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Toward a Mechanistic Source Term in Advanced Reactors: A Review of Past U.S. SFR Incidents, Experiments, and Analyses

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Toward a Mechanistic Source Term in Advanced Reactors: A Review of Past U.S. SFR Incidents, Experiments, and Analyses

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In 2015, as part of a Regulatory Technology Development Plan (RTDP) effort for sodium-cooled fast reactors (SFRs), Argonne National Laboratory investigated the current state of knowledge of source term development for a metal-fueled, pool-type SFR. This paper provides a summary of past domestic metal-fueled SFR incidents and experiments and highlights information relevant to source term estimations that were gathered as part of the RTDP effort. The incidents described in this paper include fuel pin failures at the Sodium Reactor Experiment (SRE) facility in July of 1959, the Fermi I meltdown that occurred in October of 1966, and the repeated melting of a fuel element within an experimental capsule at the Experimental Breeder Reactor II (EBR-II) from November 1967 to May 1968. The experiments described in this paper include the Run-Beyond-Cladding-Breach tests that were performed at EBR-II in 1985 and a series of severe transient overpower tests conducted at the Transient Reactor Test Facility (TREAT) in the mid-1980s.

I. INTRODUCTION

Construction and operation of a nuclear power installation in the U.S. requires licensing by the U.S. Nuclear Regulatory Commission (NRC). A vital part of this licensing process and integrated safety assessment entails the analysis of a source term (or source terms) that represents the release of radionuclides during normal operation and accident sequences. Historically, nuclear plant source term analyses have utilized deterministic, bounding assessments of the radionuclides released to the environment. Significant advancements in technical capabilities and the knowledge state have enabled the development of more realistic analyses such that a mechanistic source term (MST) assessment is now expected to be a requirement during advanced reactor licensing.

Developing an MST for a reactor design entails realistically modeling radionuclide release and transport from the source to the environment during specific scenarios, while accounting for radionuclide retention and/or transmutation phenomena along the way. Associated uncertainties must also be identified and quantified. This can be difficult due to the simultaneous occurrence of multiple chemical and physical interactions. While knowledge gaps exist in the complete process of radionuclide release and transport, past U.S. experience from reactor operations, experiments, and analyses provides a source of vital information.

In the U.S., there have been three sodium reactor incidents and two reactor experiments (see Table I) that provide specific insights into the behavior of radionuclide release and transport from the core of a metal-fueled sodium reactor. This paper will provide an overview of each incident and experiment and identify the key observations that are pertinent to MST development.

It should be noted that as part of the MST development effort at Argonne, a review of phenomena affecting radionuclide transport and retention was also conducted. That work is described in (Ref. 1), which accompanies this paper.

TABLE I. List of Relevant Incidents and Experiments

Accident/Experiment	Year
SRE Accident	1959
Fermi 1 Accident	1966
EBR-II Failed Fuel Capsule	1967-68
EBR-II Run-Beyond-Cladding-Breach Tests	1985
TREAT Transient Overpower Tests	1984-87

II. REVIEW OF PAST INCIDENTS AND EXPERIMENTS

II.A. The SRE Accident

The Sodium Reactor Experiment (SRE) was a 20 MW_{th} graphite moderated, sodium-cooled thermal reactor that began operation in 1957. The fuel, unalloyed uranium metal, was enriched to 2.8% U-235 with a NaK bond and Type 304 stainless steel cladding. SRE also contained one experimental fuel element with oxide fuel and five elements with uranium fuel alloys (U, Zr, Th, Mo combinations). The 43 fuel elements each had seven fuel

rods, as seen in Fig. 1 (Ref. 2). The core sat in a tank of sodium, with inlet and outlet piping creating a loop design.

Around July 12th, 1959, a tetralin (an oil-like hydrocarbon that was used to cool the primary pump seals) leak into the primary sodium coolant occurred, forming a particulate that restricted flow through the core region and caused overheating and damage to 13 of the 43 fuel elements. The time at which fuel damage first occurred is unknown, but the damage likely began around July 12-13, shortly after a restart, with the majority of failures occurring between July 21 and July 23 (Ref. 3). Despite abnormal temperature and radioactivity readings, the reactor was not shutdown until July 23.

