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EFFECTS OF STOICHIOMETRY ON CLADDING
REACTION IN MIXED-OXIDE FUEL

L. A. Lawrence

Initial fuel stoichiometry (oxygen-to-metal ratio, O/M) has been established as one of the major parameters influencing the character and extent of fuel-cladding chemical interaction (FCCI) in UO_2-25 wt% PuO_2 fuels clad with 20% cold worked type 316 stainless steel.[1,2,3] This afternoon I will discuss briefly how the character of the interaction changes with initial O/M and the O/M dependence that has been established for the depth of interaction. The data base for this analysis consists of 134 data sets from P-23 series fuel pins irradiated in EBR-II. The first slide (SLIDE 1) summarizes the fabrication and irradiation parameters for the P-23 series fuel pins. As-fabricated O/M's ranged from 1.938 to 1.984 with the bulk of the data in the range 1.96 to 1.98. Fuel pins operated to 9.5 atom %, with local cladding inner surface temperatures to 725°C, for exposure times to 415 equivalent full power days at heat rates of ~400 W/cm.

The nature of the FCCI was characterized through metallographic examinations of samples removed from irradiated fuel pins at numerous axial locations. The FCCI was classified as predominately matrix, evolved matrix, intergranular, or combined matrix and intergranular interaction.[1] Examples of the four types of interaction are shown in the next slide (SLIDE 2). In matrix interaction, the attack is uniform with no strong preference for attack along the grain boundaries. In evolved matrix interaction there is a definite segregation of the cladding constituents in the reaction product layer. For intergranular interaction, attack is predominantly along the grain boundaries. Intergranular and matrix type interaction sometimes occur simultaneously with the intergranular attack preceeding the matrix interaction in the reaction zone. This form of interaction is designated as combined interaction. Results of the characterization of the interaction are summarized in the next slide (SLIDE 3).

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In the case of the high O/M fuel pins, cladding interaction occurs above ~500°C. For cladding temperatures greater than 675°C intergranular attack is predominant at the lower burnups. The interaction changes from intergranular to an evolved matrix type interaction as burnup increases. Examples of the interaction at three burnup levels for an O/M = 1.984 are shown in the next slide (SLIDE 4). The intergranular form of interaction has changed to an evolved matrix type interaction at the moderate burnup of 4.7 at.% for cladding inner surface temperatures of approximately 715°C.

For moderate O/M fuel pins (SLIDE 5) the threshold temperature for the interaction was somewhat higher and the interaction was generally matrix except at the higher burnup for the higher temperatures where the interaction is predominantly combined. For the moderate burnup range the number of samples examined at cladding temperatures ≥650°C were too limited to establish whether combined or matrix type interaction was the dominant form of interaction. Examples of the interaction at 3 burnup levels for an O/M = 1.966 are shown in the next slide (SLIDE 6). In this O/M and temperature range matrix type interaction is the predominant form of interaction for burnups up to 9.5 at.%. The depth of interaction is increasing with burnup but the type of interaction is not changing. It should be noted that an O/M = 1.97 is generally considered to be the upper end of most current fuel O/M specifications.

For the lowest O/M range, no interaction was observed for temperatures to ~700°C for burnups to ~3 at.% (SLIDE 7). As burnup increased, a shallow intergranular form of interaction was observed at lower cladding temperatures. Examples of the interaction at 3 burnup levels for an O/M = 1.948 are shown in the next slide (SLIDE 8). In the case of low burnup the etched cladding shows a very slight reaction at the cladding surface on the order of 2-3 microns in depth. A shallow intergranular type of interaction, to a depth of 10 microns to 12 microns, was the predominant form of interaction at a moderate burnup of 6.7 at.% for cladding temperatures of 650°C. As burnup progress the types of interaction changes from shallow intergranular to a combination of shallow intergranular and matrix.

The significant influence of O/M on the character and extent of interaction is illustrated in the next slide (SLIDE 9) for O/M's = 1.984 (high) and 1.966 (moderate) at a peak burnup of ∿9 at.% for a peak cladding temperature of ∿675°C. In the case of the higher O/M fuel the evolved matrix type interaction characterized by the metallic reaction product layer is very uniform on the circumference of the sample and the maximum cladding thickness loss due to FCCI is 65 microns. This compares to a somewhat intermittent matrix interaction on the circumference of the sample at the lower O/M. The maximum depth of interaction for the lower O/M fuel was 33 microns which is half that of the higher O/M fuel.

The depth of interaction was obtained using total cladding thickness measurement techniques. Details of the measurement and correlation, which I do not plan to cover in this presentation, can be found in the summary and the paper presented at the recent Monterey Conference on Fast Breeder Reactor Fuel Performance.(1) The next slide (SLIDE 10) summarizes the measurements of maximum depth of interaction. Results were grouped by O/M ranges to illustrate the effects of O/M on depth of interaction, expressed as a rate, to place all the data on the same relative plot.

Several models (SLIDE 11) were selected for the O/M dependence and each was fit to the data using standard nonlinear statistical techniques. A combination of O/M and burnup was also selected for the correlation reflecting the increase in fuel O/M that accompanies an increase in fuel burnup. A linear dependence was found to fit the data best.

