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FISSION-PRODUCT-RELEASE SIGNATURES FOR LWR FUEL RODS FAILED DURING PCM AND RIA TRANSIENTS

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ABSTRACT

This paper discusses fission product release from light-water-reactor-type fuel rods to the coolant loop during design basis accident tests. One of the tests was a power-cooling-mismatch test in which a single fuel rod was operated in film boiling beyond failure. Other tests discussed include reactivity initiated accident (RIA) tests, in which the fuel rods failed as a result of power bursts that produced radial-average peak fuel enthalpies ranging from 250 to 350 cal/g. One of the RIA tests used two previously irradiated fuel rods. On-line gamma spectroscopic measurements of short-lived fission products, and important aspects of fission product behavior observed during the tests, are discussed. Time-dependent release fractions for short-lived fission products are compared with release fractions suggested by: the Reactor Safety Study; NRC Regulatory Guides; and measurements from the Three Mile Island accident. Iodine behavior observed during the tests is discussed, and fuel powdering is identified as a source of particulate fission product activity, the latter of which is neglected for most accident analyses.

INTRODUCTION

Safety analyses conducted for the purposes of licensing nuclear power plants require estimates of the dose consequences from postulated accidents. However, the magnitude and duration of fission product release during accidents are highly sensitive to fuel behavior, the accident scenario, and the chemical form and inventory of fission products. Because large (conservative) uncertainties usually exist in consequence estimates, in-pile tests that define actual consequences are important.

Five simulated accident tests on single, light-water-reactor (LWR)-type fuel rods were conducted in the Power Burst Facility (PBF) at the Idaho National Engineering Laboratory.[1-3] This paper presents fission product behavior results from these tests and compares measured fission product releases with NRC Regulatory Guides,[4-6] Reactor Safety Study estimates,[7] and TMI accident data.[8,9]

The PBF is a specialized reactor designed to test nuclear fuel and components under off-normal operating conditions. This facility is operated by EG&G Idaho, Inc., for the U.S. Nuclear Regulatory Commission, under contract

with the U.S. Department of Energy, and consists of an open-pool reactor with a flux trap and a separate in-pile coolant loop. Using on-line gamma spectroscopy, the PBF fission product detection system[10] monitored fission product activity released to the coolant loop during the tests. Gamma spectra acquired during the tests were analyzed to determine the relative concentrations of identifiable fission products. Release fraction histories were developed by comparing the measured releases with the inventories calculated by the ORIGEN computer code.[11]

TEST PCM-1

Test PCM-1 evaluated the behavior of a single pressurized-water-reactor(PWR)-type fuel rod subjected to a power-cooling-mismatch (PCM) event in which a high-temperature film-boiling operation occurred for 400 s beyond cladding failure. Large local regions of molten fuel provided a potential for energetic molten fuel/coolant interaction upon failure. The rod was operated in film boiling for a total of 900 s, and rod failure occurred 500 s after the first indication of film boiling, due to cladding embrittlement and fragmentation. No energetic reactions as a result of the failure and rod breakup were observed. Shutdown of the reactor and ensuing quench of the rod caused severe rod fragmentation.

RIA TESTS

Fuel rod failure and fission product release to the coolant loop occurred during four of the PBF reactivity initiated accident tests (RIA-ST-1, RIA-ST-2, RIA-ST-4, and RIA 1-1). The short-lived fission product inventory is produced during a natural power burst that spans ~50 ms. The thermal transient initiated by the power burst affects fuel and fission product behavior for approximately 30 s.

Fuel rod conditioning was performed during Test RIA-ST-1 to promote cracking and relocation of the fuel pellets and to build up a fission product inventory. An axial-peak, radial-average fuel enthalpy of 185 cal/g was achieved in the first power burst of RIA-ST-1. No indication of fuel rod failure was observed. The second power burst of RIA-ST-1 caused an axial-peak, radial-average fuel enthalpy of 250 cal/g and rod failure.

