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UNITED STATES OF AMERICA NUCLEAR REGULATORY COMMISSION DEFICE OF SECRETARY DOCKETING & SERVICE Before the Atomic Safety and Licensing ANOBER In the Matter of) Philadelphia Electric Company) Docket Nos. 50-352 0 (

(Limerick Generating Station, Units 1 and 2

RELATED CONTROL DNDENCE

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TRANSMITTAL OF REFERENCE 9 TO APPLICANT'S TESTIMONY RELATING TO CITY-18 AND CITY-19

On November 1, 1984, Applicant transmitted to the Atomic Safety and Licensing Board and involved parties its testimony relating to Contentions City-18 and City-19 and references to that testimony. Applicant is hereby transmitting to the Board and involved parties copies of "Fission Product Behavior During the First Two PBF Severe Fuel Damage Tests" by D.J. Osetek, R.R. Hobbins, A.W. Cronenberg and K.Vinjamuri, which is Reference 9 to the testimony which had not been previously available.

> Respectfully submitted, CONNER & WETTERHAHN, P.C.

Mark J. Wetterhahn Counsel for the Applicant

December 7, 1984

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UNITED STATES OF AMERICA NUCLEAR REGULATORY COMMISSION

In the Matter of	1	
Philadelphia Electric Company) Docket Nos. 50-	352
(Limerick Generating Station, Units 1 and 2))	555

CERTIFICATE OF SERVICE

I hereby certify that copies of "Transmittal of Reference 9 to Applicant's Testimony Relating to City-18 and City-19," dated December 7, 1984 in the captioned matter have been served upon the following by deposit in the United States mail this 7th day of December 1984:

- * Helen F. Hoyt, Esq. Chairperson Atomic Safety and Licensing Board U.S. Nuclear Regulatory Commission Washington, D.C. 20555
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- * Dr. Jerry Harbour Atomic Safety and Licensing Board U.S. Nuclear Regulatory Commission Washington, D.C. 20555

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Fission Product Behavior during the First Two PBF Severe Fuel Damage Tests

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July 15-19, 1984

ANS Topical Meeting on Fission Product Behavior and Source Term Research

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FISSION PRODUCT BEHAVIOR DURING THE FIRST TWO PBF SEVERE FUEL DAMAGE TESTS*

D. J. Osetek, ** A. W. Cronenberg, # R. R. Hobbins ** and K. Vinjamuri**

ABSTRACT

The results of the first two severe fuel damage tests performed in the Power Burst Facility are assessed in terms of fission product release and chemical behavior. Un-line gamma spectroscopy and grab sample data indicate limited release during solid-phase fuel heatup. Analysis indicates that the fuel morphology conditions for the trace-irradiated fuel employed in these two tests limit initial release. Only upon high temperature fuel restructuring and liquefaction is significant release indicated. Chemical equilibrium predictions, based on steam oxidation or reduction conditions, indicate I to be the primary iodine species during transport in the steam environment of the first test and CsI to be the primary snecies during transport in the hydrogen environment of the second test. However, the higher steam flow rate conditions of the first test transported the released iodine through the sample system; whereas, low-hydrogen flow rate of the second test apparently allowed the vast majority of iodine-bearing compounds to plateout during transport.

INTRODUCTION

As a result of the accident in the Three Mile Island Unit-2 pressurized water reactor (PWR) on March 28, 1979, the U.S. Nuclear Regulatory Commission has initiated an international Severe Fuel Damage Research Program.^{##} The principal in-pile testing portion of this program is being conducted at the Idaho National Engineering Laboratory in the Power Burst Facility (PBF), where a bundle of 32, 0.9-m-long, PWR-type fuel rods is brought to coolant boiloff conditions, severe cladding

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Sponsors of the program include Belgium, Canada, Federal Republic of Germany, Italy, Japan, Netherlands, United Kingdom, and United States. oxidation, molten alpha-zircaloy dissolution of UU_2 , release of fission products, and ultimate rod destruction. Four bundle experiments are presently planned in the PBF Severe Fuel Damage (SFD) program. This paper summarizes results with respect to the fission product behavior observed during the first two experiments [SFD Scoping Test (SFD-ST) and SFD 1-1]. Details of the test program are described in Reference (1).

