Examination of Disks from the* IPNS Depleted Uranum Target

by

Robert V. Strain** and John M. Carpenter***

**Energy Technology Division
***Intense Pulsed Neutron Source
Argonne National Laboratory
9700 S. Cass Avenue
Argonne, IL 60439-4838

October 1995

This paper was presented at the 13th Meeting of the International Collaboration on Advanced Neutron Sources, October 11-14, 1995, Paul Scherrer Institut, 5232 Villigen PSI, Switzerland

*Work supported by the U. S. Department of Energy, IPNS Program, under contract W-31-109-Eng-38.

ICANS-XIII
13th Meeting of the International Collaboration on
Advanced Neutron Sources
October 11-14, 1995
Paul Scherrer Institut, 5232 Villigen PSI, Switzerland

EXAMINATION OF DISKS FROM THE IPNS DEPLETED URANIUM TARGET

Robert V. Strain* and John M. Carpenter†

*Energy Technology Division
†Intense Pulsed Neutron Source
Argonne National Laboratory, Argonne, IL 60439, USA

ABSTRACT

This paper describes the results of visual, gamma ray scanning, and destructive metallurgical examination of two disks from the depleted uranium target of the Intense Pulsed Neutron Source.

1. Introduction

This report describes the results of examining the Zircaloy-2 clad depleted uranium disks (with emphasis on the first two) from the Intense Pulse Neutron Source (IPNS) Target. That target operated from August, 1981 to June, 1988 and from September, 1991 to September, 1992 at 450 MeV, pulsing at 30 Hz with a time average proton current of about 15 μA. The target was removed from service when the presence of fission products (^{135}Xe) in the coolant cover gas indicated a failure in the Zircaloy-2 cladding. Altogether, the target had absorbed about 240 mA hours of proton current, and endured between 50,000 and 100,000 thermal cycles. The purpose of the examination was to assess the condition of the disks and determine the cause of the cladding failure.

The IPNS depleted uranium target consisted of eight water-cooled uranium alloy disks, 10 cm in diameter and 2.7 cm thick, clad with Zircaloy-2. The peak centerline temperature during operation was ~225 C in the second disk. The first, third, fifth, and seventh disks contained stainless steel sheathed thermocouples in diffusion bonded Zircaloy-2 wells to measure the temperature near the center of the disk.

The uranium alloy (450 ppm carbon, 250 ppm iron, and 350 ppm silicon) core material is often referred to as Springfield adjusted uranium and has superior performance in the form of reduced swelling (compared to high-purity uranium) in the 400 to 600 C temperature range. The results of the original design study[1] indicated an expected total dimensional change ($\Delta L/L$/year) estimated to be 0.014, most of it resulting from thermal-cycling-induced growth (0.006) and thermal expansion (0.005). The design study also indicated that the cladding on the disk subject to the highest stress was predicted to fail after about 500 days of operation as a result of thermal cycling fatigue. Other failure mechanisms that were considered included 1) irradiation-induced swelling, 2) failures of the seam weld at the circumference of the each disk, 3) failures in the welds at the thermocouple well, and 4) separation between the uranium alloy and the cladding as a result of inadequate bonding. Such a separation could result in higher than normal temperatures and acceleration of the thermal-cycling-induced growth and irradiation swelling, which might lead to early failure of the cladding.

Keywords: Argonne, Uranium, Targets, Metallurgy

2. Non-destructive Examinations

Non-destructive examinations of the eight disks consisted of visual examination (including macro-photography), dimensional measurements (diameter and thickness), and gamma scanning.

Figure 1. Rear Face of Disk #1.

2.1 Visual Examination

Both faces of all 8 disks were visually examined and photographed. The condition of the disks has been described by Carpenter and Hins[2]. That description with some modification is as follows: Witness of the flow channel guide bars (flow separators) was apparent on the faces of all the disks. The marks left by the flow separators result from reduced discoloration of the Zircaloy-2 cladding that was in contact with the spacers compared to the cladding exposed to the coolant. However, the spacers appear to have prevented expansion of the disk surface and to have produced sharply defined indentations across the center regions of the first three disks.

