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R. A. Karnesky, J. W. Jost and I. Z. Stone

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## CESIUM MIGRATION IN LMFBR FUEL PINS

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### INTRODUCTION

The factors affecting the axial migration of cesium in mixed oxide fuel pins and the effects of cesium migration on fuel pin performance are examined in this paper. The development and application of a correlated model which will predict the occurrence of cesium migration in a mixed oxide (75 w/o  $UO_2$  + 25 w/o  $PuO_2$ ) fuel pins over a wide range of fabrication and irradiation conditions are described.

### CESIUM MIGRATION PHENOMENA

The fission product cesium has been extensively studied in mixed uranium-plutonium oxide fuels because of its mobility within the fuel column and its influence on fuel pin behavior. Cesium has been determined to be one of the primary fission products associated with cladding corrosion.<sup>1,2</sup> High localized cesium concentration, resulting from axial migration of cesium have been found to be associated with several potentially deleterious phenomena. These phenomena include: areas of localized cladding strain,<sup>3,4,5</sup> fragmentation of insulator pellets,<sup>3,4</sup> restriction of central void,<sup>3,4</sup> plutonium in the outer regions of the fuel,<sup>9</sup> and reduced permeability of the insulator region to fission gas.<sup>6</sup>

### Factors Influencing Cesium Migration

In fuel pins similar in design and operating under conditions similar to the Fast Flux Test Facility driver fuel,<sup>7</sup> cesium migrates rapidly to the fuel cladding gap, where it is relatively stable. At burnups greater than ~50 MWd/kg cesium is typically observed to migrate axially and form localized concentration peaks.

Postirradiation examination of over 1000 mixed-oxide fuel pins irradiated in EBR-II has established that the burnup at which the axial migration of cesium first occurs, and the amount of cesium that migrates are largely influenced by two factors, cladding inner surface temperature and fuel oxygen-to-metal ratio. Fuel pins operating with a high cladding inner surface temperature will exhibit axial cesium migration at a lower burnup

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than pins having the same fuel smeared density, cladding size, and fuel oxygen-to-metal ratio operating at a low cladding temperature. Pins containing fuel with low oxygen-to-metal ratios exhibit axial cesium migration at the low burnups. It has also been observed that the cesium concentration in the fuel column tends to decrease with increasing cladding inner surface temperature, except for localized peaks.

Effects of Local Cesium Concentrations

Localized cesium concentration peaks, formed as a result of the axial migration of cesium, occur predominantly at the ends of the fuel column. They usually occur in the first 6 mm of the UO<sub>2</sub> insulator and are often associated with areas of localized cladding strain. Cesium peaks can also occur within the fuel column and result in areas of localized cladding strain. Figure 1 shows a typical example of cesium-induced localized cladding strain in both the fuel and insulator regions. The fuel pin, PNL-8-24, had been irradiated in EBR-II to a burnup of ~9 at.%.