The correlation of depth of interaction (SLIDE 10) predicts no cladding interaction for an O/M $\leq$1.942. The other terms in the equation such as the development of the factor K for the confidence levels for maximum depth of attack are discussed briefly in the summary for the paper and were discussed in detail at the Monterey Conference.(1)

The measured and predicted depths of interaction are compared in the next slide (SLIDE 13) for the different O/M's in the data base. The differences between the measured and predicted depths of interaction are rather evenly distributed about zero indicating that the simple linear dependence is a good statistical approximation for the data base. This summary plot also illustrates the number of data sets for any given O/M in the data base. The largest number of data sets occur for the higher O/M's (O/M = 1.97 and greater).

The depth of interaction was calculated for a typical breeder reactor fuel pin operating at a peak cladding inner surface temperature of 700°C to a peak burnup of 8 atom % (SLIDE 14). Axial burnup and cladding temperature profiles are shown in the upper curves. The maximum depth of interaction is ∿50 microns at the 95% confidence level (K = 12.23) for an O/M = 1.97, considered to be the upper end of most fuel specifications. The maximum depth of attack occurs approximately 3/4 of the way up the fuel column where a combination of burnup (or heat rate) and cladding temperature produces the most severe conditions. For an initial O/M = 1.95 the maximum depth of interaction is in the range of 10 microns to 15 microns, which is a factor of ∿4 reduction in depth of interaction compared to an O/M = 1.97.

In summary, (SLIDE 15) substantial reductions in the extent of interaction have been established for corresponding reductions in initial fuel O/M. Similarly, significant changes in character and temperature for initiation of FCCI were observed to accompany reductions in O/M.

REFERENCES

1. L. A. Lawrence, J. W. Weber, J. L. Devary, "Fuel-Cladding Chemical Interaction in Mixed-Oxide Fuels," Proceedings of the International Conference on Fast Breeder Reactor Fuel Performance, Monterey, CA, March 5-8, 1979, pp. 432-444.

2. L. A. Lawrence, J. W. Weber, J. L. Devary, "Effects of Stoichiometry on Cladding Reaction in Mixed-Oxide Fuel at High Burnup," Trans. Am. Nucl. Soc., 28, p. 210 (1978).

3. L. A. Lawrence, D. C. Hata, J. W. Weber, "Effects of Stoichiometry on Cladding Attack in UO_2-PuO_2," Nuclear Technology, pp. 195-206, February, 1979.

FABRICATION AND IRRADIATION PARAMETERS

FUEL 75 WT% UO_2-25 WT% PuO_2
U ENRICHMENT: 65 WT%

O/M = 1.938 TO 1.984
SMEARED DENSITY = 85%

CLADDING TYPE 316 - 20% COLD WORKED STAINLESS STEEL
5.84 MM (0.230 INCH) OD x 5.08 MM (0.200 INCH) ID

IRRADIATION REACTOR: EBR-II
PEAK BURNUP: TO 9.5 ATOM %
LOCAL CLADDING INNER SURFACE TEMPERATURE: TO 725°C
PEAK HEAT RATES: ~400 W/CM

