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# CESIUM RELOCATION IN MIXED-OXIDE FUEL PINS RESULTING FROM INCREASED TEMPERATURE REIRRADIATION*

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# CESIUM RELOCATION IN MIXED-OXIDE FUEL PINS RESULTING FROM INCREASED TEMPERATURE REIRRADIATION

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Mixed-oxide fuel pins from EBR-II test subassemblies PNL-3 and PNL-4 were reirradiated in the GETR to study effects of increased fuel and cladding temperatures on chemical and thermomechanical behavior.[1] Radial and axial distributions of cesium were obtained using postirradiation nondestructive precision gamma-scanning techniques.[2] (Slide 1) Data presented here relate to the dependence of cesium distribution and transport processes on temperature gradients which were altered after substantial steady-state operation. Cesium is known to move both radially and axially in mixed-oxide fuel pins during normal operation [3] and has been identified as one of the primary fission products involved in fuel-cladding attack.[4] Cesium has also been associated with local cladding deformation or rupture due to cesium-insulator ($UO_2$) chemical interaction at the ends of the fuel column.[5]

The fuel pins which were reirradiated in GETR are summarized in the next slide. (Slide 2). Fuel pins can be grouped according to their prior EBR-II burnup (low and moderate) and heat rating (low and intermediate). Sibling pins were also obtained to establish the conditions of the fuel pins prior to the higher temperature reirradiation in GETR.

## Radial Cesium Behavior

Radial cesium profiles at selected axial locations on the fuel pins were obtained using two dimensional scanning techniques. A typical two dimensional profile is shown in the next slide as diameter profiles at 0° and 90° (Slide 3). The two diameter profiles are then unfolded mathematically to obtain a two dimensional profile. These two dimensional profiles can be represented as density plots or isometric projections as shown on the next slide (Slide 4). Data can also be represented as a contour plot if desired.

The radial distributions of cesium were quite similar for all pins in the series. Results of the radial profile measurements are summarized in the next slide (Slide 5). They are:

(A) No significant differences occurred between any of the fuel pins, sibling or reirradiated.

(B) Cesium was generally rather evenly distributed around the circumference of the fuel even though there were circumferential temperature variations in GETR.

(C) Cesium was generally concentrated in the outer approximately 1 mm of fuel although some cesium appears to remain in the inner regions of the fuel.

Results indicate that in all the fuel pins in the test, sibling or reirradiated, operated a temperatures and radial thermal gradients which were sufficient to move the cesium to the outer cooler regions of the fuel pellet.

Axial Cesium Behavior

Axial cesium distributions were compared to similar data from the sibling pins following EBR-II irradiation to determine how the cesium was redistributed as a result of the imposed temperature profiles. Axial temperature profiles for the moderate burnup, intermediate power pin (PNL-4-14/RCT-1A) for both EBR-II and GETR are shown in the next slide (Slide 6). The corresponding axial cesium profile for this fuel pin is compared to the sibling pin profile in the next slide. (Slide 7).

Cesium ($^{137}Cs$) was found to move to both ends of the fuel column but predominantly toward the cooler lower end. Cesium was peaked over the lower $\sim$ 1/3 of the reirradiated fuel pin. Also, there is an absence of the large activity peaks seen in the sibling pin and associated with pellet to pellet interfaces. The scales for cesium counts for the two scans are not the same but were optimized for sensitivity to local variations in concentration. Therefore, a direct comparison of the two profiles (sibling and reirradiated) is not possible.

Similar $^{137}Cs$ profiles for the corresponding low-burnup case are shown in the next slide. (Slide 8). Again, cesium moved predominantly to the cooler end of the column with the concentration at the top quite similar at both burnups. At the lower end of the column cesium is concentrated over approximately the first inch of fuel rather than the lower $\sim$ 1/3 of the column.

A comparison of the two intermediate power pins (low and moderate burnups) reirradiated at similar conditions (Slides 7 and 8) suggests that the concentration profits are dependent upon prior EBR-II burnup.

Similar cesium data for the low burnup, low power pin reirradiated in GETR is shown in the next slide. (Slide 9). Results in this case are different from the low burnup intermediate power pin. Cesium is concentrated toward the cooler end of the column in a manner more like the moderate burnup pin (Slide 7) than the corresponding low burnup pin. This comparison of relative profiles for the two low burnup pins suggests a dependence on prior EBR-II heat rating.

Axial relocation data for cesium in the corresponding low power moderate burnup pin was not available. This pin failed during the reirradiation in GETR. The cladding breach resulted in significant quantities of NaK entering the fuel pin. The data suggest a significant perturbation to the distributions of cesium within the pin as a result of the NaK intrusion.

The axial redistribution of cesium for these pins reirradiated at increased temperatures in GETR is summarized in the next slide. (Slide 10).

