relative humidity, high efficiency particulate air (HEPA) filters to remove particulates, charcoal adsorbers to remove iodine, and additional HEPA filters to remove excess charcoal dust. According to Regulatory Guide 1.52 (Ref. 41), charcoal adsorber beds can be designed to remove from 90% to 99 % of elemental iodine and from 30% to 99% of the organic iodide.

Revised insights on accident source terms may have implications for ESF filtration systems for SFRs. Current ESF filtration systems are not sized to handle the mass loadings of nonradioactive aerosols that might be released as a result of the ex-vessel release phase, which could release significant quantities of nonradioactive as well as radioactive aerosols. NUREG-1465 (Ref. 1) suggests some possible strategies for reducing aerosol mass loads upon the filter system in LWRs (such as BWR suppression pools and water overlying core debris) that will not be useful for SFRs. This is an area that needs to be studied more carefully for SFRs. The ability of sodium to capture the aerosols and the rates at which aerosols are released from the sodium could be a major contributor to the ability of the containment to meet regulatory requirements.

Another issue that must be addressed is the impact of the chemical form of iodine within the containment for SFRs. Current LWR filtration systems include charcoal adsorber beds to trap and retain elemental iodine, which is the assumed primary form of iodine. With proper pH control, these adsorber beds may not be necessary. This will depend on the answers to two key questions:

1. To what degree will CsI and NaI retained on particulate filters decompose to elemental iodine?

2. What effect would hydrogen burns have on the chemical form of iodine within containment?

The answer to the first question seems to depend heavily upon the action of the demisters that are installed in the filtration system. Preliminarily, it appears that CsI retained on particulate filters as an aerosol is chemically stable provided that it is not exposed to moisture. However, if it is exposed to moisture, then CsI would decompose and form ionic iodine (I^-), which would lead to the evolution of elemental iodine. If the demisters and heaters do not function adequately, this process could become significant. It is also unclear that the demisters themselves will not trap some CsI aerosol. This issue needs to be explored more fully.

4.2 Aerosol Behavior

In the unlikely event of a HCDA for an SFR, some of the metals in the sodium coolant will be vaporized. Aerosols are formed when these vaporized metals (sodium, cesium, manganese, chromium, iron, uranium, and plutonium) are oxidized. Aerosol behavior is important to the transport of fission products and other highly radioactive materials because it impacts the leakage into the environment. In general, the behavior of aerosols is important inside the primary heat transport system and in the containment building, as well as during atmospheric transport outside containment. The effect of aerosols on fission product transport is most apparent in the containment building.

4.2.1 Primary Heat Transport System

The formation of aerosols is unlikely in the reducing atmosphere in the primary heat transport system of a SFR unless there is ingress of air. In this case, the sodium and other vaporized material will likely burn and produce high concentrations of fine aerosols. Early experiments with burning sodium indicate that these fine aerosols will quickly agglomerate into large sizes (Refs. 55 and 56). Without the ingress of air, most of these vapors will condense on the colder surfaces, such as stainless steel in the upper internals of the reactor. Clement (Ref. 57) modeled these phenomena of cooling of saturated vapor-aerosols on the wall surfaces and concluded that the process is controlled mostly by heat transfer and not as much by wall surface area.

In the case of unsaturated vapor, saturation will occur near the cold surfaces, and deposition will begin. Large wall deposits found on the walls in small experiments conducted under LWR conditions are indications of this phenomena (Refs. 58 and 59). It is expected that the same phenomena will take place in an SFR.

Experiments conducted at Oak Ridge National Laboratory (ORNL) indicate that UO_2 aerosols are suppressed when passing through sodium pools. This is because the large rising bubbles of hot UO_2 vapor will collapse or break up as they move upward through the pool of sodium. This breakup is a result of a combination of phenomena including condensation of sodium vapor, Taylor and Helmholtz instabilities, mechanical shearing on the upper internals, and reflected shock waves at the bottom of the reactor head. Therefore, the amount of radioactive fuel aerosols released to the cover gas in the reactor vessel or pool following a severe accident can be expected to be significantly less than in the case of releases in water pools such as would occur in an LWR. Those aerosols that do reach the cover gas region will produce a jet of coolant that will scrub the aerosols from the gas and rapidly cool the gas to coolant temperatures (Refs. 21 and 54).

4.2.2 Containment Building

By far the largest amount of information on aerosol behavior has been focused on the behavior of aerosols in the containment building and subsequent potential leakage from the containment building. In the case of a severe SFR accident, large amounts of sodium combustion aerosols will always accompany any release of fuel aerosols into the containment. Numerous experiments have been conducted that show that fuel and sodium aerosols co-agglomerate and form large, non-spherical, “fluffy,” oddly shaped particles (mixed aerosols). These have been shown to fall out more rapidly than either of the aerosols composed of sodium or uranium alone (Refs. 21, 56, and 60). This further reduces the air borne material available for release through leakage from the containment. These experiments form an empirical basis for several codes used to predict aerosol and mixed aerosol behavior (Refs. 56, 61, and 62). A series of experiments was conducted at Hanford Engineering Development Laboratory (HEDL) in order to compare test results in a large containment simulation experiment (Refs. 53, 63, and 64). Results indicate that agglomeration and plate out reduced the amount of suspended material by five orders of magnitude in 2000 seconds. Although significant differences existed, most of the codes used to analyze the results generally agreed with the test results, some as close as 20-70% over the entire time of the test run (400,000 seconds). It should be noted that most codes used in this analysis date back to the early- to mid-1980s. For instance, HAARM-3 (Ref. 65), the NRC sponsored code, is available through the Radiation and Safety Information Code Center (RSIC), but is only capable of running on a CRAY 7600. Since most funding for this type of SFR research ended in that time frame, it is likely most of codes will need modification to allow them to run on today’s desktops or work stations. The most likely exception is the CONTAIN code (Ref. 66, 76) and MELCOR (Ref. 74) that are used today for LWR source term analyses. Versions of these codes have been kept up to date by SNL.

Generally, it appears that behavior of sodium and fuel aerosols within containment can be modeled with existing codes and has a significant empirical data basis.