RESIDENCE TIME: TO 415 EFPD*

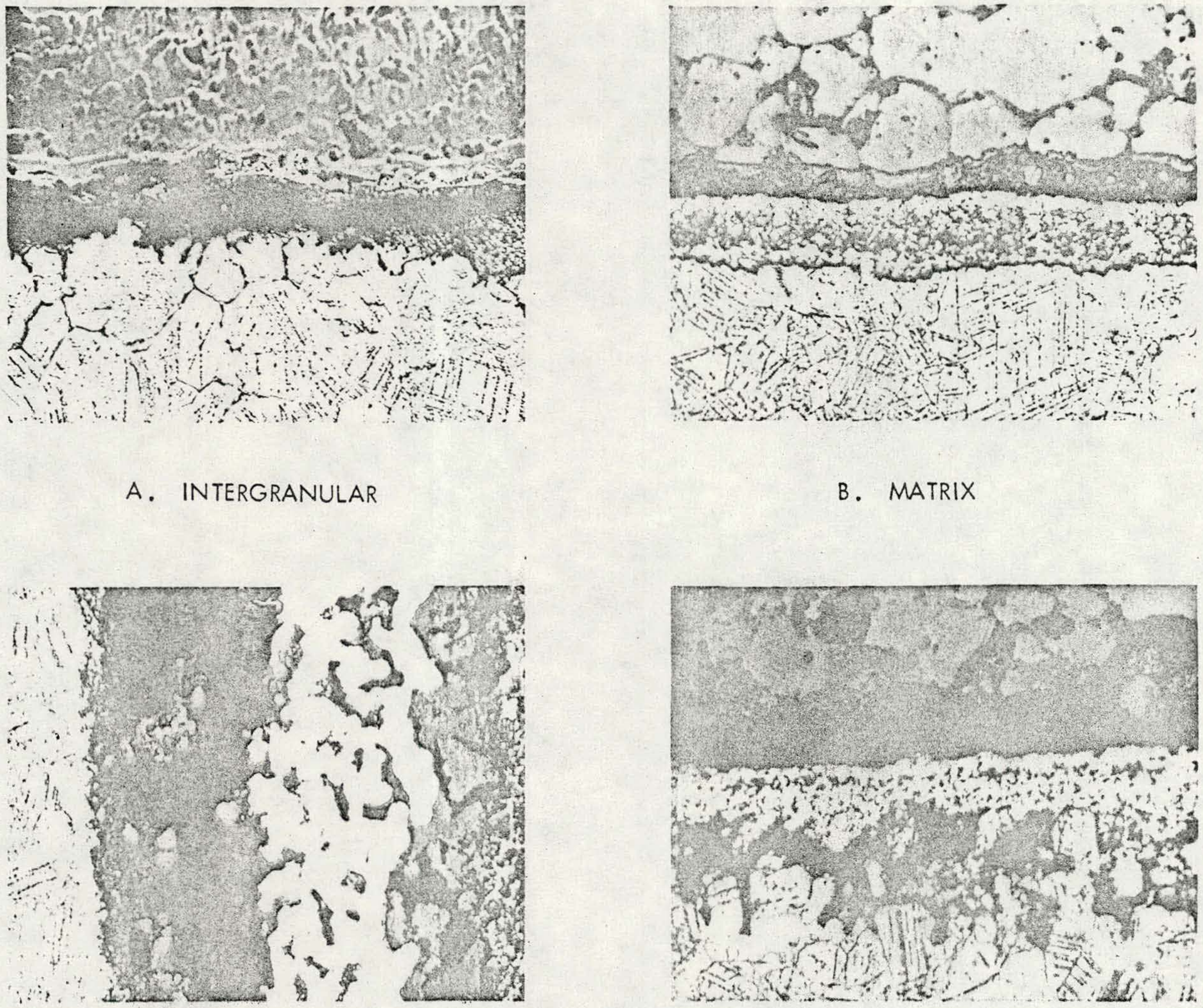
*EQUIVALENT FULL POWER DAYS

(SLIDE 1)

TYPES OF INTERACTION

FUEL

ETCHED CLADDING



A. INTERGRANULAR

B. MATRIX

C. EVOLVED MATRIX

D. COMBINED

25 MICRONS

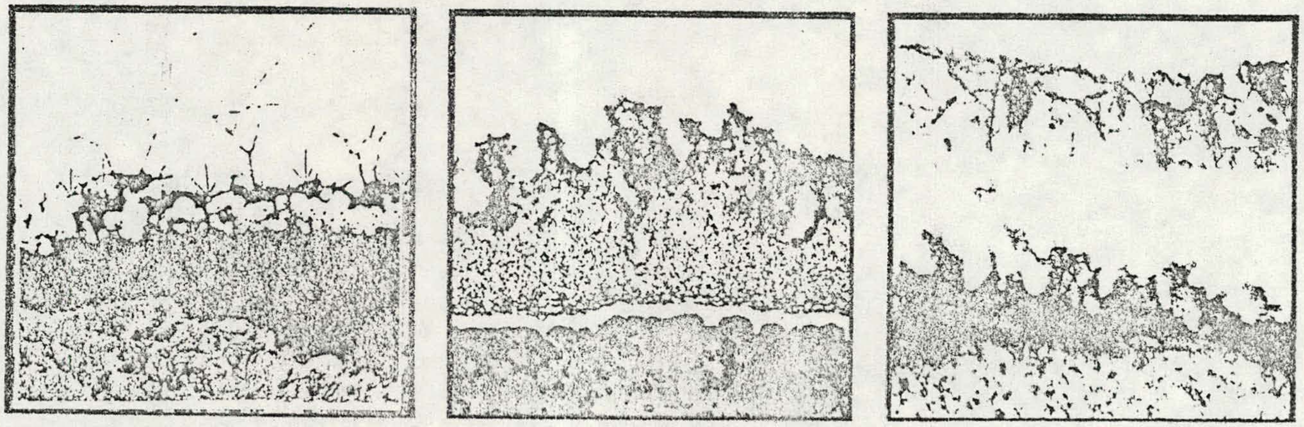
CHARACTERIZATION OF INTERACTION

O/M	BURNUP		
	LOW (0-3 at.%)	MODERATE (3-6 at.%)	HIGH (6-10 at.%)
HIGH (1.98-1.99)	>675°C** INTERGRANULAR <675°C MATRIX <500°C NO FCCI 7 PINS *	>675°C EVOLVED MATRIX <675°C MATRIX <500°C NO FCCI 2 PINS	>500°C EVOLVED MATRIX <500°C NO FCCI 4 PINS
MODERATE (1.96-1.97)	> 550°C MATRIX < 550°C NO FCCI 10 PINS	> 550°C MATRIX < 550°C NO FCCI 3 PINS	> 650°C COMBINED > 525°C MATRIX < 525°C NO FCCI 4 PINS
LOW (1.94-1.95)	>700°C SHALLOW MATRIX <700°C NO FCCI 3 PINS	>625°C SHALLOW INTERGRANULAR <625°C NO FCCI 4 PINS	>600°C SHALLOW INTERGRANULAR AND MATRIX <600°C NO FCCI 4 PINS

* NUMBER OF FUEL PINS EXAMINED WITH INDICATED O/M's AND BURNUPS

** CLADDING INNER SURFACE TEMPERATURE

EFFECTS OF BURNUP ON CHARACTER OF FCCI, O/M=1.984



CLADDING INNER SURFACE TEMPERATURE ≃ 715°C

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(SLIDE 4)