A. Substantial amounts of cesium relocated toward cooler end of fuel column.

B. Redistribution dependent upon prior EBR-II burnup and heat rating.

C. Cesium which moved, in part, originated in pellet-to-pellet interfaces.

I would like to conclude this discussion with a plausible explanation of these observations of cesium relocation. (Slide 11). The cesium formed at any axial location can either remain at that location or migrate down the temperature gradient. Migration down the gradient is affected by formation of compounds with the fission products and/or fuel. Cesium as an iodide retains some of its mobility at the fuel surface temperatures of this test and therefore moves down the gradient until the vapor pressure precludes further movement. As a telluride, movement is restricted and the cesium remains in the regions of high cladding temperature where it can promote cladding chemical interaction.

Cesium movement down the fuel pin temperature gradient is also slowed by the formation of compounds with the fuel. In the case of the low and moderate burnup intermediate power pins (PNL-4's),differences observed in the axial cesium profiles could result from:

- A higher oxygen potential at the fuel surface in the high burnup pin, and

- a somewhat higher threshold energy for formation of cesium fuel compounds (> -135 Kcal/mole) in the low burnup pin due to a higher fuel surface temperature.

The higher oxygen potential in turn augments the chemical interaction of the cesium with the fuel thereby retarding movement down the temperature gradient.

Similarly, in the case of the two low burnup pins reirradiated, the observed differences in cesium profiles could be due to lower fuel surface temperatures in the low power pin. These lower temperatures result in a more negative value for the threshold energy for fuel cesium compound formation. Therefore, fuel cesium compounds are formed at axial locations further from the bottom of the fuel column in the low power pin when compared to the corresponding intermediate power pin.

I would like to summarize in the last slide (Slide 12). Cesium was found to redistribute itself significantly during increased temperature reirradiation in GETR. The observed differences in axial relocation can be explained in terms of differences in the local oxygen potential, and the fuel-cladding gap temperatures.

References

1. L. A. Lawrence, E. T. Weber, "Reirradiation of Mixed Oxide Fuel Pins at Increased Temperatures," *Trans. Amer. Nucl. Soc., 23* (1976). (Also HEDL-SA-75-1084)

2. J. R. Phillips, "New Techniques in Precision Gamma Scanning," LA-5260-T, July 1973.

3. R. A. Karnesky, et al., "Cesium Migration in Mixed-Oxide Fuel Pins," *Trans. Amer. Nucl. Soc., 22,* 229 (1975).

4. R. W. Ohse and M. Schlechter, "The Role of Cesium in Chemical Interaction of Austenitic Stainless Steels with Uranium Plutonium Oxide Fuels," EUR 4893e, August 1972.

5. J. D. B. Lambert, et al., "A Failure Mechanism in Mixed-Oxide Fuel Elements," *Trans. Amer. Nucl. Soc., 17,* 193 (1973).