The RIA-ST-2 fuel rod was exposed to a single power burst, with no significant steady state operation. The axial-peak, radial-average fuel enthalpy of 260 cal/g achieved during this single power burst caused rod failure. The posttest appearance of the RIA-ST-2 rod was similar to that of the RIA-ST-1 rod; no evidence of fuel melting or grain growth was observed. Massive oxidation, oxide spalling, cladding splitting and fracture, and cladding ridging occurred. Approximately 10 to 15% of the fuel was lost to the coolant loop, due to fuel grain separation or powdering.

Fission product release data were obtained during Test RIA-ST-4; however, the primary objective of the test was to assess the safety of the PBF during

severe fuel rod failure tests in which a potential for energetic molten fuel/coolant interactions existed. The axial-peak, radial-average fuel enthalpy caused by the RIA-ST-4 power burst was 350 cal/g at the time of rod failure. A power transient of this magnitude is greater than that possible in a commercial reactor during an RIA. Severe fuel fragmentation occurred during RIA-ST-4; a total of 155 g of fuel fragments were collected from within the test train. Fuel lost to the coolant loop was estimated to be <1%. Approximately 90% (570 g) of the fuel melted. Most of the particles found in the test train were spherical or rounded, suggesting that the fuel was molten at the time it fragmented. Detailed posttest analyses[12] suggest that 3500 K was the probable maximum fuel temperature.

Test RIA 1-1 included four fuel rods, two previously irradiated (4.6 MWd/kg) and two unirradiated. The test included a preconditioning phase during which the fuel rods were cycled through several power ramps. The rods were then subjected to a single power transient, causing an estimated axial-peak, radial-average fuel enthalpy of 285 cal/g. Posttest examinations confirmed that all four test rods failed as a result of the RIA power burst. Approximately 43% and 72%, respectively, of the two previously irradiated test rods were fragmented during the test transient; however, no fuel was lost from the region within the flow shrouds, due to rapid formation of complete coolant flow blockages by relocation of rod fragments and molten UO_2. Approximately 34% and 44%, respectively, of the two previously unirradiated fuel rods in Test RIA 1-1 were fragmented, and up to 27% of the fuel debris was dispersed to the PBF coolant loop. Fuel melting occurred in less than 1% of the irradiated and unirradiated fuel.

FISSION PRODUCT BEHAVIOR

Since the fission product detection system was still in its developmental stages when most of these data were collected, the absolute calibration of the spectrometer was uncertain until Test RIA 1-1. Therefore, to provide the most conservative estimate of the fission product release fractions, it was necessary to normalize the data from Test PCM-1 and the RIA-ST series to a value of 1.0 for the largest release fraction measured in each test. Data are presented as a listing of the normalized or absolute isotopic release fractions of the principal fission products measured after the isotopes reached equilibrium concentration. This procedure ensures that the normalized release fractions presented represent a maximum upper limit of the isotopic releases experienced by the UO_2 fuel in each test. The normalization was applied uniformly to each release fraction in Test PCM-1, RIA-ST-1, RIA-ST-2, and RIA-ST-4 to generate relative magnitudes for isotopic release fractions. Although this technique complicates direct comparison of absolute release fractions between different tests, it permits comparison of various ratios of relative release fractions from one test to another. The numbers shown for Test RIA 1-1 are not normalized; these data represent absolute release fractions.

Table I summarizes the important fuel behavior parameters from each test. The fuel fragmentation mentioned in the table is illustrated in Figure 1, which shows a cross section of the fuel from RIA-ST-1. Grain separation is apparent across the entire radius of the fuel from near the centerline to the periphery of the pellet.

TABLE I. A COMPARISON OF FUEL BEHAVIOR PARAMETERS DURING THE PBF TESTS

	PCM-1	RIA-ST-1	RIA-ST-2	RIA-ST-4	RIA 1-1[b]
Transient condition[a]	900 s in film boiling	250 cal/g UO_2 burst	260 cal/g UO_2 burst	350 cal/g UO_2 burst	285 cal/g burst
Fuel melted (%)	25	0.	0.	>90	<1.
Fuel lost to loop (%)	24	10.	15.	<1	8.5
Maximum fuel temperature (K)	3100	3000.	3000.	3500	3100.
Fragmentation (% of rod)	38	16.7	27.8	>90	48.

a. RIA burst values are radial average fuel enthalpy at the axial flux peak.

b. Values for RIA 1-1 are the mean for the four test rods.