CHARACTERIZATION OF INTERACTION

O/M	BURNUP		
	LOW (0-3 at.%)	MODERATE (3-6 at.%)	HIGH (6-10 at.%)
HIGH (1.98-1.99)	>675°C** INTERGRANULAR <675°C MATRIX <500°C NO FCCI 7 PINS *	>675°C EVOLVED MATRIX <675°C MATRIX <500°C NO FCCI 2 PINS	>500°C EVOLVED MATRIX <500°C NO FCCI 4 PINS
MODERATE (1.96-1.97)	> 550°C MATRIX < 550°C NO FCCI 10 PINS	> 550°C MATRIX < 550°C NO FCCI 3 PINS	> 650°C COMBINED > 525°C MATRIX < 525°C NO FCCI 4 PINS
LOW (1.94-1.95)	>700°C SHALLOW MATRIX <700°C NO FCCI 3 PINS	>625°C SHALLOW INTERGRANULAR <625°C NO FCCI 4 PINS	>600°C SHALLOW INTERGRANULAR AND MATRIX <600°C NO FCCI 4 PINS

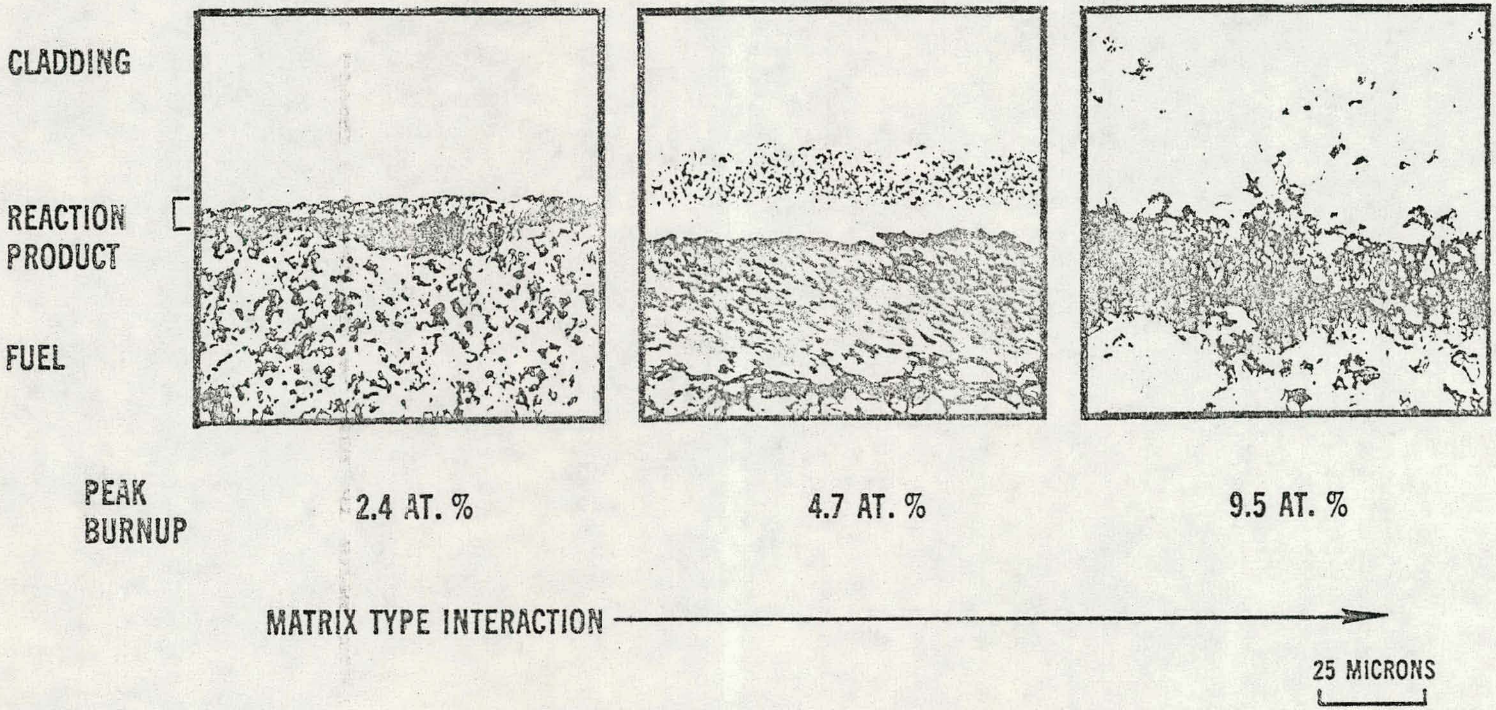
* NUMBER OF FUEL PINS EXAMINED WITH INDICATED O/M's AND BURNUPS

** CLADDING INNER SURFACE TEMPERATURE

HEDL 7901-096.1

(SLIDE 5)

EFFECTS OF BURNUP ON CHARACTER OF FCCI, O/M = 1.966



CLADDING INNER SURFACE TEMPERATURE ≃ 650°C

HEDL 7805-153.2

(SLIDE 6)