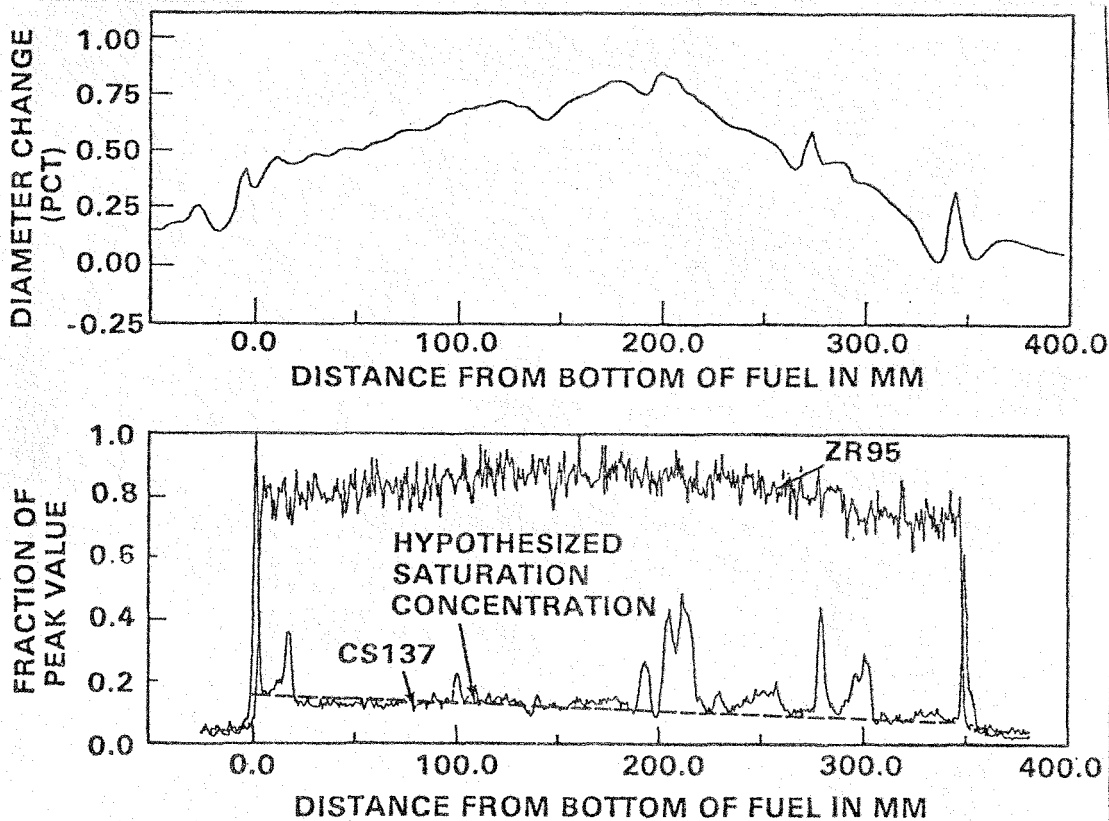


Figure 1. Profilometry and Gamma Scan Data from PNL-8-24.

The magnitude of the cesium peak and the amount of cladding strain observed are usually not proportional. The amount of cladding strain is dependent on many factors, e.g., insulator-to-cladding (or fuel-to-cladding) gap size, pellet density, cladding temperature, fuel temperature, and cladding strength. These areas of cladding strain are believed to be the result of mechanical interaction as a consequence of fission product cesium reacting with uranium-plutonium to form a low density cesium-uranate complex.

## MODEL DEVELOPMENT

Prior analytical efforts have been primarily concerned with describing the qualitative radial and axial distributions of cesium, and evaluating the effects of localized cesium concentration peaks on fuel pin performance. Recent efforts at the Hanford Engineering Development Laboratory (HEDL) have been directed at developing a model that will predict, quantitatively, the occurrence of axial cesium migration in mixed oxide fuel pins similar in design and operating under conditions similar to the FFTF driver fuel.

### Hypothesis

Numerous observations of axial cesium migration led to the formation of a hypothesis concerning the conditions necessary for axial cesium migration. It was hypothesized that an upper limit exists to the concentration of stable (nonmigrating) cesium in the fuel cladding gap. The gamma scan data in Figure 1 show an example of the hypothesized cesium saturation concentration. The localized cesium peaks are seen to be superimposed on the saturation concentration. This saturation limit is controlled by the cladding inner surface temperature and fuel oxygen potential. As the concentration of cesium in the fuel cladding gap increases with increasing burnup no axial migration will occur until this saturation limit is exceeded. After the saturation limit is exceeded, the excess cesium in the fuel cladding gap migrates axially to local sinks. The axial migration of cesium to these localized sinks results in local concentration peaks, which are superimposed on the hypothesized saturation concentration. The occurrence of localized cesium peaks is not treated in the current modeling effort.