SLIDE 1

# CESIUM RELOCATION IN MIXED OXIDE FUEL PINS

- RADIAL AND AXIAL CESIUM DISTRIBUTIONS MEASURED BY GAMMA SCANNING
- CHANGES IN CESIUM PROFILES DUE TO

  CHANGES IN FUEL TEMPERATURES DURING REIRRADIATION

  DIFFERENCES IN PRIOR EBR-II BURNUP AND HEAT RATING

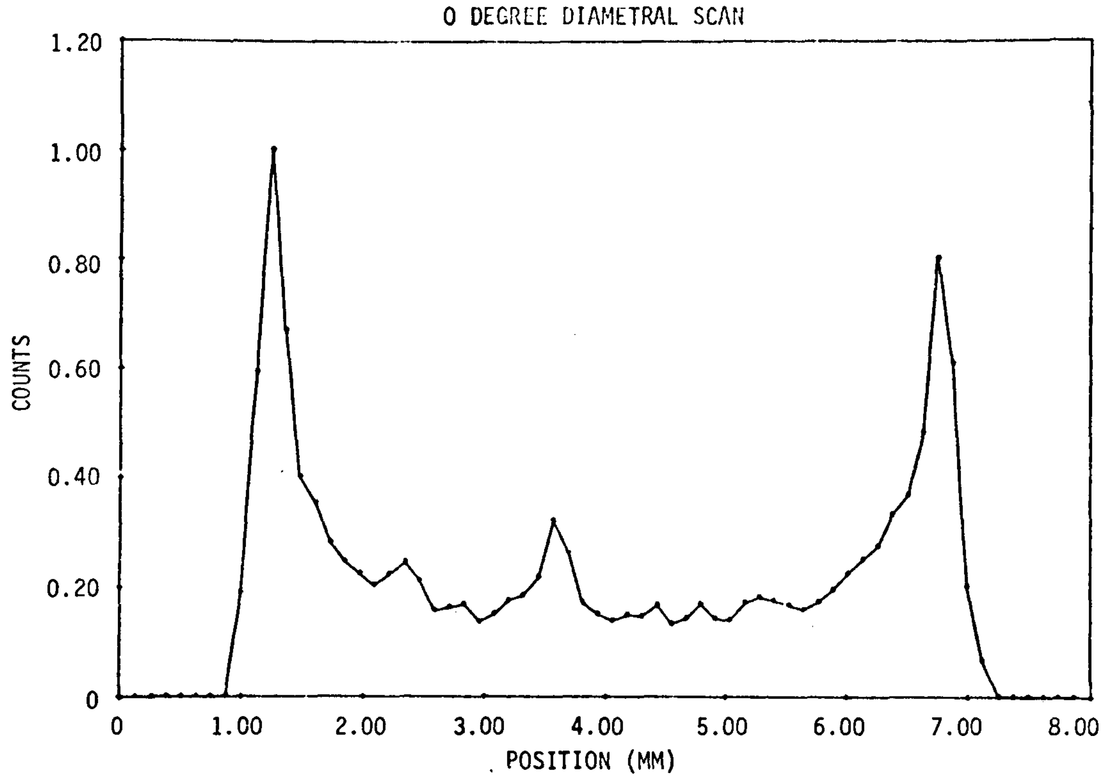
HEDL 7605-148.1

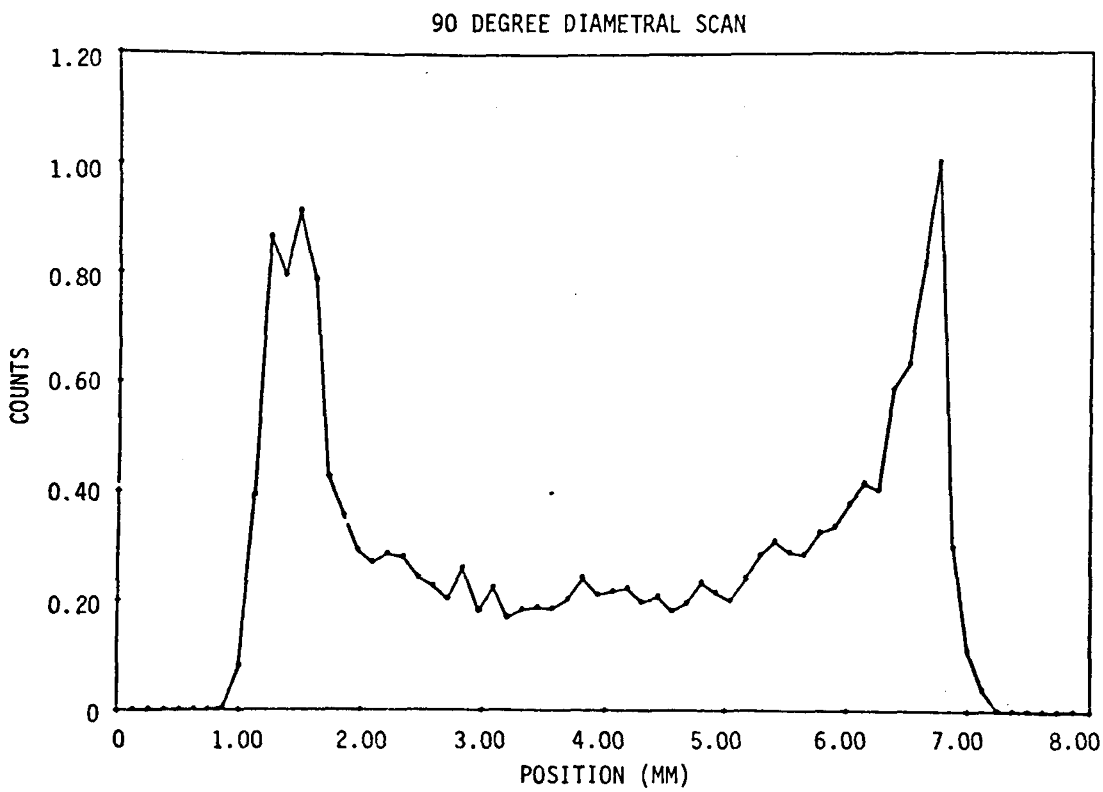
REIRRADIATION OF EBR-II PINS IN GETR

# SUMMARY OF CAPSULES AND FUEL PINS

| | | EBR-II BURNUP PRIOR TO REIRRADIATION | |
|---|---|---|---|
| EBR-II HEAT RATING PRIOR TO REIRRADIATION | | LOW BURNUP<br>PNL-3 (2.1 at. %)<br>PNL-4 (2.4 at. %) | MODERATE BURNUP<br>PNL-3 (4.7 at. %)<br>PNL-4 (6.4 at. %) |
| | LOW POWER<br>170 - 188 w/cm<br>5.2 - 5.7 kw/ft | RCT-2 (PNL-3-41)<br>SIBLING: PNL-3-26 | RCT-4 (PNL-3-14)<br>RCT-5 (PNL-3-16)<br>SIBLING: PNL-3-15 |
| | INTERMEDIATE POWER<br>274 - 294 w/cm<br>8.4 - 8.9 kw/ft | PCT-3 (PNL-4-61)<br>SIBLING: PNL-4-60 | RCT-1A (PNL-4-14)<br>SIBLING: PNL-4-12 |