4.2.3 Containment Leakage

Early work on sodium aerosols (Ref. 67) indicate that small leak paths are quickly plugged, sometimes reducing gas flow rates by as much as three orders of magnitude over distances of 3-4 cm. A review of sodium aerosol leak rate data was carried out by Morewitz (Ref. 68). His analysis showed that for the usual containment leak rate of 0.1 vol%/day, a reduction of respirable aerosol of more than four orders of magnitude is possible over the cases where the aerosol is assumed to leak as a gas (no plugging). If accounted for, this has a great potential of reducing the severe accident source term for an SFR.

4.2.4 Atmospheric Dispersion

Experiments were conducted on dispersal of sodium combustion products in the atmosphere (Ref. 69). Both fan distribution and jets of liquid sodium were released across the wind from heights ranging from 6 m to 30 m. The sodium combustion products quickly agglomerated to rather large diameters, up to 600 µm. More than 50% of the aerosol mass was deposited within several hundred meters of the release point. In addition to the sprays, pools of sodium were burned with the wind blowing across the burning pool. In all tests, the principal fallout occurred near the release point, decreasing exponentially with distance down wind.

4.2.5 Impact of Sodium Aerosols on Safety Related Equipment

Outside of public health and safety issues associated with sodium aerosols and fuel/fission product aerosols, another important consideration is the sodium aerosol concentration at the air intake ports used for air-cooled decay heat removal (Ref. 70). This can lead to restricted flow and cause reductions in heat transfer rates under high aerosol concentrations. Several modeling codes were modified to predict the transport of aerosols to the air intake ports and subsequent deposition on the surfaces of the ducts and heat exchanger fins. Larger uncertainties exist in modeling these phenomena than for modeling aerosols within containments. Small-scale tests to reduce the uncertainty in the models were being planned at the time the ALMR design program was terminated.

4.2.6 Conclusion

Testing and modeling of sodium aerosols, along both LWR and SFR fuel/fission product aerosol behavior, has been ongoing for over four decades. Both small- and large-scale tests involving transport of aerosols within the primary heat transfer system, within containment, as well as tests measuring leakage from containment and dispersion in the atmosphere have been conducted. Models have been developed based on the empirical data from these tests. Results indicate that for severe accident situations, proper treatment of aerosol behavior can have significant impacts on the off-site dose. Sodium aerosols tend to agglomerate into rather large fluffy particles that quickly fall out. Sodium chemically reacts with several fission products, forming compound aerosols that also fall out rather quickly and act as scrubbers. Because sodium aerosols tend to agglomerate into large particles, plugging of small leaks such as might occur in the containment building has been observed during testing. Sodium aerosols tend to fall out close to the point of release during atmospheric dispersion, again resulting in a reduction in offsite consequences.

Sodium aerosols resulting from fires may have impacts on safety-related equipment, especially air-cooled decay removal systems. Models have been developed that indicate that rather large amounts of sodium aerosols are required to reduce the efficiency of these systems; however, the uncertainty associated with these models is rather large. Experiments are needed in order to reduce these uncertainties.

5. SFR Accident and Release Codes

As full-scale tests are not reasonable or cost effective, accident analyses are generally performed using computational codes to determine the consequences associated with hypothetical accidents. Accurate model depiction of phenomena is paramount in providing a mechanistic source term or predetermining fission group release rates to the containment, such as those proposed in the AST. Furthermore, accident analysis codes can predict the driving potential transporting fission products throughout the reactor system and to the environment. While the thermal-hydraulic conditions throughout the reactor are viewed as being more reliably and readily attained, the modeling requirements necessary to determine the fission product inventory throughout the reactor system are quite complex. A discussion on the currently available code suites and the necessary modeling practices, uncertainties, and individual codes necessary to calculate a mechanistic source term is presented.

5.1 Currently Available Safety Analysis Codes

The determination of the radioactivity escaping to containment and the evolving nature of the in-containment source term requires analysis of the evolving thermal-hydraulic state throughout the reactor system. While this can be performed numerous ways, the NRC currently applies lumped parameter codes when performing verification analysis on submitted designs; therefore, the focus of this section will reflect this practice to a reasonable extent.

Source term determination is dependent on the following:

- thermo-physical properties and the chemical reactions of the fission products and sodium throughout the reactor coolant system and containment building;
- the thermal-hydraulics of the system as it pertains to the accident analysis and transportation of the radionuclides; and
- the deposition characteristics of the fission products and aerosol dynamics throughout the accident.

As such, in many ways lumped parameter codes simplify model implementation and limit the necessary runtimes for accident analyses. Phenomenological models developed to predict these dependencies are incorporated into accident analysis codes to readily quantify the consequence of an accident and, therefore, are a predominant source in producing mechanistic source terms. To fulfill risk analysis requirements in past LMR/LMFBR reactor programs, the SFR codes developed during the CRBR Fast Flux Test Facility and/or the IFR programs were the SAS4A/SASSYS-1 (Ref. 42), SIMMER-II/III/IV (Ref. 44), and CONTAIN-LMR (Ref. 76). These computational codes may be considered the current domestic codes available for analyzing severe accidents and can provide necessary insights when determining the in-containment source term for SFRs.

SAS4A and SASSYS-1 were both developed at Argonne National Laboratory (ANL) to perform computational analysis of severe accidents. SAS4A was developed to perform transient overpower accident (TOP) and transient undercooling accident (TUC) analyses through the coupling of mechanistic models for thermal-hydraulics, neutronics, core material performance, and fuel melt/relocation. SASSYS-1 was developed for decay heat removal and later expanded for passive safety system analysis during ATWS. Similar to SAS4A, SASSYS-1 includes the same modeling functionality as SAS4A, but, as may be expected, the dependency on the entire plant necessary to model passive safety system performance resulted in expanding the capabilities of the code to capture the thermal-dynamic states of the secondary sodium loop and containment building. A recent SASSYS-1 application example was the preliminary safety evaluation of the Advanced Burner Test Reactor (ABTR) (Ref. 45).

The SIMMER series (I-II) was initiated at Los Alamos National Laboratory (LANL) to model the transition phase of severe LMFBR accidents. The code performs this function through the coupling of space- and time-dependent neutronics and multiphase, multicomponent Eulerian fluid dynamic equations to predict the two-dimensional motion of LMFBR core materials during a CDA (Ref. 43). The latest revision has been expanded to model three-dimensional motion of core material during core disruption (Ref. 44).

The accident analysis codes SAS and SIMMER are capable of determining the chronology of accident events, the severity of the melt event, and, possibly, the blowdown source to containment. However, beyond this, tracking the behavior of fission products following the released initialization and throughout the transient is not characterized.