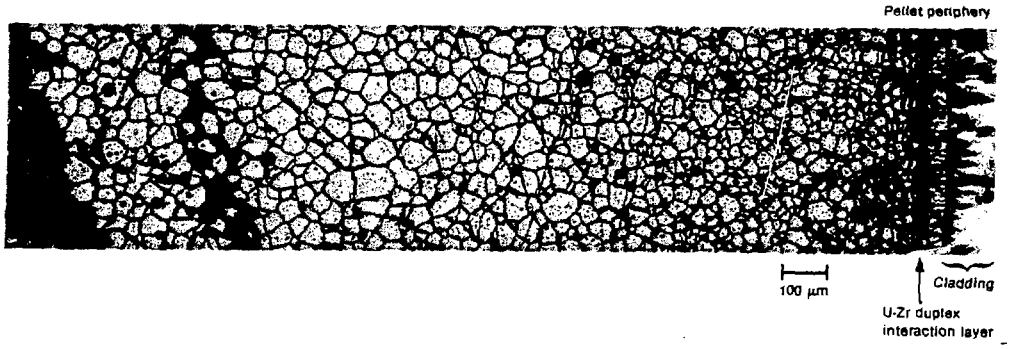


Figure 1. Fuel fragmentation produced during Test RIA-ST-1.

Table II summarizes the measured fission product release fractions for each test. Fission product behavior during PBF tests is generally characterized by:. large noble gas release fractions; medium to high rubidium release fractions; low to medium iodine release fractions; and widely varying cesium, barium, and lanthanum release fractions. Whereas a noble gas isotope demonstrated the largest release during Tests PCM-1, RIA-ST-2, and RIA 1-1, ^{142}La exhibited the largest release during RIA-ST-1 and RIA-ST-4. It should be realized that for realistic analysis of an accident scenario, the entire time-dependent release fraction should be considered. Only equilibrium release fractions are summarized in Table II.

Time-dependent release fractions of a few selected isotopes from the RIA 1-1 and RIA-ST-4 signatures are shown in Figures 2 and 3, respectively. These figures illustrate that the release fraction histories vary for different fission product isotopes and fuel rod conditions. The histories of all the isotopic release fractions measured during the PBF tests, with the exception of RIA 1-1, are given in Reference 13.

TABLE II. FISSION PRODUCT RELEASE FRACTIONS TO THE PBF TEST LOOP

	PCM-1		RIA-ST-1		RIA-ST-2		RIA-ST-4		RIA 1-1	
Isotope	RRF	Uncertainty	RRF	Uncertainty	RRF	Uncertainty	RRF	Uncertainty	RF	Uncertainty
^{85m}Kr	1.00	0.07	--[a]		--[a]		--[a]		1.05[e]	0.20
^{87}Kr	0.92	0.08	0.55	0.15	0.44	0.06	0.26[b]	0.03	0.28	0.05
^{88}Kr	0.93	0.07	0.49	0.06	0.46	0.08	0.62[b]	0.08	0.30	0.06
^{88}Rb	0.90	0.07	0.64	0.09	0.63	0.12	0.81[b]	0.10	0.33	0.06
^{89}Rb	0.020	0.001	0.56	0.11	0.67	0.10	0.18	0.02	0.34	0.06
^{131}I	--[a]		0.23	0.03	--[c]		0.048	0.009	0.45	0.08
^{132}I	0.012	0.001	0.23	0.02	--[c]		0.053	0.007	0.43	0.07
^{133}I	0.017	0.001	0.21	0.02	0.34	0.08	0.042	0.006	0.39	0.08
^{134}I	0.015	0.001	0.32	0.04	0.36	0.06	0.064	0.008	0.24	0.05
^{135}I	0.024	0.007	0.62	0.09	--[c]		0.056	0.007	0.27	0.05
^{135}Xe	0.86	0.05	0.15	0.02	0.30	0.06	0.82[b]	0.10	0.29	0.05
^{138}Xe	0.86	0.07	0.75	0.11	1.00	0.14	0.85	0.11	0.40	0.07
^{137}Cs	--[a]		--[a]		--[a]		--[a]		0.65[f]	0.14
^{138}Cs	0.15	0.04	0.43	0.04	0.55	0.08	0.45	0.06	0.27	0.05
^{139}Cs	0.020	0.004	0.75	0.15	1.3[d]	0.3	0.19	0.02	0.32	0.06
^{139}Ba	0.0075	0.0013	0.62	0.40	0.87	0.16	0.52	0.07	1.77[d]	0.33
^{140}Ba	--[a]		0.13	0.02	--[c]		0.021	0.003	0.15	0.03
^{141}Ba	0.0024[e]	0.0003	0.32	0.04	0.67	0.08	0.40	0.04	0.52	0.10
^{142}Ba	--[a]		0.31	0.06	0.66	0.10	0.47	0.06	0.15	0.04
^{140}La	--[a]		0.13	0.02	--[c]		0.021	0.002	0.022	0.004
^{142}La	0.0019[e]	0.0004	1.00	0.13	0.62	0.08	1.00	0.12	0.20	0.04
^{143}Ce	--[a]		--[a]		--[c]		0.012	0.003	0.0068	0.0016