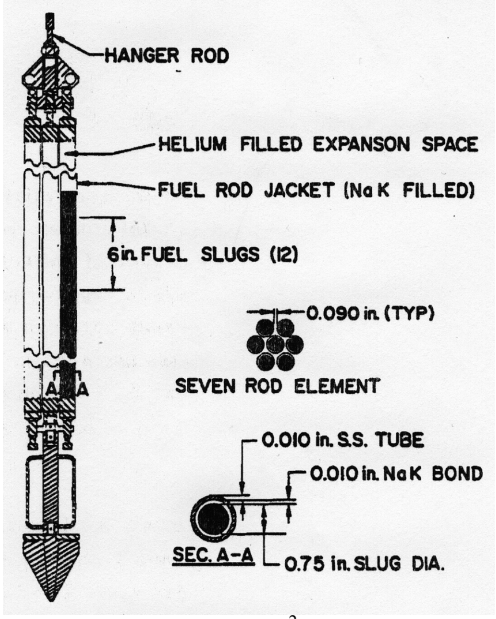

Fig. 1. Typical SRE fuel element[2].

While cladding failures occurred in 13 fuel elements (11 unalloyed uranium fuel elements, and two uranium alloy fuel elements)[2], no significant uranium melting was found beyond that which occurred as part of the eutectic formation with iron[3]. Fig. 2 (Ref. 2) shows the fuel damage in one of the fuel elements (channel 55) containing the experimental uranium alloy fuel.

Through an examination of the failed elements and thermocouple readings, it is thought that 11 of the fuel elements failed due to eutectic cladding penetration, while two failed due to repeated cycling through the α-β phase transformation temperature for uranium, which caused the fuel to expand until the cladding burst (as was the case for channel 55)[2]. Another possibility is that boiling of the NaK fuel bond, which has a lower boiling point than sodium (785°C vs. 883°C at atmospheric pressure), may have lead to higher pressure loadings on the cladding. It is estimated that peak fuel temperatures reached ~940°C, although the highest thermocouple reading was ~800°C (Ref. 2). The low burnup of the fuel at the time of failure (less than 0.1% core average) likely limited the release of fission gas, since the gas bubbles had not yet agglomerated[4].

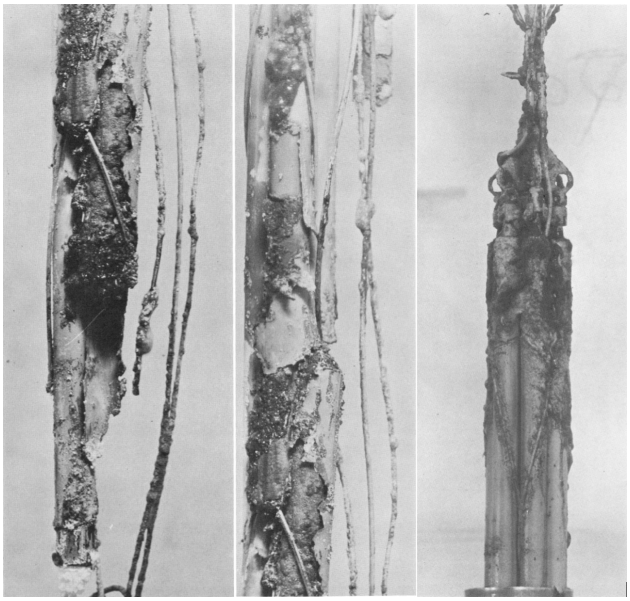

Fig. 2. Fuel damage in channel 55 of SRE (left: bottom, center: middle, right: top)[2].

The release at SRE is somewhat unique due to the presence of large quantities of carbon in the reactor. It is estimated that 7 to 70 lbs of carbonaceous material may have been deposited in the primary system as a result of the tetralin leak[3]. The carbon likely acted as a filter, removing some of the radionuclides within the primary sodium, as subsequent radioactive analysis of the carbon fragments showed a much higher concentration of fission product contamination than in the primary sodium. Subsequent analysis of the primary system also showed substantial plateout of Sr and Ce on primary piping, but not of Cs, since it is a fellow alkali metal of sodium[3].