The front (upstream, toward the proton source) face of disk #1 shows a 25 mm-long crack running across the disk in the direction of the thermocouple. The crack occurs near the center of the disk and appears to be imbedded in a narrow depression channel that is visible on the disk surface. The depression channel extends from near the center of the disk to the thermocouple tube exit point (over 50 mm long). The central region of the disk appears to exhibit a general bulging (except under the coolant spacers). A pattern of wrinkles and apparent blistering about 50 mm diameter is also visible near the center of the disk.

Fig. 1 shows the rear face of disk #1. In this view the disk is still housed in the flow guide cup. A small blunt crack is present near the center of the disk on a outward projection (blister) of the surface. The imprint of a flow spacer is quite close to this defect. The dial indicator measurements indicate that the projection extends to about 2 mm above the plane of the face. Undulations in the surface occur randomly over the entire surface, but are most pronounced in the central region of the face. A wide depression channel is visible, which extends from the exit point of the thermocouple to the distorted central region, and a narrow depression channel extends diametrally from the central region to the edge in line with the thermocouple.

The front face of disk #2 shows a surface projection near its center that appears to coincide with the location of the projection on the back face of disk #1. No cracks were visible on either face of disk #2, but wrinkles were present near the center of these faces. These undulations appear to be less severe and cover a smaller area than those on disk #1. Both faces of disk #3 and the front face of disk #4 show some visual evidence of wrinkles in the central region. The disk faces further back in the target stack show very little evidence of surface distortion.

2.2 Dimensional Measurements

Diameter (0° [aligned with the thermocouple well direction] and 90° orientations) and thickness measurements (eight locations) were made on each of the disks using a micrometer. These measurements indicate that significant diameter changes occurred in disk #1 (~0.25 mm increase) and slight diameter changes occurred in disk #2 (~0.05 mm increase). The measurements also indicate significant thickness increases near the center of disk #1 (~0.5 mm) and slight increases near the center of disk #2 (~0.08 mm). A dial indicator was used to measure the maximum deviation across each face of the disk. These measurements showed maximum distortions of 1. mm and .025 mm on the front faces of disks #1 and #2, respectively. The maximum distortion was found on the back face of disk #1 at the short crack (2 mm).

2.3 Gamma Scans

Gross beta and gamma activity measurements at the front and back face of each disk show peak values at the back face of disk #1 (120 R/hr) with slightly lower values at the front of disk #1 and both faces of disk #2 (110 R/hr). The activity diminished progressively in the remaining disks (from 105 to 4 R/hr). Diametral scans across the disks were performed at four orientations and for several nuclides. Scans for ^{137}Cs and ^{95}Nb/Zr fission products at the front face of disk #1 are shown in Fig. 2. The shape of these curves reflects the distribution of fission events and shows that most of the damage occurred near the center of the disks (but not perfectly centered) in a roughly circular pattern. The activity outside a radius of about 20 mm was less than half the peak activity. The area of measurable gamma activity is in general agreement with the observation of distortion of the cladding in disks #1 and #2.

