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Comments on the Possible Roles of Volatile Fission Products (Cesium) in CABRI Tests

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An investigation of information within the CABRI program that relates to the possible roles of volatile fission products (as represented by cesium) will be described. This study was partially motivated by the observation of localized ^{137}Cs concentration peaks in the axial gamma scans of pins pre-irradiated to about 5% burnup (B.U.) level. In order to evaluate potential effects of such concentrations, a re-examination of the existing test data for the 1% B.U. pins was performed. A comparison of CABRI hodoscope fuel motion results and the pre-CABRI ^{137}Cs axial concentration profiles revealed an approximate spatial correlation between the initial points of fuel dispersal and cesium concentration enhancements (seven of eight cases).

INTRODUCTION

1. Recently the CABRI program staff completed the test matrices for the fresh UO_2 and low pre-irradiated (1% burn-up) mixed oxide fuel pins and began preparation for the H-series tests using pins pre-irradiated to about a 5% burn-up (B.U.) level. It was anticipated that the increased formation of fission products and their net retention in the latter fuel would be several times higher than that of the 1% B.U. case (J-series). The main aspects of study were to be the effects of burn-up in pin behavior under similar test conditions. In particular, the role of retained fission gases was to be evaluated. However, initial non-destructive exams and subsequent destructive examinations revealed that the total retained fission gases in the 5% B.U. fuel was comparable to that of the 1% B.U. fuel, although the volatile fission product seemed to scale with the increase in burn-up.[1] As a consequence, the relative roles of volatile fission products (such as Cesium) to fission gas effects should be magnified. In addition there were observed localized axial concentrations of Cesium as indicated by the axial gamma-scan data for Cs-137 (e.g. Fig. 1) and Cs-134. These concentrations were a few centimeters in axial extent and often had an intensity two to three times greater than the mean activity.

2. In order to evaluate potential effects of such concentrations in the 5% B.U. case, a re-examination of the existing low burn-up pin test data was performed. The approach was to determine whether there was a Cs axial concentration pattern and how it related to observed phenomena for the 1% B.U. cases (e.g. Fig. 2). Volatile fission products might provide an additional pressure source for the pin or promote cladding failure by local gap pressures, altered fuel temperature profiles, or some kind of attack (fuel adjacency effect). Such effects might manifest themselves in lowered pin failure thresholds (enthalpy-wise, b) unexpected failure locations, c) increases in fuel ejection, and/or d) an increased extent (magnitude, time) of fuel motion. Since the CABRI hodoscope data can provide input to all four points, test-by-test comparisons of observed fuel motion events and the Cs-137 pre-CABRI axial distribution as given by the gamma-scan data were performed. The gamma scans were performed at Cadarache at the hot cell laboratory (LECA) and at the CABRI facility (SES). When possible the CABRI data were used to corroborate or extend the comparison.

3. It should be mentioned as background that the possibility of volatile fission product (VFP) effects on irradiated fuel behavior has been under discussion (debate) for several years[2,3], and Cs is a representative of that class. Since Cs lies on the peak of the fission yield curve and its boiling point is 680°C, it is in fact one of the most likely VFP contributors. More specifically, the potential of Cs related test pin pressurizations in recent SANDIA in-pile tests was indicated by observations of Cs and Rb release in the vapor/gas clouds at the time of fuel swelling, cladding rupture, and fuel break-up. Post test examinations also showed a depletion of the Cs

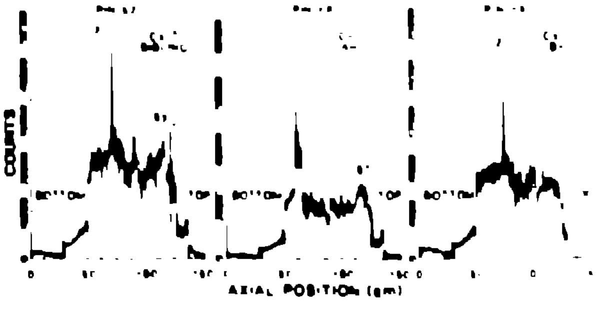

Fig. 1. (Cs^{137}) axial gamma scans for three 5% B.U. pins

inventory in the fuel. It had been suggested in the past that the current CABRI ramp rates were too fast for Cs effects to be important relative to fission gas. This opinion was partly supported by an appeal to the random distribution of Cs-137 enhancements in LECA's test pin gamma scans and the lack of an identified correlation with observed phenomena.

