

**IRRADIATION TESTS OF <sup>99</sup>Mo  
ISOTOPE PRODUCTION  
EMPLOYING URANIUM METAL FOILS\***

G. L. Hofman, T. C. Wiecek, E. L. Wood, and J. L. Snelgrove  
Argonne National Laboratory  
Argonne, Illinois 60439-4841 USA

and

A. Suario, H. Nasution, D. Lufti-Amin, and A. Gogo  
Badan Tenaga Atom National, Indonesia

Presented at the  
1996 International Meeting on  
Reduced Enrichment for Research and Test Reactors

October 7-10, 1996  
Seoul, South Korea

The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. W-31-109-ENG-38. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes.

\*Work supported by the US Department of Energy  
Office of Nonproliferation and National Security  
under Contract No. W-31-109-38-ENG

# IRRADIATION TESTS OF $^{99}\text{Mo}$ ISOTOPE PRODUCTION TARGETS EMPLOYING URANIUM METAL FOILS

G. L. Hofman, T. C. Wiencek, E. L. Wood, and J. L. Snelgrove  
Argonne National Laboratory  
Argonne, Illinois 60439-4841 USA

A. Suripto, H. Nasution, D. Lufti-Amin, and A. Gogo  
Badan Tenaga Atom National, Indonesia

## ABSTRACT

Most of the world's supply of  $^{99\text{m}}\text{Tc}$  for medical purposes is currently produced from the decay of  $^{99}\text{Mo}$  derived from the fissioning of high-enriched uranium (HEU). Substitution of low-enriched uranium (LEU) metal foils for the HEU  $\text{UO}_2$  used in current target designs will allow equivalent  $^{99}\text{Mo}$  yields with little change in target geometries. Substitution of uranium metal for uranium alloy and aluminide in other target designs will also allow the conversion of HEU to LEU.

Several uranium-metal-foil targets have been fabricated at ANL and irradiated to prototypic burnup in the Indonesian RSG-GAS reactor. Postirradiation examination of the initial test indicated that design modifications were required to allow the irradiated foil to be removed for chemical processing. The latest test has shown good irradiation behavior, satisfactory dismantling and foil removal when the U-foil is separated from its containment by metallic, fission-recoil absorbing barriers.

## INTRODUCTION

The RERTR Program has during the last three years continued its effort to develop use of low-enriched uranium (LEU) to produce the fission product  $^{99}\text{Mo}$ . This work comprises both target and chemical processing development and demonstration. Two major target systems are now being used to produce  $^{99}\text{Mo}$  with highly-enriched uranium (HEU) -- one employing research reactor fuel technology (either uranium-aluminum alloy or uranium aluminide-aluminum dispersion) and the other using a thin deposit of  $\text{UO}_2$  on the inside of a stainless steel (SST) tube. The latter was used by Cintichem until 1989 and is presently used by BATAN, Indonesia. Both a target and its associated chemical processing must be developed to replace the deposited  $\text{UO}_2$  target. This paper summarizes progress in irradiation testing of targets based on uranium metal foils. Several targets of this type have been irradiated in the Indonesian RSG-GAS reactor operating at 22.5 MW, and postirradiation examinations have been performed in the adjacent

BATAN hot cell facility, under a cooperative research agreement between BATAN and Argonne National Laboratory. Chemical processing of foil targets will be discussed in a later paper at this meeting.

## **URANIUM FOIL TARGET**

(Differential Thermal Expansion - DTE - Target)

The design and pre-irradiated test result of the experimental target have been reported at previous RERTR meetings.<sup>[1]</sup> Briefly, the test target shown in Fig. 1 is externally identical in dimensions (except length) and appearance to the presently used UO<sub>2</sub>-coated "Cintichem" target. The fissile part of the target is a thin (125- $\mu$ m-thick) uranium metal foil sandwiched between slightly tapered inner and outer tubes. The inner tube is made of a material with a larger thermal expansion coefficient than the outer tube material in order for the differential thermal expansion to assist in maintaining good thermal contact between the foil and the tubes. The taper and the greater shrinking of the inner tube upon cooling after irradiation facilitate disassembly. Thin oxide layers were produced on the inner and outer tubes to serve as diffusion barriers to inhibit diffusion bonding of the uranium to the inner and outer tubes. Our original concept was to separate the irradiated foil from the tubes, so that only the foil need be dissolved to recover the molybdenum. The target is equipped with a swaglok fitting that allows evacuation and back filling with He prior to irradiating as well as fission gas removal after irradiation.