CONTAIN-LMR (Ref. 76) was developed at SNL as a modification to CONTAIN 1.11 (Ref. 66). The recent LWR CONTAIN version 2.0 (Ref. 46) is currently employed for NRC containment performance analysis. In addition to performance analysis, this computational code employs several radiological tracking models to predict the radiological conditions inside containment. The LMR modification applies aerosol tracking (MAEROS), sodium burn models (NACOM and SOFIRE), sodium concrete interaction (SLAM), core-concrete interaction and debris resulting in aerosol production (CORCON/VANESA), and an architect for handling two condensable mediums (water and sodium).

While the codes currently available for severe accident analysis meet some of the necessary requirements for producing an alternative source term, a vacancy exists among domestic codes for the phenomenological tracking of fission products within the reactor coolant.

5.2 Future Code Development Requirements

The preparation of a code suite able to perform in-containment mechanistic source term determination should provide a level of capabilities comparable to those of the source term package and MELCOR, which were used in NUREG-1465 (Ref. 1). The source term package, used for various studies in NUREG-1465, has since been replaced entirely by MELCOR, which is treated as the repository for the state of knowledge concerning severe accident analysis of reactors. In this regard, the functionality present in MELCOR should be captured either by preservation and/or expansion, where necessary, to meet the unique demands of SFR designs.

The relevant modeling needs of the eventual design may be separated by locations within the reactor system–core, reactor coolant system, and in-containment–and are presented as such.

5.2.1 Core Modeling Needs

Severe accident analysis, with regard to source term determination, traditionally starts with determining the fission product inventory. The inventory is separated into the current fuel inventory, free gas volume inventory, cover gas inventory, and reactor coolant reactivity inventory. While the fuel and free gas inventories remain contained within the cladding, no relevant consequence to public safety exists.

The initial core inventory is often determined using ORIGEN. ORIGEN's accuracy is dependent on cross-sectional data of higher energy neutron capture. The resolution of higher energy neutron absorption is difficult due to overlapping resonances that produce additional uncertainty in the cross-sectional data used and, likewise, the eventual core inventory determined and applied in consequence analyses.

The fuel gas inventory often determined by a fuel-to-gap release model is captured to provide a portion of the activity released into the reactor coolant system given a fuel pin failure. A model for fission product release from the fuel will likely be determined empirically, requiring sensitivity to burn-up effects (fuel restructuring), temperature, and the state of the receiving sodium, which will be based on the transient conditions. The exclusion of more sophisticated physical models for the release of fission gases and transport of fission products, such as BUBL, GRASS codes, or more recent models that capture fuel restructuring or defect diffusion, is due to the integration requirement of traditional lumped parameter codes. Since fuel restructuring is simplistic compared to LWR fuels, some modeling necessities can be simplified in fast reactors.

In the event of a hypothetical accident, the timing of the accident and release rates of the free gas volume and fuel inventories must be established to determine the means by which fission products disperse throughout the reactor system. In a deterministic sense, gap release initiates to the coolant at the moment of fuel pin rupture, given that the pressure of the fuel pin is greater than that of the coolant. The timing of fuel pin rupture is often determined based on failure criteria, pin pressure, and/or cladding/fuel temperature, etc. Fuel behavior models exhibit inconsistency in methodology for calculating gas release from solid and molten fuel during transient analyses (Ref. 47). Better agreement would assert more confidence in the failure criteria established; therefore, conservative failure criteria is strongly suggested as fuel behavior model agreement is not often satisfactory. To reduce the complexity of modeling release rates, empirical correlations such as CORSOR and BOOTH models are often employed. Parametric studies similar to the LWR experiments used to determine the coefficients applied in the CORSOR and BOOTH models could be employed for LMR release models. Caution is suggested as dependencies are hidden when applying this approach, and the applicability of the results are limited to the scope of the experiments. Furthermore, detailed analyses will be necessary to determine when fuel failure or clad rupture occurs with respect to fuel characteristics, such as potential metallurgical effects, sodium corrosion, evolution of fuel and gap properties with time, etc., which is more difficult under transient conditions. This is necessary to portray good understanding of the fuel in general.

The complexities of melt progression require more thorough investigation for general accident analysis let alone source term analysis due to fuel performance uncertainties. Developing release models that are accurate throughout the melt progression must be available to perform accurate source term calculations. No such model is currently available.

5.2.2 Reactor Coolant System

The release of the fission products into the coolant stream affects the equilibrium coolant activity through entrainment of fuel and fission product as well as the dissolution of fission products. Processes relevant to the removal of fission products within the reactor coolant system are dictated by thermochemistry, which provides the phase and chemical species within the reactor system and fission gas transport to the cover gas. The interactions that will impact the source term include the interactions of vapors, vapors and surfaces, condensation of vapors on surfaces (or particulates), aerosol physics (which is discussed in the next section), resuspension, revaporization, and the processes involving pool evaporation and boiling discussed in Section 3.6.1.

Vaporization, condensation, and chemisorption of fission products are driven by the difference in partial pressures and the equilibrium partial pressure for the fission product species being considered. Two common methods for determining the partial pressure for all chemical species are (1) to use equilibrium chemical modeling to first determine the chemical species and their concentrations and phase state for saturation conditions and (2) to implement preexisting knowledge on fuel chemical species release along with the supporting curve fits for the partial pressures (Ref. 79). The complexity of this problem is associated with establishing an accurate thermodynamic database for all of the anticipated chemical forms and quantifying the unknown error being introduced. The large operating regimes within the reactor are capable of producing low-to-high temperature and pressure regimes where physical properties of the chemical species need to be accurate. Additional uncertainty can be introduced as the transient may result in increases to the temperature or flow causing vapors and aerosols that have been removed through deposition or condensation to be vaporized or suspended, neither of which are currently modeled (even in MELCOR) and may require consideration.

SFR reactors transport fission products to the cover gas through vaporization and/or bubble transport. As discussed in Section 3.6.1, aerosol suspension may occur from sparging. This is anticipated to be similar to other liquid sparging systems where sufficient data has been gathered. Bubbles may form from the gap release or very high temperatures. The energetic bursting of the bubble at the pool surface may suspend aerosols containing fission products into the cover gas.