Following the accident, samples of the primary sodium were taken to determine the radionuclides released from the fuel. Using this data, SRE staff developed the release fractions seen in Table II. However, the first primary sodium sample analysis was delayed approximately seven days following the accident due to high Na-24 activity. During this delay some portion of the original fission product activity decreased. In (Ref. 3), an attempt was made by SRE staff to account for these factors, although no details on the calculation are provided. The revised release estimates were Cs/Sr ~1%, I/Ba-La/Ce ~0.7%, Zr-Nb ~0.4%, and Ru ~0.2%. In addition to the isotopes listed in Table II, the following isotopes were identified, but not quantified in the analysis: Xe-133, Kr-85 (both were detected in the cover gas).

An investigation into the SRE accident noted several key conclusions[3]; of particular interest are the following:

1. Although significant fuel melting did not occur, the release fraction of the various fission products to the primary sodium indicated some degree of volatile fission product release.
2. Only Xe and Kr isotopes were identified in the reactor cover gas system.
3. The iodine release fraction was smaller than expected, and deposition in the primary system or escape to the cover gas occurred in undetectable quantities.
4. The carbonaceous particulate material that resulted from the tetralin leak proved to be an effective fission product scavenger.

TABLE II. SRE Estimated Radionuclide Release Fraction

Isotope	Estimated Release Percentage of Damage Fuel Inventory[3]
Cs-137	1.05%
Cs-134	0.66%**
Sr-89	0.92%
Sr-90	0.87%
I-131	0.32%
Ce-141	0.25%
Ce-144	0.22%
Ba-La-140	0.22%
Zr-95 + Nb-95	0.18%
Ru-103	0.09%

*From neutron capture in Cs-133; estimated

SRE was later repaired and resumed operations in late 1960, and continued to operate until 1964. Boeing, who later purchased the SRE site, was the target of a 2004 class action lawsuit claiming harm to local residents due to the 1957 accident. In particular, the plaintiffs argued that significantly more iodine had been released from the plant than what was documented in official estimates. The court case resulted in renewed interest in metal-fuel radionuclide release phenomena, and analyses prepared by expert witnesses on behalf of Boeing[4,5] concluded that there was no significant release of iodine due to retention within the fuel and primary sodium.

II.B. The Fermi I Accident

The Fermi 1 unit, located at the current site of the Enrico Fermi Nuclear Generating Station outside of Detroit, was a $200MW_{th}/66MW_e$ sodium fast breeder reactor operated by Detroit Edison. The intent of Fermi 1 was to demonstrate the commercial viability of a fast reactor power station. The reactor went online in 1963 and began generating power in mid-1966. Fermi 1 contained U-10wt% Mo fuel pins with zirconium cladding that were enriched to 25.6% U-235. The core consisted of 105 core subassemblies with 140 pins per square subassembly[6]. The core sat in a sodium-filled vessel, with inlet and outlet piping as part of a loop SFR design.

On October 5th, 1966, two pieces of a Zircaloy baffle broke loose from the melt-down section liner below the core. This Zircaloy device for retaining molten core had been a late addition to the Fermi 1 design[6]. The pieces of Zircaloy were carried upward by the primary sodium flow and were lodged at the entrance of the core region. The blockage starved coolant flow to the core and resulted in damage to four of the 105 subassemblies. Significant fuel melting occurred in two subassemblies, where it is theorized that flow was reduced to approximately 3% of nominal[6]. The other damaged subassemblies likely had flow reduced to 10% and 30% of nominal[7]. The entire incident evolved in one hour during a rise in reactor power (to 31 MW_{th}) as part of a plant test[6]. All four subassemblies had been in the reactor since the original loading program.

Subsequent analyses were performed to assess the recorded sodium temperature rise[8], and to estimate the approximate fuel temperature during the incident[9]. Although there are no recorded fuel temperatures from the melted assemblies (M-127 and M-098), based on analyses of the fuel behavior, it is thought that fuel pins failed soon after exceeding ~1100°C, with a maximum temperature of ~1400°C, before slumping and being cooled by lower structure or an infusion of sodium vapor[9]. The subsequent analyses also determined that once melting in M-127 and M-098 penetrated the subassembly can walls, stagnant sodium present between the subassemblies provided convective, two-phase cooling to the subassembly interior and prevented further damage[7]. It should be noted that no propagation of fuel melting to neighboring assemblies was observed[6].