### The Data Base

In developing an empirically derived model to describe the hypothesized cesium saturation limit, a set of 33 fuel pins was selected. These pins ranged in peak burnup from ~2 at.% to ~9 at.% and covered a wide range of fabrication and irradiation conditions. Gamma scan data from these pins were quantified and statistically analyzed. Quantification was performed by calculating the total cesium generated in the pin and equating this with the integrated "Compton corrected" Cesium-137 gamma activity profile (area under Cesium-137 gamma scan profile). Total cesium was

determined based on the EBR-II run history and the RIBD<sup>10,11</sup> fission product inventory code. By assuming that all the cesium was distributed in proportion to the axial Cesium-137 gamma activity profile and that all the cesium was deposited on the cladding inner surface, concentration profiles were determined for all 33 pins. Each pin was then divided into 30 segments with those segments containing large localized cesium concentration peaks deleted from the data set. Other parameters included in the analysis were: local cladding inner surface temperature, local burnup, fuel smeared density, cladding inner radius, fuel oxygen-to-metal ratio, fuel pin linear power, and fuel column weight.

Visual inspection of these data (Figure 2) showed that the relationships between the cesium concentration on the cladding inner surface and burnup was markedly different, at burnups greater than ~6 at.%. At low burnups the cesium concentration appeared to be insensitive to changes in temperature and oxygen-to-metal ratio. At high burnups the cesium concentration was found to be relatively insensitive to burnup as would be the case should the hypothesized saturation limit exist.