FISSION PRODUCT DETECTION SYSTEM

The on-line fission product sampling and monitoring system is shown schematically in Figure 1. The test effluent, consisting of steam, hydrogen, and fission products, is drawn from the fuel bundle and routed to the monitoring system through a 1.3-cm (1/2-in.) diameter stainless steel pipe. Six effluent steam samples are remotely opened at various times during the test to provide samples containing condensed steam, fission products, and hydrogen for 'posttest analysis. The remaining steam is condensed and cooled to a temperature below 340 K. The effluent then enters a separator vessel, where a continuous nitrogen gas purge sweeps hydrogen, fission gases, and other noncondensables from the separator, past a gamma spectrometer and a hydrogen monitor, into a collection tank. The liquid from the separator



Figure 1. SFD Fission Product and Hydrogen Monitoring System for Test 1-1.

vessel was monitored during the SFD-ST by two gamma spectrometers located upstream and downstream of a 5-micron filter. Downstream of the filter the liquid effluent passed through one of six remotely operated flow-through liquid grab samples into the collection tank. The gamma spectrometer located downstream of the filter was relocated to monitor the steam line upstream of the condenser during Test SFD 1-1.

TEST RESULTS

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Table 1 presents an overview of the test parameters and basic results for the SFD-ST and Test SFU 1-1. Both tests employed fresh fuel, which was preconditioned prior to the transient to ensure the generation of both short- and long-lived fission products. At the end of preconditioning, the high temperature transient was initiated at an effective burnup of =0.008 to 0.009 at.%. The SFD-ST was conducted on Octuber 29, 1982. The 32-rod bundle was subjected to a slow heating ramp of =0.13 K/s to peak fuel temperatures in the range of 1700 K in an oxygen-rich environment, followed by oxidation-driven heatup at =10 K/s to 2400 K, and was terminated by a rapid quench and coolant reflood. Considerable cladding oxidation and melting, fuel liquefaction, and fuel fragmentation occurred.

Test SFD 1-1 was conducted on September 8, 1983. This test was designed to simulate the fuel heatup during a small-break loss-of-coolant accident without emergency core cooling. The initial heating ramp was =0.45 K/s to cladding temperatures of =1300 K, followed by an exothermic oxidation driven heatup rate of =1.3 K/s to 1700 K, with a subsequent runaway oxidation ramp of =30 K/s to 2400 K. The test was terminated by a gradual decrease in power and fuel cooldown. An argon purge was used during part of the cooldown to sweep fission products from the bundle.

Figure 2 illustrates the noble gas and iodine behavior for both tests, as a function of peak fuel temperatures. A small burst of activity (fractional release rate 10^{-7} min^{-1}) is noted at fuel temperatures of $\approx 1100 \text{ K}$, which corresponds to the approximate time of rod failure and release of the fuel-cladding gap inventory of noble gases. Only a small amount of fission product release was detected in either test until the fuel temperatures reached 1700 K. Fractional release rates increased from 10^{-6} to 10^{-3} min^{-1} as fuel temperature increased to 2400 K. Fission product release rates increased sharply in the SFD-ST when the fuel bundle was quenched from high temperature by rapid injection of reflood water. Release

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Nominal Coolant Fuel Cooldown Flow Burnup Comments Mode (g/s)(at.%) Heating Rate Test Oxygen rich Quench 16.0 Highly oxidized bundle 0.0089 U.13 K/s to 1700 K SFD-ST Fuel liquefaction 10.0 K/s to 2400 K 375 g H₂ generated Steam starved Slow 0.67 0.0079 0.45 K/s to 1300 K Less oxidation SFD 1-1 1.3 K/s to 1700 K Fuel liquefaction 72 g H₂ generated 30 K/s to 2400 K 10° Liquidus period SFD 1-1 Noble gas 10' Frectional Release Rate (min⁻¹) 10-2 NUREG-0772 10.3 104 SFD 1-1 lodine 10-8 10. SFD-ST Noble gas 10" SFD-ST lodine 10." leflood Cooldown Heatup 10. 1000 1700 2400 1700 1000 PS 010-684-08 Fuel Temperature (K)

SUMMARY OF THE SFD-ST AND SFD 1-1 TEST CONDITIONS



rate constants for noble gas and iodine reached 10⁻¹ min⁻¹ momentarily during the quench. Grain boundary shattering that occurs as a result of such quenching is believed to be the reason for the enhanced release rates.