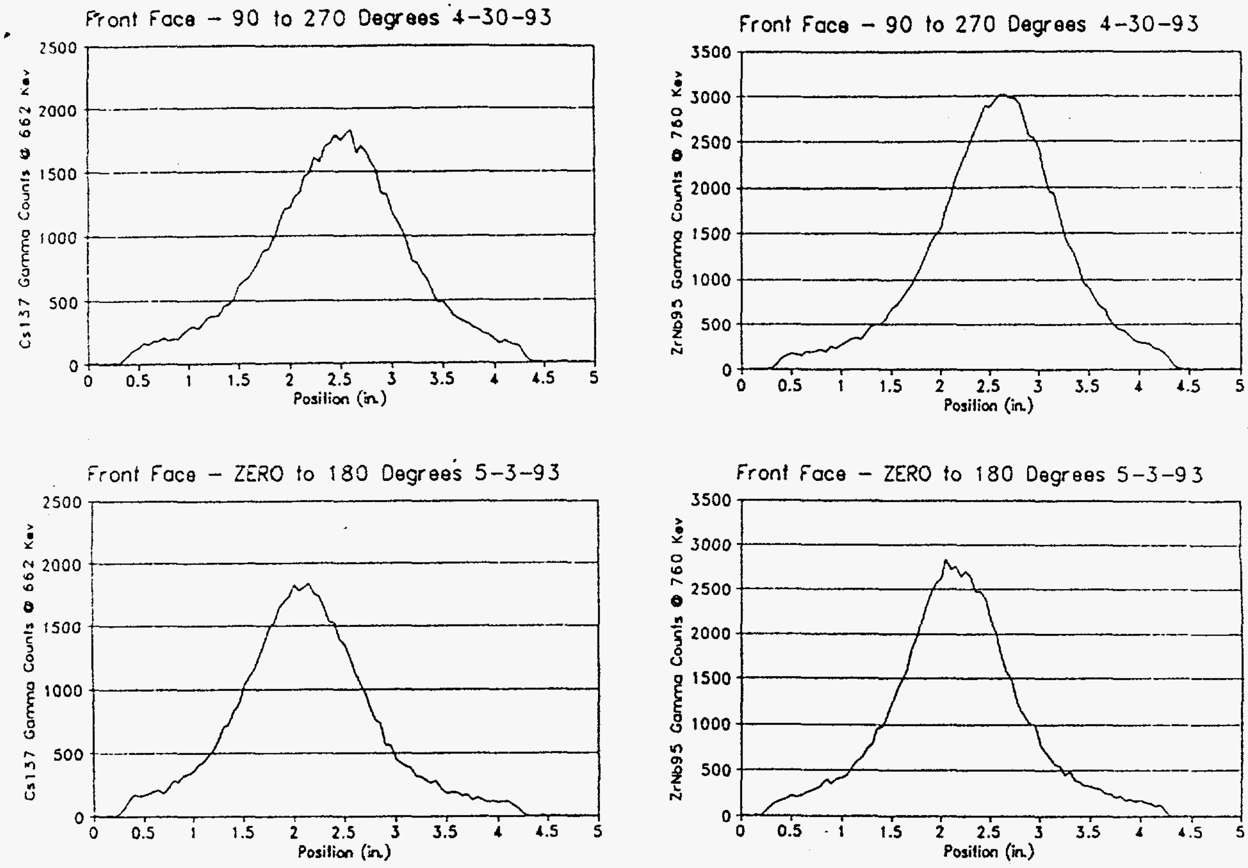

Figure 2. Gamma Activity Across the Diameter of Disk #1, Typical Shapes.

3. Destructive Examinations

3.1 Disk #1

Disk #1 was sectioned approximately perpendicular to the thermocouple about 12 mm from its centerline. The dominant feature revealed by the cut was a large crack through the core (uranium) at the thermocouple locations (Fig. 3) that was filled with powder.

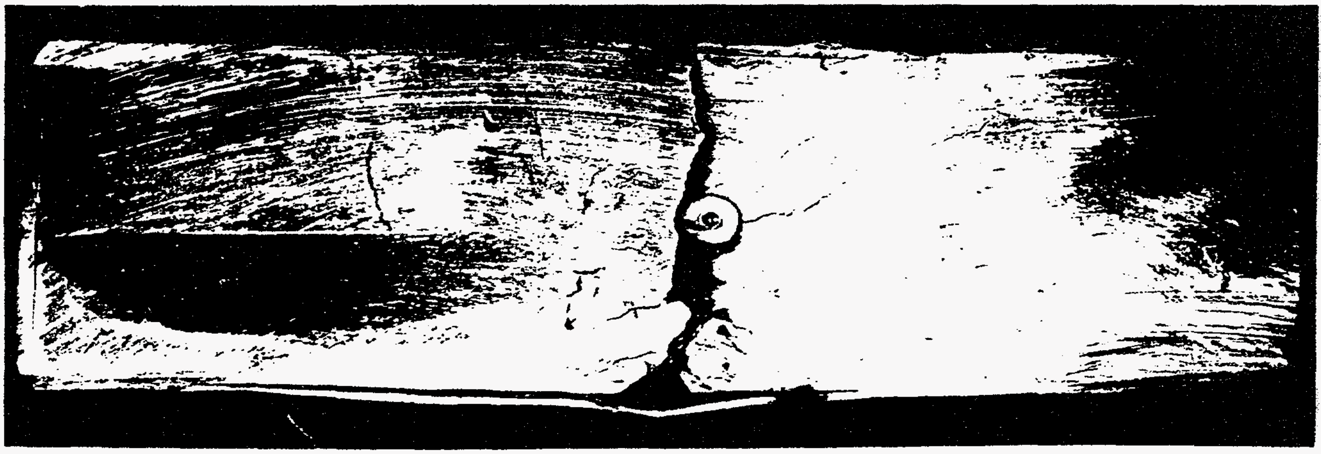

Figure 3. Section Perpendicular to the Thermocouple in Disk #1.