The TRACER code (Ref. 80) developed by the Power Reactor and Nuclear Fuel Development Corporation (PNC) has some prior validation (Ref. 81-82) for reactor coolant system fission product transport. The code calculates the transportation of fission products to the cover gas due to bubble transport and evaporation from the pool surface, transport within the cover gas, and transport and some attenuation mechanisms within the reactor coolant system. TRACER may be the most comprehensive SFR in-vessel code to date.

Appropriate representation of the phase states in the primary and intermediate sodium systems will aid in determining fission product relocation. The state of the coolant will likely impact other pertinent variables for accident analysis, such as fuel relocation, exposed fuel surface area, release characteristics, etc. These will impact fuel progression modeling as well as the release rates anticipated for various fuel states.

5.2.3 Containment

Determining the in-containment source term is largely performed by good bookkeeping of aerosol and volatile fission product masses throughout the reactor system up to the break site and cover gas and then throughout the containment volume. This is performed by tracking the transportation of the aerosols and volatile fission products. The thermal-hydraulics calculations performed between the containment control volumes, typically by a lumped parameter code, determines the bulk fluid driving potential that transports aerosols and volatile species. Given that the general transport of aerosols and fission products is accurate, aerosol physics and vaporization of volatile species remain. Determining vaporization of volatile species is performed via partial pressure calculations as described above.

Aerosol tracking is important as aerosol will host individual fission product species. Aerosols also provide a site for condensation of fission product vapors to occur. Since aerosols transport fission products, the removal of lofted aerosols becomes important in determining the in-containment source term that may be passed to the environment. The primary mechanisms that affect aerosol size include condensation and evaporation to and from the aerosol and agglomeration; the removal mechanisms include deposition and gravitational setting. Effects on the aerosol size are driven by condensation/evaporation and agglomeration of aerosols. Agglomeration mechanisms include Brownian, differential gravitational settling, turbulent shear and inertial forces, and the removal mechanisms of aerosols including diffusiophoresis, thermophoresis, gravitational settling, and diffusion to surfaces.

Assuming the physical aerosol phenomena modeling practices applied today are adequate, the aerosol sources in containment characteristics must be resolved. Aerosol sources in SFR include sodium sprays from the reactor coolant system, cover gas releases, sodium-concrete interactions, and other ex-vessel interactions.

The effects of spray and pool fires were introduced in Section 3.6. Fires produce both aerosols and energy as byproducts. The increase in energy is handled by a thermal-dynamic code, and recent examples include SOFIRE-MII and ASSCOPS (Ref. 80). Aerosols released from fires are traditionally portrayed in computational codes as log-normal distributions from the guidance given in Ref. 83. Spray fires can freely distribute aerosols to the atmosphere, but for pools a fraction will remain on the pool surface with the remaining amount lofted to the atmosphere. The SPHINC computer code (Ref. 84) is a good example of a numerical code that includes the energy analysis and aerosol release fractions from pools through response surface modeling (Ref. 85).

Transportation of the cover gas and the contained fission products will be similar to the previous discussion on general transportation performed by a lumped parameter code. Fission products contained in the cover gas were presented in Section 3.6. Bubble burst suspension is pertinent here.

If it can be assumed that Monju is a good example of future SFR designs, the concrete basemat will likely be lined with stainless steel. The liner is used to potentially eliminate sodium-concrete interactions. If a puncture exists in the liner, the hole will simply slow the interaction for a short period until the liner is lifted away and eventually splits due to the chemical interaction, exposing the damage below the liner. No models were identified for aerosol production due to sodium-concrete interaction. This can be extended to corium-sodium-concrete interactions and cavity releases.

Overall, as it pertains to in-containment source term determination, current LWR codes capture general trends well, but commonly produce results with an error on the order of one magnitude or more. This should be considered during the evaluation and establishment of any code suites used to determine source terms mechanistically for the ABR.

6. Science Path Forward

Do sufficient experimental data and modeling capabilities currently exist to allow for a mechanistic source term to be developed for new SFRs in the United States under the current regulatory environment? A significant amount of experimental work and modeling was performed over the past several decades to advance SFR design technology and source term development. Much of the documented results, however, probably lack the desired pedigree required to be of use in the validation of computer models. Experimental work is needed to determine release fractions for the fuel, dispersion into liquid sodium, and releases from sodium pools for particular fuels of interest and actinide loadings. An important aspect of the experimental work is to ensure well-documented results of experiments that meet the quality requirements expected for use in today's regulatory environment. This includes designing and performing well-controlled and documented experiments that generate reproducible results with uncertainty quantification. These experiments can then be used not only for model development but also for code validation, which is of high importance for current software quality engineering requirements.

6.1 In-Pile Testing

In-pile source term experiments will develop a database for gap releases and fuel failure releases for a variety of postulated transient conditions. An experimental program can attain the same degree of technical risk regarding source term for SFRs as is available for LWRs. Since fuel type (metal versus oxide), actinide loading, and burnup are important variables in the design of SFRs, the in-pile experiments must include enough parameterization studies and range in these variables to decrease the source term uncertainty and allow for computer model validation. Experiments should include fission product gap releases for failed cladding, fission product and actinide fuel melt releases into liquid sodium, and fission product and actinide debris bed releases to liquid sodium. In-pile/in-situ fission product and actinide releases from the sodium at different temperatures and accident conditions should also be studied.

6.2 Sodium Testing

A significant amount of work was performed from the 1960s to the 1980s on fission product and fuel solubility and retention in sodium pools under non-boiling and boiling conditions and pool and spray fire conditions. However, a significant experimental program needs to be reinstated in the United States to study release mechanisms of fission products and actinides from liquid sodium. Specific fission products and actinide release experiments need to be developed that can be used to model and validate computer codes. With the little information available for metal fuels and actinides, experimental work for solubility and release of these elements is of vital importance for the Advanced Fuel Cycle Initiative (AFCI) program and SFRs in general.

Solubility and release studies are needed for additional fuel types and elements in addition to the few that have been experimentally studied, namely UO_2, NaI, cesium, and SrO. There are many elements described earlier in the report that have significant radiological consequences for which there is no experimental information. It is critical to use realistic concentration levels in sodium pool experiments for both fission products and actinides. If fission products or actinides were soluble to a small extent, yet the solubility limit was exceeded in the experiment, then a nonconservative RF would result. Realistic sodium chemistry is also a need in future experimental testing. Experiments are needed for not only high-purity sodium, but sodium-sodium hydroxide mixtures, and sodium-concrete reactions as well.