4. However, the author believes an approximate spatial correlation has been noted in most of the cases, although a more detailed study of fuel motion is needed. Practically speaking this should be done at a later time when modeling and hodoscope signal-to-mass conversion procedures become more routine. In addition, the issues of initial Cs inventory, Cs survivability under irradiation, and depletion of the inventory will be addressed via the data on hand. It should also be noted that a detailed discussion of the complex physical chemistry issues related to volatile fission produce migration, compound formation, release rates, pressurization potential, etc. are beyond the scope of this note. In fact, much fundamental research is still needed. However, the prospects are good for taking advantage of the low fission gas retention in H-series pins to assess fission product effects relative to I-series pins as proposed in the fourth section.

EXPERIMENTAL BACKGROUND

5. It is appropriate to briefly address test data that provide input on fission gas (Xe) and volatile fission product (Cs) inventory depletion, transient release, and concentration survivability. These are addressed partly through specific Sandia test program results and two reference tests of the CABRI I-series matrix, AI1 and BI1.

Sandia test information

6. The fuel disruption test program at SANDIA has been a useful source of data bearing on the issues of Xe and Cs inventory depletion and transient release.[2] In one particular series electron microprobe analysis was used to evaluate the Xe and Cs inventories after steady state (PNL 10-12) and transient irradiation (FD 1.7 and FD 1.8). The PNL 10-12 pin (5.4% B.U.) was measured to have retained Xe and Cs of 0.14 and 0.4 wt %, respectively. The FD 1.7 pin (5.4% B.U.) retained Xe and Cs of 0.07 and 0.35 wt %, respectively. However, the FD 1.8 pin (4.7% B.U.) retained the same amount of Xe but relatively much less Cs. Table 1 shows this in terms of depletion. The measured Cs release (in wt %) was about our times that of Xe in the FD 1.8 test. Both transient irradiations involved an energy injection of 1.5 kJ/g, but the FD 1.8 ramp rate was faster.[3]

Table 1 Xenon and Cesium Inventory Depletion in Sandia Tests FD 1.7 and FD 1.8

TEST	Xenon (Wt % Released)	Cesium (Wt % Released)*	Energy Injected (kJ/g)	Ramp Rate
FD 1.7	[illegible]	[illegible]	1.5	Slow
FD 1.8	[illegible]	[illegible]	1.5	Faster

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*An integration of electron microprobe data

7. Secondly, an absorption spectrometer technique with a high speed film camera was used to monitor transient Cs release. The FD 4.3 test data show Cs and Rb releases at the time of pin break-up, but the amount was not determined. Such a timing of the Cs release indicates a participation in the pin break-up.

CABRI reference tests

8. An important open issue for the CABRI tests involved the Cs concentration stability under nominal power, loss-of-flow (LOF) and/or transient-over-power conditions (TOP). For the I-series tests nominal power meant 550 W/cm at the peak power node for several minutes. In the CABRI test matrix A-type tests are pure TOP's and the B-type tests involve a LOF plus a TOP except for BI1, a pure LOF.

9. An important part of the CABRI cesium survivability story is revealed by the AI1 test since the pin did not fail. In effect, the 0.6 kJ/g energy injected during the TOP was a lead-in to the AI2 and AI3 tests (see next section) and the establishment of the 1% B.U. fuel pin failure threshold.

10. From the gamma scans one can deduce that most of the Cs concentrations survive a) nominal power at about 550 W/cm, b) a 0.6 kJ/g transient energy injection. So A-type test availability for Cs and B-type test availability up to B.O are indicated. One would like to establish that some Cs was released during this test, but it is not clear. However, if this result is coupled with the AI2 and AI3 experiments, we can establish a window in energy deposition threshold (0.6 - 1.3 kJ/g) when Cs distributions are drastically altered. Note the before and after Cs-137 profiles in Figs. 2 and 3. The microprobe data on the radial profile also demonstrated that even after the 0.6 kJ/g test, there was about a 75% Cs retention of the total formation, Cs was still in the fuel-including the melt region (but possibly depleted), Cs was in the fuel cladding gap, and Xe was completely released from the melt region.

