

2

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

FAILURE EXPERIENCE IN REFRACTORY ALLOY-CLAD FUEL PINS APPLICABLE TO SPACE NUCLEAR POWER

D. S. Dutt
C. M. Cox
May 1984

AMERICAN NUCLEAR SOCIETY

June 3-8, 1984

New Orleans, Louisiana

HANFORD ENGINEERING DEVELOPMENT LABORATORY
Operated by Westinghouse Hanford Company, a subsidiary of Westinghouse Electric Corporation,
P.O. Box 1970, Richland, Washington 99352
under U.S. Department of Energy Contract No. DE-AC06-76FF02170
Work supported by the Assistant Secretary for Nuclear Energy
Office of Breeder Technology Projects

COPYRIGHT LICENSE NOTICE

By acceptance of this article, the Publisher and/or recipient acknowledges the U.S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering this paper.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED
[Signature]

MASTER

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

FAILURE EXPERIENCE IN REFRACtORY ALLOY-CLAD FUEL PINS
APPLICABLE TO SPACE NUCLEAR POWER

D. S. Dutt and C. M. Cox

Westinghouse Hanford Company

Numerous in-reactor tests were conducted in the 1960's and early 1970's to develop fuel elements for space nuclear reactors. Most of the tests emphasized refractory metal-clad UN, UC, and UO₂. Previous reviews were provided by Weaver and Scott⁽¹⁾ and by Gluyas and Watson⁽²⁾. More recently, these data were reviewed for supporting information concerning the technical feasibility issues as they relate to the current reactor designs.⁽³⁾ This paper will focus on the fuel pin failure experience to obtain insight into design options which will lead to a fuel system with the greatest potential for success.

The fundamental requirements for a space nuclear reactor are relatively low weight and long lifetime (Figure 1). For the reactor core, this has been translated into a compact, liquid-metal-cooled fast reactor with fuel goal burnups of 3 to 5 atom % and fast neutron fluences of 2 to 3×10^{22} n/cm² (Figure 2). Specific powers are modest by comparison with terrestrial reactors, but maximum coolant temperatures are much higher -- up to 1500°K versus about 800°K. The high operating temperatures are required to increase thermal efficiency and to minimize the size and weight of the heat rejection system. This has a significant effect on the reactor design, e.g., Li rather than the more conventional Na or NaK must be used as the liquid metal coolant because of its lower vapor pressure. Refractory metals must be used as fuel pin cladding rather than more conventional ferrous alloys, and fuel melting temperatures must be high, which leads to the choice of ceramic fuel material. In selecting a fuel system there are several considerations that must be addressed, including: fissile atom density, fuel operating temperature, fuel swelling, fission gas release, and fuel-cladding compatibility.

In excess of 360 fuel elements^(4,5,6) were fabricated and irradiated in support of fuels development (Figure 3). The fuels were primarily UC and UN with UZrC and UO₂ included to a lesser extent. UZrC is not currently of interest

to the SP-100 program, thus is not included in this review. UN and UO_2 fuels are the primary fuel candidates, but UC is a possible alternative to UN due to its high fissile density, and it has extensive irradiation experience. The refractory claddings of interest are tungsten, molybdenum, tantalum, and columbium based alloys (Figure 4). W-Re, T-111, Cb-1Zr, and PWC-11 have been irradiated with one or more of the fuels currently under consideration. ASTAR-811C and Mo-Re alloys do not have the same irradiation experience, but are prime cladding candidates because of chemical compatibility and strength-to-weight considerations.

A more detailed breakdown of the fuel and cladding combinations for which there is irradiation experience (Figure 5) shows the predominant experience is with UN fuel. W, Ta, and Cb alloys were used extensively with UN. UC experience is the next most extensive, with W the most frequently used alloy. The UO_2 experience is the most limited and is often obtained from fuel swelling samples (Reference 7). W and Cb-1Zr were most frequently used with UO_2 .

All of these irradiations were conducted in a thermal neutron environment. The concern with extrapolating the experience of thermal reactor irradiations to fast reactor environment is the increased possibility of cladding damage from a fast neutron environment and an increased uncertainty in the operating conditions due to self shielding in thermal reactors. Many of the pins in the data base were "samples" and not typical of a true pin design where the length-to-diameter ratio is 10 or greater. Small length-to-diameter-ratios introduce the potential for greater uncertainty in operating temperatures due to end effects such as thermal flux peaking. Finally the fission rate was higher than those in the designs currently under consideration, which potentially affects fission gas release and fuel swelling.