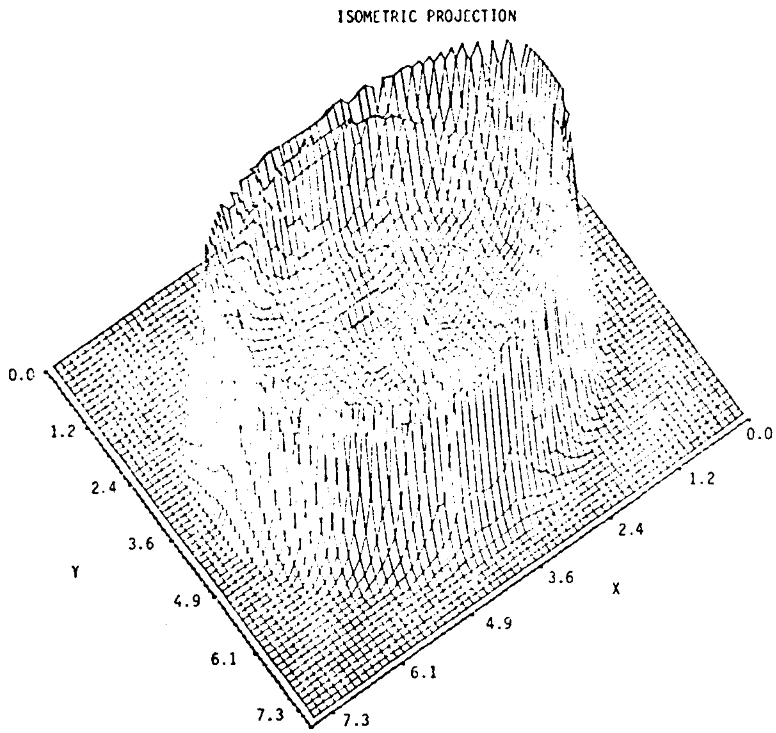
HEDL 7604-40-25

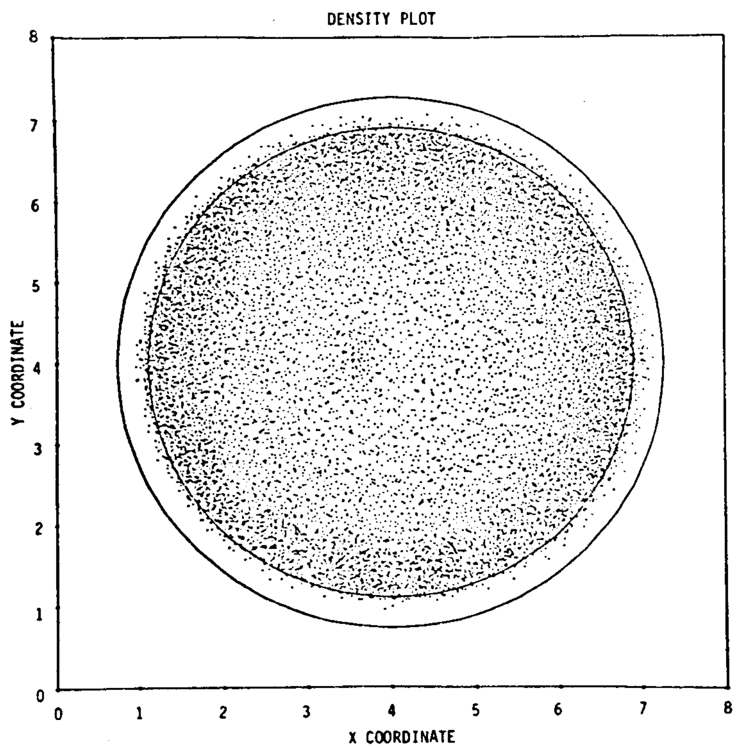



HEDL 7507-1.7

SLIDE 3




HEDL 7507-1.6

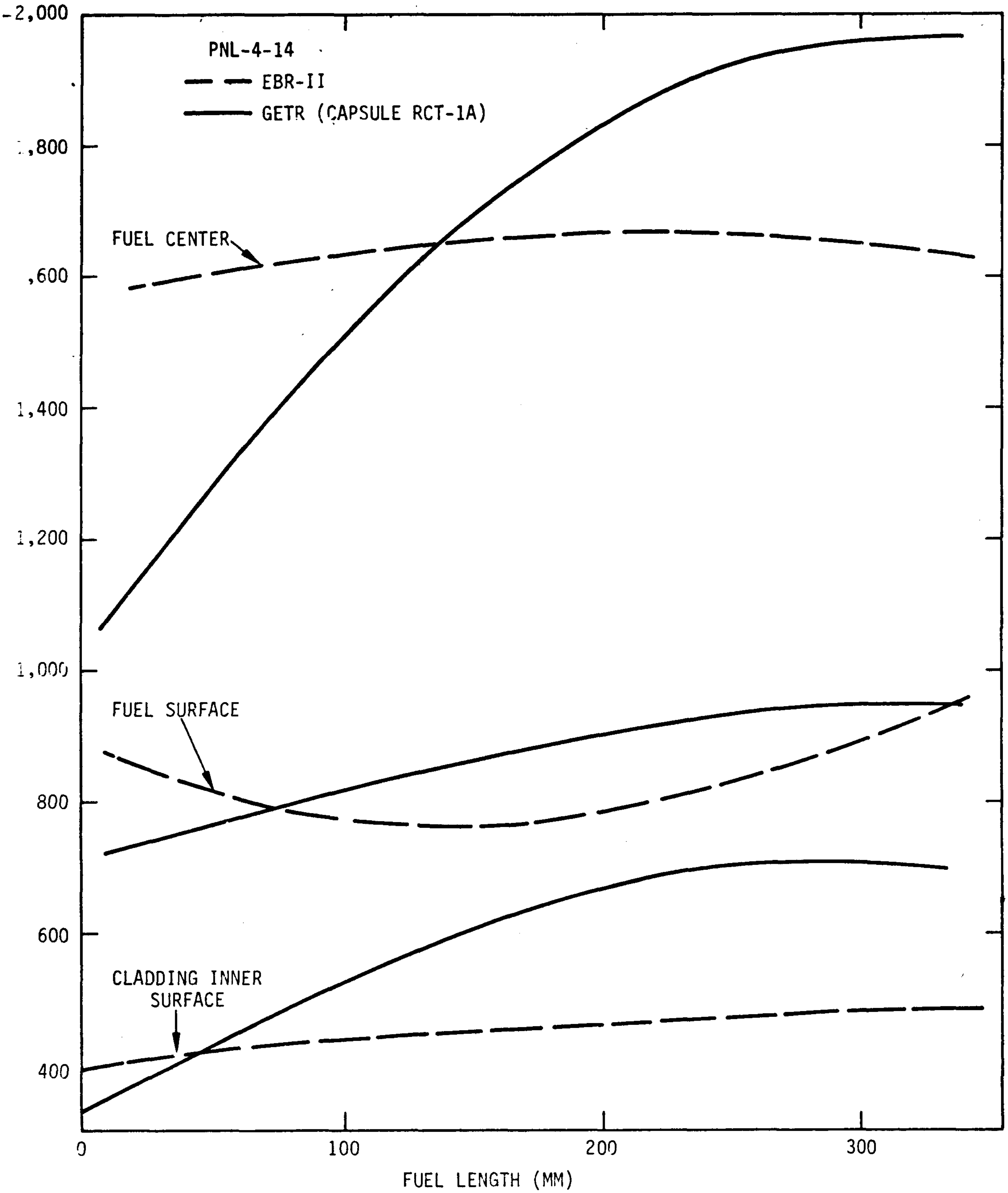
SLIDE 4

SLIDE 5

# SUMMARY OF RADIAL CESIUM BEHAVIOR

- CONCENTRATED IN OUTER LOW TEMPERATURE REGIONS OF FUEL PIN
- RATHER EVENLY DISTRIBUTED AROUND FUEL CIRCUMFERENCE
- PROFILES INDEPENDENT OF ANY DIFFERENCES IN FABRICATION OR IRRADIATION PARAMETERS SIBLING OR REIRRADIATED