Fractional release rates of noble gas during Test SFD 1-1 peaked at 3 x 10^{-2} min⁻¹ during the cooldown period. This is several orders of magnitude higher than the

release rates during heatup at the same temperature. This lack of a direct dependence on fuel temperature of fission product release is indicative of the importance of fuel behavior to release modeling. Although the bulk temperature in the Test SFD 1-1 fuel bundle was decreasing, the probable formation of a low melting point (1400 K) metallic U, Zr-phase at grain boundaries and pellet surfaces, as noted in Reference (2), could account for the continued release of fission products until temperatures dropped below \sim 1400 K.

Iodine was measured during heatup in the SFD-ST and found to agree with the noble gas release data, but iodine was not measured in Test SFD 1-1 until after slow cooldown was initiated. This difference may be due to iodine transport effects, since the effluent flow rate was higher (=16 g/s) in the SFD-ST and was predominately steam; whereas, the flow rate from the SFD 1-1 bundle was slower (=0.08 g/s) and principally hydrogen. Iodine transport in an oxidizing steam environment is predicted to be in the atomic iodine form, which undergoes little reversible (condensation) plateout. This is consistent with the measured iodine release for the SFU-ST. However, for a highly reducing environment, CsI, HI, and I species are predicted, where CsI undergoes rapid plateout such that little iodine would be detected, as indicated by the SFD 1-1 data. The influence of oxidizing versus reducing environments on steam-iodine chemistry is discussed later in this paper. Fission product washout occurred in both tests during reflood; iodine and cesium release rates from the bundle peaked during reflood at $\sim 3 \times 10^{-2} \text{ min}^{-1}$.

The measured fission product fractional release rates are compared in Figure 2 with the correlations for noble gas release presented in NUREG-0772 ($\underline{3}$). During the fuel heatup phase of the SFD tests, the release data were found to be generally lower than the out-of-pile data by a factor of $\approx 10^3$. Only when temperatures in the bundle were held above the UO₂-Zr liquefaction temperature (≈ 2170 K), did the release rates increase and eventually exceed those predicted in NUREG-0772. Release rates measured during quench and reflood phases were also consistently larger than those predicted from NUREG-0772 based on temperature alone. Details of fission product release from fuel and transport behavior in either the steam (during bundle boiloff) or water (upon bundle requench) carrier, are presented in the following section.

FISSION PRODUCT RELEASE BEHAVIOR

To interpret the noble gas release behavior for the SFD-ST and Test SFD 1-1, analysis was performed with the FASTGRASS code ($\underline{4}$), which is a mechanistic model for predicting fission gas and volatile species (I and Cs) behavior in UO₂ fuels during steady state and transient conditions. A comparison of calculated (FASTGRASS and NUREG-0772) and SFD-ST measured noble gas release rates as a function of fuel temperature is shown in Figure 3(a). As indicated, the FASTGRASS-predicted and ST measured release rates agree quite well, while a discrepancy of 10³ is noted relative to the predicted rates based upon the temperature correlations in NUREG-0772. Such a discrepancy is explainable based on the morphology characteristics of the trace-irradiated fuel employed in the SFD tests. The correlations in NUREG-0772 were developed primarily from release experience for medium-to-high burnup conditions, while the fuel tested in both SFD tests was essentially fresh, except for the develcoment of a small inventory of fission products at an effective burnup level of *0.008 to 0.009 at.*.