There further exists the need for studying the reaction and dissolution of cesium and tellurium with stainless steel to form compounds that are soluble in sodium or which can be immobilized in the metallic matrix. This could be important for metal fuels where sodium is maintained within the gap between the fuel matrix and the cladding.

Sodium-concrete interactions need further experimental study and modeling work to develop the radionuclide release under these conditions. Though numerous studies of sodium interactions with concrete have been performed, definitive data on radionuclide release during such interactions have not been quantified. Mechanistic models of the ablation process and melt chemistry have been formulated and these can be used to provide boundary conditions for the mechanistic calculation of radionuclide release.

New technologies have substantially improved fire computer modeling capabilities. Applying these tools to a sodium fire will require some additional model development and validation work. Unfortunately, most of the experiments performed in the past cannot be used to support model development today. Clear definition of the experimental boundaries and initial conditions are necessary to create the modeled conditions, and most of the experimental results lack this information. Reports of precise conditions in experiments are rare in the literature, so the heat transfer evaluations have been almost impossible to determine.

Additional experimental studies need to be performed for aerosol fragmentation, comminution, plateout, and resuspension in the containment for model validation. Experimental studies of containment filtration systems and methods for effectively decreasing the radionuclide concentration in a HCDA event are needed. Sodium aerosols resulting from fires may have impacts on safety related equipment, especially air-cooled decay removal systems. Uncertainty associated with these models is large and requires experiment validation.

6.3 Code Development

A code development and maintenance program is needed for SFRs to integrate accident analysis with release mechanisms and the development of a realistic source term. Current best-estimate codes need to be modified, as needed, to incorporate latest models. The codes need to be integrated, validated, and maintained using current software quality engineering requirements. This may include restructuring the codes using today's programming techniques, methods, and tools. Code development with verification and validation is important to ensure that the results are meaningful and have quantifiable uncertainty.

Some work is being done at SNL on a code framework called Burner Reactor Integrated Safety Code (BRISC). It is hoped that this can be further developed into a code that will allow the user to assess BDBEs and possibly produce a realistic source term for SFRs. However, there appears to be no experimental data on two-phase flow of sodium. This data will be needed in order to assess how any isotopes will be dispersed in the event of a HCDA (Ref. 86).

6.4 Two-Phase Flow

For BDBEs, the idea that the sodium coolant is heated to the boiling point (883º C) must be considered. There appears to be no experimental data related to two-phase flow of sodium. This is an area that should be heavily considered for future experimental work.

7. Test Facilities

Do sufficient test facilities exist to perform the necessary work to advance SFR development in the United States with respect to developing a mechanistic source term?

The Advanced Test Reactor (ATR) at INL is currently used to perform burnup testing of fuel for proposed SFRs in the AFCI program. Transient reactor test facilities do exist in the United States including the ACRR at SNL and the TREAT Facility at INL. The ACRR is currently operational but has yet to be used for testing in the AFCI program. TREAT is in a standby mode and would require significant funding for restart. With the closure of FFTF, no fast reactor test facility exists in the United States.

Sodium test facilities required to perform radionuclide release experiments or other large-scale experiments are non-existent in the United States.

7.1 Transient Reactor Facilities

A significant amount of in-pile transient testing work was performed in the 1980s on both metal and oxide fuels and documented previously in this report. The work was performed at both the TREAT Facility at INL and the ACRR Facility at SNL. Fuel failure testing and debris bed coolability was performed with and without sodium coolant in order to better understand the failure mechanics associated with a HCDA.

No in-pile testing work has been performed using fast reactor fuel with the sole purpose of studying the source term for HCDAs in sodium coolant. LWR source term experiments were performed in the 1980s and 1990s in the ACRR. In order to fully understand the behavior of the fission product release into the coolant, the fuel fracturing mechanics and release of the actinides into the coolant, and the release of nuclides from the coolant into the vapor region carefully crafted in-pile experiments will need to be designed and performed. This work must include both metal and oxide fuel forms with different levels of burnup and potential actinide loadings. Different configurations also must be studied that represent possible post accident geometries. Short-lived isotopes of krypton, xenon, iodine, and tellurium can be generated in a steady-state operation in ACRR prior to performing the transient to allow the behavior of these fission products to be observed.

Only two reactors exist in the United States that are capable of performing transient testing conditions for in-pile experiments: the ACRR and TREAT. The ACRR is currently operational and can operate in a steady-state mode at 2 MW, a pulse mode, or a transient rod withdrawal mode. Pre-irradiation of fuel is possible since the reactor can run at 2 MW for an indefinite period of time. TREAT is in a non-operational standby condition. TREAT could not operate in a long duration steady-state mode but could operate in a pulse-transient mode.

7.2 Sodium Test Facilities

During the 1970s and 1980s, a number of sodium facilities existed in the United States and throughout the world for performing small-, intermediate-, and large-scale testing. Descriptions of these facilities and other pertinent information are presented in Sienicki and Grandy (Ref. 5). Some of these facilities included the capability to study sodium fires and aerosol production.

The Aerosol Behavior Code Validation and Evaluation (ABCOVE) program was developed in accordance with the LMFBR safety program during the 1980s, and was a joint effort by the NRC and Department of Energy (DOE) (Ref. 53). The program involved analytical modeling work and large-scale testing. The large-scale testing was performed at the Containment Systems Test Facility (CSTF) at the HEDL in the 850 m^3 containment vessel. Sodium fire testing was also performed at the CSTF.

Coupled aerosol physics-bubble dynamic phenomena were studied in the 1980s at the Fuel Aerosol Simulant Test (FAST) facility at ORNL (Ref. 54). The experiments focused on UO_2 aerosol release in a 0.46 m^3 vessel using capacitor discharge to vaporize UO_2 pellets.

Other experiments were conducted at ANL East, LANL, Atomics International, General Electric Nuclear Engineering Division, and Westinghouse Electric Corporation Advanced Reactors Division.

Release of fission products from the sodium coolant has been studied since the 1970s (Ref. 21). A significant amount of experimental work was performed in the study of iodine, cesium, rubidium, tellurium, antimony, noble metals, and uranium oxide.

Work needs to be restarted in the United States in the area of sodium aerosol generation, sodium fires, sodium/stainless steel interactions at the small-, intermediate-, and large-scale. State-of-the-art facilities for sodium experiments and testing need to be constructed if the United States is ever to make resurgence in the area of SFR development.