The BI1 case

11. The BI1 test was a pure LOF with the scram about six seconds after B.O. During the test, the cladding melted from a large axial zone, and there were two minor fuel releases of 2 and 4g into the channel at 46 and 51 cm, respectively. It is interesting to report that the broad Cs peak at 51 cm in Fig. 4 is at the same location as the earlier and larger fuel ejection event. The Cs-137 inventory is again shown to be highly depleted in the final state in Fig. 4. Also, the Cs profile structure includes a cluster of peaks on either side of the 2-cm long interpellet gap at about 63 cm BFC. The Cs signals in the axial gap are non-zero (approx. 15%) and may be due to Cs deposited on the cladding (a measure of the effect) and/or Cs gamma-ray emanations from the exposed surfaces of the two pellets bounding the gap. This latter contribution is probably small since the Zr-95 profile shows such little signal in the gap (the collimation is evidently good). So a large fraction of the Cs is in the fuel.

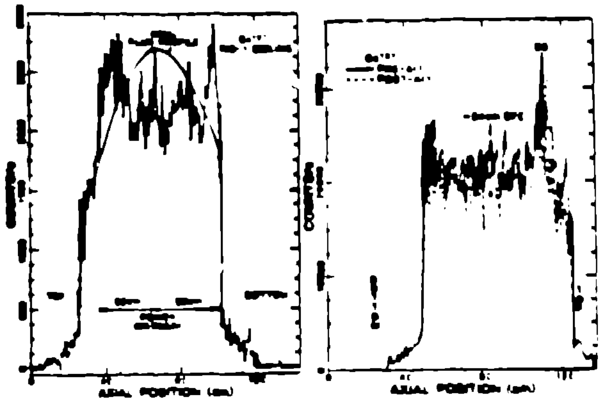

Fig. 2. Cesium-137 axial gamma scans for two 1% B.U. pins

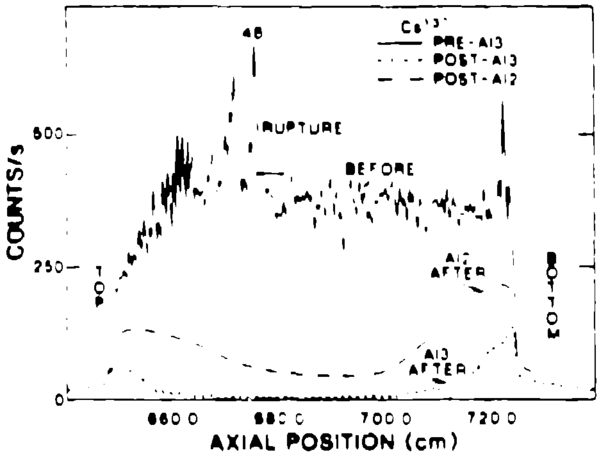

Fig 3. Cesium-137 axial gamma scans for pre-AI3, post-AI3, and post AI2 states

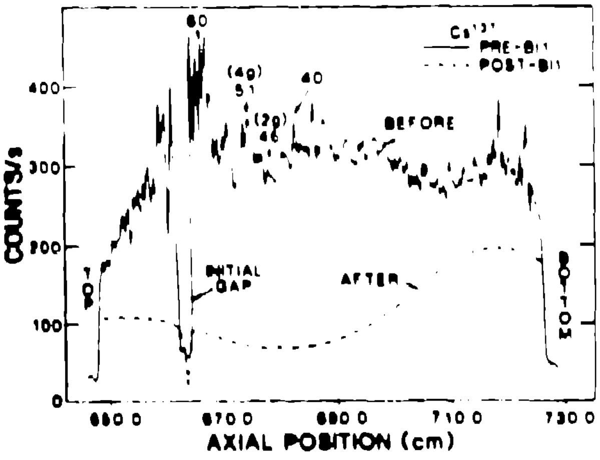

Fig. 4. Cesium-137 axial gamma scans for pre- and post-BI1 states

COMPARISONS OF CS^{137} GAMMA SCANS AND HODOSCOPE-OBSERVED PHENOMENA FOR 1% B.U. PINS (I-SERIES)