Out-of-reactor thermal tests had shown that a thin aluminum oxide layer prevented any interdiffusion between the uranium foil and the aluminum tube during a six-day test at 400°C<sup>[1]</sup> -- a temperature substantially higher than expected to occur during irradiation of the target. The initial irradiation test has shown that radiation rendered the oxide layer ineffective and also enhanced interdiffusion, resulting in what appears to be a substantial conversion of uranium to UAl<sub>3</sub>. As will be detailed in the following section, several modifications led to a successful design in which thin (10- $\mu$ m) non-fissile foils were installed between the uranium foil and the target tubes.

## **IRRADIATION TEST RESULTS**

To date three irradiations have been completed in the isotope irradiation rig situated in the core center of the RSG-GAS reactor. Each irradiation lasted six days, after which the targets were allowed to cool in the reactor basin for one day. They were then transported to the hot cell for dismantling and postirradiation examinations. The various test targets with their salient attributes are listed in Table 1.

**TABLE I  
TEST TARGET MATRIX**

Irrad.	Outer Tube		Inner Tube			
Test No.	Material	Surface Condition	Material	Surface Condition	Target Foil	Result
1	Zr	air oxidized	Al	anodized	U <sup>(1)</sup>	inextractable U-Al interaction
2	Zr	air oxidized	Al	Zr coated	U <sup>(1)</sup>	extractable U-AlZr bonding
	Zr	air oxidized	Mg	as-machined	U <sup>(1)</sup>	extractable U-Mg bonding
	Zr	air oxidized	Zr	air oxidized	U <sup>(1)</sup>	inextractable U-Zr bonding
3	Zr	air oxidized	Al	as-machined	Ni-U <sup>(2)</sup> -Ni	extractable Al-Ni bonding
	Zr	air oxidized	304SST	as-machined	U <sup>(2)</sup>	inextractable
	Zr	air oxidized	304SST	as-machined	U <sup>(2)</sup> -Ni	extractable U-SST bonding
	Zr	air oxidized	304SST		Cu-U <sup>(2)</sup> -Cu	extractable foil easily removed

(1) Pure uranium,  $\beta$  treated

(2) Adjusted uranium,  $\beta$  treated

## TEST NUMBER ONE

A single target was irradiated in this test. This target consisted of an anodized aluminum inner tube, an air-oxidized zirconium outer tube, and a  $\beta$ -treated pure uranium foil.

Out-of-reactor heating tests had shown that during six days at 400°C no interdiffusion between either aluminum or zirconium and uranium foil occurred. It was hoped that this simple surface treatment would also prevent diffusion during irradiation and that the more complicated addition of diffusion barriers would not be needed.

The first operation in the hot cell involved the collection of fission gas from the target. This standard procedure for the production targets at BATAN is accomplished by cooling the target in liquid N<sub>2</sub> to precipitate the gas, after which the swagelok seal is opened and connected to a copper vessel that in turn is cooled while the target is reheated. The fission gas is thus transferred from the target to the copper vessel. No measurable gas pressure was observed when this operation was performed in the test target, confirming our prediction that only a very minimal fraction of the fission gas would be released from the U foil by recoil. Therefore, the gas collection procedure was omitted for the subsequent test targets.

Dismantling of the test target is performed with a lathe-type machine by cutting through the zirconium outer tube with a plumber's tube cutter. The top and bottom end plugs are thus removed and the inner tube is to be pressed out with a mandrel toward the top (the larger) end of the tapered tube. It was not possible to remove the inner tube from the first test target with the force available in the hot cell. The target was then sectioned and metallographically examined. As shown in Fig. 2, there was substantial interaction between the uranium foil and the aluminum inner-tube. The anodized layer has evidently been destroyed, and radiation (recoil)-enhanced diffusion has locked the target assembly, making extraction of the inner tube impossible. Interdiffusion of uranium and aluminum results in the formation of, primarily, UAl<sub>3</sub>, an intermetallic compound with a much-lower density than uranium. However, there appears to be no indication of interaction having occurred between the uranium foil and the outer zirconium tube.

## TEST NUMBER TWO

Three targets were irradiated in this test, each one with a different inner tube. In one target the aluminum inner tube was retained but it was coated with zirconium by means of flame spraying. This choice was based on the apparent non-bonding of the uranium foil and the outer zirconium tube in the first test. A second target contained a zirconium inner tube for the same reason; however, this combination lacked the thermal expansion difference. The third target contained a magnesium inner tube. Magnesium has all the desirable properties of aluminum but does not form compounds with uranium. The zirconium-coated aluminum and magnesium inner tubes could both be extracted from the outer tube without difficulty. The uranium foils, however, were firmly bonded to both, (see, for example, Fig. 3) and could not be removed. The zirconium inner tube could not be extracted from the all-zirconium target. It is clear that the differential thermal expansion feature is needed for easy extraction.