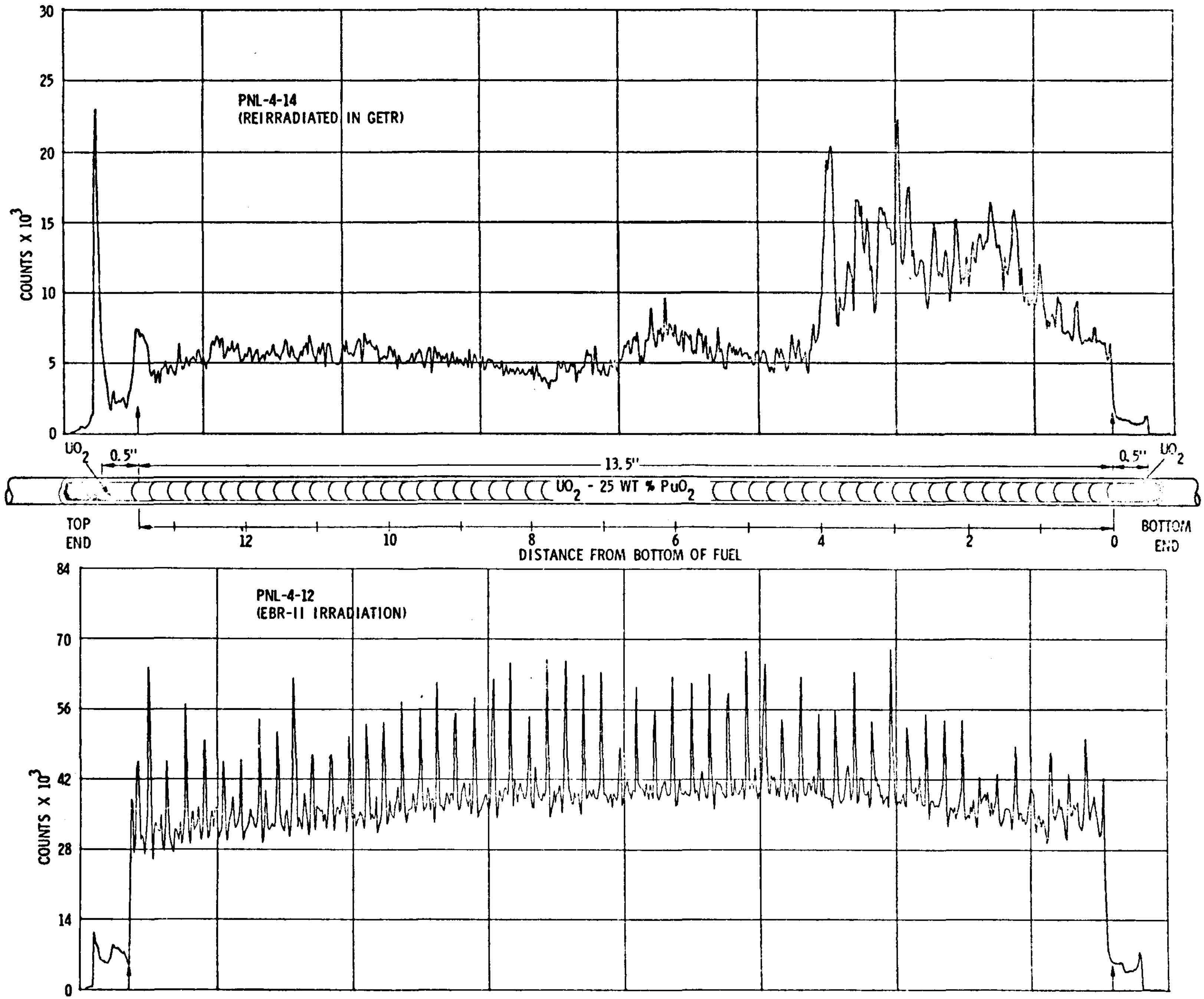
HEDL 7605-148.2




HEDL 7602-164.9

SLIDE 6

SLIDE 7

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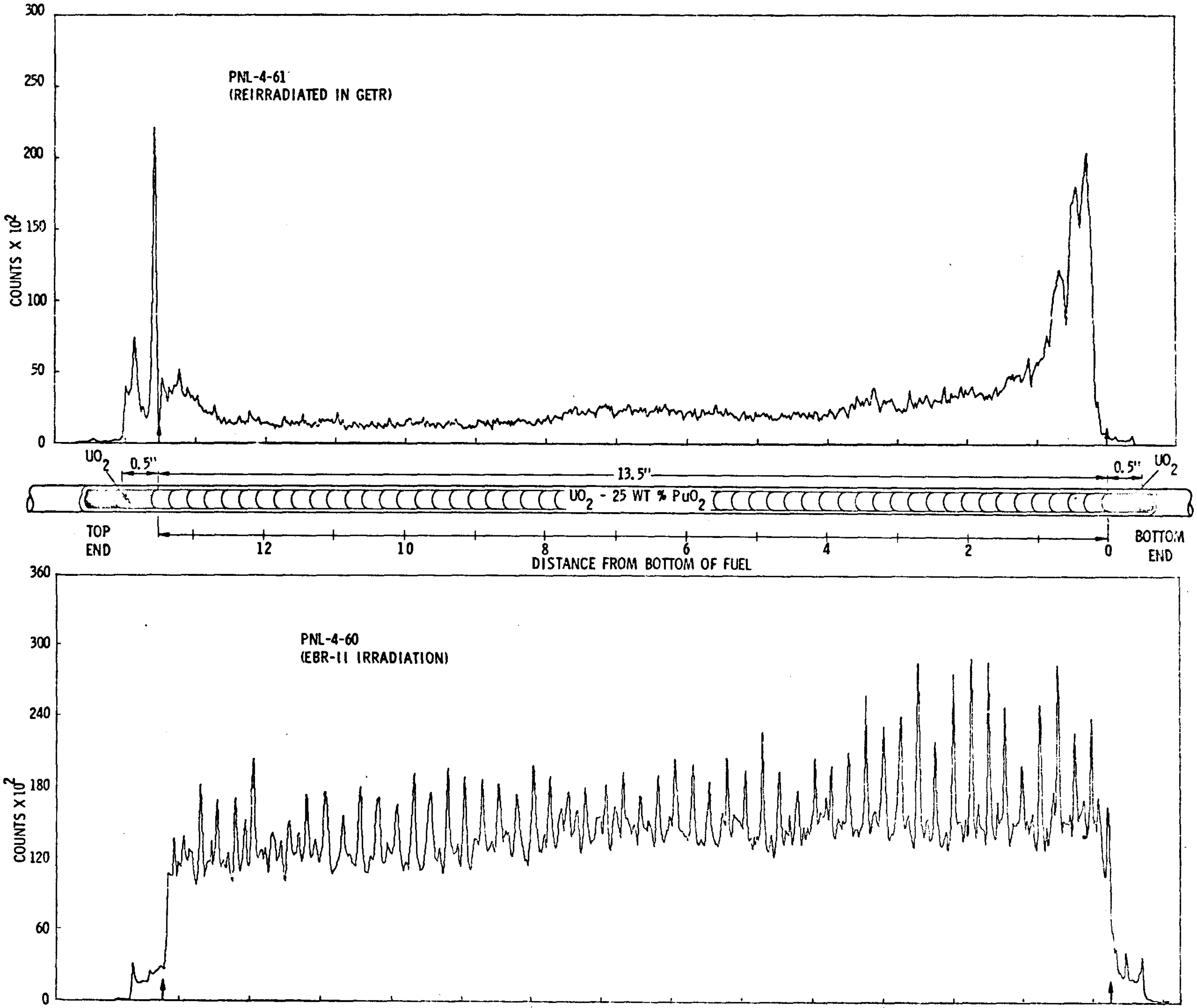
SLIDE 8