General Comments

12. The discussions in this section are concerned principally with the comparison of axial gamma scan data of the I-series (RIG-1, 1% B.U.) pins and the transient hodoscope data . The cesium distributions in the pre-CABRI state are approximately measured by the detection of the gamma rays emitted by the Cs-137 and Cs-134 radiosotopes. Since these two isotopes each have different precurseurs, they can have different distribution profiles from each other, or from the total. The fact of cesium migration during the preirradiation period from a high temperature region to lower temperature regions (axially and radially) is illustrated for the axial case in Fig. 2 for pin #34, one of the I-series siblings. The axial profile of Cs is quite different from the axial flux profile shown by the solid line. Although this particular Cs profile exhibits relatively symmetric migration (subject to interpretation of the large fluctuations in the data), many of the other profiles can be qualitatively characterized (in the author's opinion) as having a general flatness from BFC to midplane, concentration enhancements in the axial region from about 45-65 cm BFC with additional well-defined peaks often also evident in this zone, and a rapid decrease relative to the midplane values from 65-75 cm BFC. This latter region seems to match the flux profile, however.

13. If one proposes the hypothesis that these volatile fission products (such as Cs) can contribute to pin pressurization or fission product attack (weakening) of the cladding, the standard hodoscope fuel motion representation can be considered as an appropriate source for comparison (e.g. Fig. 5 for BI2). The evidence for cladding rupture or pin failure is the redistribution of the fuel at that location and a sudden signal increase. The fuel ejection and fuel motion in the channel also can be tracked via the hodoscope signals. The initial point of fuel pin failure often then becomes the "source" of the waves of fuel motion in space and time. Potential information on the magnitude, velocity, and time duration of fuel movements could be useful to modeling/identifying a previously uncalculated effect in the CABRI tests.

14. It should be remembered that the gamma-scan data only show the pre-and post-Cabri distributions of the Cs radioisotopes and the hodoscope only detects the fissioning fuel during the CABRI reactor operations (although ms time samples are possible in the TOP). Thus, we do not have a direct coupling of observations (such as the SANDIA absorption spectroscopy measurements during the transient.

The AI2 and AI3 Cases

15. For the AI2 test, I will note that the failure at 230 ms after trigger was unexpected (energy injection 0.9 kJ/g), and its axial location was 46 ± 3 cm BFC. Post-test exams indicated the fuel shell was intact except from

45-49 cm BFC. The Cs-137 gamma scan showed that there were clusters of concentration peaks at 7-8 cm and 60-63 cm BFC and a relatively broad peak/enhancement centered at 50 cm BFC. The latter's proximity to the failure location might allow participation in the fuel ejection as well as possible partial pressurizing of the pin.

16. In the AI3 test, the pin was reported to have failed 82 ms after TOP trigger (energy injected ∼ 0.85 Kj/g) and at 45 ± 3 cm. The Cs-137 concentration peaks are evident in Fig. 3 at 2, 48, and 52 cm BFC with a small cluster of peaks a little higher than the last. The peak at 48 cm is relatively narrow (∼ 6 mm) with an intensity about 70% greater than the mean value. This pre-test Cs peak position is suggestively close (within the above quoted errors) to the observed cladding rupture location. Fig. 3 also shows the post-AI2 Cs inventory is greater than that of AI3.

The BI2 case

17. The BI2 test involved a TOP triggered before boiling onset so we anticipate a mechanical cladding rupture which was reported at 51 ± 1.5 cm. BFC and 79 ms after TOP trigger. As seen in Fig. 5, the abrupt changes in the hodoscope signals are consistent with this report. We anticipate the Cs-137 concentration to be roughly the pre-CABRI state as shown in Fig. 6. The LECA sampling resolution limits the relative intensity information, but well-defined peaks at 5, 50, 55, 60 and 63 cm BFC are evident. Again within the spatial measurement error, there is a correlation between a Cs peak and the rupture location (as well as the fuel motion "source" location). The second event about 30 ms after rupture as labelled in Fig. 5 that causes fuel to move both upward and back towards the midplane is in the same axial zone as the Cs enhancements.

The BI3, BI4, and BI5 cases

18. The BI3, BI4, and BI5 tests involving fuel swelling, a second dispersive event, and simultaneous fuel signal changes at three axial locations, respectively, will only be addressed in Table 2 due to space constraints.