Metallography of the extracted inner tubes revealed no obvious interaction between either uranium and magnesium, or uranium and the zirconium coating on aluminum (see Figs. 4 and 5). Intimate contact and an intense flux of fission recoil atoms at the uranium surface apparently results in sufficient atomic mixing at the interface to affect a strong metallurgical bond.

### TEST NUMBER THREE

The first two tests led to the following conclusions:

1. The thermal expansion difference between inner and outer tube is necessary.
2. Recoil-atom flux needs to be eliminated from the uranium foil-tube interface.

Several metals were evaluated for recoil barriers. The principal requirement for these metals is primarily of a chemical nature. Because the barrier metal will bond to the uranium foil it should not interfere with the dissolution and purification part of the  $^{99}\text{Mo}$  process.

Four metals were thus selected as barrier candidates: copper, nickel, iron, and zinc. The first three are suitable for the acid dissolution process while zinc can be used for a base process as well. For the current test copper and nickel were selected. Since the recoil range in Cu and Ni is approximately 7  $\mu\text{m}$ , a 10- $\mu\text{m}$ -thick barrier should stop all recoil atoms from reaching the target tube surfaces.

At this stage of the experiment another modification was introduced. Accepting the fact that uranium foil will bond to any material by the recoil mixing mechanism described above, a possible workable design could be the following. Change the inner tube to an inert material that may be immersed in the dissolver, allowing only the attached uranium foil to be dissolved. The obvious material is a 300-type stainless steel, a material similar to that of the dissolver vessel. Types 304 and 316 stainless steel have a higher coefficient of thermal expansion than zirconium, satisfying that criterion as well.

Another concern was raised during the examination of the second test. The uranium foils (albeit bonded to the inner tubes) appear to be rather granular. The concern was that even if the foils were removable, they would be very fragile. Metallography of the as-fabricated pure uranium foils showed them to have a rather large-grained microstructure. Individual irradiation growth of such large grains would indeed result in the observed coarse granular macrostructure of the uranium foils. It was decided to use so-called "adjusted" uranium for the third test. This required addition of small amounts of aluminum and iron to the uranium, which drastically reduced the grain size in the finished uranium foils.

The third test was thus comprised of the following four targets. Three targets contained a type 304 stainless steel inner tube, of which one had no recoil barriers (serving as a control sample), a second had a 10- $\mu\text{m}$  foil at the uranium-outer zirconium interface (this target represents the option of immersing the inner tube together with the uranium foil), and a third had copper foils on both inner and outer foil-tube interface (a copper-uranium-copper sandwich).

To round out the test, an aluminum inner tube with nickel foils at both interfaces (a nickel-uranium-nickel sandwich) was added. Time constraints did not permit anodizing the aluminum inner tube. All four targets were irradiated in one 6-day period by coupling the targets in pairs, connecting them at their swaglok connectors.

The two prime test targets proved to be entirely successful. The target with a nickel foil at the uranium-zirconium outer tube interface was easily extracted. There was no evidence of bonding between the nickel foil and the outer zirconium tube. The foil (uranium-nickel combination) was firmly locked to the stainless steel inner tube as expected. The target with copper on both sides of the uranium foil was also easily extracted and the copper-uranium-copper foil (sandwich) could be easily slid off the inner stainless steel tube (see Figs. 6 and 7). Small samples were cut from both targets for chemical dissolution studies. The target with a stainless steel inner tube, without recoil barriers on either side could not be extracted. The target with an aluminum inner tube and (nickel-uranium-nickel) foil combination was easily extracted; however, the nickel inner foil appeared bonded to the aluminum inner tube. A small piece of the foil could be dislodged, indicating that the bond may not be very sound.

Review of the literature reveals that nickel-aluminum interdiffusion may have occurred to a minor extent (this is not the same as the before-mentioned recoil mixing), and it is believed that anodizing the aluminum inner tube surface will prevent this interdiffusion.

## **CONCLUSIONS**

Low-enriched-uranium metal foils can be conveniently removed from tubular target assemblies after irradiated, when recoil-atom absorbing barriers are placed between the uranium foil and the target tubes to prevent bonding. The coefficient of thermal expansion of the inner tube material should be larger than that of the outer tube to facilitate assembly of the target.

