RELEASE AND DEPOSITION OF FISSION PRODUCTS FROM SHEATHED CANDU® FUEL SAMPLES IN HOT-CELL EXPERIMENT 5 TGT TESTS

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ABSTRACT

Three tests were carried out to investigate deposition and retention of fission products, released at high temperatures in steam and inert atmospheres, on the furnace tube and on two deposition sections made of CANDU-reactor-relevant structural materials. Trace-reirradiated segments of a fuel element discharged from Gentilly-2 were heated to peak temperatures of up to 2055 K in flowing carrier gas environment. The elements Te, I and Cs showed some preferential deposition from steam or inert gas containing a low concentration of oxygen on Zr-2.5Nb deposition sections compared to carbon steel or Inconel 600 deposition sections. In an inert-environment test, the released fission products were not deposited preferentially on either carbon steel or stainless steel.

1. INTRODUCTION

Three tests designated TGT1[1], TGT2 and TGT4[2] in Hot-Cell Experiment 5 (HCE5) were carried out at AECL's Chalk River Laboratories in 1999 March under the sponsorship of the CANDU Owners Group (COG). The primary objective of these tests was to measure deposition and retention of fission products (FP) released at high-temperatures from irradiated CANDU-type fuel on CANDU-reactor-relevant structural materials in steam and inert atmospheres. Another objective was to investigate release of FP from CANDU fuel samples in these conditions. The data obtained from the TGT1, TGT2 and TGT4 tests will be useful for validation of FP release and transport computer codes such as SOURCE IST 2.0 [1] and SOPHAEROS-IST 2.0 [2].

2. EXPERIMENTAL APPARATUS AND MEASUREMENT TECHNIQUES

Trace-reirradiated segments of a CANDU-6 fuel element were heated to peak temperatures up to 2055 K in flowing carrier gas environment in a 15 kW vertical tube furnace apparatus [3]. The furnace had zirconia main heating elements, molybdenum disilicide preheating elements and a

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[1] TGT = Thermal Gradient Tube

[2] The planned test TGT3 was not performed because the furnace tube assembly could not pass the leak test.

single-piece vertical yttria-stabilized zirconia guide tube. Figure 1 shows the furnace loaded with a TGT furnace tube assembly. The assembly consisted of a single-piece yttria-stabilized zirconia furnace tube, and a lower O-ring sealed fitting that provided a gas inlet and supported an yttria sample boat on a zirconia support post. The distance from the sample position to the top of the furnace tube was 476 mm. The furnace tube assembly was inserted through the top of the furnace, and was suspended from the top of the furnace casing. A graphite sleeve fitted over the top of the guide tube guided a flow of inert gas (Ar/2%H_2) downward, through the annulus between the furnace tube and the guide tube, to help prevent ingress of oxygen through the furnace tube wall.