CHARACTERIZATION OF INTERACTION

O/M	BURNUP		
	LOW (0-3 at.%)	MODERATE (3-6 at.%)	HIGH (6-10 at.%)
HIGH (1.98-1.99)	>675°C** INTERGRANULAR <675°C MATRIX <500°C NO FCCI 7 PINS *	>675°C EVOLVED MATRIX <675°C MATRIX <500°C NO FCCI 2 PINS	>500°C EVOLVED MATRIX <500°C NO FCCI 4 PINS
MODERATE (1.96-1.97)	> 550°C MATRIX < 550°C NO FCCI 10 PINS	> 550°C MATRIX < 550°C NO FCCI 3 PINS	> 650°C COMBINED > 525°C MATRIX < 525°C NO FCCI 4 PINS
LOW (1.94-1.95)	>700°C SHALLOW MATRIX <700°C NO FCCI 3 PINS	>625°C SHALLOW INTERGRANULAR < 625°C NO FCCI 4 PINS	> 600°C SHALLOW INTERGRANULAR AND MATRIX < 600°C NO FCCI 4 PINS

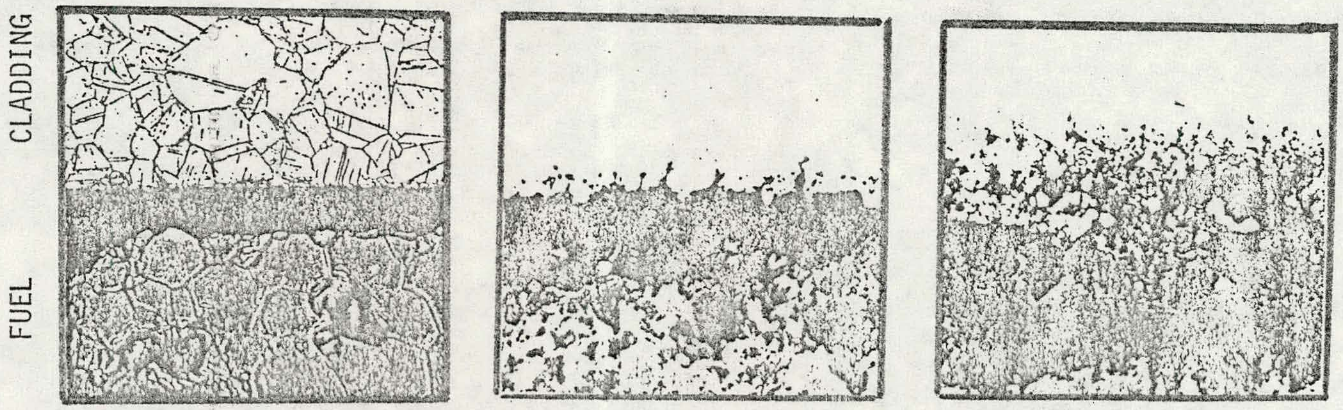
* NUMBER OF FUEL PINS EXAMINED WITH INDICATED O/M's AND BURNUPS

** CLADDING INNER SURFACE TEMPERATURE

HEDL 7901-096.1

EFFECTS OF BURNUP ON CHARACTER OF FCCI, O/M = 1.948

(ETCHED CLADDING)