## **REFERENCES**

1. T. C. Wiencek et al, "LEU <sup>99</sup>Mo Target Fabrication and Testing," Proceedings of 1994 International Meeting on Reduced Enrichment for Research and Test Reactors, Williamsburg, VA USA (1994).

TAPERED ZIRCALOY-AL TARGET WITH U FOIL FOR <sup>99</sup>MO PRODUCTION

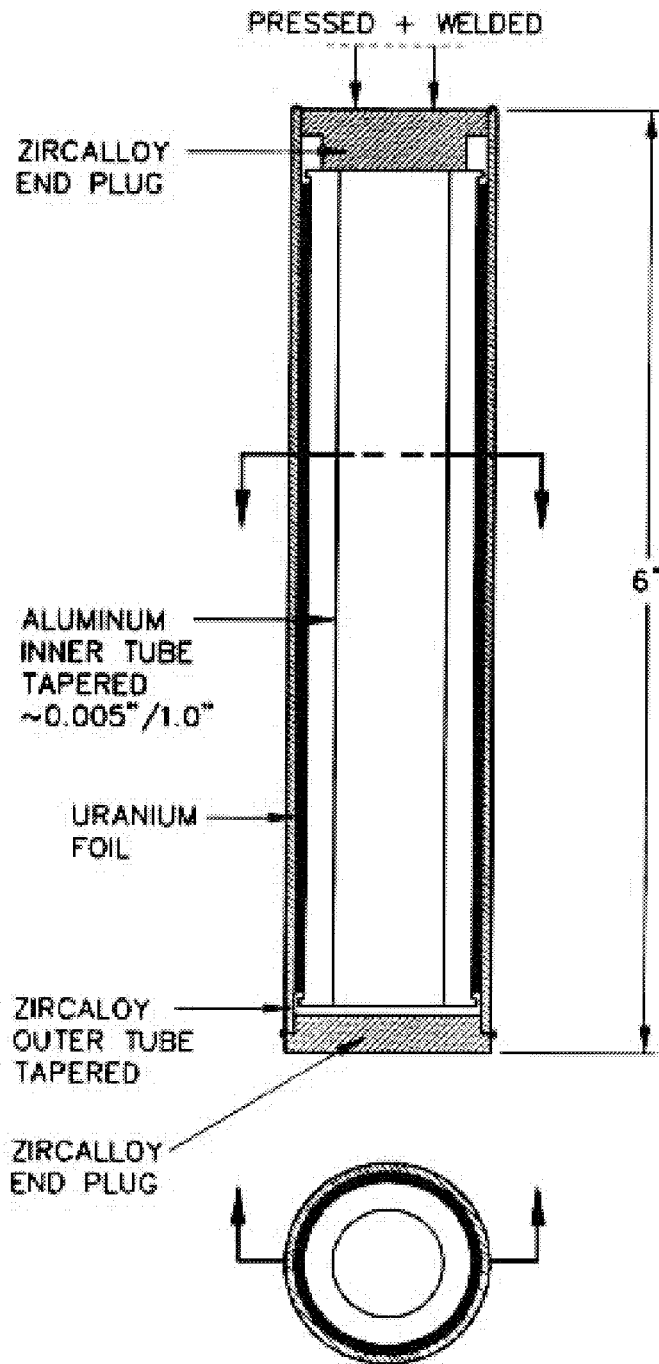
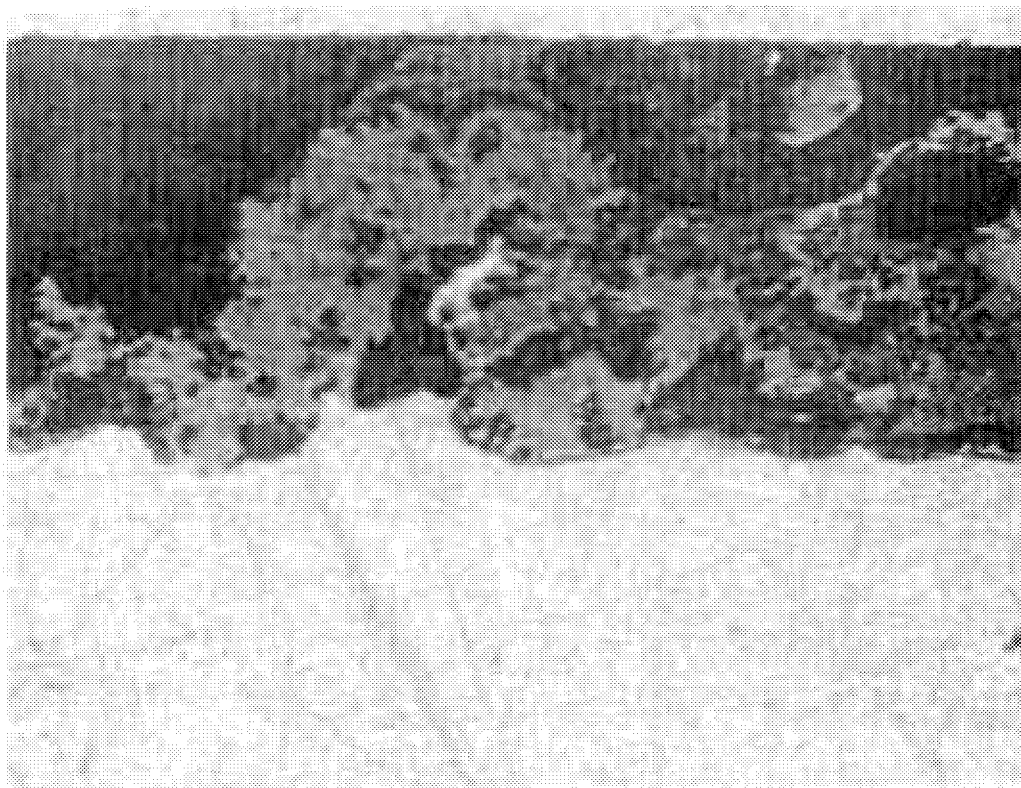