In addition to the major crack, smaller cracks radiate from the thermocouple location, and small cracks were visible at several other locations in the core material. The cladding was separated from the uranium near the major crack on both the front and back of the disk. Analysis of the material in the major crack with scanning electron microscopy (SEM) and neutron diffraction showed that it was primarily UO_2 (~70%) and UH_3 (~30%).

Figure 4. Micrographs of a Sample from Disk #1.

The powder in the major crack was assumed to be the result of interaction between the cooling water and the core material. The corrosion of uranium by water produces uranium oxides and under certain conditions uranium hydride. The formation of these reaction products results in large volume increases since the specific gravity of the most dense corrosion product (UO_2) is about 11 g/cm^3 compared to about 19 g/cm^3 for alpha phase uranium. We believe the volume increase of the relatively large amount of corrosion product to be the cause of the protrusion near the center of the back face of the failed disk. The location of the protrusion (with the short crack) of the cladding on the back face of the disk corresponds to the location with a large amount of the corrosion product.

A sample for optical microscopy was cut at a location 12 to 25 mm from the center of the disk. This sample was chosen because it included several smaller cracks, but was away from the major crack filled with corrosion product. Low magnification composite photographs of the sample from disk #1 show that the cladding has separated from the core material and a network of cracks is present in the uranium. Higher magnification photographs show that every crack examined contained a gray-colored material (Fig. 4) that is assumed to be corrosion product as a result of reaction between the uranium core material and the coolant water. In most areas the corrosion product appears to be a single phase, medium gray, monolithic material that is assumed to be uranium dioxide. At the end of cracks (where the crack is extending into the core material) the corrosion product appears to consist of two phases (the medium gray phase and a lighter gray phase). The lighter gray phase may be uranium hydride (if its not an optical illusion). The formation of hydride is promoted by high hydrogen partial pressure. The crack tip would tend to trap hydrogen and is a logical location for the formation of the hydride.

3.2 Disk #2

Reasoning that because corrosion had not taken place in the disk adjacent to the failed disk it must show cleaner evidence of the cause of failure than the failed disk, we performed a thorough destructive examination of disk #2. Disk #2 was initially sectioned through its center. Visual examination of the cut surfaces showed readily observable cracks in the core material at a distance of about 25 mm from the center. Two samples were cut from this area. A low magnification composite photograph of the first sample shows one large crack system in the uranium and several short cracks some distance from the main crack. Higher magnification micrographs (Inset in Fig. 5) revealed no evidence of corrosion products or other material in these cracks (the material that appears in the micrographs is epoxy that was used to fix the sample in a mount). The cracks in the uranium indicate that the failure of the cladding on the first disk initiated in the uranium. We conclude that the cracks in the uranium were the result of anisotropic swelling from a combination of thermal cycling and irradiation.

A second sample was cut that included the back face of disk #2. A low magnification composite micrograph, Fig. 6, of this sample shows that the main crack system extended through the entire thickness of Disk #2 to the back face. Again the cracks contained no corrosion product. At the intersection of the main crack with the cladding on the rear face of Disk #2, the core material appears to have pulled away from the cladding. The uranium on the two sides of the crack appear to be displaced from each other parallel to the direction of the crack. There appears to be a second phase present at the inner surface of the cladding in the region where the core material has separated from the cladding. Etching of the cladding indicates that the microstructure of the cladding is altered (the addition of fine precipitates) to a depth of about 150 μm in the region of the separation.