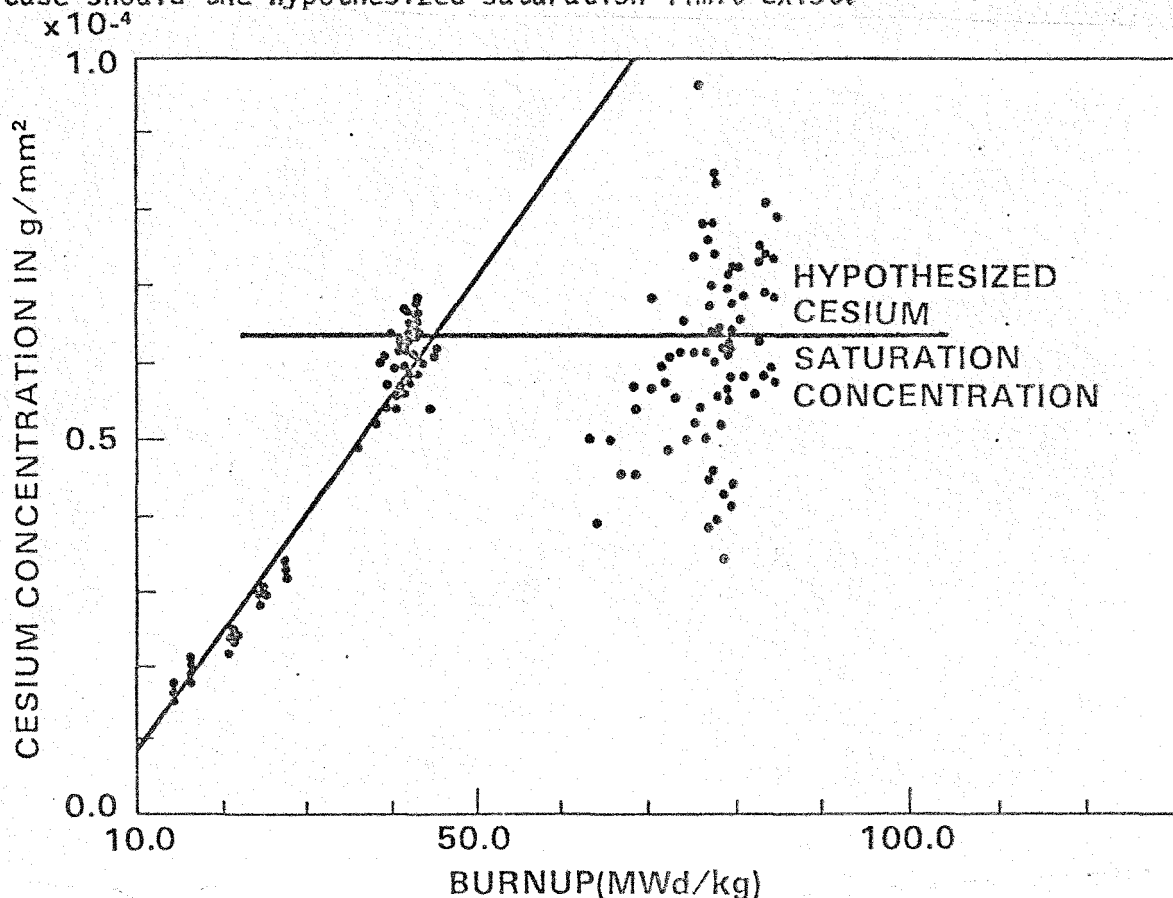


Figure 2. Cesium Concentration on Cladding Inner Surface in the Temperature Range of 425°C to 525°C.

## The Correlation

These data were statistically analyzed using a multiple linear regression technique as outlined by Draper.<sup>12</sup> Both backwards and forwards stepwise procedures were used to eliminate nonsignificant parameters. The entire data set was analyzed using these procedures, and the major terms [cladding inner surface temperature (T), oxygen-to-metal ratio (O/M), and local burnup (B)] were identified. Three interaction terms (O/M x T, O/M x B and B x T) seemed to support the hypothesis that two different processes were controlling the cesium concentration. Based on these observations the data were divided into two burnup groups for analysis.

Analysis of the low burnup data showed burnup and linear fuel weight to be the most significant parameters influencing cesium concentrations on the cladding inner surface. The following equation was derived to describe the cesium concentrations on the cladding inner surface.

$$Y_1 = \frac{0.1314 \times B \times W}{2\pi R} \quad (1)$$

where:

- $Y_1$  = The cesium concentration on the cladding inner surface (without axial migration) (g/mm<sup>2</sup>)
- B = Local burnup (MWd/kg)
- W = Linear fuel weight (total heavy metal divided by fuel column length) (kg/mm)
- R = Cladding inner radius (mm)

This equation describes the accumulation of cesium on the cladding surface as a function of burnup, pin geometry and fuel linear density, with the correlated constant thought to be the cesium yield (g/MWd). Examination of the data used to derive this equation showed that virtually no axial cesium migration had occurred in the low burnup pins. This equation, therefore, predicts the concentration of cesium in the fuel-to-cladding gap in the absence of axial cesium migration.

The high burnup data showed little dependence on burnup. A strong correlation was found with temperature and the oxygen-to-metal ratio. The following equation was derived for the cesium concentration on the cladding inner surface from these data.

$$Y_2 = -1.2473 \times 10^{-3} + 7.3666 \times 10^{-4} (O/M) - 2.620 \times 10^{-7}(T) \quad (2)$$

where:

- $Y_2$  = Cesium saturation concentration (g/mm<sup>2</sup>)
- O/M = Fuel oxygen-to-metal ratio
- T = Cladding inner surface temperature (°C)

Examination of the data base used for the high burnup correlation showed that axial cesium migration had occurred in all pins. The correlated equation showed no burnup dependence, indicating that Equation 2 describes the hypothesized saturation concentration, and confirms its existence. The equation shows that the concentration of stable cesium increased with increasing oxygen-to-metal ratios and decreased with increasing temperatures, (Figure 3).