Uranium carbide fuel elements were irradiated at quite high temperatures (Figure 6). Most of these UC fuel pins and all of the failed UC fuel pins had fuel pellet densities in excess of 94% of theoretical. Four of the pins operating at cladding temperatures above 1600°K failed prior to achieving 1 atom % burnup. W-26Re was used as cladding for these high temperature irradiations. Large diameter increases, 16 to 28%, were measured on the failed pins, most

probably due to the fuel swelling. Most UC irradiations with cladding temperatures in the range 1200 to 1600°K used columbium alloys, usually with tungsten diffusion barriers. The only failures at burnups less than 2.5 atom % occurred in Cb-1Zr clad fuel pins without diffusion barriers. Fuel-cladding reactions were observed in their postirradiation examinations. The W-26Re-clad UC fuel pins which operated at temperatures above 1400°K and to burnups in excess of 3 atom % all failed and showed diameter increases of 28 to 46%, possibly indicating an increase in the UC swelling rate with burnup. The observed cladding strains associated with these UC fuel pins (Figure 7) indicate considerable ductility in W when tests were conducted at temperatures in excess of 1600°K. These large strains are not inconsistent with the projected UC swelling at these high temperatures. The failure of pins at 1400°K and at a burnup of approximately 4 atom % is also probably due to high fuel swelling. These data can be used to establish guidelines for operation with UC fuel. UC probably cannot achieve the SP-100 burnup requirements at temperatures above 1400°K. In the temperature range of most interest (up to 1500°K), it appears UC fuel will operate satisfactorily up to 3 atom % burnup if a barrier is provided to prevent fuel/cladding chemical interaction in those alloys susceptible to chemical attack.

About half the uranium nitride fuel pins failed when operated at cladding temperatures above 1600°K, probably fission gas loading (Figure 8). Tungsten alloys were generally used for cladding at these high temperatures and demonstrated significant in-reactor ductility, with postirradiation diameter increases of typically 5 to 10%. Uranium nitride irradiations in the 1200 to 1550°K temperature range primarily used columbium or tantalum alloys, with tungsten diffusion barriers between fuel and cladding. These irradiations, to burnups up to 2.5 atom %, and with several to 4 atom %, were generally successful. Most failures seem to correlate with fuel pellet densities in excess of 90% of theoretical, fuel oxygen contents in excess of 1000 ppm, and possibly anomalous hydrogen embrittlement of Ta alloy cladding. The failures associated with T-111 clad UN are possibly the only test sequences which can be used to guide the selection of fuel fabrication parameters. This test series had high and low density fuel. All failures were associated with the high density fuel. Some of the fuel elements had a fabricated central void

in the fuel, presumably to reduce the mechanical loading on the cladding. The central void had no effect on the failure rate of the pins with high fuel density. W-26% Re clad fuel pins exhibited high ductility (Figure 9).

The 65 UO_2 pins, none of which failed, included a fuel smear density range of about 85 to 90% of theoretical and burnups up to 6 atom % (Figure 10).

Thirty-five of these fuel pins used Cb-1Zr cladding and operated with cladding temperatures up to about 1250°K (Figure 10). Two UO_2 pins used T-111 cladding at similar temperatures. Twenty-seven fuel pins clad with W-26Re operated with cladding temperatures of 1500 to 2200°K. Cladding strains were typically less than 1% Δd for the high temperature, W-26Re clad fuel pins. Additionally, 259 specimens of Cb-1Zr clad UO_2 -BeO were irradiated over a considerable temperature range with the highest being 1500°K.⁽⁸⁾ It was concluded that the UO_2 -BeO was compatible with the Cb-1Zr, but burnup at these temperatures should be limited to 1.25 atom % due to UO_2 -BeO swelling. At a lower temperature, 1270°K, the burnup limit was considerably higher (4.5 atom %).

Results of these experiments, although having such nonprototypic features as thermal reactor irradiation rather than fast reactor irradiation, and in many cases very short fuel columns, indicate that all three fuels can operate satisfactorily. Burnups up to 4 atom % can be achieved if fuel densities and impurities are properly controlled and diffusion barriers are used with some fuel/cladding combinations. Considerations of fuel/cladding chemical compatibility and fuel swelling indicate that UN and UO_2 are more promising than UC for higher burnup applications. W barriers will be required for Ta and Cb alloys if used with UN and will be required for Ta alloys if used with UO_2 . Cladding temperatures are probably limited to 1500°K for Mo, Ta, and Cb alloys and 1600°K for W alloys.