As a result of the accident, Xe and Kr were both present in the reactor cover gas and Cs, I, Sr, and Ba-La were detected in the primary sodium. A small amount of Xe and Kr was released into the containment building through ordinarily insignificant leaks in the primary cover gas system, but radioactivity levels were not dangerously high, and no significant plant personnel exposure occurred. The containment building isolation system automatically activated and prevented any significant release through the waste stack[10].

Immediately following the accident, it was unknown how many fuel elements failed or melted. Table III presents an initial analysis of radionuclide release conducted by Fermi 1 staff based on the measured activity of the system[6]. A range of release fractions was assumed to provide bounds on the number of melted pins. In addition to the isotopes listed in Table III, the following isotopes were identified, but not quantified in the analysis: Xe-135, Sr-90, Ba-La-140, Ce-141, Ce-144, I-133, Ru-103, Zr-Nb-95. An explanation for the high Sr-89 readings was not provided.

Once the damaged fuel assemblies were removed, initial analysis appeared to show fuel-melting equivalent to a little greater than one fuel assembly worth of pins (140 pins)[6]. However, subsequent reports describe the number of melted fuel pins as equivalent to two fuel assemblies (280 pins)[11]. Table IV presents the estimated release fractions based on both cases. The release fractions in Table IV likely err on the conservative side, as all radionuclide releases are attributed to melted fuel pins, even though additional fuel assemblies suffered fuel pin damage.

Further analysis conducted by the operators at Fermi 1 found high plateout losses of Sr and Ba-La in the primary sodium, as shown in Table V. As expected, plateout of the alkali metal Cs was comparably low. These findings are similar to what was observed at SRE. However, no significant plateout in the heat exchangers was found[13].

A significant cleanup and repair operation was initiated after the Fermi 1 accident, and the metallic U-Mo fuel was replaced with an oxide core. Fermi 1 was restarted in 1970 and continued to operate until 1972.

TABLE III. Fermi 1: Fuel Failure Analysis for Radioactivity

Region	Isotope	Activity in System (Ci)[6]	Activity in Average Pin (Ci)[6]	Number of Fuel Pins Melted per Assumed Release Fraction[6]		
				10%	50%	100%
Cover Gas	Kr-85	1.43	0.02		142	71
	Xe-133	6.4	0.118		109	54
					240**	170*
Primary Na	Cs-137	2.75	0.17	162	32.4	16.2
	Sr-89	1570	5	3140	628	314
	I-131	5.04	0.33	153	30.6	15.3

* 100% release of Xe-133 and 0% release of I-133 (which would lead to additional Xe-133)
** 50% release of Xe-133, 10% release of I-133

TABLE IV. Fermi 1: Estimated Release Fractions Based on Findings in (Ref. 6)

Isotope	Estimated Release Fraction to Primary Sodium[6]	
	140 Pin Melt	280 Pin Melt
Kr-85	~50%	~10%
Xe-133	~100%*	<50%**
Cs-137	~10%	~5%
Sr-89	Inconclusive	Inconclusive
I-131	~10%	~5%

* Assuming 0% I-133 release
** Assuming 10% I-133 release

TABLE V. Fermi 1: Estimated Plateout Losses

Isotope	Estimated Plateout Loss[12]
Sr-89, Sr-90	93%
Cs-137	8%
Ba-La-140	75-84%

II.C. The EBR-II Fuel Failure Incident

EBR-II was a 62.5 MW_{th} SFR built and operated by Argonne National Laboratory at Argonne West located in Idaho. The reactor operated from 1963 to 1994. From November 23, 1967, to March 1968, a series of radionuclide releases occurred due to the melting of a U-Pu-Zr fuel element within an experiment capsule.

Capsule BC02, pictured in Fig. 3 (Ref. 14), contained an experimental ternary metallic alloy fuel element (U-15Pu-10Zr). Due to two flaws that likely occurred in the manufacturing process, sodium was lost from the capsule bond during irradiation. Adequate cooling could not be maintained due to the inability to transfer heat from the fuel[15]. On November 23, 1967, during the first reactor startup with BC02 in core, a spike in Xe-135 activity in the cover gas was detected, as seen in Fig. 4 (Ref. 14). However, the reading for Xe-133 did not increase significantly. This indicated a fresh fuel failure, since Xe-133 is a longer-lived radionuclide compared to the short-lived Xe-135.

