CONF-841105--57

BEHAVIOR OF Cs, I, AND Te IN THE FISSION PRODUCT RELEASE PROGRAM AT ORNL*

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TI85 005369

To be presented at the American Nuclear Society 1984 International Conference, November 11–16, 1984, Washington, D.C.

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*Research sponsored by the Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission under Interagency Agreement DOE 40-551-75 with the U.S. Department of Energy under contract DC-AC05-84OR21400 with the Martin Marietta Energy Systems, Inc.

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SUMMARY

Experiments have been conducted at ORNL with highly irradiated light-water reactor (PWR and BWR) fuel rod segments to investigate fission product release in steam in the temperature range 500 to 2000°C.[1-3] The primary objectives were to quantify and characterize the releases under conditions postulated for loss-of-coolant accident (LOCA) and severe accident conditions. In all, 26 experiments have been conducted — 24 with high burnup and 2 with low burnup fuels. To aid in the interpretation of fission product release, 12 implant and 18 control experiments were also conducted; the behavior of HI, I_2, Cs_2O, CsOH, Te, and TeO_2 (individually and in different combinations) was studied. In this paper only the observed behavior of cesium, iodine, and tellurium is discussed.

The experimental apparatus consisted of a furnace assembly where the fuel rod segments were heated and a fission product collection system. During a test, the volatile fission products were released from fuel rod segment into a flowing steam—helium atmosphere. The fission products released from the furnace were transported to the collection system which included a platinum thermal gradient tube (TGT) where the volatile species deposited, and a filter pack which collected particulates and the more volatile and penetrating species such as I_2, HI, and CH_3I. The analytical methods used in these studies were gamma spectrometry, neutron activation analysis, spark source mass spectrometry (SSMS), x-ray diffractometry, and energy dispersive x-ray analysis (EDX).

The relative data for 22 tests of irradiated fuel are summarized in Table 1.[4] The percentage values were estimated by assuming the iodine which deposited on surfaces at temperatures above 200°C was CsI, the iodine that collected on the charcoal was I_2 (also HI and CH_3I), and the fraction on the first filter was particulate. These data show that the percentage molecular iodine was high only in tests conducted in steam where the releases of iodine were very low, <71 μg. In fact, in tests where the percentage of molecular iodine was >11%, the mass of iodine released was <17 μg, an amount that could be lost in the percent error in tests where the releases were high (>500 μg iodine). When total iodine release for all the tests is considered, only 0.3% of the iodine was apparently collected as molecular iodine. Figure 1 gives example TGT deposition profiles for cesium and iodine. These results, when considered with TGT fission product deposition behavior[5] and SSMS data, indicate that the released iodine behaved like cesium iodide.

In the gas purge tests, BWR-4 and HUB-12 (see Table 1) the available cesium and iodine (not tied up in the fuel matrix) was purged by purified helium while the fuel segments were heated in steps up to 1200°C. The results showed that cesium and iodine were collected as cesium iodide and cesium oxide (probably Cs_2O).

In the tests where the test atmosphere was steam–helium, cesium (excluding CsI) behaved like cesium hydroxide. The released cesium reacted energetically with several apparatus materials; with quartz at temperatures <1000°C, with zirconia in the temperature range 800 to 1200°C, and with 304-stainless steel at temperatures between 600 and 900°C (not determined above 900°C).

An effort to determine and characterize tellurium release (by SSMS) has been made in the recently conducted HI series of tests. The only significant release (estimated to be ~50%) was obtained in test HI-2.[2] In that test, unlike the others, the Zircaloy cladding was totally oxidized by the steam. This suggests that steam oxidation of the Zircaloy cladding played an important role in tellurium release. If so, the likely mechanism was that the tellurium was retained by the unoxidized cladding as less volatile $ZrTe_2$. As the cladding became more completely oxidized, the tellurium was released because zirconium oxide is thermodynamically more stable than $ZrTe_2$. The collection behavior for tellurium in test HI-2 suggested that it deposited mainly as telluride.

In conclusion the results showed that cesium and iodine were released primarily as CsOH and CsI, and that tellurium release was controlled by steam oxidation of the Zircaloy cladding. The released form of tellurium is not certain at this time.

References

1. R. A. Lorenz, J. L. Collins, M. F. Osborne, R. L. Towns, and A. P. Malinauskas, Fission Product Release from BWR Fuel Under LOCA Conditions, NUREG/CR-1773 (ORNL/NUREG/TM-388), July 1981.

2. M. F. Osborne, R. A. Lorenz, J. R. Travis, C. S. Webster, and K. S. Norwood, Data Summary Report for Fission Product Release Test HI-2, NUREG/CR-3171 (ORNL/TM-8667), February 1984.

3. M. F. Osborne, J. L. Collins, R. A. Lorenz, K. S. Norwood, J. R. Travis, and C. S. Webster, Data Summary Report for Fission Product Release Test HI-4, NUREG/CR-3600 (ORNL/TM-9011), June 1984.

4. Technical Bases for Estimating Fission Product Behavior During LWR Accidents, NUREG-0772, U.S. Nuclear Regulatory Commisision, pp. 4.10-4.11, June 1981.

5. K. S. Norwood, J. L. Collins, M. F. Osborne, and R. A. Lorenz, "Characterization and Chemistry of Fission Products Released from LWR Fuel Under Accident Conditions," presented at the ANS Topical Meeting on Fission Product and Source Term Research held at Snowbird, Utah, June 1984.