REFERENCES

1. S. C. Weaver, J. L. Scott, R. L. Senn, B. H. Montgomery, Effects of Irradiation on Uranium Nitride Under Space-Reactor Conditions, ORNL-4461, October 1969.
2. R. E. Gluyas and G. K. Watson, Materials Technology for an Advanced Space Power Nuclear Reactor Concept: Program Summary, NASA-TN-D-7909, March 1975.
3. C. M. Cox, D. S. Dutt, R. A. Karnesky, "Fuel Systems for Compact Fast Space Reactors," 1st Symposium on Space Nuclear Power, January 1984.
4. J. T. Mayer, et al., EXFILE: A Program for Compiling Irradiation Data on UN and UC Fuel Pins, NASA TM X-68226, May 1973.
5. M. Kangilaski, et al., High Temperature Irradiation of Niobium-1% Zirconium-Clad UO₂, BMI 1730, June 28, 1965.
6. D. L. Keller, Progress on Development of Fuels and Technology for Advanced Reactors During July 1971 Through June 1972: Final Report, Task 6 and Task 7 Annual Report, In-Reactor Creep Program, BMI 1927, June 30, 1972.
7. W. Chubb, V. W. Storhok, D. L. Keller, "Factors Affecting the Swelling of Nuclear Fuels at High Temperatures, Nuclear Technology 18, pp. 231-256, June 1973.
8. M. S. Freed, M. A. DeCrescente, R. M. Kuhns, Development of UO₂-BeO Fuels, PWAC436, July 14, 1965.

Figure 1.

DESIGN CONSIDERATIONS

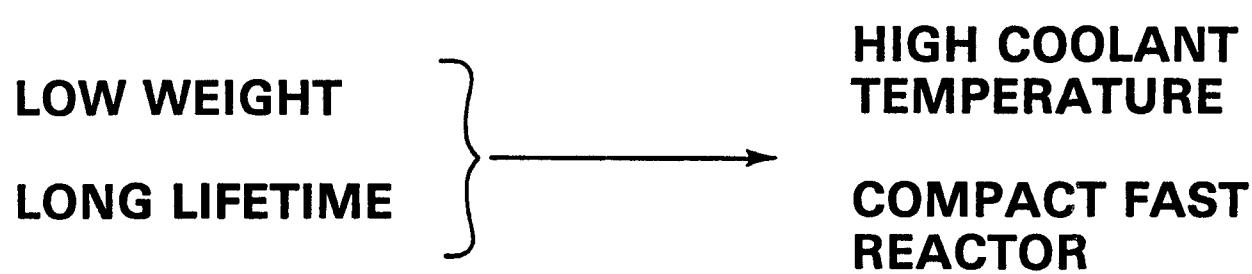


Figure 2.

FUEL SYSTEM REQUIREMENTS

CLADDING TEMPERATURE — 1300 TO 1500°K

PEAK BURNUP ————— 3 TO 5 ATOM PERCENT

POWER DENSITY ————— 900 TO 1300 WATTS/cc

**FAST NEUTRON FLUENCE — 2 TO 3 x 10²²
NEUTRONS/ cm²**

Figure 3.