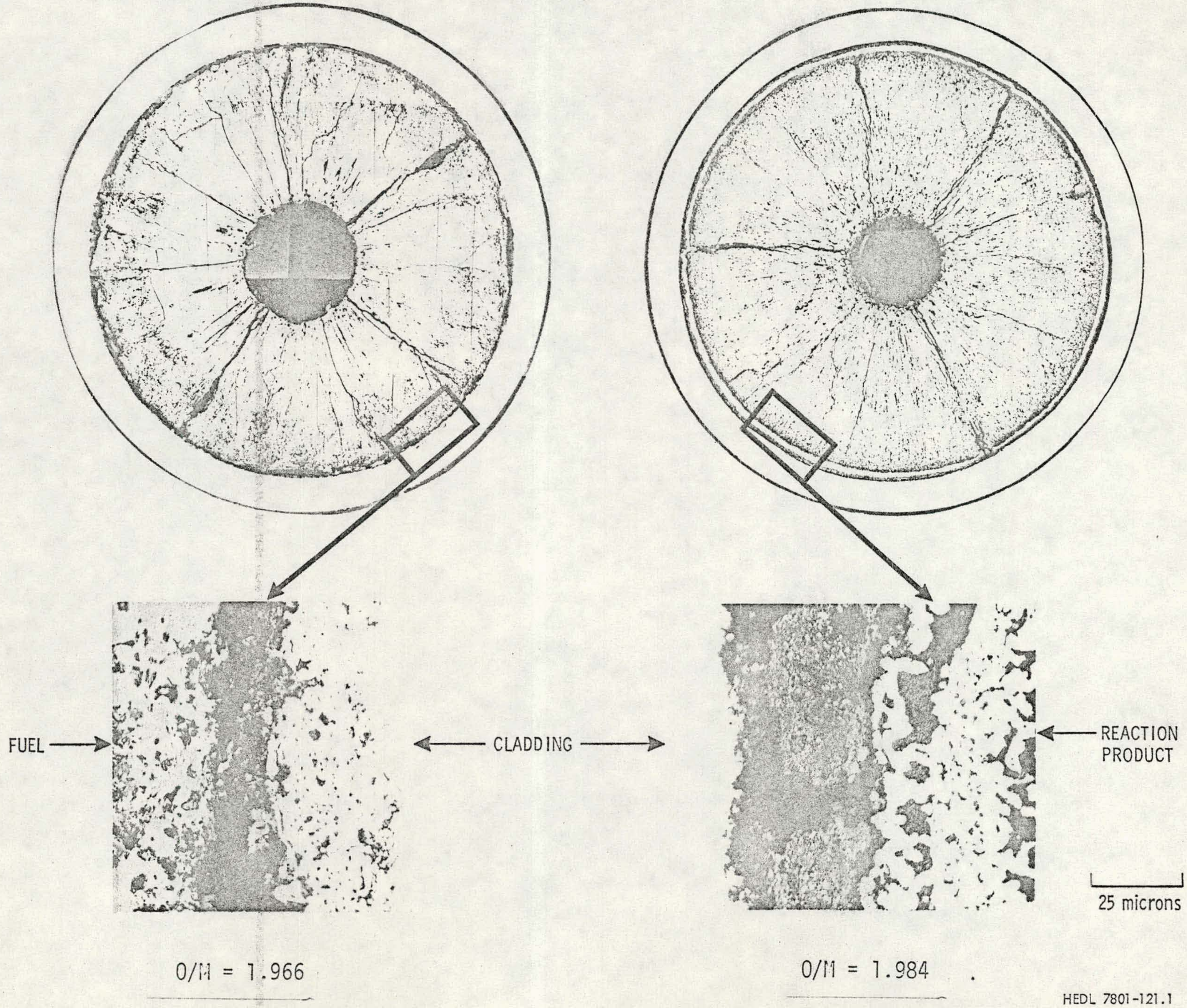


	2.3 at.%	6.7 at.%	8.3 at.%	
Peak Burnup	2.3 at.%	6.7 at.%	8.3 at.%	
	No Interaction	Shallow Intergranular Interaction	Shallow Intergranular and Matrix Interaction	25 microns

CLADDING INNER SURFACE TEMPERATURE ≈650°C

(SLIDE 8)

EFFECTS OF O/M ON FCCI



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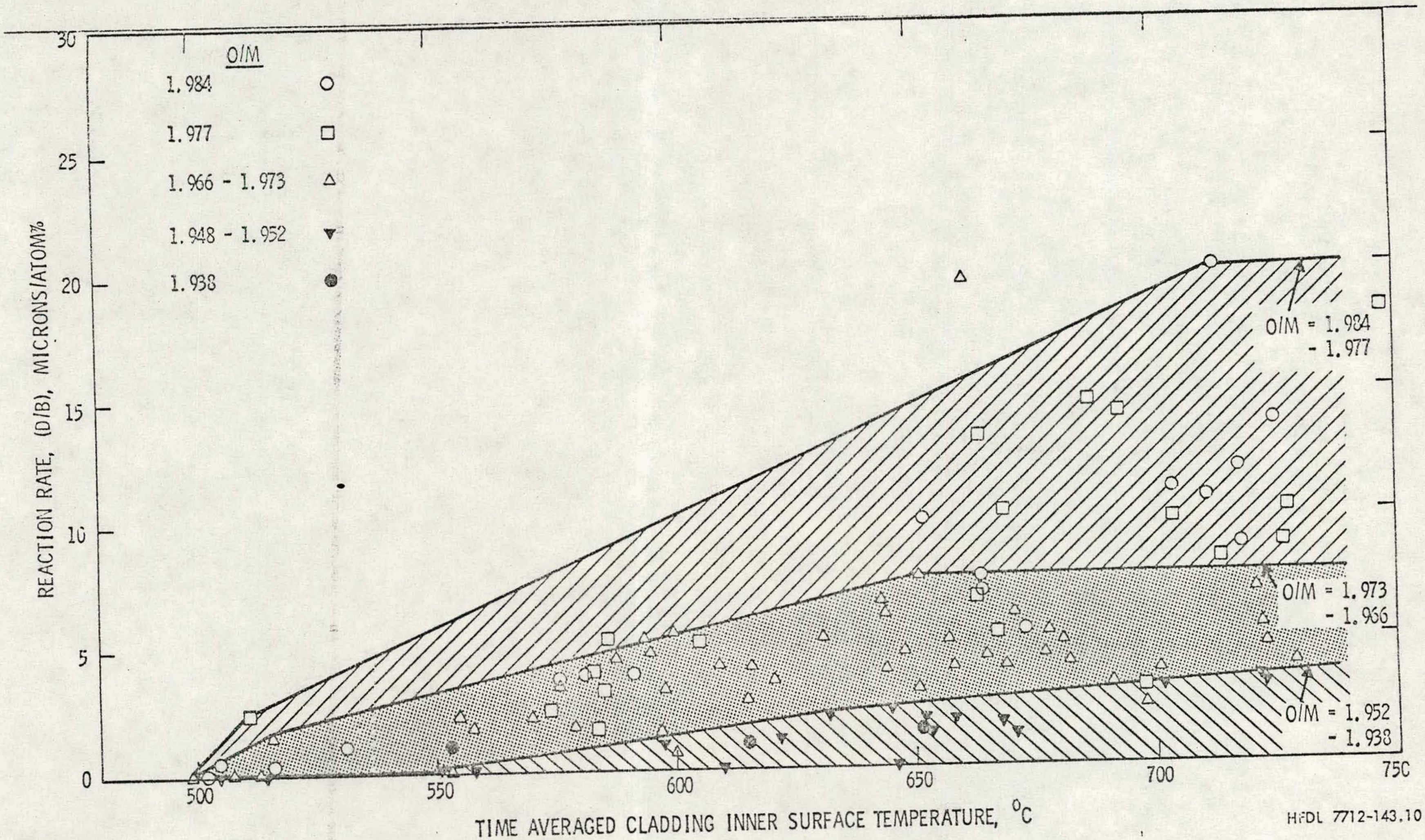
Peak Cladding Inner Surface Temperature of ~675°C. Peak Burnup ≈ 90.0 MWd/kgM.