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Appendix A – Ranking for Fission Products and Actinides

Table 9 shows a ranking of fission products and actinides in order of importance to downwind dose for an unmitigated release. The color scheme represents the order of magnitude (factor of 10) for the importance to the downwind dose. The color scheme is as follows for the worst contributor to the least contributor: 4 - purple, 3 - red, 2 - yellow, 1- green, 0/blank - white.

The results were found by running ORIGEN2.2 for a LMFBR spectrum for a three-year burn-up with U-235/U-238 fuel. The complete inventory was then assumed to be released to the environment (unmitigated release). The MELCOR Accident Consequence Code System (MACCS) calculations at 3 km for Technical Area V at SNL were then used to determine the downwind dose. The results are presented for comparative purposes to allow for qualitative identification of the most important fission product and actinide nuclides. Table 10 shows the ranking of the fission products by elements and chemical grouping.

Table 9. Ranking of Fission Products and Actinides in Order of Importance to Downwind Dose for an Unmitigated Release.

Nuclide	Half-Life	Importance Ranking	Most Important
Nobles			
Kr-85m	4.48 H	1	
Kr-87	1.27 H	2	x
Kr-88	2.84 H	3	x
Xe-133	5.24 D	1	
Xe-133m	2.19 D		
Xe-135	9.10 H	2	x
Xe-135m	15.3 M	1	
Xe-138	14.1 M	2	
Halogens			
Br-82	1.47 D	1	
Br-83	2.40 H		
Br-84	31.8 M	1	
I-131	8.04 D	3	x
I-132	2.28 H	3	x
I-133	20.8 H	3	x
I-134	52.6 M	3	x
I-135	6.57 H	3	x
Light			
Ge-77	11.3 H		
Rb-86	18.7 D		
Rb-88	17.7 M	1	
Rb-89	15.4 M	2	
Sr-89	50.5 D	3	x
Sr-90	29.1 Y	3	x
Y-90	2.67 D	2	x
Sr-91	9.50 H	3	x
Y-91	58.5 D	4	x
Y-91m	49.7 M	2	
Sr-92	2.71 H	3	x
Y-92	3.54 H	2	x
Y-93	10.2 H	2	x
Y-94	18.7 M	2	
Y-95	10.3 M	1	
Zr-95	64.0 D	3	x
Nb-95	35.0 D	3	x
Nb-95m	3.61 D		
Nb-96	23.4 H		
Zr-97	16.8 H	3	x
Nb-97	1.23 H	2	
Mo-99	2.75 D	3	x
Tc-99m	6.01 H	2	x
Mo-101	14.6 M	2	
Tc-101	14.2 M	1	
Ru-103	39.3 D	3	x
Tc-104	18.2 M	2	

Ru-105	4.44 H	2	x
Rh-105	35.4 H	2	x
Nuclide	**Half-Life**	**Importance Ranking**	**Most Important**
Ru-106	1.02 Y	4	x
Ag-111	7.47 D	1	
Ag-112	3.13 H	1	
Cd-115	2.23 D	1	
Cd-115m	44.6 D	1	
In-115m	4.49 H		
Heavy			
Sn-121	1.13 D		
Sn-123	129.2 D	1	
Sn-125	9.63 D	1	
Sb-125	2.76 Y	1	
Te-125m	58 D		
Sb-126	12.4 D	1	
Sb-127	3.84 D	2	x
Te-127	9.4 H		
Te-127m	109 D	1	
Sn-128	59.1 M	2	
Sb-128m	10.1 M	1	
Sb-129	4.40 H	2	x
Te-129	1.16 H		
Te-129m	33.6 D	2	x
Sb-131	23.0 M	2	
Te-131	25.0 M	1	
Te-131m	1.35 D	2	x
Te-132	3.26 D	3	x
Te-133	12.4 M	2	
Te-133m	55.4 M	2	
Te-134	42 M	2	
Cs-134	2.06 Y	2	x
Cs-136	13.2 D	2	x
Cs-137	30.2 Y	2	x
Cs-138	32.2 M	2	
Ba-139	1.40 H	1	
Ba-140	12.8 D	3	x
La-140	1.68 D	3	x
Ba-141	18.3 M	2	
La-141	3.90 H	2	x
Ce-141	32.5 D	3	x

Nuclide	**Half-Life**	**Importance Ranking**	**Most Important**
Ba-142	10.7 M	2	
La-142	1.54 H	2	x
La-143	14.1 M	1	
Ce-143	1.38 D	3	x
Pr-143	13.6 D	3	x
Ce-144	284.6 D	4	x
Pr-144	17.3 M	2	
Pr-145	5.98 H	2	x
Pr-147	13.4 M	1	
Nd-147	10.98 D	3	x
Pm-147	2.62 Y	3	x
Pm-148	5.37 D	2	x
Pm-148m	41.3 D	2	x
Nd-149	1.72 H	1	
Pm-149	2.21 D	2	x
Pm-151	1.18 D	2	x
Sm-151	90 Y	1	
Sm-153	1.93 D	1	
Eu-155	4.71 Y	1	
Eu-156	15.2 D	1	
			48
TRUs			
U-234			
U-235		1	
U-236		1	
U-237		2	x
U-239		2	x
Np-237		1	
Np-238		2	x
Np-239		4	x
Pu-238		4	x
Pu-239		4	x
Pu-240		3	x
Pu-241		2	x
Am-241		1	
Cm-242		1	
Cm-244			

Table 10. Ranking of Fission Products Elements in Order of Importance to Downwind Dose for an Unmitigated Release.