SPACE REACTOR FUELS DATA BASE

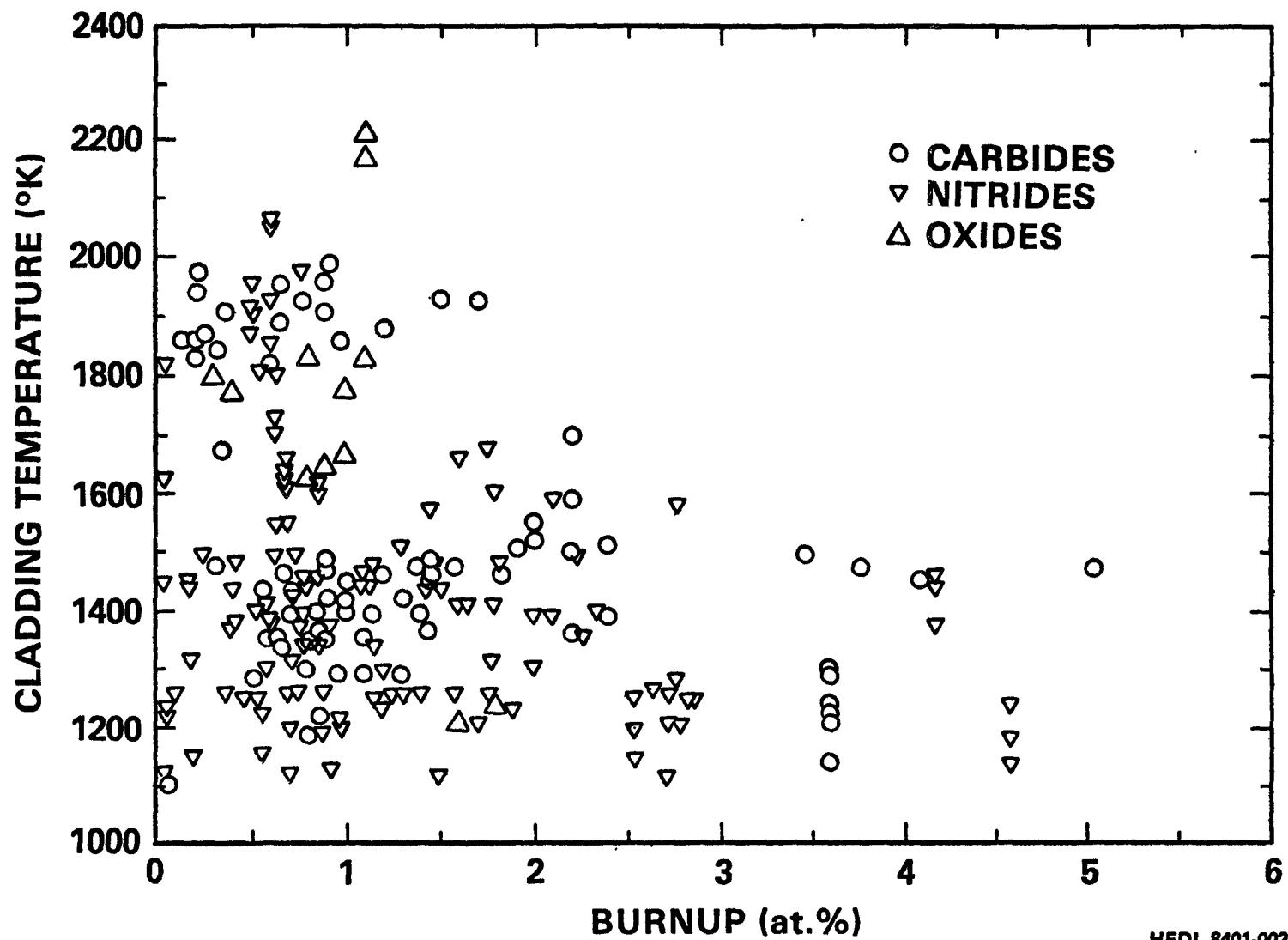


Figure 4.

REFRACTORY CLADDING MATERIALS WITH IRRADIATION EXPERIENCE

- W-26Re
- VAPOR DEPOSITED W
- Ta-8W-2Hf (T-111)
- Mo-0.5Ti-0.09Zr-0.03C (TZM)
- Cb-1Zr
- Cb-1Zr-0.1C (PWC-11)
- Cb-9W-1Z (D-43)
- STAINLESS STEEL

Figure 5.