(SLIDE 9)

DATA BASE FOR MAXIMUM DEPTH OF ATTACK



(SLIDE 10)

CORRELATION OF DEPTH OF INTERACTION

MODELS FOR O/M DEPENDENCE SELECTED FOR THE DATA CORRELATION

- $O/M - C$
- $(O/M - C)^2$
- $\left[(O/M)^{1/2} - C\right]^2$
- COMBINED O/M AND BURNUP (EFFECTIVE BURNUP)

$$C_1(O/M) + C_2(B) + C_3$$

(SLIDE 11)

CORRELATION OF DEPTH OF INTERACTION

$$D = .4521\,(B + K)(O/M - 1.942)(T - 728) \quad \text{for } B > 0,\ O/M > 1.942,\ T > 728$$

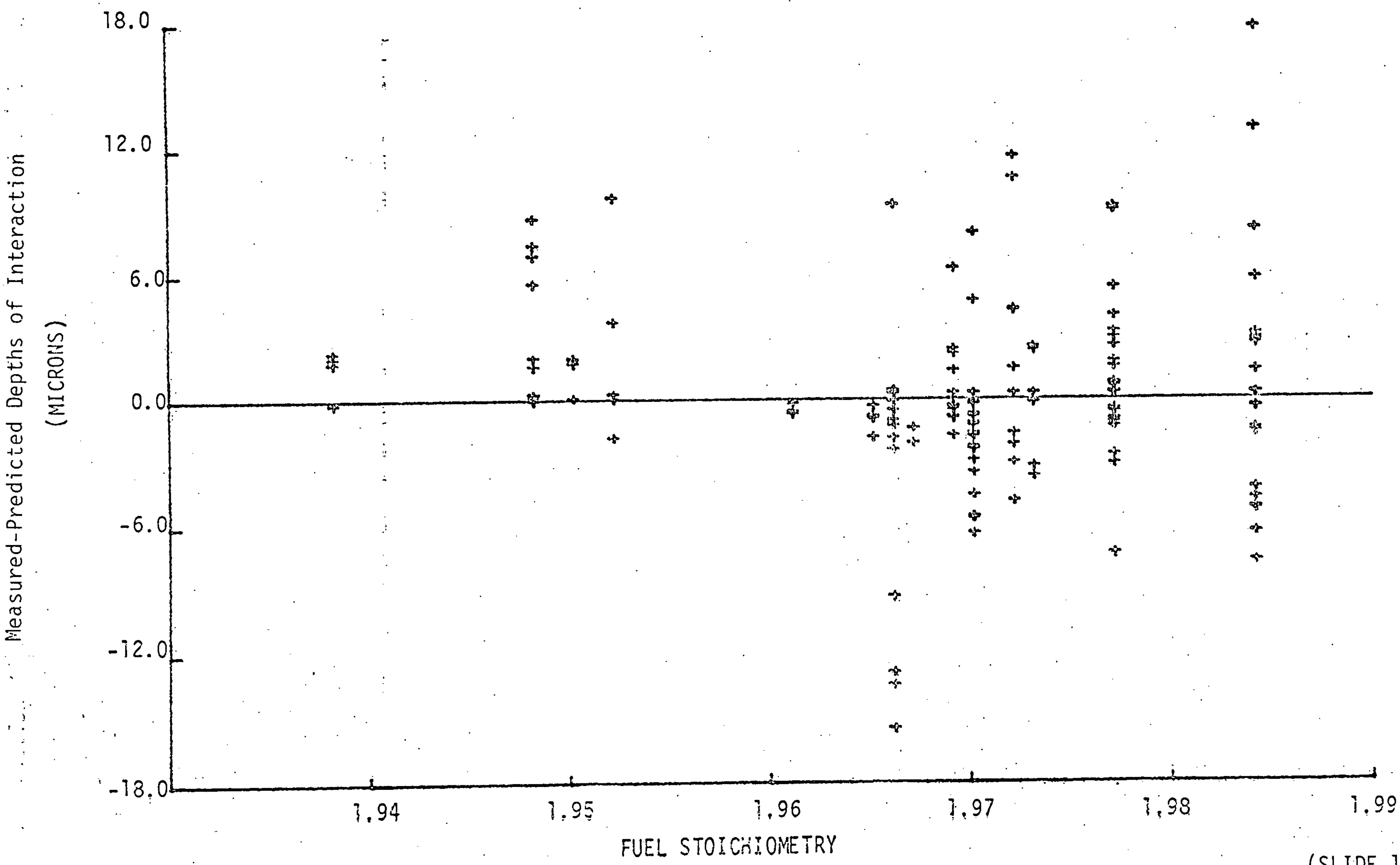
FOR CONDITIONS OUTSIDE THIS RANGE D IS ZERO

WHERE:

D = DEPTH OF INTERACTION (MICRONS)
B = LOCAL FUEL BURNUP (ATOM %)
K = CONSTANT (FUNCTION OF CONFIDENCE LEVEL FOR MAX. DEPTH OF INTERACTION)
O/M = INITIAL FUEL OXYGEN-TO-METAL RATIO
T = LOCAL TIME AVERAGED CLADDING INNER SURFACE TEMPERATURE, °C

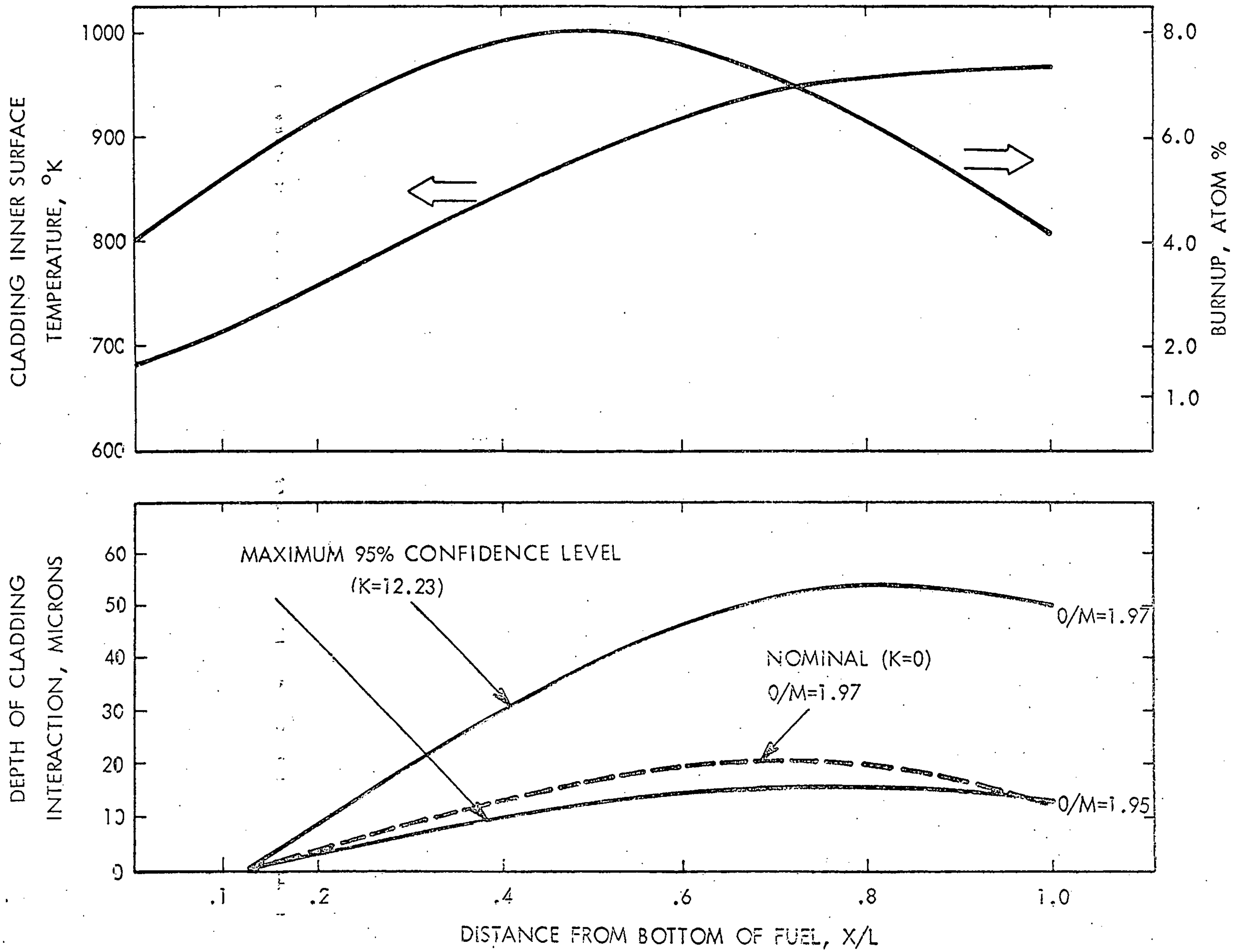
K	CONFIDENCE LEVEL
12.23	95%
9.45	90%
6.66	80%
5.04	70%
3.00	50%

COMPARISON OF MEASURED AND PREDICTED DEPTHS OF INTERACTION



(SLIDE 13)

PREDICTED DEPTH OF INTERACTION FOR A BREEDER REACTOR FUEL PIN



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(SLIDE 14)

SUMMARY

- REDUCTIONS IN INITIAL FUEL O/M RESULT IN SUBSTANTIAL REDUCTIONS IN EXTENT OF INTERACTION.

- SIGNIFICANT CHANGES IN CHARACTER AND TEMPERATURE FOR INITIATION OF INTERACTION ACCOMPANY REDUCTIONS IN O/M

(SLIDE 15)