Element	Importance Ranking	Elemental Categorization
Kr	3	1–Noble Gas
Xe	2	1–Noble Gas
Br		4–Halogen
I	3	4–Halogen
Ge		11–Main Group Metal
As		11–Main Group Metal
Rb		2–Alkali Metal
Sr	3	3–Alkaline Earth
Y	4	9–Trivalent
Zr	4	8–Tetravalent
Nb	3	7–Transition Metal
Mo	3	7–Transition Metal
Tc	2	7–Transition Metal
Ru	4	6–Platinoid
Rh	2	6–Platinoid
Ag		11–Main Group Metal
Cd		11–Main Group Metal
In		11–Main Group Metal
Sn		11–Main Group Metal
Te	3	5–Chalcogen
Sb	2	11–Main Group Metal
Cs	2	2–Alkali Metal
Ba	3	3–Alkaline Earth
La	3	9–Trivalent
Ce	4	8–Tetravalent
Pr	3	9–Trivalent
Nd	3	9–Trivalent
Pm	3	9–Trivalent
Sm		9–Trivalent
Eu		9–Trivalent

Appendix B – Experimental Database Review

B.1 Aerosol Experiment Review

Berlin *et al.* (Ref. 35) performed the uranium PAVE experiments that used up to 300 g of UO_2 and 4-5 kg of sodium. These tests were performed until all the sodium had vaporized. The only details that were given about the aerosol samplings were that they were taken about midway up the vessel height and that the samplings were performed using x-ray diffraction techniques. The results of these tests are shown in Table 11, which was taken directly from the report.

Table 11. Data from Berlin *et al.* (Ref. 35).
1983 PAVE Experiments.

Experiments	T (°C)	Mass of Na (kg)	Radioactive Compounds in Sodium			Average RT*
			Type	Average Size (μm)	Mass (g) and (activity)	
PAVE 2	800	4.6	UO_2	2	60	950
PAVE 3	780	4.4	UO_2	10	126	2000
PAVE 4	780	4.4	UO_2	200	95	1100
PAVE 5	Boiling	4.4	UO_2	2	148	60
PAVE 6	Boiling	4.9	85SrO 141CeO_2 103Ru	1.9 1.2 2.4	0.2 (120μCi) 1.2 (259μCi) 0.7 (58μCi)	300 100 180
PAVE 7	Boiling	5	UO_2	1.2	271	80

*RT = retention factor

Another series of experiments performed by Berlin *et al.* (Ref. 35) is labeled the FANAL experiments. These experiments involved sodium fires with iodine and cesium. The details on the composition of the iodine and cesium were not presented. Liquid sodium was heated to 550°C and poured into the combustion vessel, which was in an air atmosphere. Aerosol samples that deposited in the test apparatus were analyzed. The data taken from the report are shown in Table 12.

Table 12. Data from Berlin *et al.* (Ref. 35).
1983 FANAL Experiments.

Experiment	FP*	Initial Activities (mCi)	Emitted Activities (mCi)	RF
FANAL 11	131 I	43.4	11.07	0.66
FANAL 12	131 I	12.7	3.5	0.63
FANAL 14	131 I	54.9	13.9	0.51
FANAL 17	110m Ag	2.E-02	4.6E-03	1.3
	113 Sn	0.4	2.2E-04	550
	137 Cs	1.04	1.04	0.3
FANAL 21	110m Ag	0.43	0.12	0.6
	113 Sn	6.9	<8E-3	>150
	125 Sb	0.15	<5.1E-3	>5
	131 I	35.4	9.7	0.61
	137 Cs	8.65	5.35	0.27
FANAL 22	125m Tc	5.765	1.04E-02	76

*FP = fission product

Jordan (Ref. 49) presented experiments that were performed in the Natrium brand und Schwebstoffilter anlage (NABRAUS) test facility. The sodium and cesium were mixed mechanically in the fire pan under an inert atmosphere. After the melt was mixed, oxygen was added, and the mixture spontaneously ignited. Aerosol samples were taken throughout the test at different locations and were analyzed for cesium using neutron activation. The data from these experiments are shown in Table 13. Jordan concluded "that sodium and cesium evaporated independently of each other and that this process is governed by the temperature and vapor pressure." Jordan also thought that the "vapor pressure of the oxygen in the atmosphere as a limiting factor for cesium release."

Table 13. Data from Jordan (Ref. 49).
Cesium-Sodium Fire Experiments.

O_2 (%)	Cs-Content of Sodium Pool (ppm)	Maximum Aerosol-Concentration (Mg/m^3)	Maximum Cs-Content of Aerosols (ppm)	Mg Cs/m^3	Cs-Content of Residue in Pool (ppm)
0	100	4040	1700	--	14
1	100	3940	1300	4.5	14
11	100	7600	480	3.6	--
21	100	21400	200	4.2	37
22	100	11700	300	3.5	--
21	200	11000	310	3.3	50
21	500	18000	1500	--	58

B.2 Fission Product Release from Evaporation

A large experimental series was performed under the NALA program in Germany (Ref. 50). These tests were performed to investigate the release of fission products from hot sodium under inert atmosphere conditions. The experiments performed for uranium were greater than the other materials. The materials that were tested include UO_2 powder, cesium metal, NaI, and SrO powder. For the cesium and iodine, vaporization was the release process. For uranium and strontium, a mechanical release process was used. There were three different experimental vessels referenced throughout the data tables with the following descriptions from the report:

- Type A: "A vertical cylindrical stainless steel vessel, 7 cm in diameter, 10 cm tall, with thermocouple tube attached at the lid, and inlet and outlet tubes."
- Type B: "Similar to (A), but the thermocouple tube in the bottom and outlet tube enlarged. This vessel was used to determine fission product concentrations as a function of height in the pool."

- Type C: “Horizontal stainless steel pipe, 6 cm in diameter, 20 cm long, containing thermocouples to measure pool and gas temperature. This vessel allowed for horizontal gas flow across the pool surface, without being deflected as in the first two setups.”

The twenty-four UO_2 experiments were performed and are presented in Table 14. Experiments 1-10 used vessel type A, experiments 11-17 used vessel type B, and experiments 18-21 used vessel type C, as described above. Experiments 22-24 were performed using vessel type A open inside an inert atmosphere glove box. The sixth column labeled "v" is the flow rate of the inert gas in liters per minute.

The next group of experimental results is from the five cesium release experiments. For these experiments, vessel type A was used. The data shown in Table 15 has measured mass flow rates for both the sodium and the cesium. The cesium release columns represent both the experimental data and the calculated results from the model. For the previous experiments the release amount depended on the temperature of the liquid sodium. There is a trend with the temperature of the sodium compared to the amount of sodium and cesium released. The hotter the sodium is for the experiment, it seems the larger the release will be. The same result was shown for the above UO_2 experiments.

Table 14. Data from Sauter and Schütz (Ref. 50). Fission Product Release UO_2 Experiments.