Operations continued, and as the reactor was repeatedly cycled through power operations and shutdowns, which was common for EBR-II, the longer-lived Xe-133 became present by December 1967. By April of 1968, increases in the I-131 level in the primary sodium began to occur. A search was initiated to find the failed fuel element. Through the process of elimination, the experimental subassembly containing BC02 was removed in May 1968 (Ref. 14).

Subsequent evaluation and radiograph of BC02, seen in Fig. 5 (Ref. 14), indicated that gross fuel melting and rearrangement had occurred in the upper and lower regions of the fuel pin. The molten fuel flowed, or slumped, outward until it contacted the colder capsule walls[14]. It is likely that multiple melting events occurred over the approximately seven-month timeframe that BC02 was in the core. The increase in I-131 levels in the primary sodium, observed at the start of 1968, was likely the result of the release of any remaining bond sodium from the fuel element, which had become chemically

fixed with small amounts of iodine from the fuel during melting[14].

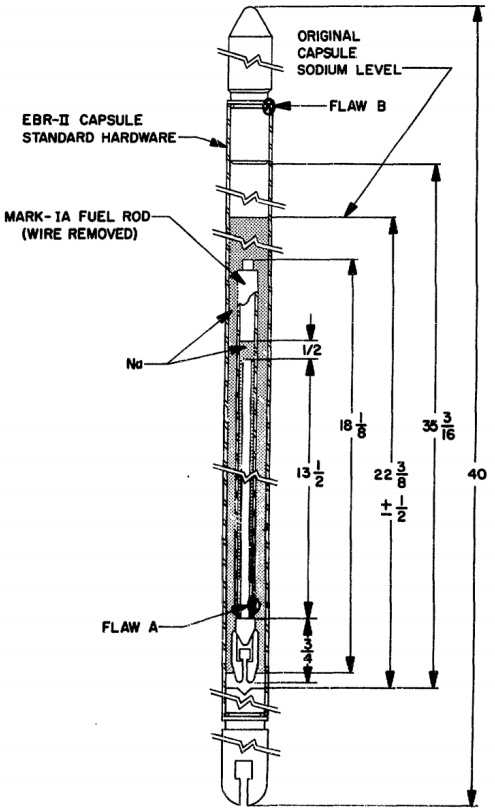

Fig. 3. Diagram of EBR-II Capsule BC02 (with two flaw locations highlighted)[14].

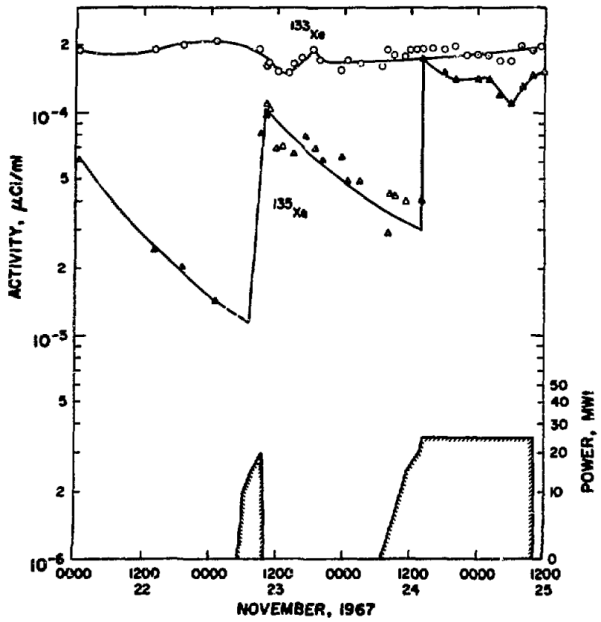

Fig. 4. EBR-II cover gas samples – November 1967 (Ref. 14).