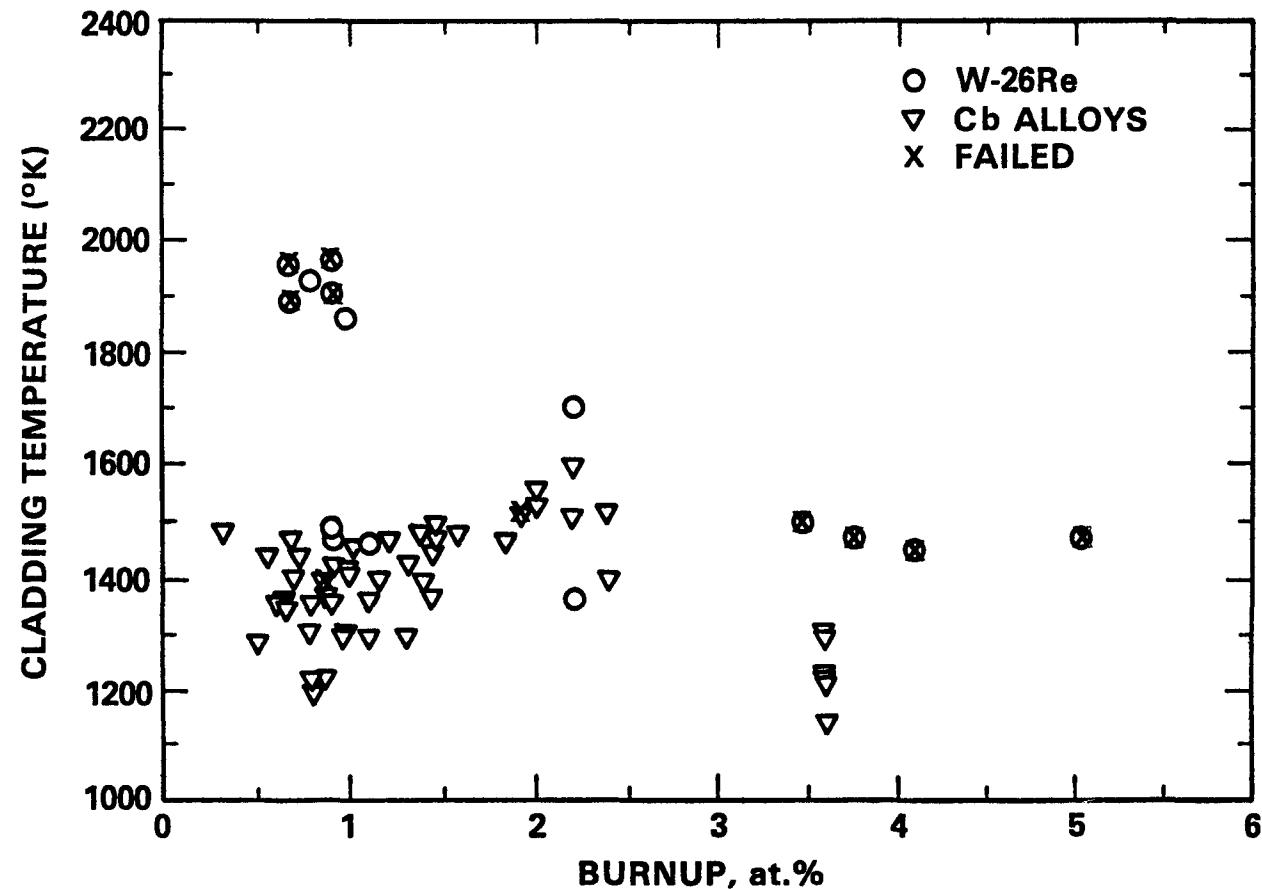
SPACE REACTOR FUELS DATA BASE

	UC		UN		UO ₂
	14	(8)*	33	(14)	27
W-26Re					
TUNGSTEN			2		1
T-111					
WITH DIFFUSION BARRIER			55	(14)	
WITHOUT DIFFUSION BARRIER			2	(1)	2
TZM					
WITHOUT DIFFUSION BARRIER			6		
Cb-12r					
WITH DIFFUSION BARRIER	47		35		2
WITHOUT DIFFUSION BARRIER	13	(2)	15		35
PWC-11					
WITH DIFFUSION BARRIER			47	(3)	
AUSTENITIC STAINLESS STEELS			6		
SUBTOTALS	74	(10)	201	(32)	67
TOTALS			342		

*NUMBERS IN PARENTHESES INDICATE FAILED PINS

Figure 6.

CARBIDE IRRADIATION EXPERIENCE



HEDL 8405-187.1

Figure 7.