FASTGRASS analysis indicates that for trace-irradiated fuel, most of the nuble gas and volatile I and Cs inventory is still retained within the interior of the fuel grains, primarily as individual atoms. Since fission products in atomic form are readily accommodated within the solid fuel microstructure, they experience little release from fuel in the solid condition. Only at elevated temperatures is there a change in morphology characteristics from one of atoms in lattice solution to bubble precipitation at grain boundaries. This situation is illustrated in Figure 3(b), where at low temperatures the vast majority of the noble gas inventory is predicted to be retained in lattice solution, while at temperatures above 1900 K



Figure 3. Comparison of the Measured Noble Gas Release Rates During the SFD-ST with FASTGRASS-Predicted Release Rates.

the noble gas and volatile fission products are swept to grain boundary edges and faces. Upon destruction of the fuel grain structure, either due to fuel liquefaction (as occurred in the SFD-ST and Test SFD 1-1) or quench induced grain-boundary shattering (as occurred in the SFD-ST), the fission products pinned to grain boundaries are released ($\underline{5}$).

In Figure 3(a), it is noted that two calculational approaches were used in the FASTGRASS analysis of the SFD-ST release data, one employing a nominal grain-growth model and the other for fuel-oxidation enhanced grain growth. Both models are of the functional form:

(1)

$$D_t^2 = D_o^2 + Aexp (Q/RT)t$$

where A is a proportionality constant, D_t is the grain size at time t, D_o is the initial grain size, Q is the activation energy, R is the gas constant, T is temperature, and t is time. The parameters A and Q are dependent upon such fuel crystal properties as atomic packing, atomic mobilities, and oxidation state. The results shown in Figure 3(a) indicate that the enhanced grain growth model more closely approximates the SFD-ST data.

Steam-induced fuel oxidation following cladding failure, can have a pronounced effect on atomic mobilities and, therefore, fission product release characteristics. Figure 4 presents a plot of the diffusivity of ¹³³xe in UO_{2+x} as a function of





fuel stoichiometry ($\underline{6}$) and shows that increased oxygen content in solution in UO₂ is observed to increase the diffusivity of ¹³³Xe. Enhanced atomic mobilities result in enhanced grain growth, which in turn causes a sweeping of once-entrapped fission products from the grain interior to grain boundaries. This situation is illustrated in Figure 5 and supported by posttest examination of the SFD-ST fuel. Figure 6 is a photomicrograph of the SFD-ST fuel indicating a change in fuel stoichiometry from initially UO_{2.0} to UO_{2+x} plus U₄O₉ precipitates, and an increase in grain size from an as-fabricated condition of =4 to 35 microns. Further discussion of the governing role that fission product morphology characteristics, grain growth, and the formation of low melting point eutectics exert on release behavior are presented in References (2), (4), (5), (6), and (7).

In summary, the sequence of events leading to fission product release for the trace-irradiated fuel employed in the SFD-ST and Test SFD 1-1 appears to be as follows:

- Initial high-fission-product retention within individual grains as a result of entrapment of gaseous and volatile fission products as individual atoms or intragranular microbubbles, with nil gas release.
- In a steam-rich environment (i.e., SFD-ST), fuel-oxidation-enhanced grain growth can result in intragranular atomic and microbubble sweeping to grain boundaries and initiation of slow gas release. Under steam starvation conditions (i.e., SFD 1-1) this effect is less pronounced.
- Destruction of the grain structure via fuel liquefaction or quench-induced grain boundary shattering, with rapid-enhanced intergranular gas release.
- Sustained gas release during cooldown as the liquefied U, Zr mixture remains molten to lower temperatures (∿1400 K).

Analysis of the SFD data indicates that the release characteristics for the trace-irradiated fuel employed in the SFD-ST and Test SFD 1-1 differ from the release characteristics predicted by NUREG-0772 which is attributed to large differences in fuel morphology for low versus medium-to-high burnup fuel. In addition to morphology, fission product behavior is also strongly influenced by steam chemistry once released from fuel, as discussed in the following section.

FISSION PRODUCT/TRANSPORT CHEMISTRY

Upon release from fuel, fission products such as Cs and I can mix and react with the steam and hydrogen produced by the steam-zircaloy reaction. The chemical



Figure 5. Illustration of Retained Noble Gas/Volatile Distribution for Trace-Irradiated Fuel Subjected to Grain-Growth and Fission-Product Sweeping Phenomena



Figure 6. Postirradiation Microstructure of the SFD-ST Fuel Remains, Indicating Grain Growth to 35 microns and UO2 Oxidation to U409-Inclusions.