Figure 5. Cracks in the Core Material of Disk #2

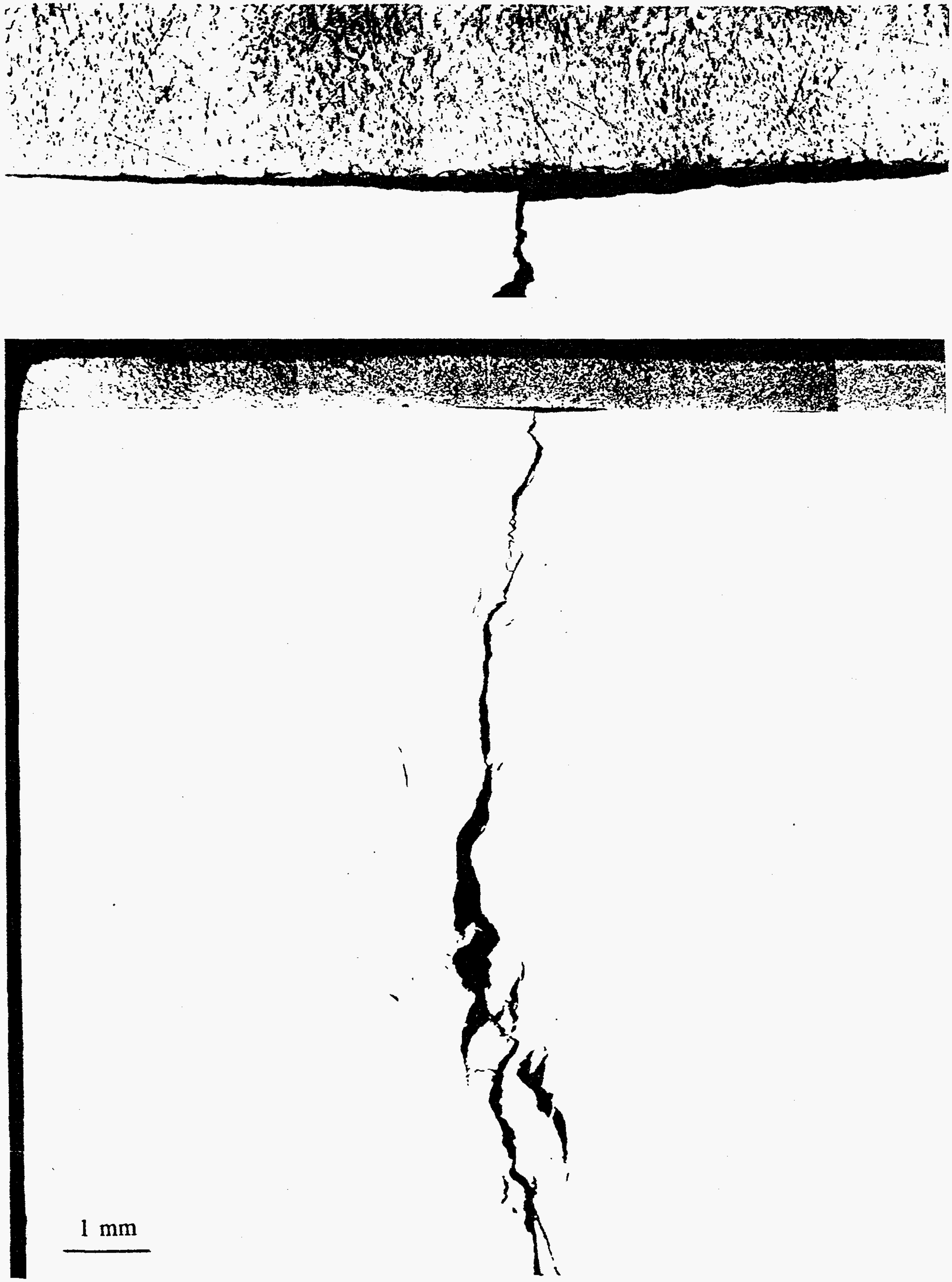

Figure 6. Intersection of a Crack in the Core Material with the Cladding, Disk #2.