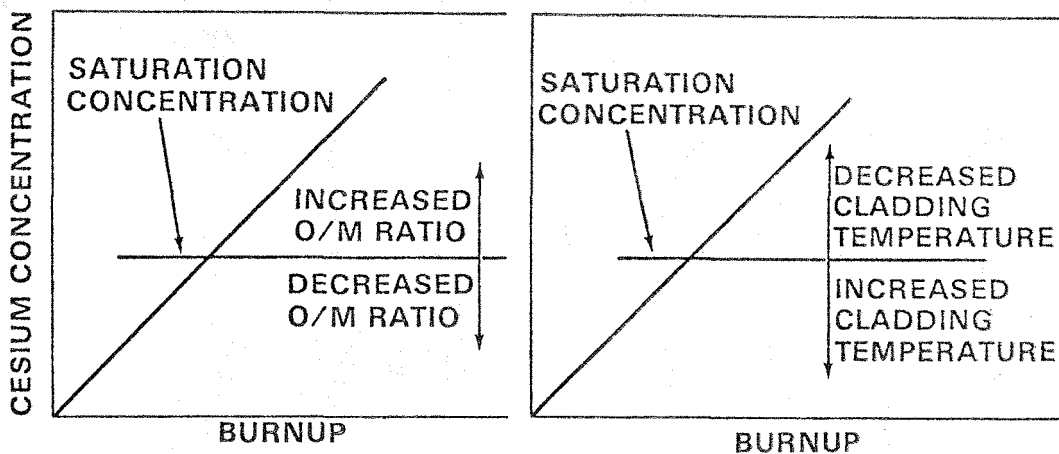


Figure 3. Effect of Cladding Inner Surface Temperature and Oxygen-to-Metal Ratio on Cesium Saturation Concentration.

Both of these predictions are consistent with earlier observations. Equations (1) and (2) can be used to predict the actual concentration of cesium on the cladding inner surface given the fabrication parameters and fuel pin operating conditions. By equating (1) and (2) and solving for the burnup, an equation can be derived that will predict the burnup at which cesium saturation will occur in a given fuel pin segment.

$$B_s = \frac{2\pi R [-1.2473 \times 10^{-3} + 7.3666 \times 10^{-4} (O/M - 2.2620 \times 10^{-7} (T))]}{0.1314 W} \quad (3)$$

where:  $B_s$  = burnup at which saturation occurs (MWd/kg).

In addition to predicting when axial cesium migration will begin (Figure 4), this equation can be used to determine whether Equation (1) or Equation (2) should be used in predicting actual cesium concentrations.

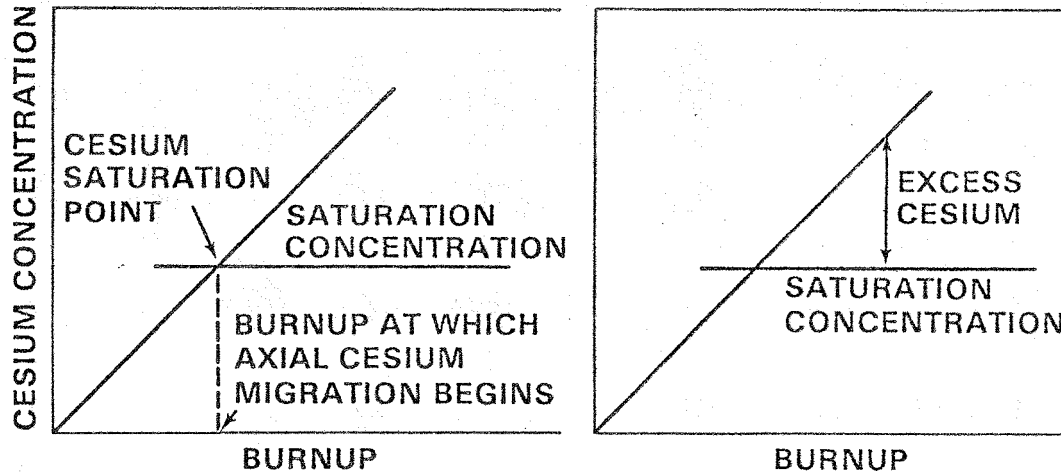


Figure 4. Calculation of Cesium Saturation Point and Amount of Excess Cesium for a Given Fuel Pin Segment.