The BI6 case

19. The initial comparison between hodoscope observed phenomena and the Cs-137 axial gamma scans was performed on the last I-series test, BI6. In addition to an interest in the two simultaneous failure points, there also existed a complementary CABRI gamma scan which exhibited a well-defined concentration peak. Figure 7 shows the hodoscope fuel signal data as a function of time and axial position. Two abrupt signal changes at 63 ms and at 42 and 58 ± 2 cm from the (BFC) have been interpreted as evidence of fuel ejection into the channel. Although the LECA scan data were suggestive of an enhancement in concentration of Cs in the upper third of the fissile column, the corresponding CABRI gamma scan in Fig. 8 clearly shows a region with 20% enhancement from 45-63 cm BFC with an obvious peak at 49 cm BFC. This peak's height is about two times the average value and the peak width is only about a single pellet height. It appears that we have no spatial coincidence of either rupture location with this peak, but it was reported that the fuel column shifted abruptly in this region at the same time as the two ruptures occurred. Returning to Fig. 7, the sources of fuel motion as indicated by the origins of the arrows on the plot, can be seen to be in proximity to the Cs enhancement as noted on the vertical axis.

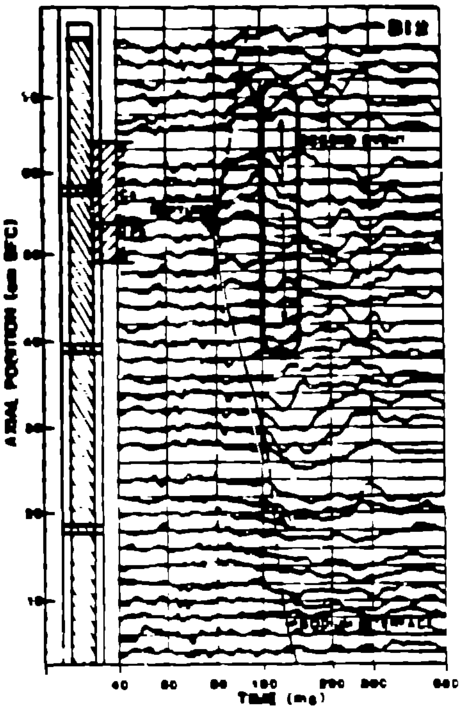

Fig. 5. Standard CABRI hodoscope data display for BI2

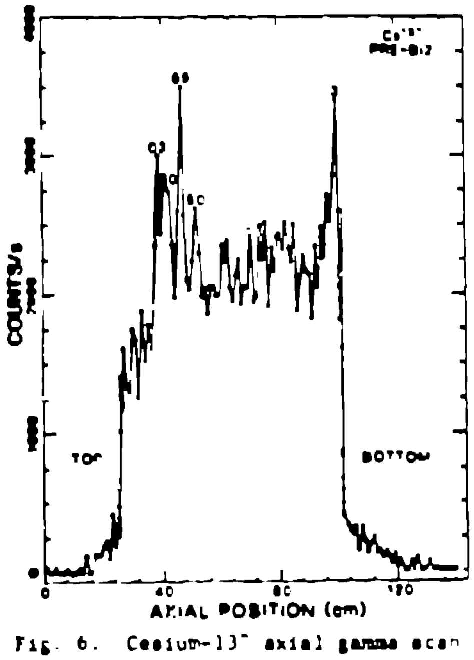

Fig. 6. Cesium-137 axial gamma scan for the pre-BI2 state

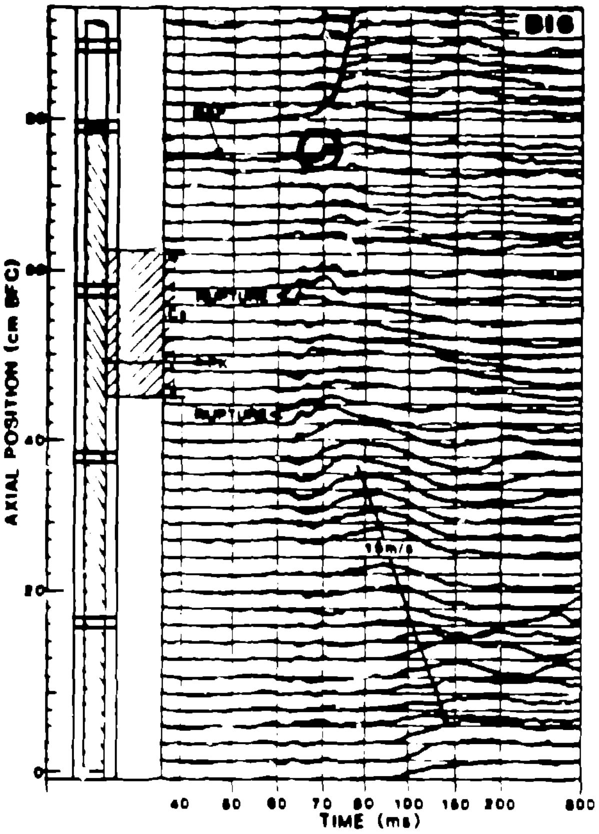

Fig. 7. Standard CABRI hodoscope data display for BI6

20. Table 2 summarizes the results of the I-series tests (RIG-1). The table includes the type of test, fuel ejection location, pre-test Cs distribution, Cs depletion, and a qualitative assessment of the spatial correlation. In seven out of the eight tests involving pin failure there appears to be an approximate spatial correlation between the Cs enhancement and initial fuel