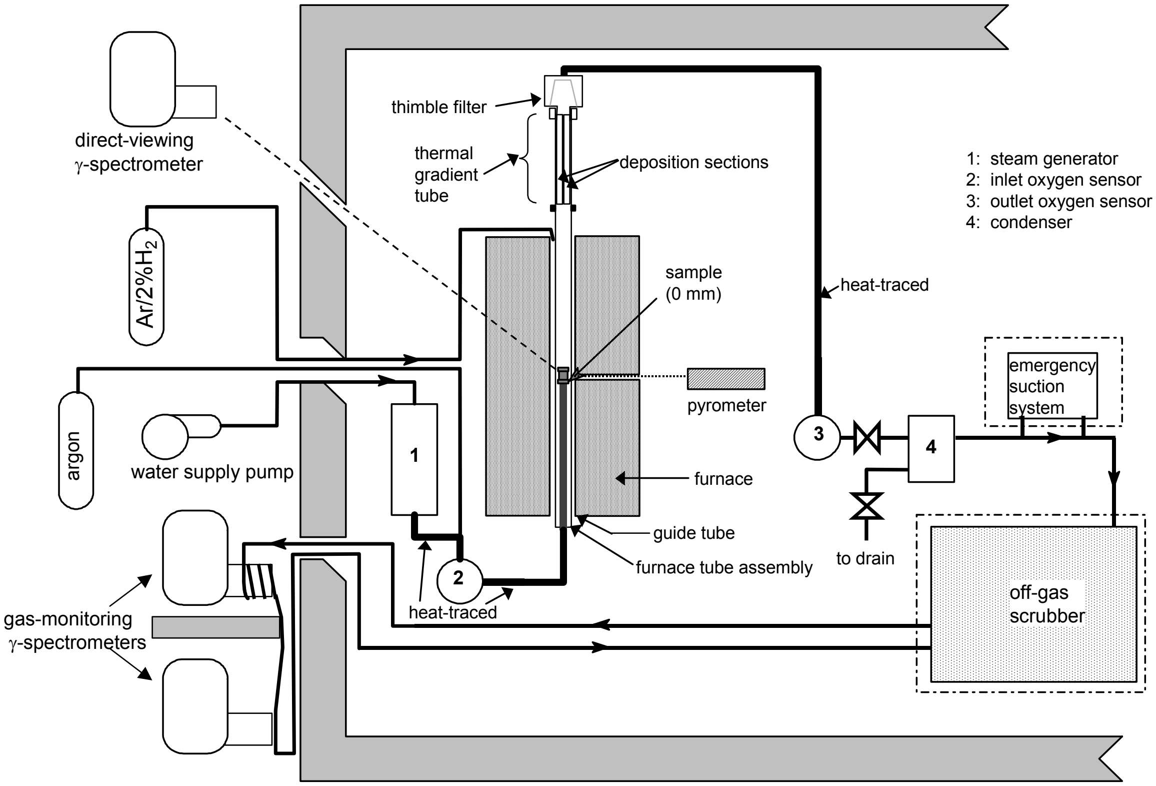

Figure 1: HCE5 Experimental Equipment for the TGT Tests

A TGT was attached to the furnace tubes in these tests. Two 168.3 mm long deposition sections chosen from four different CANDU-relevant materials (stainless steel SS-403, carbon steel, Inconel 600 and Zr-2.5Nb alloy) were inserted into the TGT housing. The internal surfaces of the deposition sections were maintained at desired temperature using two resistance heaters enclosing the thermal gradient tube. Another heater was provided to minimise FP deposition onto the downstream end of the furnace tube. The thermal gradient tube outlet was connected to the thimble filter via an end cap with an O-ring seal. Both the thimble filter and tubing were trace-heated to prevent steam condensation.

Typical argon flow rates during TGT tests were 400 mL/min for purging and 200 mL/min during the high-temperature part of the test. Distilled deionized water was fed at a controlled rate of

60 g/h from outside the hot cell to a small steam generator on the furnace chassis. The steam and argon were mixed and fed through heat-traced lines into a zirconia oxygen sensor [4] upstream of the furnace tube; a similar oxygen sensor monitored the oxygen partial pressure downstream of the filter assemblies. The oxygen sensors were operated at temperatures in the range 930 K to 1040 K, and were used to determine sample oxidation rates. Downstream of the second oxygen sensor, the steam was condensed and the non-condensable gas was passed through high-efficiency filters before flowing into the delay coil for γ-spectrometric monitoring.

Two optical pyrometers, arranged to view opposite sides of the outside of the guide tube, were employed for measurements of furnace hot zone temperatures. The fuel sample temperatures were estimated with an uncertainty of ±20 K using empirical correlations obtained from pre-experiment tests performed to determine the sample temperature as a function of the preheater temperature readings or the temperatures read by the optical pyrometers. The wall temperatures in the TGT region were determined from correlations determined from pre-test wall temperature measurements and the control thermocouples for the heaters in this region of the apparatus.

Three γ-spectrometers were used to measure fission-product releases during the tests (Figure 1). A direct-viewing γ-spectrometer was located outside the hot-cell and sighted through a collimated port to view the fuel sample through the wall of the furnace. Each spectrum was collected over a period of about 300 s. Fission-product release percentages were calculated from changes in count rates of selected peaks in the direct-viewing γ-spectra using methodology described in Reference [3]. Two γ-spectrometers monitored the noble gas count rates in the carrier gas flowing in two shielded delay coils connected serially outside of the hot cell [5-9].

Post-test scanning γ-spectroscopy was used to determine FP release and deposition characteristics of the deposition sections and furnace tube assembly components exposed to the carrier gas flow during the test. For post-test measurements, a scanning table was placed inside the hot cell [3,8,9] in front of a hot-cell port with a lead collimator with a slit width of about 10 mm. Spectra of the sample were measured at 6 mm or 5 mm intervals. The Aptec γ-spectrometer used for γ-scanning was located outside the hot-cell at the 6" round port. The distance between the spectrometer and the face of the hot cell was increased for some spectra to decrease the count rate. The γ-scanning count rates were converted to linear activities (Bq/cm) and fractions of the sample inventory using the methodology described in Reference [3]. The furnace tubes and deposition sections were leached using room-temperature water (adjusted to pH 10 to simulate water in the primary heat transport system), and the leachate samples were analyzed using ICP-MS (Inductively Coupled Plasma - Mass Spectrometry).

3. FUEL SAMPLES

The samples used in the TGT tests were segments 20 mm long cut from a Gentilly-2 CANDU power reactor fuel element. This fuel element was irradiated at a linear power of 47 to 57 kW/m to a burnup of 193 MWh/kgU and discharged about 24 months before the experiment. The segments were trace-reirradiated in the NRU reactor at low power just prior to the experiment to build up inventories of short-lived fission products.

4. CONDITIONS OF TEST TGT1

The peak sample temperature in the TGT1 test was 2055 K, and the high-temperature test atmosphere was steam. The deposition sections in the TGT housing were made of carbon steel and Zr-2.5Nb alloy.

The sample was heated to 1773 K at a rate of 5 K/min in argon flowing at a rate of 0.2 L/min, at which point a steam flow of 1 g/min was added to the argon flow while heating continued to 2055 K. The sample temperature was maintained near 2055 K for 3600 s, and then decreased at a rate of 6 K/min. When the sample temperature decreased below 1773 K, the steam flow to the sample was turned off, and the sample was cooled to room temperature in flowing Ar.

5. CONDITIONS OF TEST TGT2

The sample was heated at a rate of 5 K/min in flowing argon to the maximum temperature of 1963 K. The temperature plateau was maintained for 3800 s, then the sample was cooled to room temperature in flowing argon at a rate of 6 K/min. The deposition sections employed in this test were made of stainless steel SS-403 and carbon steel. The TGT2 test procedure was similar to that in test TGT1.