If the local burnup is less than the saturation burnup ( $B_s$ ) then Equation (1) is used to predict the cesium concentration. If the local burnup is greater than  $B_s$ , Equation (2) should be used to predict the saturation concentration. The amount of cesium that will tend to migrate out of a given fuel pin segment (Figure 4) can be estimated by taking the difference between Equation (1) and Equation (2), and multiplying by the cladding surface area in a given fuel pin segment.

$$C_y = (Y_1 - Y_2) 2\pi RL \quad (4)$$

where:

$C_s$  = grams of excess cesium (or, if negative, capacity to absorb excess cesium) in a segment of length  $L$  (mm)

If the value is negative it is the number of grams of excess cesium a given segment can absorb. If  $C_s$  is positive it is the number of grams of cesium that will tend to migrate from that segment.

These techniques have been successfully used to predict cesium concentrations in irradiated fuel pins.

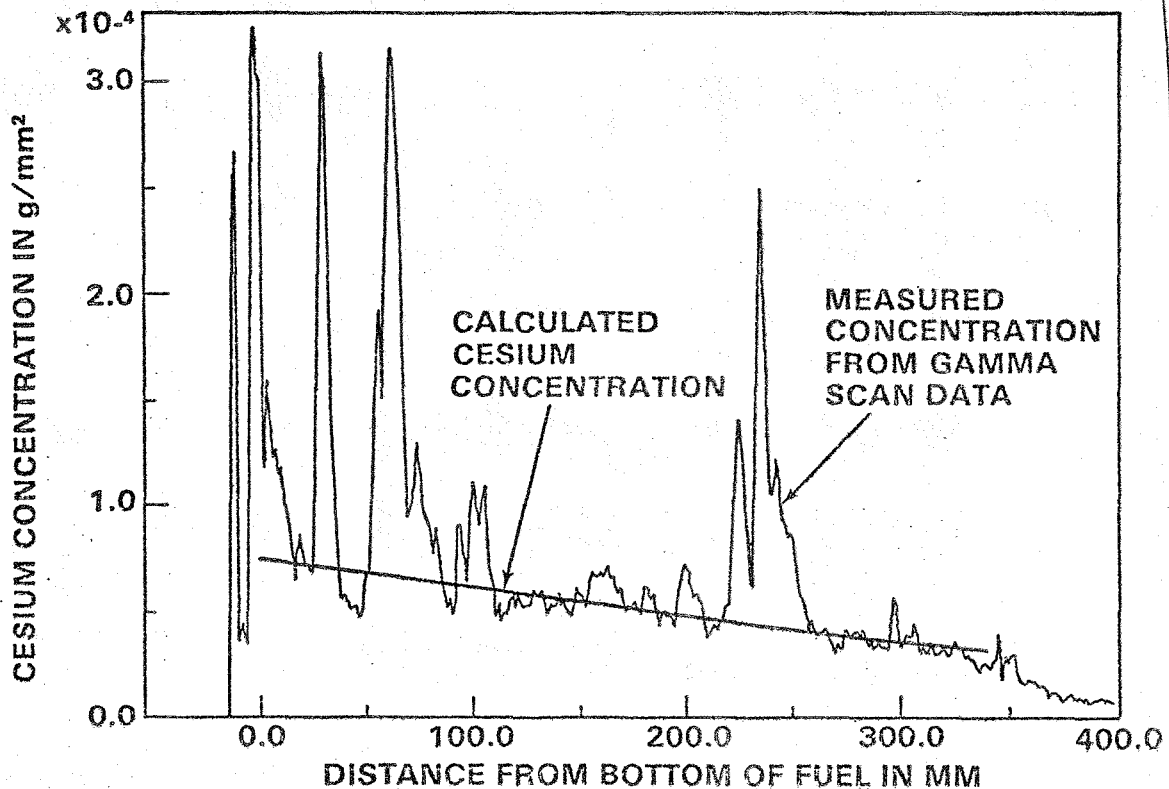


Figure 5. Comparison of Calculated and Measured Cesium Concentrations in P-14-52.

