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RELEASE OF FISSION PRODUCTS DURING CONTROLLED LOSS-OF-COOLANT ACCIDENTS AND HYPOTHETICAL CORE MELTDOWN ACCIDENTS*

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DISCLAIMER

Introduction

A few years ago the Projekt Nukleare Sicherheit joined the United States Nuclear Regulatory Commission in the development of a research program which was designed to investigate fission product release from light water reactor fuel under conditions ranging from spent fuel shipping cask accidents to core meltdown accidents. Three laboratories have been involved in this cooperative effort. At Argonne National Laboratory (ANL), the research effort has focused on noble gas fission product release, whereas at Oak Ridge National Laboratory (ORNL) and at Kernforschungszentrum Karlsruhe (KfK), the studies have emphasized the release of species other than the noble gases. In addition, the ORNL program has been directed toward the development of fission product source terms applicable to analyses of spent fuel shipping cask accidents and controlled loss-of-coolant accidents, and the KfK program has been aimed at providing similar source terms which are characteristic of core meltdown accidents.

In this report we describe the ORNL results for fission product release from defected fuel rods into a steam atmosphere over the temperature range 500-1200°C, and the KfK results for release during core meltdown sequences.

The ORNL Program

The ORNL Program is designed to proceed through six phases, each comprised of a characteristic series of experiments. The first of these, the Control Test Series, was performed primarily to test the operation of the apparatus and the analysis techniques and to investigate the behavior of selected chemical forms of iodine and cesium under various test conditions. The results of these tests have been reported elsewhere.[1]

The second series of experiments, the Implant Test Series, involved tests with Zircaloy-4 clad fuel rod segments which contained unirradiated UO_2 pellets coated with CsOH, CsI, and TeO_2. These experiments, which are described in greater detail elsehwere,[2] are summarized in a later section of this report.

The third set of experiments, the Low Burnup Fuel Test Series,[3] consisted of two experiments which were performed with fuel capsules that had been irradiated to 1000 MWd/MT at high heat rating (560-660 W/cm). These experiments were followed by the High Burnup Fuel Test Series;[4-10] this test series utilized fuel irradiated to 30,000 MWd/MT at low heat rating (175-320 W/cm) in the H. B. Robinson pressurized water reactor.

The four sets of experiments described above have been completed. The fifth phase has only recently been started; this phase, which is designated as the High Temperature Test Series, involves an extension of the tests conducted in the high burnup test series from maximum test temperatures of 1200°C to 1600°C. The concluding series of experiments, the BWR Fuel Test Series, will involve tests with full burnup fuel rods obtained from the Peach Bottom-2 boiling water reactor; these tests will be conducted over the temperature range 700-1200°C.

Experimental Apparatus for the ORNL Program

All six series of experiments which form the ORNL program involve the apparatus displayed in Fig. 1. The tests reported here were conducted in flowing 80% steam-20% helium or 80% steam-20% argon gas mixtures at atmospheric pressure. Heating of the test component was provided by either a resistance-heated furnace (as is shown in Fig. 1), or by an induction coil.

Two types of experiments were conducted. One type involved rupturing the cladding of the test piece by pressurizing the rod internally with inert gas (helium or argon) at a preselected temperature, whereas in the second type of experiment the cladding was defected prior to the test by carefully drilling a 0.159 cm hole through the cladding at the midsection of the test component.

Material released from the test fuel rods which does not deposit on the quartz furnace tube components is transported by the flowing gas mixture through a gold foil-lined thermal gradient tube. Volatilized chemical species which condense at temperatures above 150°C deposit in this region of the apparatus. Particulate material is collected on a filter pack which is comprised of three HEPA borosilicate fiberglass filters and which is located at the terminus of the thermal gradient tube. Volatile forms of iodine, such as I_2, HI, and CH_3I, are collected on impregnated charcoal and silver-exchanged zeolite sorbers, whereas the noble gas fission products are collected from the flowing gas stream using charcoal traps which are maintained at 195K. Prior to entering these traps, the gas stream is dried using an ice-bath condenser and a freeze trap.