Figure 1. Differential Thermal Expansion (DTE) Target





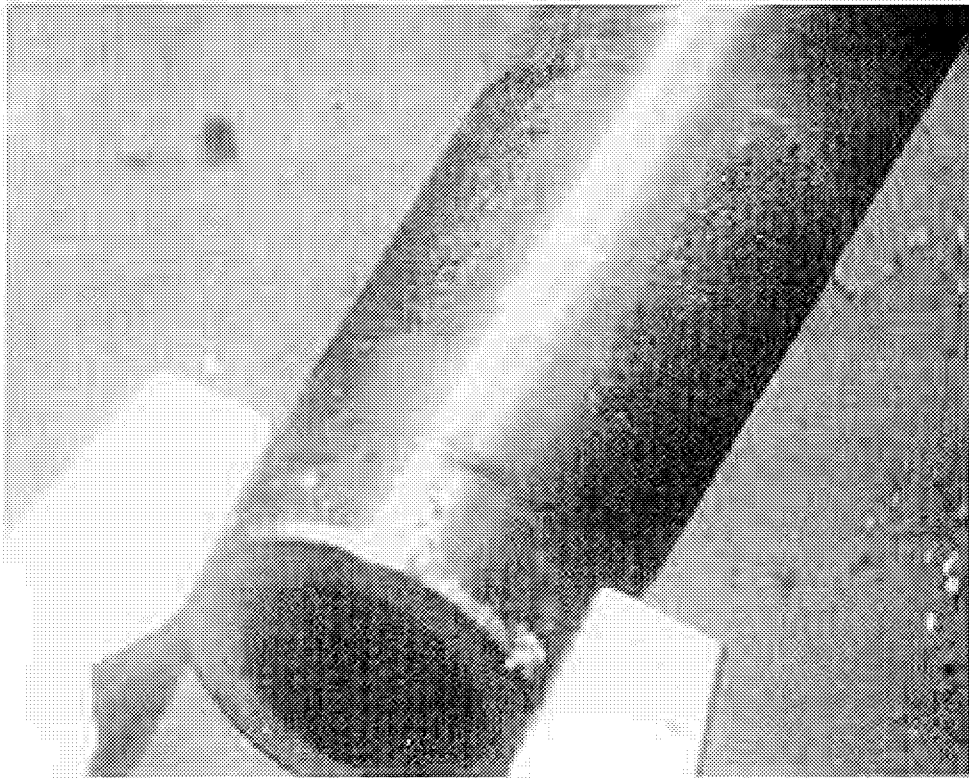
← Zr

← U-foil

← Al

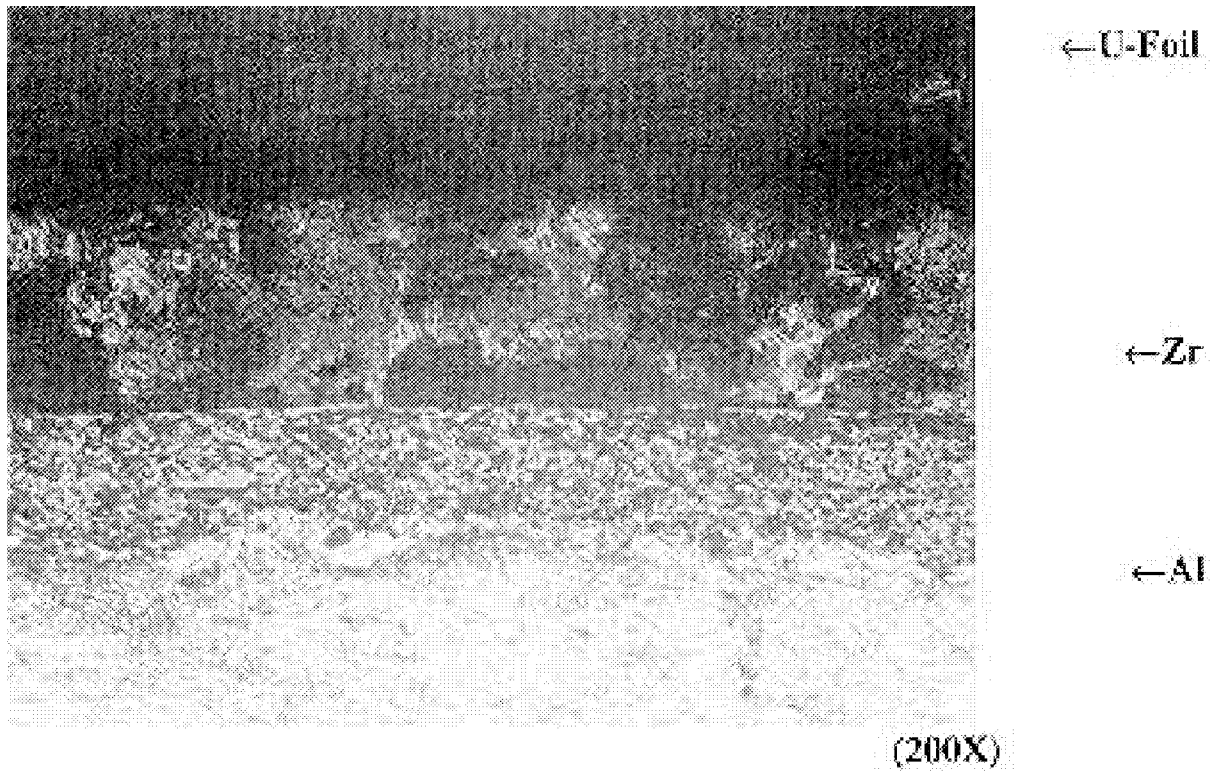
(500X)

Figure 2. Cross Section of First Target Showing Interaction of U-foil and Al Inner Tube