6. CONDITIONS OF TEST TGT4

The peak sample temperature in the TGT4 test was 2058 K, and the high-temperature test atmosphere was argon. The deposition sections placed in the thermal gradient tube were made of Inconel 600 and Zr-2.5Nb alloy. The TGT4 test procedure and operational parameters during fuel sample heating were similar to those in tests TGT1 and TGT2 except that the inert gas flow in the annulus between the furnace tube and the guide tube could not be maintained. Fibrous insulation was packed around the top of the annulus in order to decrease the amount of air passing through the annulus due to natural convection. The temperature plateau was maintained for 3100 s. Near the end of the temperature plateau, the failure of a furnace main heater element was detected and the test was terminated earlier than planned. Oxygen permeated through the zirconia furnace tube into the inert carrier gas due to the absence of inert gas purging of the annulus between the furnace tube and the guide tube, so the results of test TGT4 should not be interpreted as fuel behaviour in an inert atmosphere.

7. RESULTS

Post-test γ-scanning data were used to determine fractional FP releases to complement direct-viewing γ-spectrometry observations (especially for isotopes with low inventories or low fractional releases), and to estimate upper limits on the fractional releases of isotopes that could not be observed by γ-scanning. The total percentages deposited for all the isotopes observed in the TGT tests by γ-scanning are given in Table 1. The final fractional releases of the isotopes detected by direct-viewing γ-spectroscopy in these tests are also presented in Table 1.

Table 1: Deposited Fractions and Fractional Releases of FP Sample Inventories, with 1σ Uncertainties

Isotope	Test TGT1 T_{max}=2055 K Steam		Test TGT2 T_{max}=1963 K Argon		Test TGT4 T_{max}=2058 K Argon	
	Fraction Deposited (% of sample inventory)	Direct-Viewing Fractional Release (%)	Fraction Deposited (% of sample inventory)	Direct-Viewing Fractional Release (%)	Fraction Deposited (% of sample inventory)	Direct-Viewing Fractional Release (%)
Zr-95	<0.12	8 ± 5	<0.09	-0.05 ± 8 (≤ 17)	<0.14	3 ± 6
Nb-95	<0.08	-1.1 ± 1.1 (i.e. ≤ 1.0)	<0.06	-0.03± 1.4 (≤3)	<0.10	-0.8 ± 0.9 (≤ 1)
Mo-99	96 ± 11	90 ± 4	N/A	-1.6 ± 12 (≤23)	3.6 ± 0.4	N/A
Ru-103	<0.04	7 ± 1	<0.03	0.9 ± 1.1	<0.05	-1.5 ± 1.5 (≤ 2)
Ru-106	---	8 ± 7	---	-1.9 ± 1.5 (≤ 1)	---	5 ± 0.9
Ag-110m	7.7 ± 0.8	N/A	15.4 ± 1.6	N/A	9.7 ± 1.0	N/A
Sb-125	79 ± 9	N/A	---	N/A	63 ± 7	N/A
Te-132	22 ± 2	82 ± 17	---	-22 ± 21 (≤ 20)	4.8 ± 0.5	N/A
I-131	66 ± 7	81.8 ± 1.2	2.5 ± 0.3	5.3 ± 1.6	12.6 ± 1.3	42 ± 3
I-132[3]	21 ± 2	79.8 ± 1.4	---	-0.27 ± 3.1	4.1 ± 0.4	30 ± 28
Cs-134	62 ± 6	81 ± 2	12.7 ± 1.3	10 ± 4	52 ± 5	47 ± 3
Cs-137	59 ± 6	81 ± 6	11.4 ± 1.2	5 ± 8	49 ± 5	50 ± 5
Ba-140	5.9 ± 0.6	2.5 ± 1.6	0.06 ± 0.01	3.9 ± 1.5	7.9 ± 0.8	5 ± 2
La-140[4]	3.0 ± 0.3	6 ± 2	0.08 ± 0.01	4.1 ± 1.9	7.2 ± 0.7	9 ± 4
Ce-144	<0.11	N/A	<0.05	N/A	<0.08	N/A
Nd-147	<0.8	-1.2 ± 29 (≤ 56)	<0.6	17 ± 14	<1.2	-10 ± 36 (≤ 62)
Eu-154	<6	N/A	<1	N/A	<7	N/A
Eu-156	---	N/A	---	-21 ± 14 (≤ 7)	---	N/A
Np-239	<0.14	-7 ± 20 (≤ 33)	<0.4	14 ± 12	<3	N/A

---	Not observed in γ-scanning spectra
<##	In the “Fraction Deposited” column, this is the estimated upper release limit for this isotope in this test, based on non-observance of this isotope in γ-scanning spectra
≤##	In the “Direct-Viewing Fractional Release” column, the release values calculated to be negative are provided as the calculated R ± 1σ, with an upper limit ≤(R+2σ) shown for interpretation purposes. Minor apparent disagreements between these values result from rounding.
N/A	Not observed in direct-viewing γ-spectra in this test

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[3] I-132 deposition in γ-scanning originates from decay of deposited Te-132.

[4] La-140 deposition in γ-scanning originates from decay of deposited Ba-140

In test TGT1, the final fractional release of Mo-99 was about 90%, and releases of I and Cs isotopes were about 80%. Releases of about 80% of Sb and 22% of Te isotopes, along with 8% of Ag and 5% of Ba, were also indicated by γ-scanning of the deposition sections and furnace tube. The Cs and I release kinetics in the TGT1 test (Figure 2) were similar to those observed in the HCE5 iodine volatility tests [3] in steam environment. Rapid release of Cs occurred at a temperature between 1743 K and 1773 K (after the introduction of steam) at approximately the same time as releases of noble gases (Kr-85, Xe-133 and Xe-133m).