pin rupture location and/or sources of fuel motion. It is noted that AI2 and BI1 which both had marginal failures have only moderate Cs depletion. Since AI2 was a pure TOP and BI1 was a pure LOF, this could be an interesting test of modeling. Both the AI1, AI2, and AI3 and the BI1, BI3, and BI5 sets show progressive Cs depletion with increased TOP energy injection. The former involve pure TOP's and the latter were LOF-TOP's with the TOP triggered 5-6 seconds after coolant boiling onset (B.O.).

PROPOSED TESTS OF FISSION PRODUCT EFFECTS IN CABRI

21. The previous section's comparisons are consistent with the hypothesis that Cs related pressure may have been present in the RIG-1 tests, but the probabilities appear to be higher for participation in the fuel ejection and motion than for triggering the rupture. The observed depletion of the Cs inventory is qualitatively similar to that reported from the SANDIA tests[3]. In the absence of detailed information on the radial distribution of the Cs, the involved chemical forms, intragranular versus intergranular disposition, etc., the magnitude of the roles in CABRI are not easily determined. However, it is suggested that one can utilize the RIG-3 (5% B.U.) and RIG-1 (1% B.U.) tests to provide a cleaner in-pile test of Cs significance than previously anticipated.

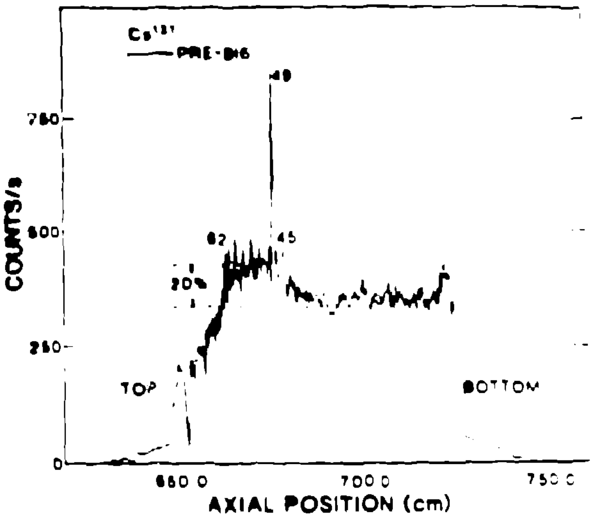

Fig. 8. Cesium-137 axial gamma scan for the pre-B16 state

22. Due to the unexpectedly low fission gas retention in the higher burn-up RIG-3 pins and the measured 4-5 times larger volatile fission product inventory in RIG-3 than RIG-1, one has a relative magnification of the VFP (Cs) potential contributions. Other investigators have suggested that the Cs-related pressures are comparable to those of fission gas in the SANDIA tests (FD4.2, FD.4.3)[2-3]. It is instructive to apply such an hypothesis to the CABRI tests with the AH3-AI3 case as an example.

Hypothesis: Cs-related pressures were comparable to those of fission gases in AI3.