← U-foil

**Figure 3. Extracted Al-Zr Coated Inner Tube with Bonded U-Foil**



**Figure 4. Cross Section of Extracted Al-Zr Coated  
With U-Foil Bonded to Zr Coating**

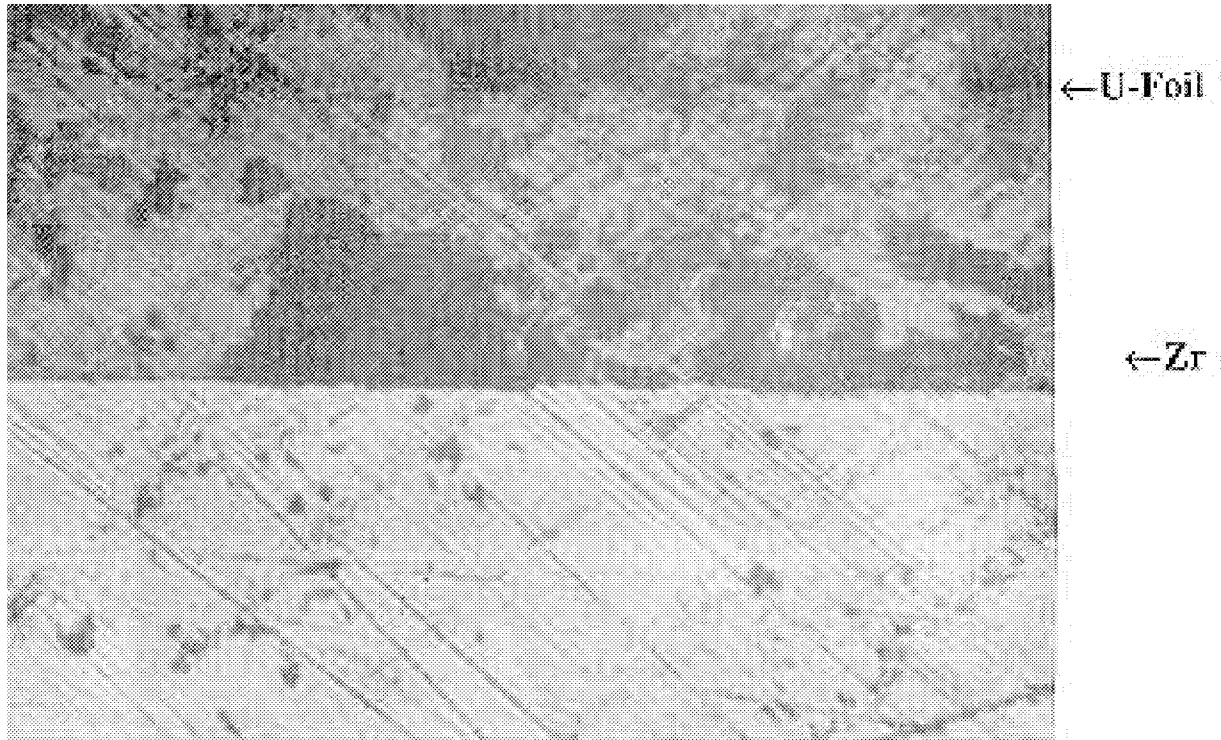
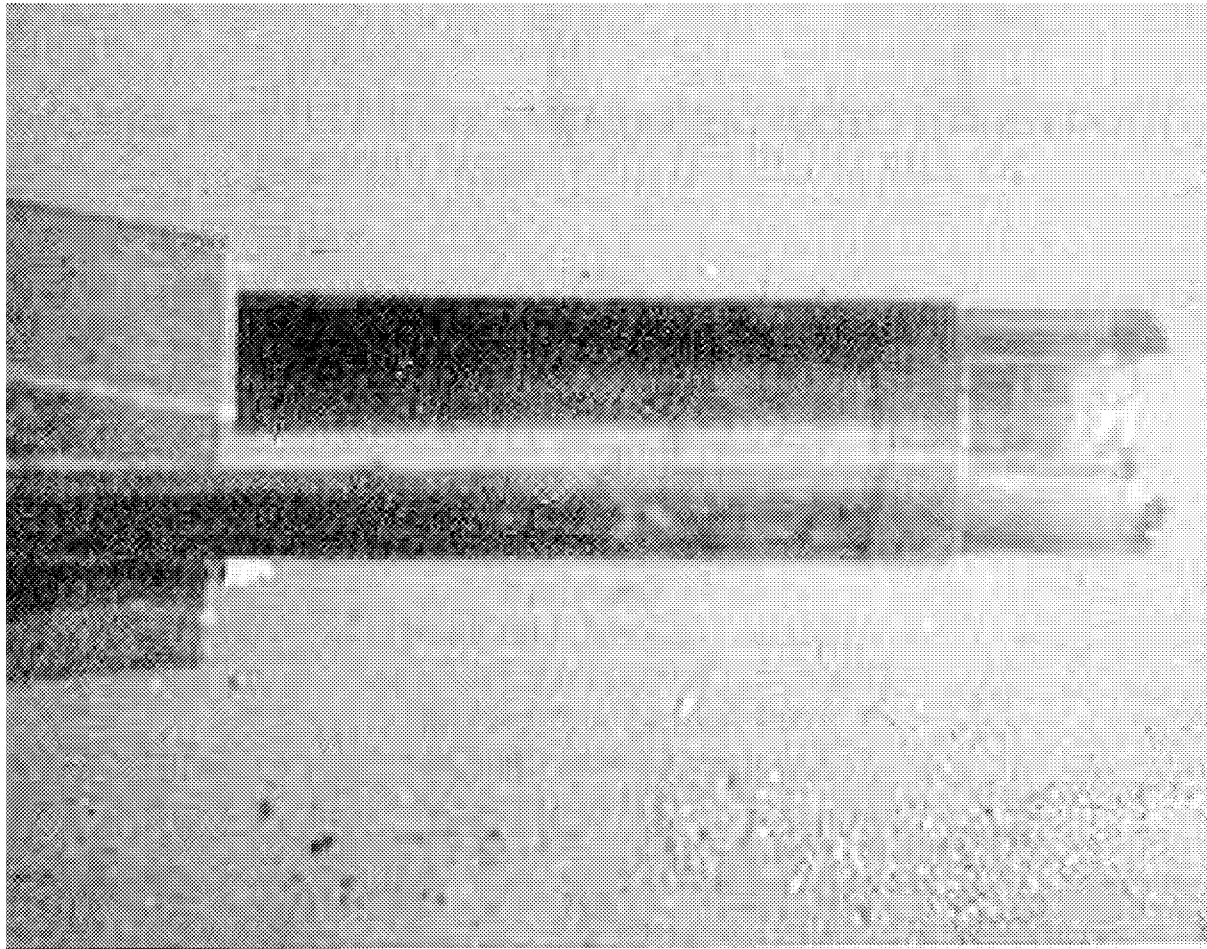
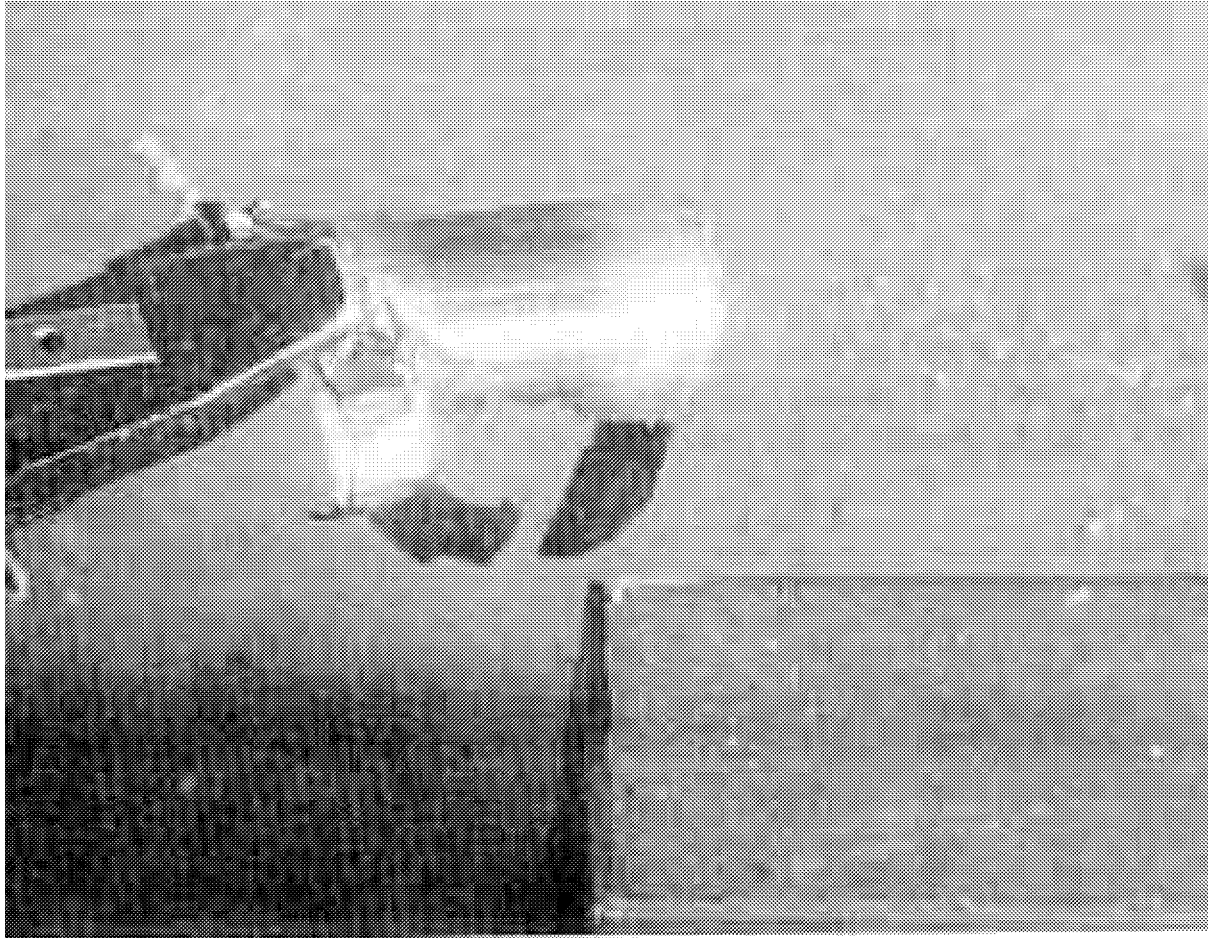


Figure 5. Cross Section of Zr-U Interface in all-Zr Target



**Figure 6. Cu-U-Cu Foil Partially Extracted**



**Figure 7. Cu-U-Cu Foil Removed from Target Tube**