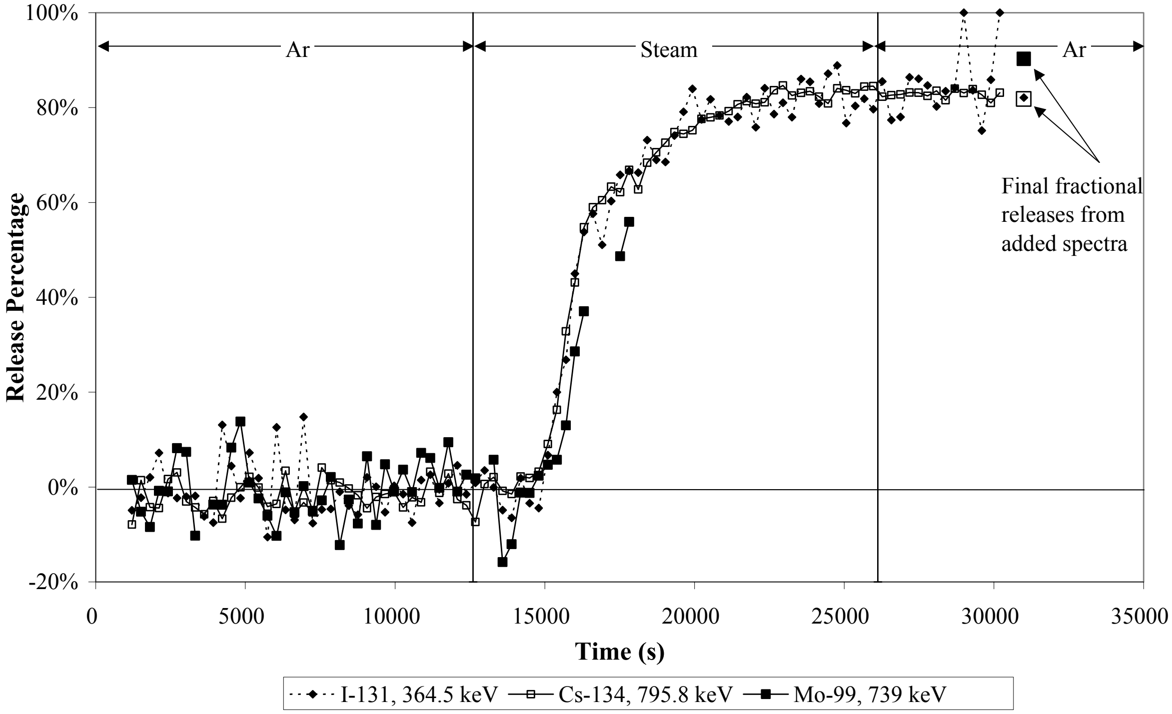

Figure 2: Mo-99, I-131 and Cs-134 Release Kinetics for Test TGT1

No Mo release was detected in test TGT2 from direct-viewing or γ-scanning observations, and relatively small releases (between 2% and 13%) of Kr, Xe, I and Cs isotopes were detected. The lack of Te release in inert environment was probably due to reaction of the Te with the unoxidized sheath [10].

In test TGT4, slow release of Xe began when temperature reached 1943 K. Rapid release of Xe, I, and Cs was observed at a temperature of 1996 K (Figure 3) and was probably associated with oxygen permeation into the inert carrier gas.

Deposition of Mo-99, I-131 and Cs-134 on the furnace tube assembly from test TGT1 are plotted in Figure 4. The “Linear Percentage” variable used on the deposition graphs is the percentage of the sample inventory of that isotope per mm of scanned length. Of the elements observed on the deposition sections, the Te, I and Cs showed some preferential deposition at temperatures above 760 K (positions between 510 mm and 530 mm) on the Zr-2.5Nb deposition section compared to the carbon steel deposition section (e.g., Figure 5).