OBSERVED DIAMETER CHANGE IN UC FUEL PINS

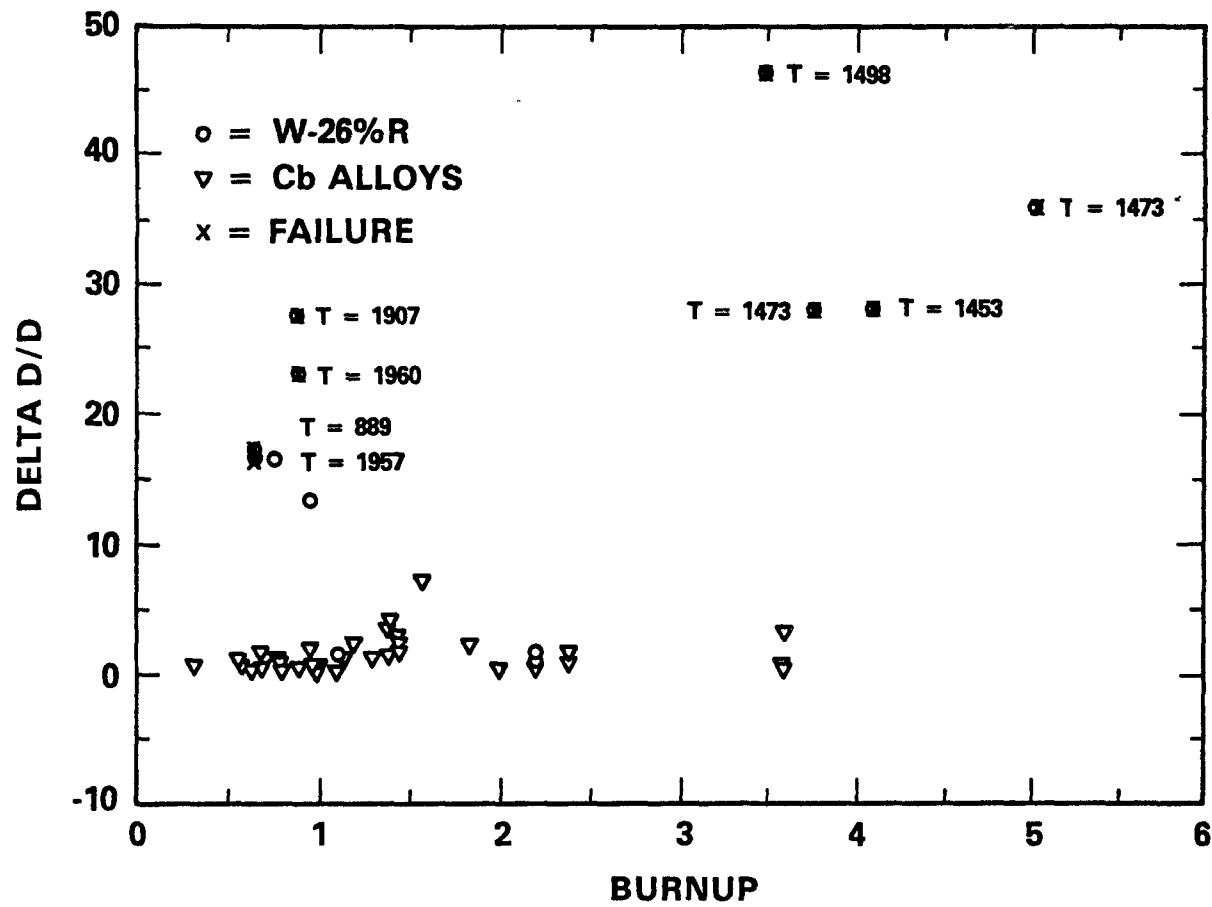


Figure 8.