A NaI(Tl) crystal which is shielded from the fuel rod source is employed to monitor ^{134}Cs and ^{137}Cs activity in the thermal gradient tube and filter pack.

Implant Test Series

The implant tests were conducted using specially fabricated test components. In these tests, gap inventories of the volatile fission products cesium, iodine, and tellurium were simulated by coating unirradiated UO_2 pellets with suitably radiotracer-tagged amounts of CsOH, CsI, and TeO_2. By this means it was possible to precisely define the gap inventories of these fission product species in

the test component. Similarly, mass balance determinations could be made to demonstrate the ability of the collection/characterization system to contain and account for the volatilized species completely. A detailed description of this series of experiments, as well as an extensive presentation of the test data, has been given elsewhere;[2] a summary of the results for cesium and iodine release is presented in Table 1. Material balances for all of the implant tests were within 99.8 $\pm$ 5.6% of total inventory for iodine, and 89.1 $\pm$ 9.7% of total inventory for cesium. The lower value for cesium probably resulted from the method employed to estimate initial cesium inventories and a systematic error due to counting geometry.[2]

Low Burnup Fuel Test Series

Twelve UO_2 fuel rods were irradiated in the General Electric Test Reactor (GETR) over the period September-November 1967 as part of the Prompt Fission Product Release Program at ORNL. Two of these rods were retrieved from storage for use in the Low Burnup Fuel Test Series. In both of the tests the cladding was defected prior to testing by drilling a 0.159 cm hole through the cladding. The results of these tests, which are presented in greater detail elsewhere,[3] are summarized in Table 2.

High Burnup Fuel Test Series

The High Burnup Fuel Test Series was conducted using rod segments which were machined from two fuel rods of bundle B05 of the H. B. Robinson pressurized water reactor. The rods were irradiated over the period October 1971-May 1974 and attained a calculated average burnup of 28,000 MWd/MT and a calculated peak burnup of 31,400 MWd/MT. Detailed characteristics of both the rods and the rod

segments have been published previously.[4,12] Similarly, the experimental results which are summarized in Table 3 are discussed in consdierably greater detail in previous reports of this work.[4-10]

ORNL Models for Cesium and Iodine Release

It became evident during the course of the experimental work that two mechanisms control the release of cesium and iodine nuclides from defected fuel rods. The first of these, designated the "burst release" component, is due to convection of the fill and fission product gases as these are vented from the rod at the time of rupture. This type of release is sudden; it persists only for the minute or so that the gases in the interconnected void volume are vented through the rupture in the cladding.

The second component, termed "diffusional release," is due to gas-phase transport of the fission product species along the interconnected voids to the point of rupture as a result of a gradient in concentration. This time-dependent component is unimportant relative to burst release at low temperatures, but becomes significant at about 1200°C even for short time periods.

The experimental data presented in Tables 1-3 could be correlated by assuming that the two mechanisms are independent, and are therefore additive. In mathematical form, the total mass of a given fission product species that is released upon cladding rupture of a fuel rod is assumed to be given by

$$M = M_B + M_D, \tag{1}$$

where M is the total mass of species released, M_B is the mass released at the time of rupture, and M_D represents the mass of material which escapes due to diffusional release.

For cesium, the burst release component over the temperature range 700-900°C can be described by the mathematical relation[13]

$$M_B = [V_B G^{0.8}/7.74]\exp[6.06 - (7420/T)]; \quad (2)$$

in which M_B is expressed in μg Cs released, V_B denotes the volume of plenum and other interconnected void gas vented, in cm^3 at 273K and system pressure, G is the inventory of cesium in the pellet-cladding gap regions measured per unit of cladding (in units of $\mu g/cm^2$), and T is the temperature, in K, at the rupture location.