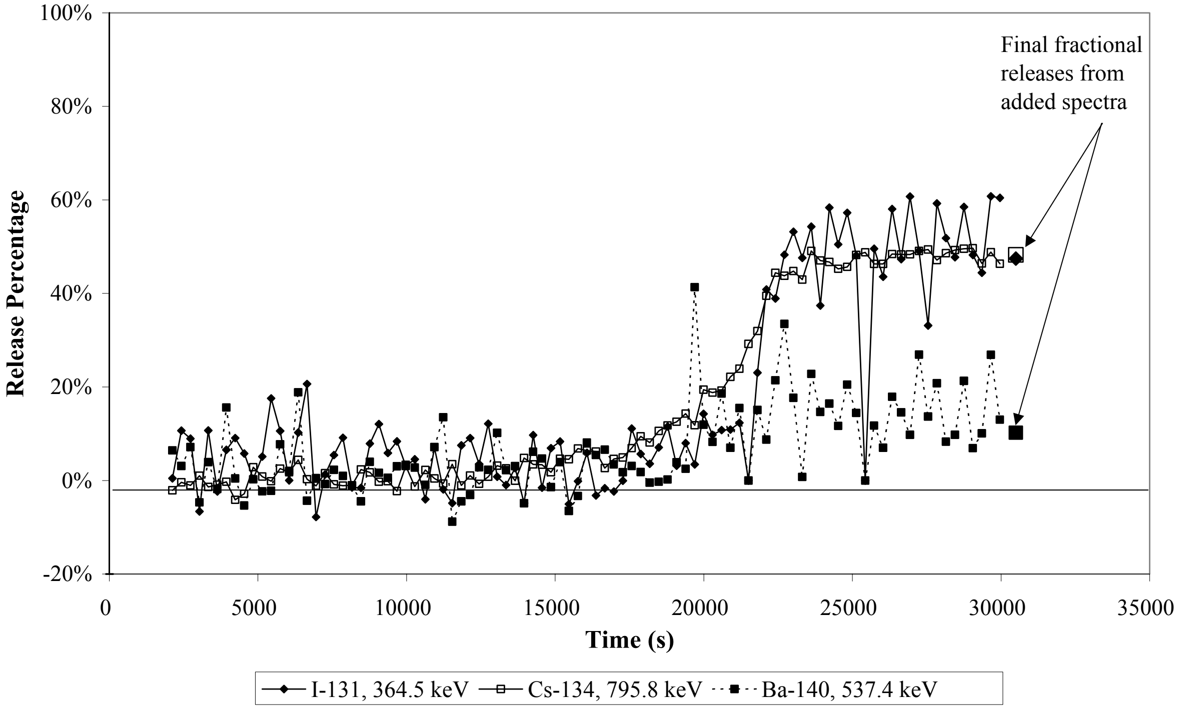

Figure 3: I-131, Cs-134 and Ba-140 Release Kinetics for Test TGT4

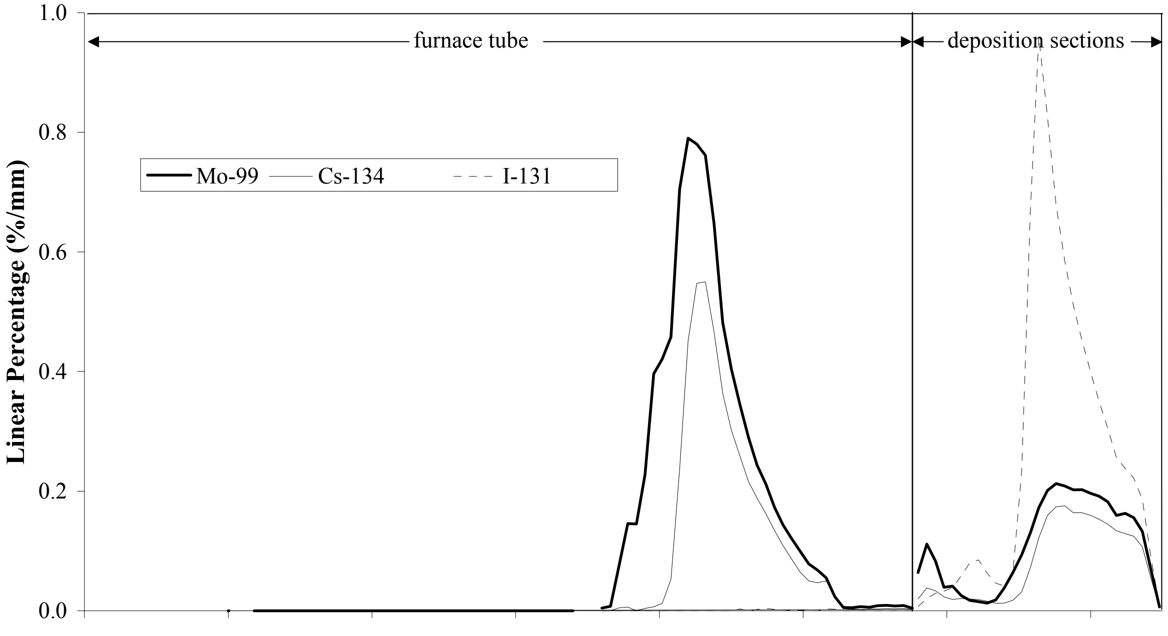

Figure 4: Linear Percentage Deposition of Mo-99, I-131 and Cs-134 as Functions of Position Along the Gas Flow Path in Test TGT1

Mo and Sb did not show any discernible preferential deposition. The preferentially deposited Te, I and Cs on the Zr-2.5Nb deposition section were not removed to any significant extent by leaching. Only about 20% of the other deposited Te was removed by leaching. The remaining deposited Mo, I and Cs were removed with good efficiency (about 95%) by leaching.

Linear percentage deposition of Cs-134 and I-131 on the furnace tube assembly from test TGT2 are plotted in Figure 6. The only elements detected on the deposition sections were I and Cs, and neither element deposited preferentially on the carbon steel or stainless steel deposition sections (e.g., Figure 7). Both I and Cs were removed with efficiencies of about 85%-90% by leaching.

The main elements detected on the deposition sections in test TGT4 were I and Cs, with a small amount of Te. Linear percentage deposition of Cs-134 and I-131 on the deposition sections in test TGT4 are plotted in Figure 8. Preferential deposition of I occurred at temperatures above 590 K (positions between 500 mm and 570 mm) on the Zr-2.5Nb deposition section compared to the Inconel 600 deposition section (Figure 9). Cesium did not deposit preferentially. Removal of I and Cs by leaching was very efficient (>95% for I, >99% for Cs).