composition of this vapor mixture depends on species concentration, temperature, pressure and oxidation/reduction conditions. From thermodynamics, the equilibrium composition, at a particular temperature and total pressure, can be found by noting that the total free energy of formation (ΔG_f°) at chemical equilibrium approaches zero, that is:

$$\sum \Delta G_{f}^{\circ}$$
 (Products) - $\sum \Delta G_{f}^{\circ}$ (Reactants) ≈ 0

For an ideal gas, ΔG_{f}° can be expressed as

 $\Delta G_{f}^{\circ} = RT \ln (p_{i})$

where p_i is the partial pressure of a particular gaseous component of the reacting mixture. Changes in the H-O ratio will produce a change in the concentration levels of the other components, such that equilibrium is maintained. The results of Sallach's equilibrium analysis (8) for the Cs-I-O-H system, was used to assess the primary chemical forms for the SFD-ST and Test SFD 1-1 conditions.

Table 2 summarizes the fission product concentration and thermal-hydraulic conditions at the time of enhanced fission product release for the SFD-ST and Test SFD 1-1; that is, the time at which fuel temperatures exceed =2000 K. The Cs-I atom ratio was estimated to be =10, based upon the ORIGEN-calculated fission product yields and associated radioactive decay chains at the initiation of the high temperature transient. The concentration and H-O ratio were determined from the experimental steam and H_2 -gas flow rates measured during each test.

Table 2

SUMMARY OF THERMODYNAMIC AND CONCENTRATION CONDITIONS FOR THE SFD-ST AND TEST SFD 1-1 AT THE TIME OF ENHANCED FISSION PRODUCT RELEASE

Parameter	SFD-ST	SFD 1-1
Steam temperature	=1400 K (=1130°C)	=1400 K (=1130°C)
System pressure	=7.0 MPa (=70 bar)	=6.6 MPa (=66 bar)
H/O mole ratio	=2.06	=45
Cs/I mole ratio	=10	=10
I/H ₂ O mole ratio	=10=9	=10 ⁻⁸
Cs/H ₂ O mole ratio	=10=8	=10 ⁻⁷

. . .

(2)

(3)

Figure 7 illustrates the approximate fractional iodine species concentrations as calculated by Sallach, in the steam/hydrogen environment for the SFD-ST and SFD 1-1 test conditions. At lower temperatures, CsI is stable in a steam environment; however, at increased temperatures the CsI molecule undergoes dissociation via the reaction:

 $2 \operatorname{CsI}(g) + 2 \operatorname{H}_{2}O(g) \longrightarrow 2 \operatorname{CsOH}(g) + 2 \operatorname{I}(g) + \operatorname{H}_{2}(g) \longrightarrow 2 \operatorname{CsOH}(g) + 2 \operatorname{HI}(g)$ (4)

resulting in increased cesium-hydroxide (CsOH) formation, where temperature, pressure, and oxidation/reduction conditions affect the distribution of iodine among I and HI species.

It should be noted that for trace-irradiated fuel, the release concentration of iodine into steam is on the order of 10^{-8} to 10^{-9} , while the lowest concentration levels presented by Sallach are on the order of 10^{-7} . Since the effect of decreasing iodine concentration is toward a diminished abundance of CsI at thermochemical equilibrium, the results plotted in Figure 7 may overpredict somewhat the CsI mole fractions for the SFD test conditions. Nevertheless, using Figure 7 as a guide, the following fractional partitioning of I and Cs species are estimated:





	SFD-ST	SFD 1-1
T _{steam} ≃ H/U ≃	1400 K 2.06	$T_{steam} \approx 1400 \text{ K}$ H/O ≈ 45
Cs/I =	10	Cs/I ≈ 10
Moles of	I as CsI = 10%	Moles of I as CsI = 48%
Moles of	I as HI = 10%	Moles of I as HI = 48%
Moles of	I as I = 80%	Moles of I as I = 4%
Moles Cs	as CsOH = 99%	Moles Cs as CsUH = 95.2%