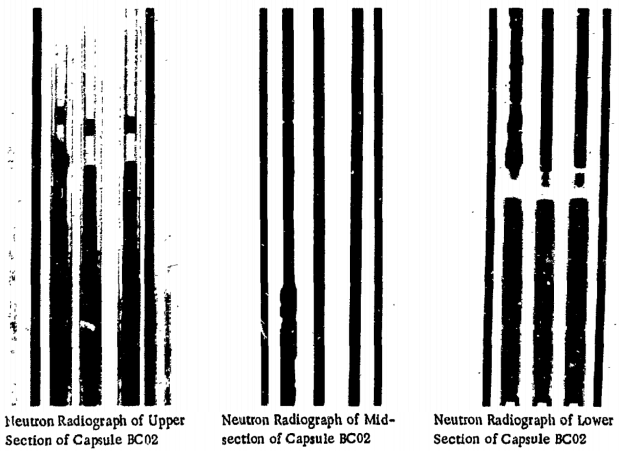

Fig. 5. Radiograph of EBR-II Capsule BC02 (Ref. 14).

II.D. EBR-II Run-Beyond-Cladding-Breach Tests

A series of experiments, referred to as the Run-Beyond-Cladding-Breach (RBCB) tests, were performed at EBR-II with metal fuel elements with intentionally weakened cladding. The goal of these experiments was to analyze the behavior of metal fuel after a cladding breach occurred and to demonstrate the compatibility between metal fuel and sodium. Tests were performed with U-Fs, U-Zr, and U-Pu-Zr fuel elements with various types of cladding[16].

The RBCB tests were performed by grinding down an area of the cladding surface of a pre-irradiated pin until only 30-40μm of the cladding remained[16]. The pin was then reinserted into the reactor, with cladding failure occurring shortly after reinsertion as pin pressure increased. The reactor would then continue to operate with the failed pin in core. A summary of the RBCB test results can be seen in Table VI (Ref. 16).

TABLE VI. Summary of RBCB Tests at EBR-II (Ref. 16)

Experiment ID Number	XY-21A	XY-24	XY-27	X-482	X482A	X482B
Composition, wt%	U-5Fs	U-19Pu-10Zr	U-19Pu-10Zr	U-19Pu-10Zr	U-10Zr	U-19Pu-10Zr
Cladding material	316SS	316SS	316SS	D9	D9	HT9
Final burnup, at%	~9.3	~7.5	~6.0	14.4	13.5	13.5
Pin diameter, mm	4.4	4.4	4.4	5.8	5.8	5.8
No. of days breached	54	233	131	168	100	150
DN signal, cps (note a)	~30-40	(note b)	(note b)	~600	~700	(note c)
Weight loss, g (note d)	2.0	2.7	2.5	4.0	3.6	3.9
Peak cladding temp(°C).	550	550	550	600	600	600

Notes: (a)Counts above background.
(b) Unavailable due to instrument malfunction.
(c) None detected, breached at startup.
(d) Expulsion of bond sodium fission gas and cesium accounts for the weight loss; fuel loss was negligible.

With the breach in cladding, fission gases and bond sodium were released from the pin. Cesium was also expelled into the primary coolant, as it had dissolved in the bond sodium. The expulsion of fission gas produced a virtually complete depressurization of the pin[16], resulting in no breach propagation (even after many days of continued operation). No fuel was extruded from the breach during any of the RBCB tests[17]. As shown in Fig.

6 (Ref. 18, 19), due to the metal fuel compatibility with sodium, no significant reaction occurs at the breach location, in contrast to oxide fuel, where the reaction with sodium exacerbates the cladding breach.