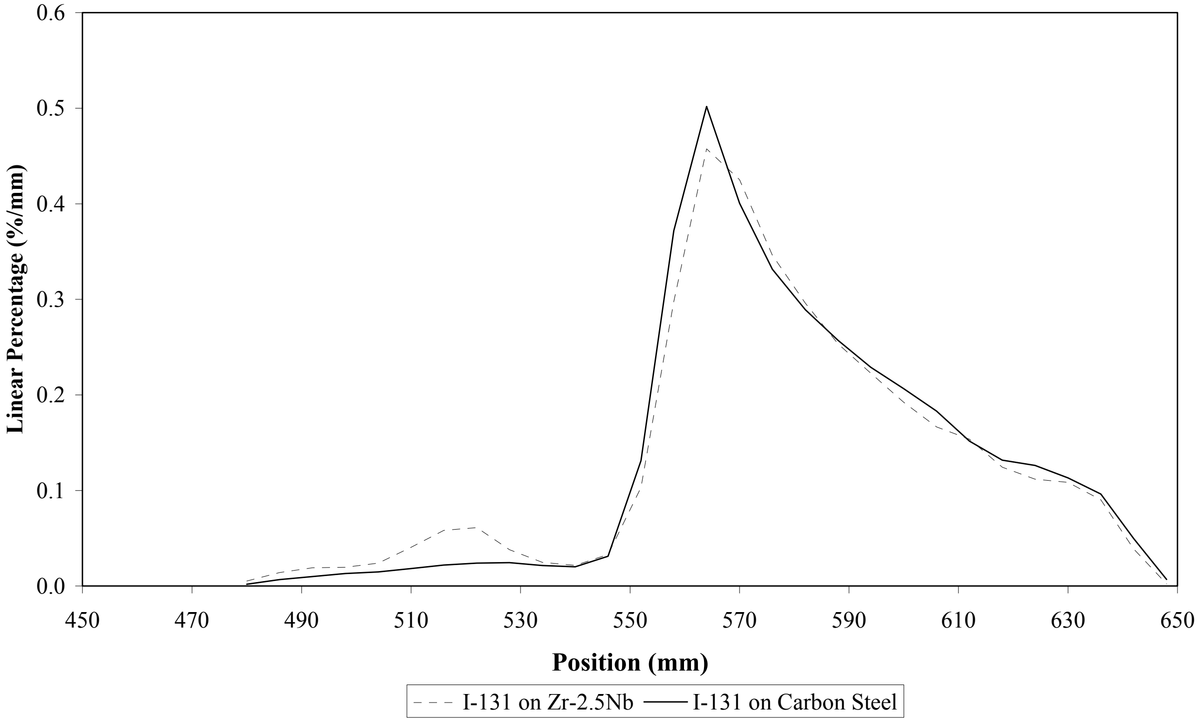

Figure 5: Linear Percentage Deposition of I-131 as a Function of Position Along the Deposition Sections in Test TGT1

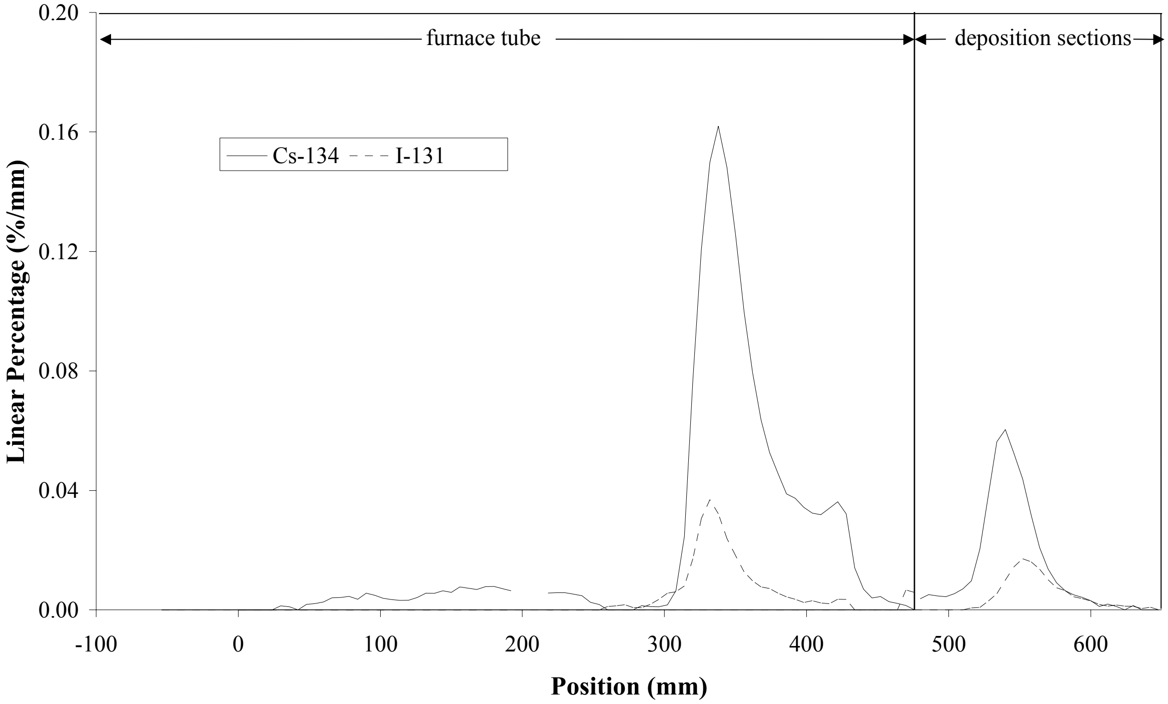

Figure 6: Linear Percentage Deposition of I-131 and Cs-134 as Functions of Position Along the Gas Flow Path in Test TGT2