Implies: AH3 should fail earlier (enthalpy-wise), at different axial location and/or exhibit different fuel motion than expected (more extensive in magnitude or duration than if one ignores Cs). Several assumptions are needed and the cleanness of the test of the hypothesis will depend strongly on the limits of validity of these proposed assumptions related to fission products, cladding and enthalpy.

23. These assumptions can generally be supported, but it should be noted that the effective gap conductance with the open hot gap for the AH3 pin is a complication. It may take a series of comparisons to resolve the issue. We can simplistically represent the hypothesis for fission gas and Cs concentrations as leading to three times the effective fission product for AH3 over AI3. This might correspond to a decrease in enthalpy threshold for failure of 100-200 J/g. It should be noted that the localized Cs peaks in the RIG-3 pin (H-series) could cause even higher pressures or a localized, specific attack. For example, destructive examination of pin 52 (the sibling)

Table 2: Summary of I-series(1%B.U.) Tests and Cs¹³⁷ Spatial correlation and depletion

TEST	Steady State Linear Power (W/cm)	Timing of LOF-TOP[a]	Energy Injected in TOP (kJ/g)[b]	Fuel Ejection Axial Location (cm BFC)	Pre-test Cs¹³⁷ Axial Distribution (cm BFC (Intensity))[c]	Approximate Spatial Correlation	Cs¹³⁷ Depletion
AI1	[illegible]0	-	0.6	-	50 (1.6)	-	weak
AI2	≈400	-	0.9	46±3(marginal) failure	45-55 (1.2)	yes	medium
AI3	≈400	-	1.3	45±3	50(1.5),61(1.7) 62(1.8)	yes	strong
BI1	≈630	B.O.+6.4s	-	46,51(marginal) failure	51(1.3),60(1.6)	yes	medium
BI2	≈630	15.5s	1.3	51±2	3(1.5),50(1.2),55(1.5) 60(1.3),63(1.3)	yes	strong
BI3	≈630	B.O.+5.1s	1.1	swelling 45-54	40-57(1.2) 64(2.2)	yes	strong
BI4	≈630	B.O.+1.5s	1.3	52±2	9(2.0),63(1.2)	no	strong
BI5	≈620	B.O.+5.0s	2.1	13-20,30-35 50-70	1(1.3),14(1.1),17(1.1) 30(1.1),48(1.2),50(1.5)	yes	strong
BI6	≈620	B.O.+2.4s	1.9	42±2 58±2	45-63(1.2),49(2.5)	yes	strong

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[a] The Loss-of-Flow follows the reduction law of $Q(t) = Q_o(1 + t/\tau)^{-1}$ with τ = 7 seconds. Typically boiling onset (B.O.) occurs 21-22 seconds after LOF initiation. Initiation of TOP is given relative to LOF initiation or B.O. BI1 was a pure LOF.

[b] The energy injected into the peak power node 190ms into the transient is given. For AI1, the time was 120ms.

[c] The general axial distribution for an I-series pin applies. Specific points are given in this column such as localised axial concentrations with the relative intensity given in parentheses.

at the axial location of the pronounced Cs peak at approximately 210 cm BFC, identified a distinct change in the fuel appearance at two azimuthal positions toward the periphery of the fuel. A cesium compound is indicated, but its effect is not determined.

Preliminary AH3 results

24. A summary of the preliminary results for the AH3 test compared to AI3 can be given as follows:

a) the AH3 failure location was higher, 54 cm BFC versus 45 cm BFC for AI3 as determined from the hodoscope data.

b) the first event in the coolant channel was more violent than in AI3 as deduced from pressure and flow rate changes.

c) the fuel dispersion from the 20-cm axial section around the failure point was faster in AH3 than in AI3 as determined from the hodoscope data.

d) AH3 voided earlier at the 10-15 cm BFC axial location than AI3 as determined from the hodoscope data. This is a region of Cs peak (Fig. 1) and fission gas concentration.

e) The fractional depletion of the Cs in AH3 was qualitatively similar as seen in the gamma scans (see Fig. 3 and Fig. 9), but of course AH3 had 5 times the initial inventory.

f) The enthalpy in AH3 at pin rupture time needs to be determined carefully (the effective gap conductance for AH3 is the main complication). Unexpected clad swelling during the irradiation may be related to AH3's lowered failure enthalpy.[4]