Figure 5 shows a comparison of the predicted and measured cesium concentrations for a fuel pin (P-14-52) irradiated in EBR-II to a burnup of  $\sim 80$  MWd/kg. Localized peaks are seen to be superimposed on the predicted

saturation concentration. Comparison of predicted and measured cesium concentrations for the entire data base (Figure 6) has shown good agreement throughout the data set. As would be expected the high burnup data shows a slightly larger scatter than the low burnup data.

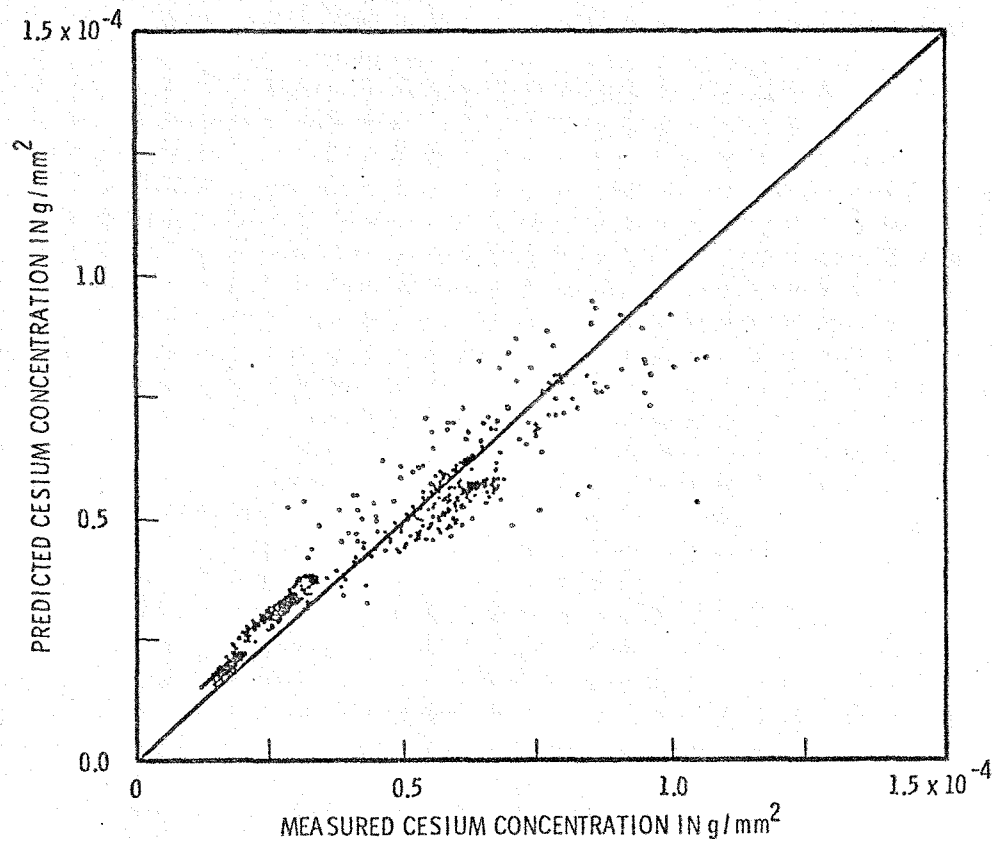


Figure 6. Comparison of Measured and Predicted Cesium Concentrations for the Data Set Used in These Correlations.

#### CONCLUSIONS

A model has been developed that will describe the concentrations of stable cesium in the fuel cladding gap over a wide range of burnup and operating conditions. This correlated model can be used to predict when cesium migration will occur and the amount of cesium that will migrate, thereby, providing a basis for evaluating the effects of cesium migration on fuel pin performance.

#### ACKNOWLEDGEMENTS

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