Although Figure 7 presents only iodine partitioning information, the CsOH mole fraction can be estimated, based on the fact that at temperatures below =2000 K little compound dissociation into atomic cesium occurs; thus, any cesium not bound to iodine is considered to be CsOH. For a Cs/I ratio of =10, a 10% mole fraction of I as CsI equates to 99% Cs as CsOH, while 48% I as CsI is equivalent to 95.2% Cs as CsOH. For the oxidizing environment of the SFD-ST, the vast majority of iodine in the high temperature bundle region is predicted to exist as low boiling point volatiles, namely 80% atomic iodine and 10% HI. As indicated in Table 3, I_2 and HI have low vaporization temperatures and, thus, are subject to limited reversible plateout (condensation) during the short transport time (~3 s) through the test train pipe network to the monitoring system. Atomic iodine is assumed to behave similar to I_2 . The high mole fractions of I and HI in an oxidizing environment, therefore, explain the fact that similar iodine and noble gas release rates were measured for the SFD-ST.

Table 3

VAPORIZATION TEMPERATURES OF I AND CS SPECIES

	Species				
	ні	12	Cs	CsOH	CsI
Tyap (K) at 1 atm	238*	457	951	1263	1553
T _{vap} (k) at 70 atm	411	860	2275	2180	2745

* At 4 atm.

For the reducing environment of Test SFD 1-1, however, approximately 48% of the iodine is predicted to be in the form of CsI and, thus, is subject to a condensation mode of plateout within the high temperature regions of the test bundle. The remaining iodine, as I and HI gases, would then be carried by steam and/or hydrogen from the bundle through the flow pipe to the fission product monitoring system. Due to the relatively low flow rate (from =0.67 g/s H_2O to 0.07 g/s hydrogen) and long transport time (\sim 600 s), cooldown of the effluent in the piping is considered to result in transformation of I and HI to CsI at lower mixture temperature (\sim 600 K), such that chemical equilibrium is maintained, as indicated by the arrow in Figure 7(b). This transformation to CsI results in additional iodine condensation and plateout on the inside piping surface, and is considered the reason for the nil iodine release measured during the high temperature phase of Test SFD 1-1. Only upon bundle reflood was significant iodine measured for Test SFD 1-1, indicative of washout of reversible CsI deposits.

Such results indicate that the observed I and Cs behavior for the PBF-SFD tests can be explained in terms of thermochemical phenomena, and again illustrate the point that an in-depth evaluation of such data can be used to confirm physical, chemical, and mechanistics models of fission product behavior for severe accident conditions. However, it should be noted that this analysis was limited to the I-Cs-H-O system, and the presence of other fission products and bundle materials could influence the chemical forms produced in these tests.

CONCLUSIONS

From the foregoing analysis, the following conclusions can be drawn relative to noble gas and volatile fission product release from fuel and transport behavior in the SFD test environments:

- Fission product release is strongly influenced by prior irradiationinduced fuel morphology characteristics. Analysis of the PBF SFD-ST and Test SFD-1-1 release data for such trace-irradiated fuel indicates limited release on heatup to temperatures up to 1900 K, since the majority of the noble gases and volatiles are retained within the grain interior as individual atoms.
- A comparison of fission product data, posttest fuel examination, and analysis indicates that in a steam-rich environment (i.e., SFD-ST), oxidation-induced grain growth and sweeping of fission products to grain boundaries can result in enhanced noble gas and volatile release for trace-irradiated fuel in the solid state. Subsequent fuel liquefaction and quench-induced grain boundary shattering result in rapid fission product release, as indicated by the SFD-ST and Test SFD 1-1 data.

- Iodine and cesium chemistry in a steam environment are strongly influenced by oxidation/reduction and concentration conditions. The high steam flow rate and low concentration conditions of SFD-ST result in predominately free iodine and CsOH transport in steam. However, for the low-flow rate reducing environment of Test SFD 1-1, the predominant chemical forms are CsI, CsOH, and HI.
- The fact that free iodine is subject to limited reversible plateout, accounts for transport and measurement of iodine during the heatup phase of the SFD-ST. However, for the reducing atmosphere of Test SFD 1-1, plateout of CsI and cooldown-induced transformation of HI to CsI account for the observation of limited iodine detection during the heatup phase of Test SFD 1-1. Only upon posttransient reflood of the Test SFD 1-1 bundle was significant iodine release observed, which is considered to be washout of reversible CsI deposits.

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NUTICE

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