The corresponding expression for the iodine burst release component over the temperature range 700-900°C is[13]

$$M_B = [V_B G^{0.8}/1.15]\exp[1.09 - (3770/T)]. \quad (3)$$

For diffusional release M_D in μg, the mathematical expression is[13]

$$M_D = M_o\,[1 - \exp(-R_o t/M_o)], \quad (4)$$

in which M_o represents the total calculated mass of the diffusing species initially in the gap, in μg, t is the time in hr over which diffusion is allowed to proceed, and R_o denotes the initial rate of diffusional release in μg/hr.

The initial rate of release of cesium over the temperature range 500-1200°C could be described by the relation[13]

$$R_o = (W/200)(G^{0.8}/7.74)(0.101/P)\exp[19.96 - (19{,}810/T)], \quad (5)$$

where W represents the width of the radial pellet-cladding gap, in μm, and P is the system pressure in MPa.

The corresponding equation for iodine is[13]

$$R_o = (W/200)(G^{0.8}/1.15)(0.101/P)\exp[15.30 - (14{,}800/T)]. \quad (6)$$

The extent to which these semi-empirical expressions correlate the experimental results is shown graphically in Fig. 2; the comparison between the release models and experiment is excellent over the entire range of mass released, which spans almost seven orders of magnitude. (The dashed lines in the figure denote factor of three error limits.)

The KfK Program

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Concluding Remarks

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Table 1. Release of cesium and iodine from defected fuel rods in steam: implant test series

Test	Type	Temp. (°C)	Time (hr)	Initial gap inventory/cm^2 cladding		Mass released	
				(μg Cs/cm^2)	(μg I/cm^2)	(μg Cs)	(μg I)
I-1	Burst	700	1.0	23.1	22.0	180.	363.
I-2	Burst	700	1.5	0.47	0.45	2.48	5.31
I-3	Burst	900	2.0	87.	8.7	486.	161.
I-4	Drilled	1100	0.8	68.	6.1	1240.	145.
I-5	Burst	700	2.0	97.	8.4	341.	109.
I-6	Drilled	500	20.0	141.	12.4	≤43.	5.8
I-8	Burst	1100[a]	0.96	127.	8.4	3310.	328.
I-10	Drilled	700	5.0	97.	9.1	5.0	12.
I-11	Drilled	1300	0.25	114.	8.0	2320.	26.
I-12	Burst	900	2.0	14.6	0.69	264.	30.2

[a]Ruptured at 900°C, then rapidly brought to 1100°C.

Table 2. Release of cesium and iodine from defected fuel rods in steam: Low burnup fuel test series

Test[a]	Temp. (°C)	Time (hr)	Initial gap inventory/cm^2 cladding[b]		Mass released	
			(μg Cs/cm^2	(μg I/cm^2)	(μg Cs)	(μg I)
LBU-1	700	5.0	69	1.47	0.046	0.11
LBU-2	900	2.0	107	2.44	19.4	20.0

[a]Both tests involved rods which were defected prior to experimentation.

[b]Estimated from analysis of noble gas fission product gap inventories.

Table 3. Release of cesium and iodine from defected fuel rods in steam: high burnup fuel test series

Test	Type	Temp. (°C)	Time (hr)	Initial gap inventory/cm^2 cladding[a]		Mass released	
				(μg Cs/cm^2)	(μg I/cm^2)	(μg Cs)	(μg I)
HBU-1	Drilled	700	5.0	13.1	1.20	0.123	0.93
HBU-2	Drilled	900	2.0	12.7	1.17	2.82	1.76
HBU-4	Drilled	500	20.0	13.1	1.20	0.017	0.105
HBU-7	Burst	900	0.02	12.7	1.17	130.	11.1
HBU-8	Burst	900	1.0	12.7	1.17	39.6	13.5
HBU-9	Burst[b]	1100	0.14	13.1	1.20	112.	16.9
HBU-10	Burst[b]	1200	0.17	11.2	1.03	279.	13.9
HBU-11	c	1200	0.17	11.3	1.05	142.	20.2

[a]Estimated from analysis of noble gas fission product gap inventories.
[b]Ruptured at 900°C, then rapidly brought to test temperature noted in table.
[c]Fuel rod previously used in test HBU-7.