a. Coolant concentration levels for these nuclides were below detectable levels.

b. During the time interval in which a normalized release fraction was determined, the values of the data points were generally monotonically increasing.

c. Background coolant concentration levels for these nuclides released during RIA-ST-1, seven days earlier, overwhelmed the magnitude of nuclide releases occurring during RIA-ST-2.

d. These release fractions appear to represent a situation in which mixing was not complete.

In RIA 1-1, the ^{137}Cs, ^{138}Cs, and ^{131}I isotopes behaved similarly, with large early releases, and steady or slightly decreasing release fractions in the five hours following the burst. The ^{140}Ba and ^{140}La isotopes were released over a longer period of time, as is illustrated by the increasing release fractions during the five-hour postburst period. In RIA-ST-4, the ^{131}I, ^{135}Xe, and ^{138}Cs release fractions each had a similar release pattern early after fuel failure, but reached different equilibrium values at different times. The ^{141}Ba release was not measured until approximately 2 min after the first indication of rod failure, and its release fraction stabilized early. The ^{142}La isotope was not measured until approximately 5 min after

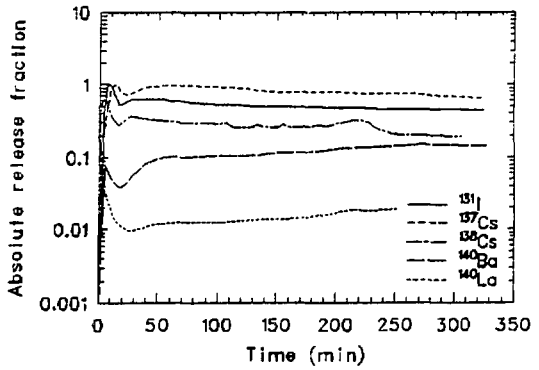


Figure 2. Fission product release signature from Test RIA-1-1.

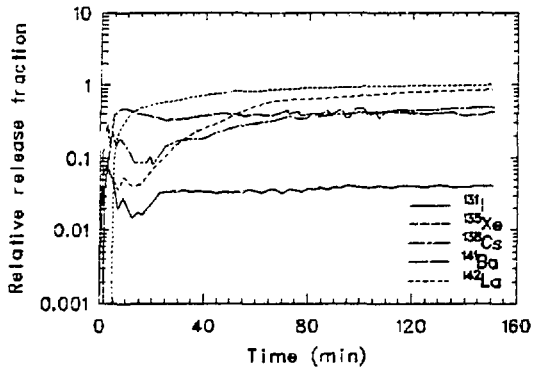


Figure 3. Fission product release signature from Test RIA-ST-4.

the first isotopes appeared, and it behaved with a monotonically increasing release fraction for more than 2 h, finally achieving the largest release fraction in this test.