4. Discussion of Results

Examination of the Zircaloy-clad uranium disks from the depleted uranium source showed that cracks were present in the cladding on the front and back face of the first disk. The faces of the first disk were distorted and measurable changes in the thickness of the disk had occurred. Observable distortion was also present on the front face of the second disk. Destructive examination of these disk showed cracking of the uranium in both disks. Corrosion products (UO_2 and UH_3) were present in the cracks in the first disk, but no corrosion products were present in the second disk. The results very strongly indicate that the cracking of the uranium preceded the cladding failure. The cracking of the uranium alloy appears to be in agreement with observation of distortion of uranium under conditions of irradiation and thermal cycling [3-5]. The presence of cracks in the uranium of disk #2 indicate that the thermocouple was not the initiator of the failure. The presence of corrosion products along the cracks, the relatively large amount of corrosion product found at the region of maximum cladding distortion, and the crack at the back face of disk #1 indicate that the reaction was limited by the availability of water in contact with the uranium. The thermocouple in the first disk may have exacerbated the corrosion by providing additional exposed surface area once water entered the cladding. The presence of hydride in the corrosion product indicates that the saturation temperature of the water was not exceeded.

5. Conclusions

Results of the detailed examination of the first two disks from the depleted uranium source assembly show that cracks occurred in the core material uranium before the cladding failed. These core material cracks were induced by anisotropic growth of the alpha-uranium caused by irradiation and thermal cycling at the operating temperature of the source. A second phase was present in the cladding at the core-cladding interface where a major crack in the uranium intersected the cladding in the second disk. If the second phase is an intermetallic compound, as was found in some cases during the manufacture of the disks[6], it is a brittle phase that could have resulted in an debonded condition. Such a bond failure could have contributed to the cracking of the cladding as a result of slightly greater amplitude of the thermal cycles, but a debond is not considered the root cause of the cracking of the core material. There was no evidence of failure of the circumferential seam weld or failure of the welds at the thermocouple well.

The widening of the cracks in the core material stressed the cladding and eventually led to its failure, probably a fatigue failure induced by the thermal cycling. Once the cladding failed, water entered through the crack in the cladding and found its way into the cracks in the core material. As the water reacted with the uranium it formed uranium dioxide and uranium hydride which have much lower densities that the metal. The volume expansion from these reactions was partially responsible for the distortion of the cladding that was observed for the first disk. The projection near the center of the back side of the first disk was caused by the formation of a pocket of corrosion product at that location. The thermocouple well in disk #1 appears to have weakened the disk structurally and enhanced the formation of corrosion product, once water had entered the cladding, by providing additional surface area in the core material.

The extensive formation of corrosion product in the core material cracks and the release characteristics of fission gas from uranium metal indicate that the source was operated for at least some time after the initial failure of the cladding with no adverse consequences. Measurable gas release is hypothesized to have occurred only after a significant amount of the corrosion products had formed.

These results are sufficient to indicate that the use of an alloy that stabilizes the uranium in the cubic (isotropic) gamma phase should greatly lengthen the useful life of the source. The examination also indicates that a second phase at the core-cladding interface may have played a secondary role in the failure of the cladding. Therefore, steps should be taken to avoid conditions that could result in the formation of intermetallic or other brittle phases at this interface.

6. References

[1] B. A. Loomis, H. R. Thresh, G. L. Fogle, and S. B. Gerber, "Design, Production, and Evaluation of a Zircaloy-clad Uranium Target for an Intense Pulsed Neutron Source Application", Nuclear Technology, Vol. 55 (December 1981), p. 617-627.

[2] J. M. Carpenter and A. G. Hins, "Experience with IPNS Targets", ICANS XII, Rutherford-Appleton Laboratory, May, 1993, Rutherford-Appleton Laboratory report 94-025, (1994), p. T-1 to T-11.

[3] L. T. Lloyd and R. M Mayfield, "Microstructural Changes of Uranium Upon Thermal Cycling", Transactions of the ASM, Vol. 50 (1957), p. 954-980,

[4] J. H. Kittel, "Preliminary Experiments on Irradiation Cycling and Partial Beta-Phase Irradiation of Uranium," ANL-5712 (April 1959)

[5] A. N. Holden, *Physical Metallurgy of Uranium,* Addison-Wesley Publishing Company, Inc., Reading, MA, p. 164-195

[6] A. G. Hins, et al., "A Study of Core/Cladding Bonding in IPNS Booster Target Discs," Proc. Second International Conference on Hot Isostatic Pressing, Gaithersburg, MD, June 9, 1989. p. 281-291