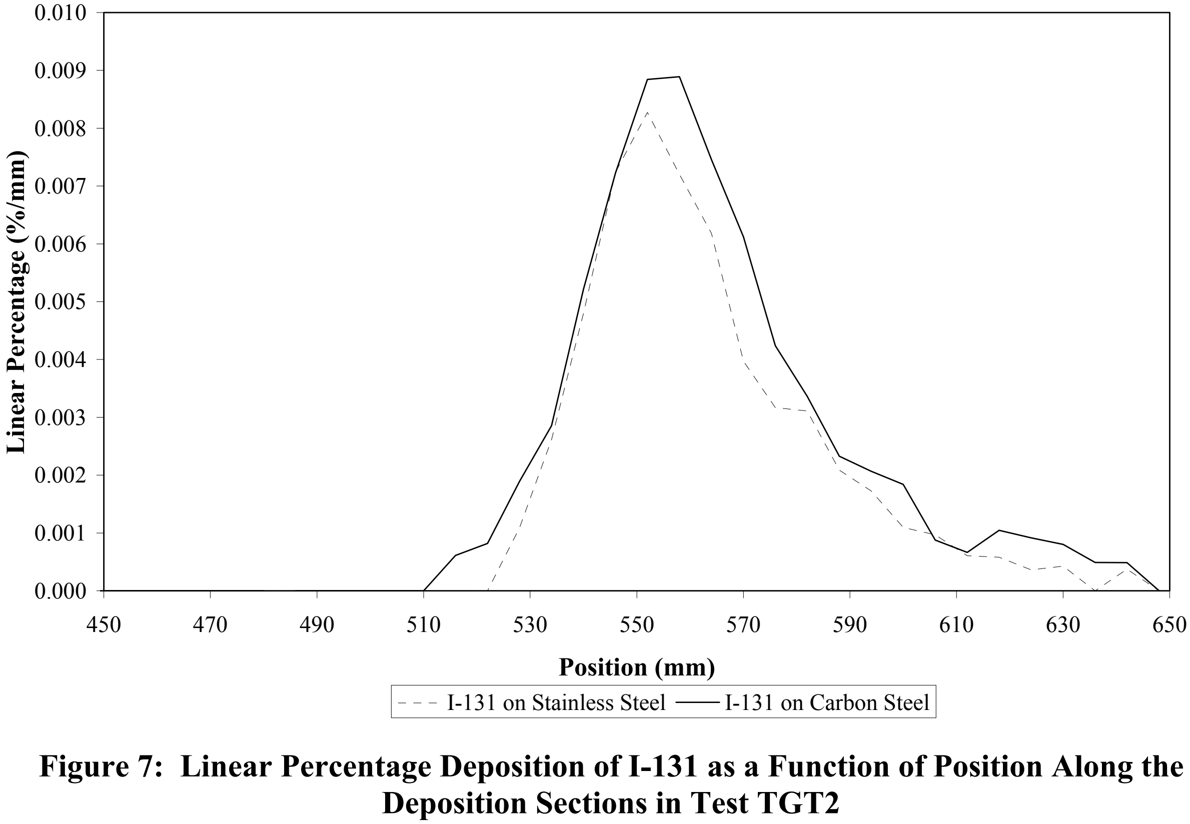

Figure 7: Linear Percentage Deposition of I-131 as a Function of Position Along the Deposition Sections in Test TGT2

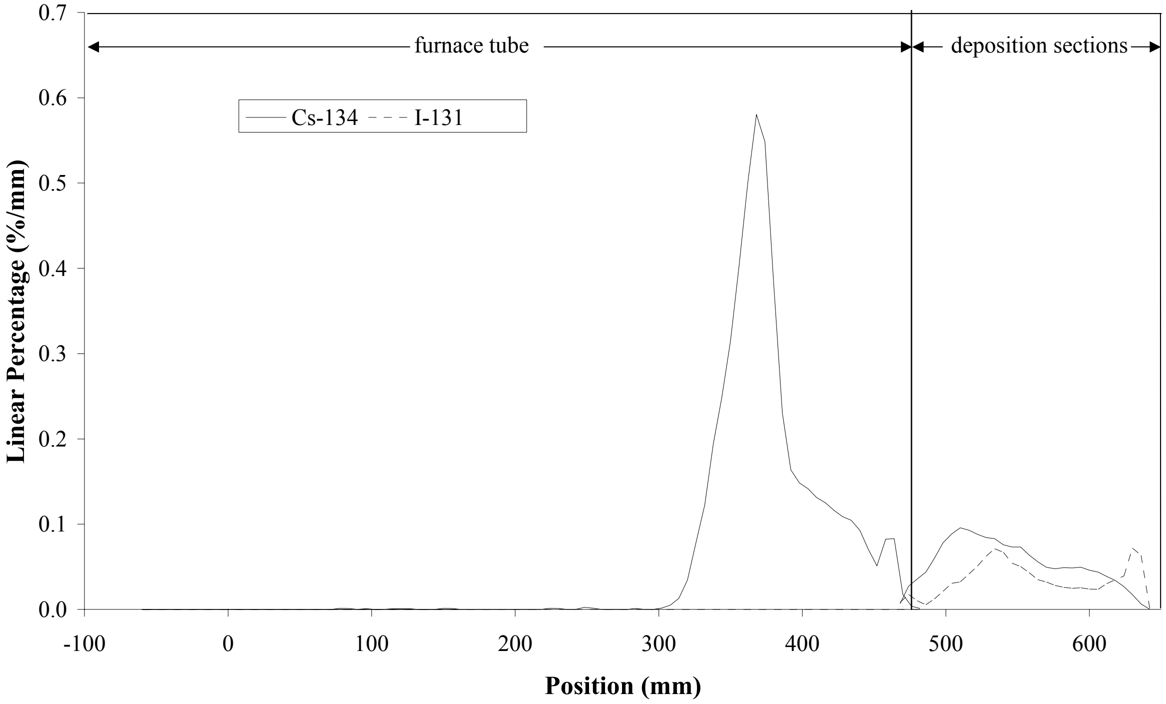

Figure 8: Linear Percentage Deposition of I-131 and Cs-134 as Functions of Position Along the Gas Flow Path in Test TGT4