UN IRRADIATION EXPERIENCE

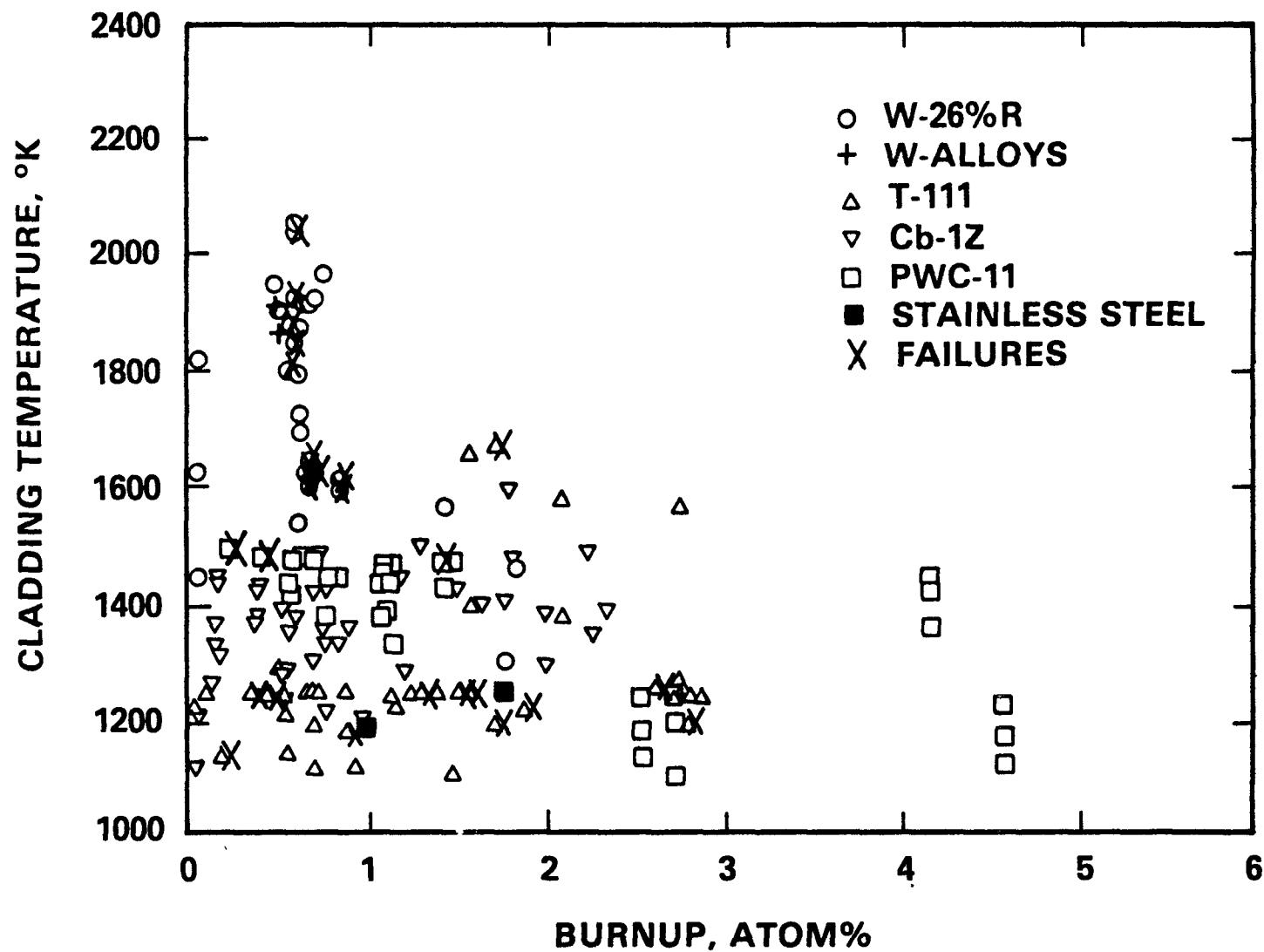
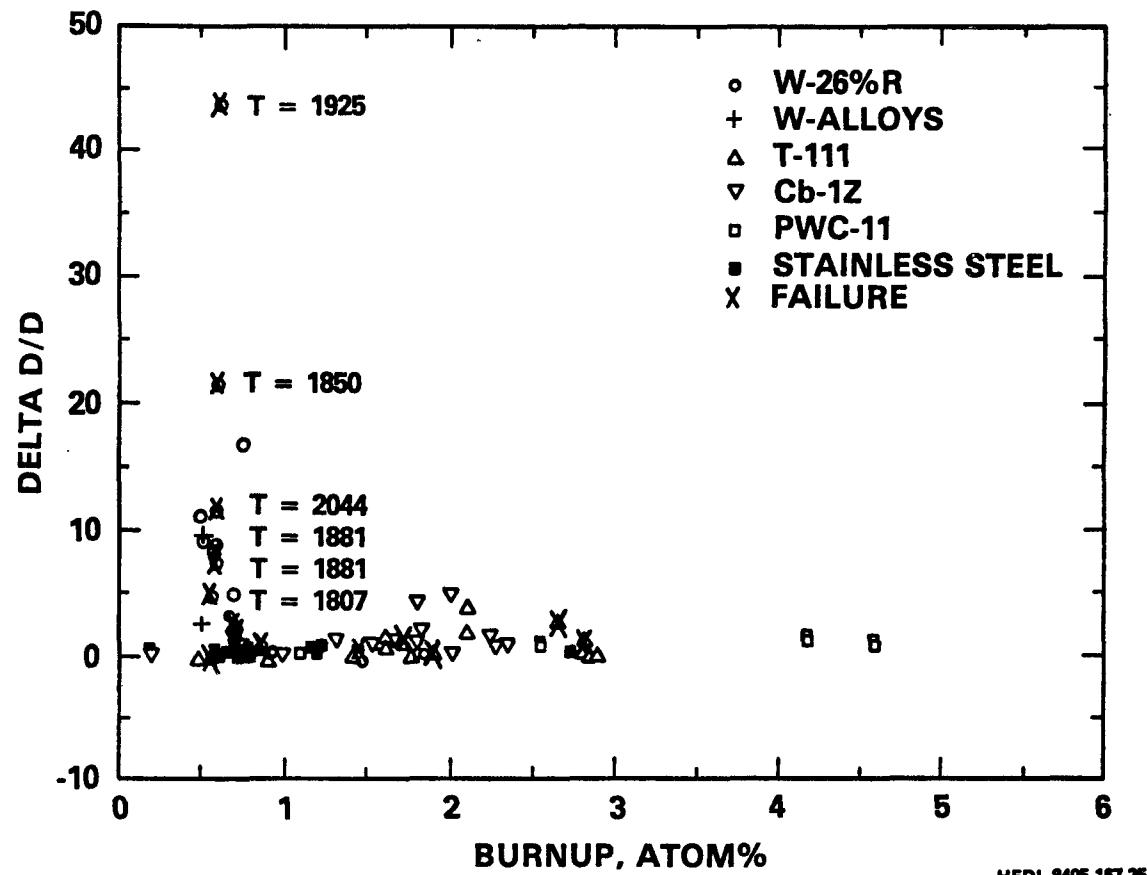