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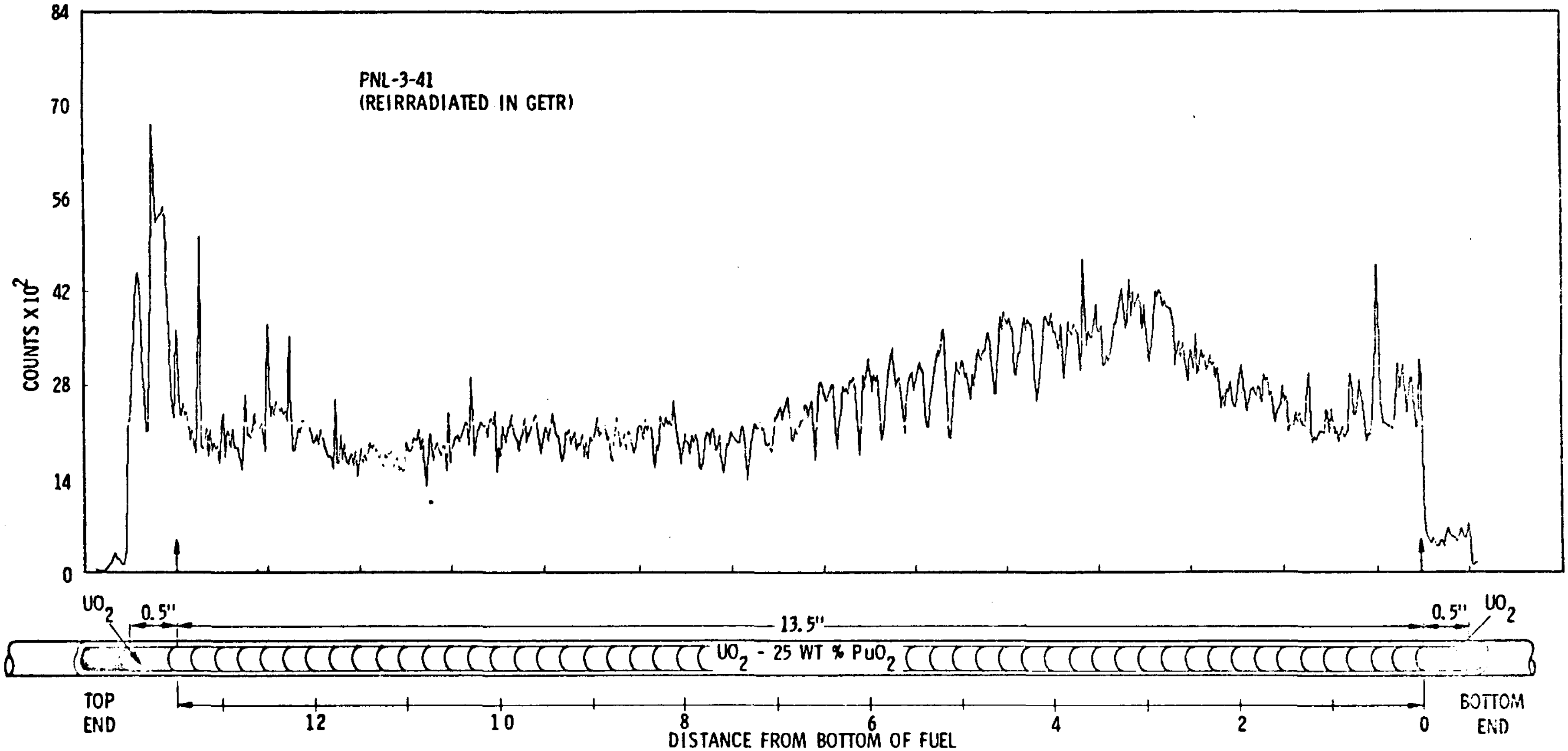
SLIDE 9