It seems surprising that a rare earth isotope would be completely released, because lanthanum and its oxide are both nonvolatile fission products having boiling points in excess of 3700 K. However, the explanation lies in the physical behavior of the 142 decay chain. Since ^{142}La has a minute independent fission yield, very little ^{142}La was present at the time of the RIA power burst and coincident rod failure. Because the ^{142}Xe, ^{142}Cs, and ^{142}Ba precursors of ^{142}La have high fission yields, they are present in large quantities. These fission products have boiling temperatures below the average fuel temperature achieved during the tests. The release of these more volatile precursor isotopes in large quantities provided the source of the large ^{142}La release fraction measured at varying later times. The nearly instantaneous generation of fission products by the power burst is coincident with the period of most extreme fuel temperature. The high-temperature transient spans a period of 25 s and, when coupled to the severe fragmentation that occurred during these tests, readily explains the large release fractions measured.

Fuel melting appeared to reduce the iodine release fraction measured in the test loop coolant. As is illustrated in Figure 4, the iodine release fractions were very small compared to the noble gas release fractions in the two tests that produced high fuel temperatures and large percentages of fuel melting (Tests PCM-1 and RIA-ST-4). The other three tests (RIA-ST-1, RIA-ST-2, and RIA 1-1), which produced extensive fuel fracturing, but little or no evidence of fuel melting, showed larger fractions of iodine release relative to the noble gas releases. The iodine may be vaporized and driven out of the fuel at these high temperatures (3100 to 3500 K), but the results suggest that the iodine quickly deposits on or reacts with the fuel rod or test train material, binding the isotope and preventing transport in the coolant. During the lower temperature tests (<3100 K), the iodine may be sufficiently volatilized to be driven out of the fuel, but the cladding or coolant conditions may be substantially different, preventing early deposition. For instance, the iodine may be present on the grain boundaries of the fuel that fractures during the course of the transient, thus exposing the iodine to the coolant.

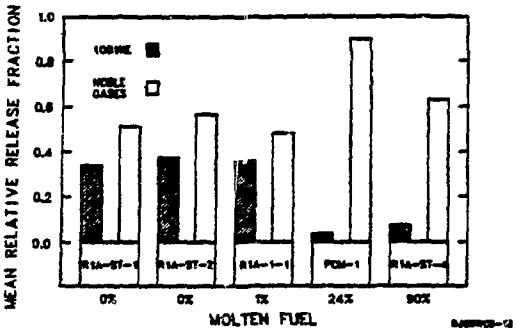


Figure 4. A comparison of the iodine-noble gas release fractions during tests with and without significant fuel melting.

A significant loss coefficient for iodine was demonstrated during the RIA-ST-1 experiment. The rate at which any isotope is removed from the circulating coolant can be estimated from the slope of the release fraction at various times after equilibrium has been established. With only a few exceptions, the fission products display very stable release fractions. As is illustrated in Figure 5, the relative iodine release fractions during RIA-ST-1 reached an equilibrium release fraction of about 25% for the period up to 300 min following rod failure. After 300 min, each of the three iodine isotopes began a dramatic decline to a new level near 3%, coincident with a reduction in the coolant temperature. The iodine was apparently being selectively removed from the circulating coolant, while the other fission products remained unaffected. This behavior illustrates the importance of fission product transport to source-term identification. The estimated release fraction could change by an order of magnitude for accidents with different primary coolant conditions.

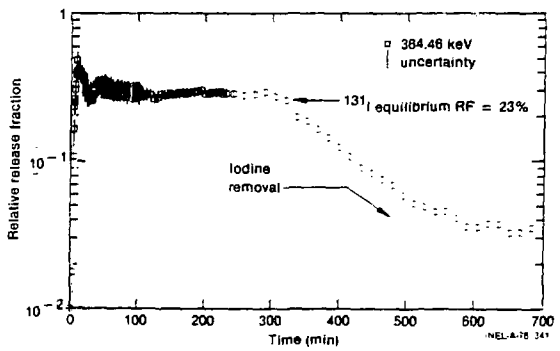


Figure 5. Release fraction history of ^{131}I illustrating rapid removal specific to iodine.