Figure 9.

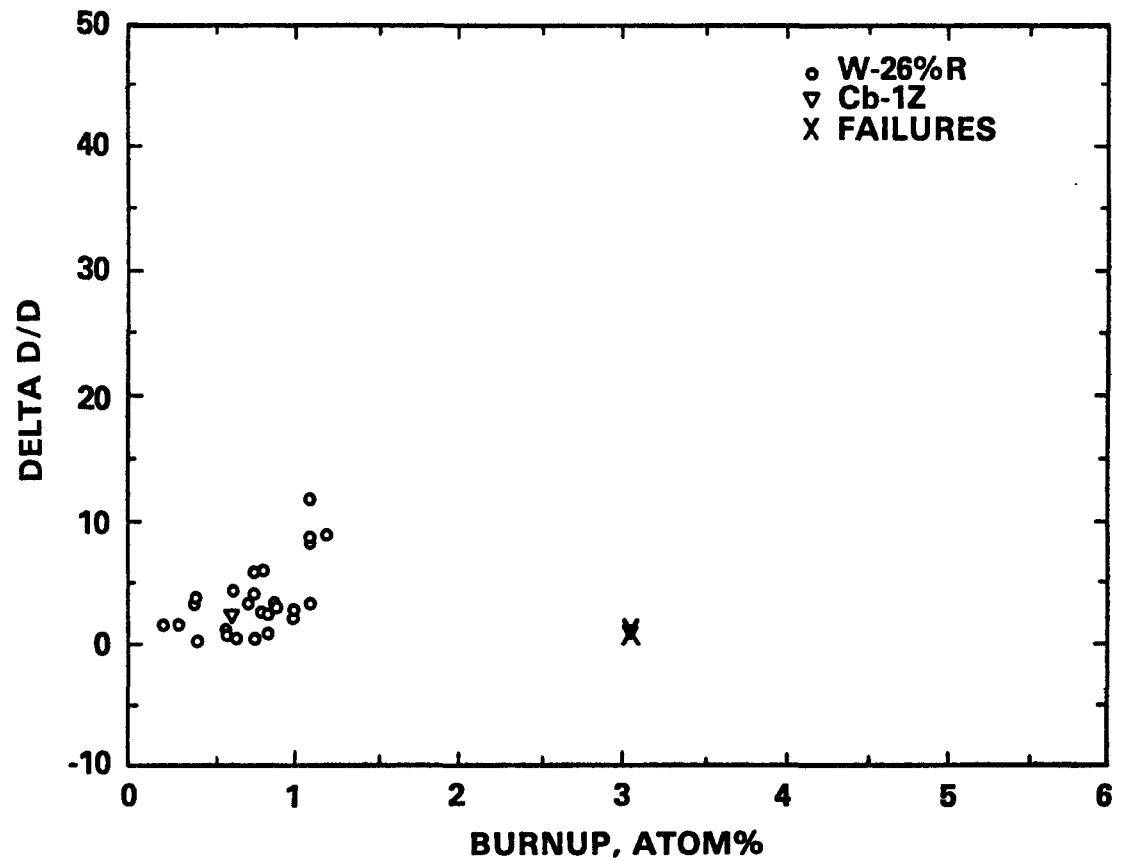
OBSERVED DIAMETER CHANGE ON UN FUEL PINS



HEDL 8405-167.26

Figure 10.

OBSERVED DIAMETER CHANGE ON UO_2 PINS



HEDL 8405-167.26