HEDL 7506-114.1

SLIDE 10

# SUMMARY OF AXIAL CESIUM REDISTRIBUTION

- SUBSTANTIAL AMOUNTS RELOCATED TOWARD COOLER END OF FUEL COLUMN
- REDISTRIBUTION DEPENDENT UPON PRIOR EBR-II BURNUP AND HEAT RATING

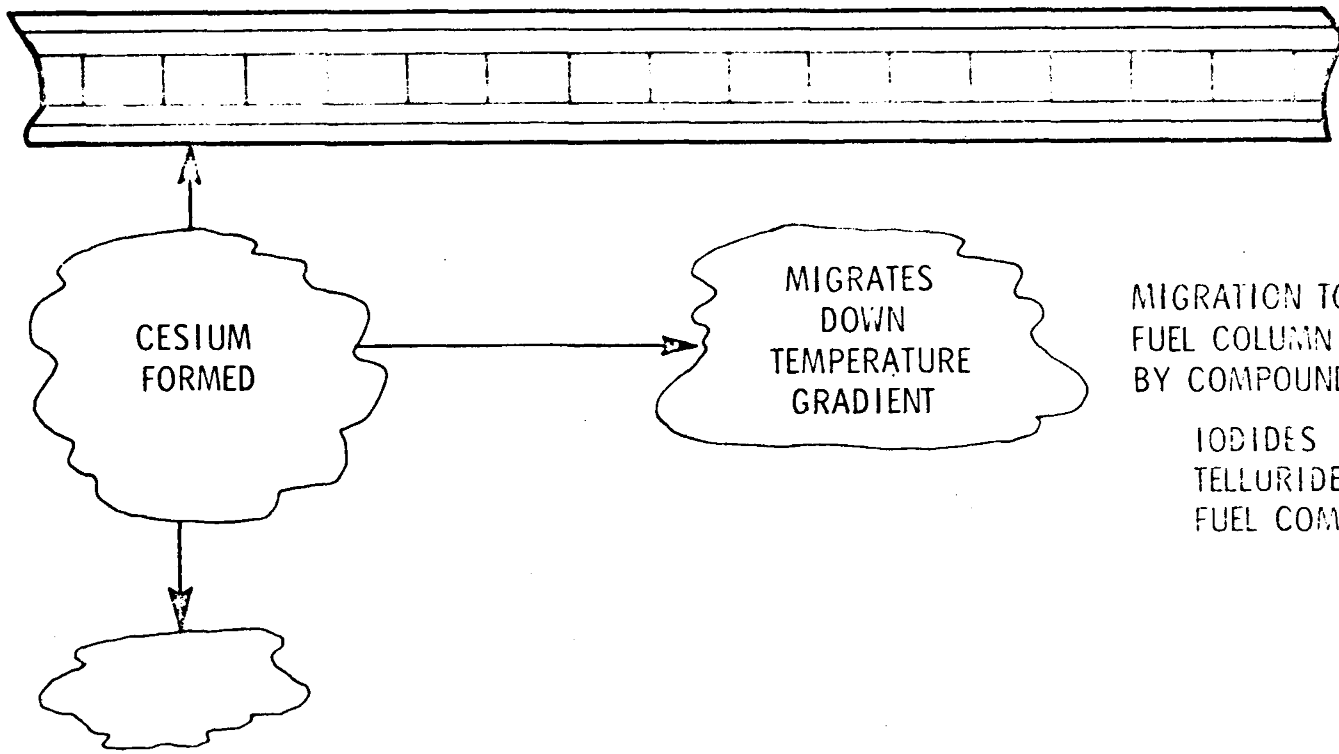
  LOW BURNUP - END OF FUEL COLUMN

  MODERATE BURNUP - LOWER 1/3 OF FUEL COLUMN
- CESIUM WHICH MOVED IN PART ORIGINATED IN PELLET-TO-PELLET GAPS

HEDL 7605-148.4

SLIDE 11

# CESIUM MIGRATION IN AN OXIDE FUEL PIN




MIGRATION TO ENDS OF FUEL COLUMN AFFECTED BY COMPOUND FORMATION

IODIDES
TELLURIDES
FUEL COMPOUNDS

REMAIN AT FUEL-CLADDING GAP

- TRAPPED IN ISOLATED POROSITY
- EXISTS AS COMPOUND WITH FUEL

# CESIUM REDISTRIBUTION

- RELOCATES TO OUTER LOW TEMPERATURE REGIONS OF FUEL PIN
- LOCAL CONCENTRATION PEAKS AT PELLET-TO-PELLET GAPS
- SUBSTANTIAL AXIAL RELOCATION TO COOLER END OF FUEL COLUMN
- AXIAL RELOCATION CONTROLLED BY:
  - LOCAL OXYGEN POTENTIAL
  - FUEL-CLADDING GAP TEMPERATURES

HEDL 7605-148.5

SLIDE 12