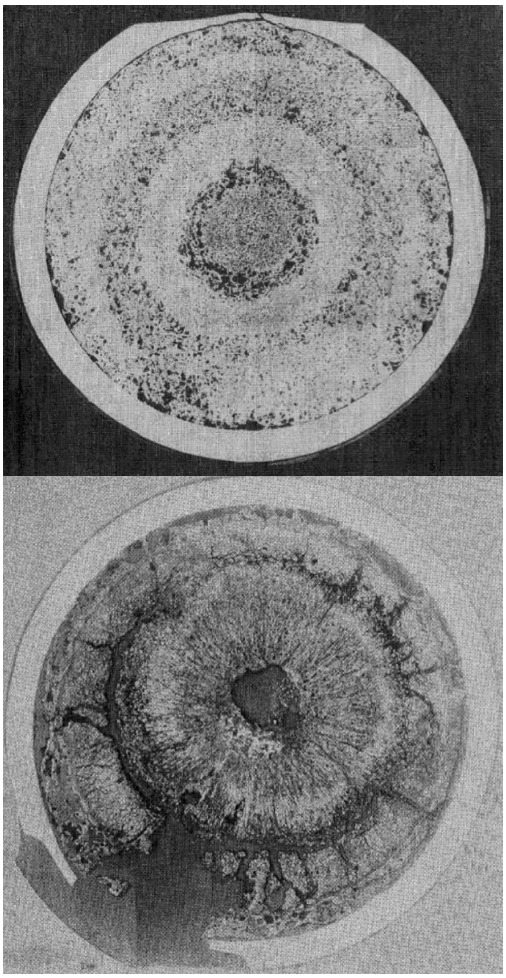

Fig. 6. Comparison of breach in cladding for metal fuel (top)[18] and oxide fuel (bottom)[19].

II.E. The TREAT M-Series Tests

In the mid-1980s, a series of safety tests were performed on metal fuel at the Transient Reactor Test (TREAT) facility at Argonne West in Idaho. These tests investigated the failure of metal fuel pins during transient overpower scenarios. The objective was to study the behavior of fuel and cladding near the cladding failure threshold for a range of burnup values and fuel/cladding combinations[16].

Beginning in 1985, tests M2 through M7 subjected metal fuel pins to a transient overpower with an eight second period. The fuel pins were placed within a tube that allowed sodium coolant flow throughout the test. Internal fuel melting occurred in all 15 of the fuel pins subjected to testing, however, only five pins were overheated to the point of cladding breach, which occurred in the range of slightly over four times nominal power. The breaches likely occurred due to the onset of rapid eutectic penetration of the cladding as fuel temperatures increased in conjunction with increasing internal pin pressure due to the expansion of fission gases within the pin or boiling of the sodium in the bond region[16].

The major findings of the tests indicated that the high thermal conductivity of metal fuel assured that peak cladding temperatures occurred near the top of the fuel column, where the coolant is the hottest. For the TREAT tests, this meant that cladding breach also occurred near the top of the fuel pin. It should be noted that this may not be the case for other metal-fuel pin designs with significantly different power profiles[20].

Of the five pins that experienced cladding breach, about half of the fuel inventory was ejected from the fuel pin as molten material through the (small) breach at the top of the pin. The fuel material was then swept out of the fuel region and deposited on surfaces of the sodium test loop upstream of the active region of the test fuel[20]. No significant flow blockages were observed from the ejected fuel.

In terms of radionuclide transport, no radionuclides were found for those tests where the cladding of all pins remained intact, despite fuel melting within the pin. For the tests where cladding breaches occurred, Rb-89, Cs-138, and Xe-138 were detected in the loop, likely as a result of the following chain[21]:

1. Fission products Br-89 and I-138, which are soluble in sodium, were released from the fuel and transported efficiently in the coolant.
2. Br-89 and I-138 decayed into noble gases, Kr-89 and Xe-138, which escaped into the gas plenum.
3. Kr-89 and Xe-138 decayed into Rb-89 and Cs-138, which settled out on the test loop wall.

III. SUMMARY

There have been three past U.S. sodium reactor incidents involving metal fuel. These incidents provide insight into the behavior of metal fuel during real core damage events. The extent of fuel damage differed in each incident, ranging from cladding failure and eutectic melting at SRE, to repeated substantial melting of the experimental fuel element at EBR-II. Perhaps the biggest takeaway from the three incidents is that no radionuclides other than the noble gases of xenon and krypton were found in the cover gas region. This implies that significant retention occurred, whether in the fuel or in the primary sodium, of many of the important radionuclides that are commonly a concern during light-water reactor core damage accidents.

Several of the important insights from the three accidents, in conjunction with the RBCB and TREAT experiments, are summarized below:

- Metal fuel cladding breach results in a release of fission gases (including Xe and Kr) and bond sodium (which may contain Cs), but does not imply a release of fuel material.
- Metallic fuel does not react with the sodium coolant, limiting the likelihood of fuel damage propagation.
- The retention of radionuclides, other than the noble gases, in the fuel pin and primary sodium appears to be very high, (even during significant fuel melting and relocation, such as at Fermi 1 and the EBR-II experimental capsule).
- All three reactor incidents involved fairly fresh fuel with relatively low burnup.
- For all three reactor incidents, cleanup operations were conducted and the reactors resumed normal operation.

ACKNOWLEDGMENTS

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