Preliminary BH1 results

25. In addition to the AH3 test the BH1 test was performed prior to the author's departure from Cadarache. BH1 did exhibit temperature oscillations during the LOF. Also, hodoscope signals implied a 2-3 cm upward displacement of the upper portion of the fissile column which created a gap at 45 cm BFC. This gap shifted downward as clusters of pellets moved upward. About seven seconds after B.O. fuel pin break-up was indicated at 40-55 cm BFC. The axial

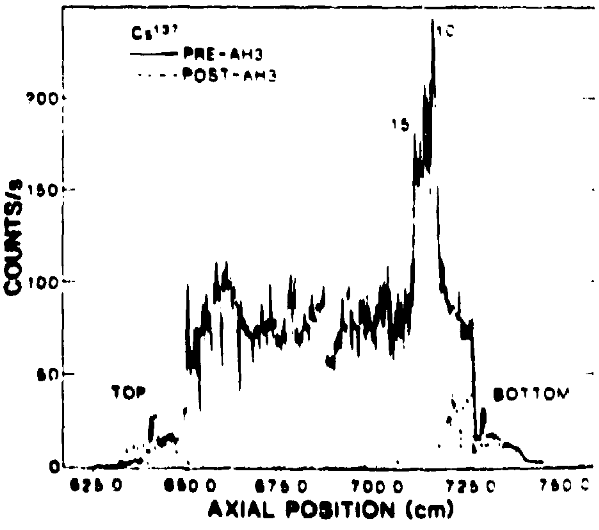

Fig. 9. Cesium-137 axial gamma scan for the pre-and post-AH3 states

extent of pin disruption and the Cs depletion are both larger than in the BI1 case. Further analysis is needed to elucidate these differences.

SUMMARY AND CONCLUSIONS

26. Some of the results of this initial study are as follows:

1. For the 1% and 5% fuel the axial profile of the Cs-137 concentration exhibits a migration away from the power maximum. For the 1% B.U. fuel this results in a final distribution with a general flatness from the bottom of the fissile column (BFC) to midplane, concentration enhancements in the axial region from about 45-65 cm BFC with additional well-defined peaks often evident in this zone, and a rapid decrease relative to the midplane values from 65-75 cm BFC (this latter region seems to match the flux profile, however). For the 5% B.U. fuel the localized Cs peaks were generally observed from 10-30 cm BFC.

2. An approximate spatial correlation was noted between the pre-CABRI Cs-137 axial concentrations deduced from the gamma scans and the hodoscope detected fuel motion sources for the I-series tests. More specifically:

a) In the eight cases involving pin failure and fuel ejection, all except one have Cs-137 concentration enhancements at/near the point of failure.

b) In all eight cases of pin failure, the Cs-137 distribution after the test indicates a notable depletion in the Cs inventory, including intact fuel shell regions. Three tests with transient energy injections in the peak power node at 190 ms of 0.6, 1.0, and 1.3 kJ/g, respectively, display the onset of significant Cs depletion. The rate and timing of Cs release are open issues.

3. In addition, a direct comparison of events in a pure transient-over-power test and a pure loss-of-flow test for the 1% and 5% B.U. cases indicated some differences in fuel motion that should be examined in more detail via modeling and destructive post-test examinations.

4. Other tests in the CABRI matrix should extend our data base for Cs-effect evaluation. Tests with an intermediate ramp rate in the H-series pins or with the newer set of pins with high fission gas retention should be useful.

27. Finally it is noted that the magnitude of volatile fission product (Cs) roles are not easily determined since the CABRI program does not monitor the Cs release during the fast transients, the SANDIA experiments (at slower ramp rates) have not quantified their transient observations of Cs release, and detailed modeling and some fundamental experiments have not yet been performed. However, the observed survivability of Cs concentrations in BI1 and AI1, the increased Cs release with increasing energy injection (AI1, AI2, and AI3; BI1, BI3, and BI5), and the approximate spatial correlation between pre-test Cs enhancements and fuel motion events are all consistent with Cs involvement in these tests. The actions of fission gas and Cs may be synergistic at a minimum, with the relative importance depending on specific circumstances. It is hoped that this preliminary study will contribute to the framework for further investigations. A wealth of information will become available in the next few years which should clarify some of these issues.

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