Table 1. Summary of iodine release species in fission product release tests

Test No.[a]	Temperature (°C)	Gaseous environment[b]	Test duration (min)	Amount of I released (μg)	% of each form		
					Presumably CsI	Presumably mainly I_2	Particulate
BWR-4[c]	700–1100	Helium	300	4800	99.99	0.004	0.01
HBU-12[c]	700–1200	Helium	480	170	91.2	0.27	8.6
HBU-1	700	Steam	300	0.9	18	72	10
HBU-2	900	Steam	120	1.8	14	73	13
HBU-4	500	Steam	1200	0.1	40	44	16
HBU-11	1200	Steam	27	20	34	8	58
HBU-7[d]	900	Steam	1	11	71	4	25
HBU-8[d]	900	Steam	61	14	4	88	8
HBU-9[d]	1000	Steam	10	17	6	88	6
HBU-10[d]	1050	Steam	11	14	26	53	20
BWR-1[d]	960	Steam	1	490	67	0.4	33
BWR-2[d]	850	Steam	1	1000	79	0.1	21
BWR-3	1200	Steam	25	1200	44	0.7	56
HT-1[e]	1300	Steam	10	71	70	10	20
HT-2[e]	1445	Steam	7	990	90	0.1	9.9
HT-3[e]	1610	Steam	3	5400	86	0.2	14
HT-4[e]	1440	Steam	0.4	750	78	0.3	22
HI-1	1400	Steam	30	514	41.7	0.5	58.8
HI-2	1700	Steam	20	13554	31.85	0.4	67.8
HI-3	2000	Steam	20	7950	36.7	0.4	62.9
HI-4	1850	Steam	20	4929	38.3	0.006	61.7
HI-5	1700	Steam	20	5803	33.5	0.07	66.4

[a]See refs. 1–3 for details.
[b]The steam environments were actually steam–(helium or argon)–hydrogen; hydrogen resulted from the Zircaloy–steam reaction.
[c]Gap purge tests.
[d]Burst release tests.
[e]Cladding was expanded before the test to provide an enlarged uniform fuel-cladding gap.

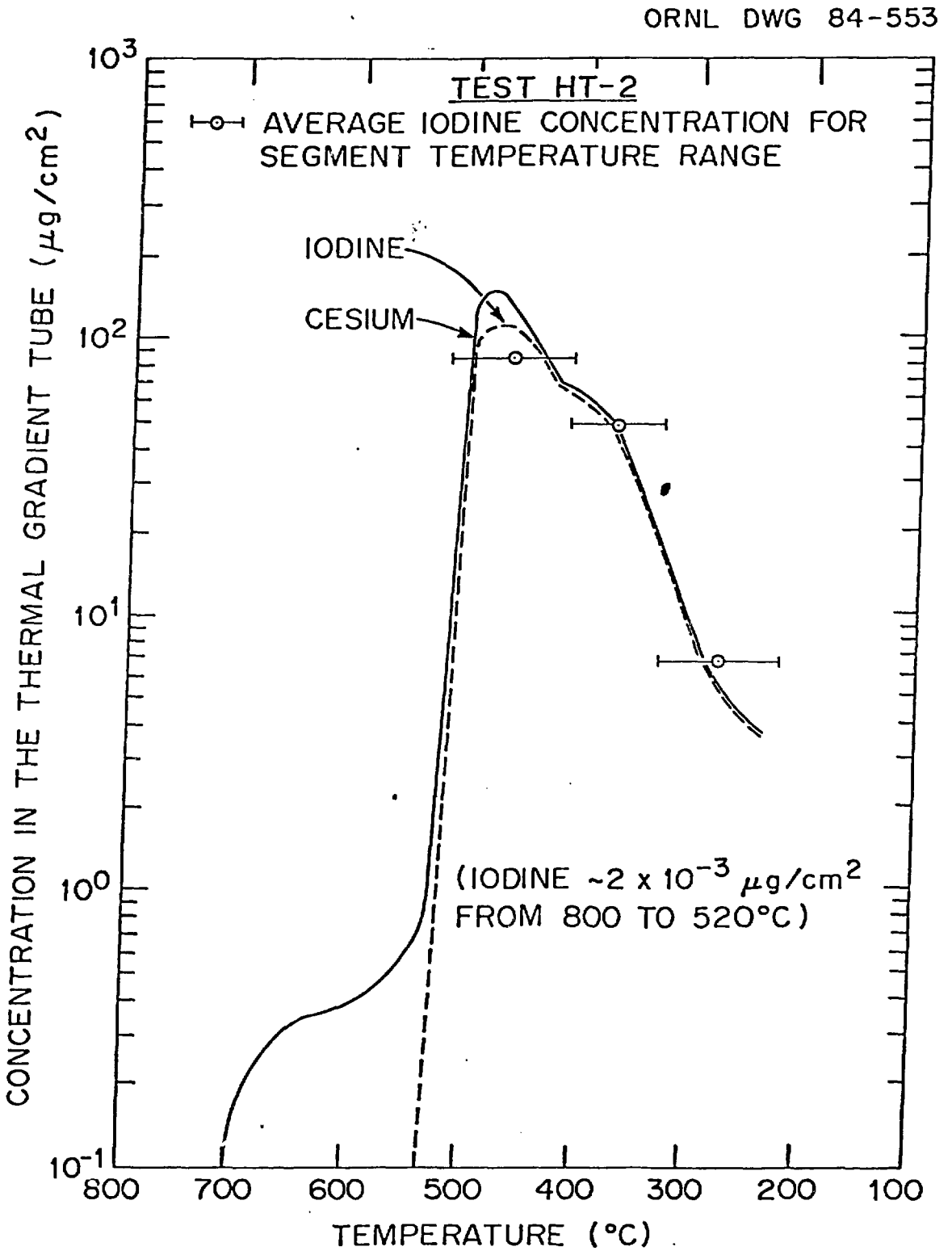

Figure 1. Deposition profiles for iodine and cesium in the TGT in test HT-2.