COMPARISON OF RELEASE FRACTIONS

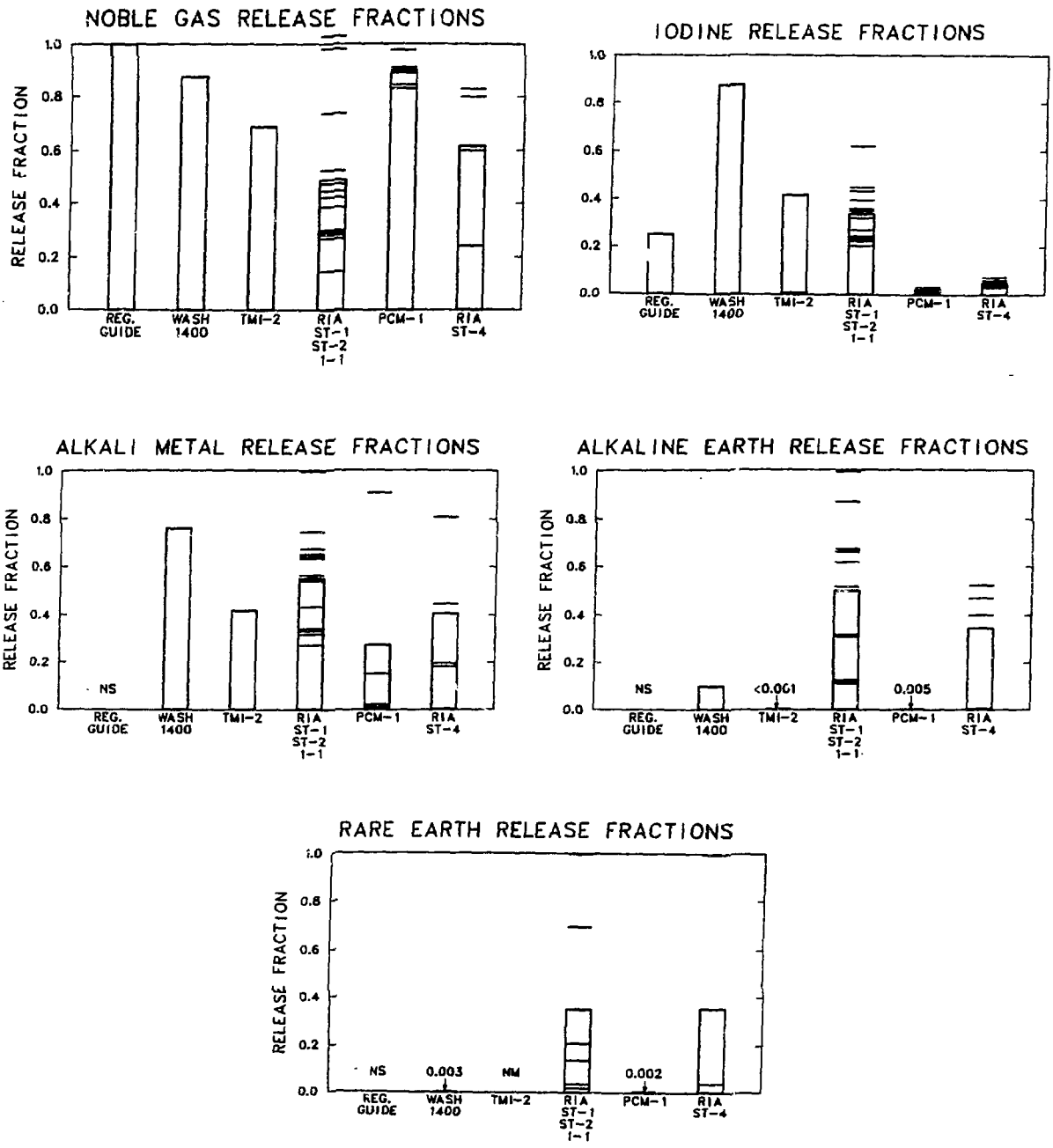
Figure 6 presents: a comparison of the release fractions suggested in the NRC Regulatory Guides for design basis accidents;[4-6] meltdown release fractions from the Reactor Safety Study;[7] release fractions measured following the TMI-2 accident;[8,9] and release fractions measured during the PBF tests. The release fractions from the various sources are compared directly. However, it should be noted that: the PBF data refer to measurements made in the coolant; the Reactor Safety Study and Regulatory Guide data refer to gaseous release in the reactor containment building; and the TMI-2 data include both gaseous and liquid releases.