Exp. No.	UO_2 (g)	d (µm)	Na (g)	T (°C)	t (min)	v (l/min)	Na released (g)	U released (µg)	RF
1	8.48	20	100	892	6	0	40.0	1580	2.0E3
2	8.68	20	100	892	6	0	23.9	1230	1.4E3
3	0.76	20	100	892	2	0.6	45.0	394	1.1E3
4	0.84	20	100	530	87	17	12.3	42	2.1E3
5	0.72	20	100	530	93	10	2.0	5	2.5E3
6	0.95	20	100	530	60	1	0.9	4	1.8E3
7	10.25	20	100	530	60	10	2.8	104	2.5E3
8	1.17	20	51	437	120	1	0.2	1	0.8E3
9	1.27	20	50	723	17	1	6.9	243	0.7E3
10	1.88	20	56	814	9	1	20.1	300	2.0E3
11	2.46	20	101	700	10	5	4.6	109	0.9E3
12	2.00	112	98	700	10	5	4.1	94	0.9E3
13	2.20	188	102	700	15	5	8.2	236	0.7E3
14	2.36	565	102	700	15	10	12.1	39	6.3E3
15	5.00	Pellets	108	620	20	15	9.5	<2	>2E5
16	5.28	Pellets	94	720	75	0.5	4.5	<1	>2E5
17	5.30	Pellets	52	825	20	0.5	36.4	<7	>4E5
18	1.41	20	178	623	15	20	11.2	1744	45
19	0.95	20	150	530	90	7.5	5.1	1083	26
20	8.35	20	164	623	20	14	7.1	7118	45
21	2.45	188	151	623	20	8.6	7.3	74	1.4E3
22	1.9	20	103	520	90	--	4.25	12	6.0E3
23	2.00	20	102	485	75	--	2.64	22	2.1E3
24	11.0	188	103	510	90	--	4.91	14	3.3E4

Table 15. Data from Sauter and Shütz (Ref. 50).
Fission Product Release Cesium Experiments.

Exp. No.	Cs (g)	Na (g)	T (°C)	t (min)	v (l/min)	Na released (g)	Cs (g) released exp.	Cs (g) released model	$\dot{m}$ kg/m^{2}h Na	$\dot{m}$ kg/m^{2}h Cs
1	0.5	101	437	90	10	0.65	0.097	0.080	0.112	3.87
2	0.5	106	530	60	5	1.46	0.125	0.107	0.378	7.35
3	0.5	102	625	17	10	3.79	0.212	0.188	2.47	34.7
4	0.5	104	723	11	8.5	11.72	0.334	0.320	16.15	111
5	0.5	99	814	8	4	25.83	0.492	0.426	45	225

For the iodine release experiments in sodium, both open and closed vessel experiments were performed. The open vessel was open to the atmospheric conditions, and the closed vessel had a controlled inert atmosphere. Table 16 presents the data taken from Sauter and Schütz (Ref. 50). The column labeled setup shows either a 1 (for closed vessel) or a 2 (for open vessel). The vessel types refer to those described at the beginning of Appendix B.2.

Table 16. Data from Sauter and Shütz (Ref. 50).
Fission Product Release Iodine Experiments.

Exp. No.	Vessel Type	Setup	NaI (g)	Na (g)	T (°C)	t (min)	v (l/min)	Na released (g)	I (mg) released	RF
1	A	1	1.67	98	522	60	10	2.5	2	17.1
2	A	1	1.04	52	603	20	9	1.55	8	2.8
3	A	1	1.43	98	725	12	10	13.34	35	3.5
4	A	1	1.16	100	786	3	10	6.33	41	1.5
5	A	1	1.57	101	838	5	1	15.18	52	4.3
6	B	1	1.00	96	510	60	3	0.39	1	3.6
7	B	1	1.00	99	703	12	11	8.36	25	3.0
8	C	1	0.10	150	450	150	9	7.15	2.8	0.9
9	C	1	0.10	201	623	15	7.5	3.50	3.5	0.4
10	A	2	1.02	104	564	45	--	3.41	8.7	3.6
11	A	2	0.10	102	450	180	--	2.22	0.9	2.0

The last chemical to be tested by Sauter and Schütz was strontium. Six tests were performed with the two setup types (1=closed vessel, 2=open vessel) using the experimental vessel type B. Table 17 represents the data that was collected.

Table 17. Data from Sauter and Shütz (Ref. 50).
Fission Product Release Sr Experiments.

Exp. No.	Setup	SrO (g)	Na (g)	T (°C)	t (min)	v (l/min)	Na released (g)	Sr (mg) released	RF
1	1	1.00	98	550	60	5	2.93	1.32	19
2	1	0.53	105	598	30	6	4.28	<0.6	<31
3	1	0.53	102	695	12	3	4.00	0.88	20
4	1	0.50	108	796	5	5	5.13	1.42	14
5	1	0.10	102	596	30	13	10.8	<0.25	<36
6	2	1.01	104	564	45	-	3.41	<0.9	<37

Morewitz (Ref. 21) also discusses some of the above-mentioned experimental work in his fast reactor source term review. Morewitz mentions more vaporization work from Stakebake and Robinson (Ref. 51) and Ettinger *et al.* (Ref. 52). Stakebake and Robinson performed experiments in both an inert atmosphere and an air combustion atmosphere with small sodium pools. These pools contained “either PuO_2 particulate or β- Na_4PuO_5.” Morewitz mentions that the results of these experiments showed that the “sodium burning release fractions for β-Na_4PuO_5 were three to four orders of magnitude smaller and the vaporization release fraction for PuO_2 particulates were 0.42×10^{-5} to 14.7×10^{-5}.” Ettinger (Ref. 52) performed small sodium pool burns that contained plutonium-cobolt-cerium alloy in a 2.76% oxygen with nitrogen atmosphere. The plutonium release fractions that were measured from these experiments were 0.9×10^{-7} to 3×10^{-7}.

B.3 Experimental Database Summary

Some general trends are made apparent in these 1960s and 1970s experimental data sets. There is an overall lack of understanding as to how the data was collected, specifically in terms of what instrumentation was used as well as how frequently or infrequently the data was collected. The majority of older experimental work in this area focused on finding retention factors, which are the amount of contaminant present at the end of a test divided by the amount released. Again, details are lacking as to how this information was collected. Another general trend present in the experimental data set is the amount of repeated experiments for validation purposes. Overall, the past experimental data is important to benchmark new experimental programs in order to advance the understanding in determining a fast reactor source term.

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