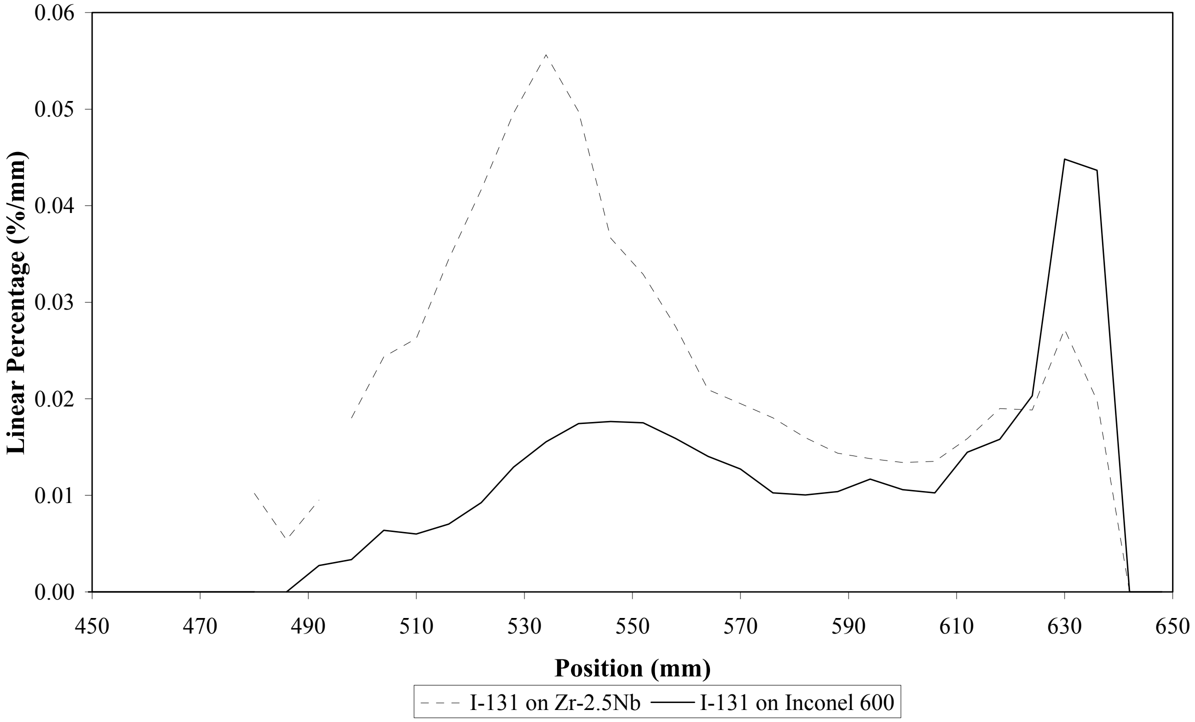

Figure 9: Linear Percentage Deposition of I-131 as a Function of Position Along the Deposition Sections in Test TGT4

8. CONCLUSIONS

The deposition and retention of fission products released from sheathed samples of irradiated CANDU fuel at high-temperatures on CANDU-reactor-relevant structural materials in steam and inert environment were investigated in the TGT tests in the HCE5 experiment.

In each test, a trace-reirradiated segment of a Gentilly-2 fuel element was heated to a peak temperature up to 2055 K for about 3600 s in a flowing carrier gas in the vertical furnace tube apparatus. Fission products and structural materials released at high temperature were deposited on the zirconia furnace tube and (at wall temperatures between 830 K and 460 K) on deposition sections placed in a thermal gradient tube downstream of the furnace tube. The remaining aerosol materials were then captured in a particulate filter maintained at a temperature below 400 K. The deposition sections were made of carbon steel, steel Type 403, Inconel 600 and Zr-2.5 Nb alloy. Direct-viewing and delay-coil on-line γ-spectrometry was used to determine releases of I, Cs, Te, Mo, Kr, Xe and other isotopes from the fuel sample. Fission products and structural materials deposited in the furnace tube, deposition sections, filters and other components exposed to the carrier gas flow were measured using scanning γ-spectrometry, and by leaching of the components followed by ICP-MS analysis of the leachate.

Of the elements observed on the deposition sections in the steam environment test TGT1, the Te, I and Cs showed some preferential deposition at temperatures above 760 K on the Zr-2.5Nb deposition section compared to the carbon steel deposition section. Mo and Sb did not show any discernible preferential deposition. The preferentially deposited Te, I and Cs were not removed by leaching, indicating that the preferential deposition was probably due to chemisorption. The fission products deposited in test TGT2 were not deposited preferentially on either the carbon steel or the stainless steel deposition section. The main elements detected on the deposition sections in test TGT4 were I and Cs, with a small amount of Te. Preferential deposition of I occurred at temperatures above 590 K on the Zr-2.5Nb deposition section compared to the Inconel 600 deposition section; Cs did not deposit preferentially.

The results on fission product releases from the fuel samples obtained from on-line direct-viewing γ-spectrometry measurement were in general agreement with the existing understanding of fission-product releases. The releases of most fission products were much higher in steam environment than in inert environment, and most of the releases occurred after complete steam oxidation of the sheath. The observed high and simultaneous releases of Mo and Cs in steam environment and the similarity of their deposition profiles is consistent with the results of reported previously HCE5 iodine volatility tests [3], and may indicate the formation of one or more compounds containing Cs and Mo.

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