There is general agreement among the noble gas release fractions; however, the halogen release fractions are substantially lower in the PBF tests that included fuel melting. The Regulatory Guides do not specify release assumptions for alkali metal, alkaline earth, or rare earth isotopes; but, significant fractions of alkali metal fission products were measured in the coolant at TMI-2 following the accident, and at PBF following fuel damage tests.

The measured release fractions for several alkaline and rare earth isotopes in the RIA tests were approximately two orders of magnitude higher than expected. This is due to the unique circumstances developed during burst-type fuel rod tests. The short periods of irradiation, with associated short temperature transients, create fission product behavior phenomena that also occur over short periods. Many of the fission product isotopes have short-lived, but volatile, parents and grandparents. The complex chemical and physical changes of the short-lived fission products play an important role in the resultant behavior. Although the release fractions of the alkaline and rare earth isotopes are large, the quantity released is small in comparison to the total inventory.

Fuel in the PBF tests did not experience total meltdown; however, most of the release fractions observed are of very nearly the same magnitude as the design basis accident and Reactor Safety Study meltdown release fractions. The principal difference is that the PBF releases were measured in the coolant, while the Regulatory Guides and Reactor Safety Study suggest gaseous release to the containment building. The large releases measured in the PBF tests may be the result of extensive fracturing and powdering of the fuel.

In addition to the fission product release associated with this grain boundary separation, fission product leaching is enhanced by the large fuel surface area exposed to the coolant. Further, fuel particles with retained fission products may themselves be released to the coolant and be available for transport. A large fraction of the measured concentrations of fission products in the PBF loop coolant may have been associated with these particles. Accident analyses generally ignore this phenomenon and assume that the worst-case behavior involves loss of coolant, fuel heatup, and melting. However, as was demonstrated in the TMI accident and in each of the PBF tests, reactor accidents can involve extensive fuel fragmentation caused by coolant injection and quenching of the hot ceramic fuel. This phenomenon enhances fission product release and produces fuel particulate source terms that may account for transport of a large fraction of fission product activity. Greater attention needs to be given to such fuel fracturing and to particulate source terms in accident analyses.



NOTE: Bars for PBF data indicate the mean value for all isotopes; lines indicate values for individual isotopes. NS = not specified; NL = not measured.

Figure 6. Comparisons of release fractions for different fission product chemical groups.

CONCLUSIONS

Fission product release from failed fuel has been experimentally measured during five PBF tests. Fission product concentrations in the PBF coolant loop were determined for a maximum of 22 short-lived fission products per test. Release fractions were calculated by comparing integrated coolant activity with fuel rod fission product inventories calculated with the ORIGEN computer code. For four of the five tests, isotopic relative release fractions were normalized to provide upper limits for absolute release fractions.

From the analysis of fission product behavior during the PBF tests, and the comparison with release fraction values suggested in the literature and reported for the TMI accident, it is concluded that:

1. Large fractions of fission products can be released from nuclear fuel during accidents that produce fuel rod fragmentation, even though significant fuel melting does not occur

2. Iodine and noble gas releases are of a similar magnitude during accidents that produce no fuel melting, and iodine appears to react or deposit during accidents that produce fuel melting, thus preventing its transport in the coolant

3. Alkaline and rare earth isotopes, not expected to be volatile during most accidents, can be released because of the volatile behavior of parent isotopes and fuel powdering that can occur during accidents.

The isotopic release fraction histories generated from this study demonstrate the importance of time-dependent release, precursor influence, and removal processes for evaluating isotopic source terms. Fission product release associated with fuel fracturing or powdering during quench from high temperatures should be considered in accident analyses. The release fractions from the core to the circulating primary coolant may be much larger during certain accidents than conventional analysis methods would predict.

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