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IRRADIATION TESTS OF ^{99}Mo ISOTOPE PRODUCTION TARGETS EMPLOYING URANIUM METAL FOILS

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ABSTRACT

Most of the world's supply of ^{99m}Tc for medical purposes is currently produced from the decay of ^{99}Mo derived from the fissioning of high-enriched uranium (HEU). Substitution of low-enriched uranium (LEU) metal foils for the HEU UO_2 used in current target designs will allow equivalent ^{99}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}Mo. This work comprises both target and chemical processing development and demonstration. Two major target systems are now being used to produce ^{99}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 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 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 has been reported at previous RERTR meetings.[1] Briefly, the test target shown in Fig. 2 is externally identical in dimensions (except length), and appearance to the presently used UO_2-coated "Cintichem" target. The fissile part of the target is a thin (125-μm-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[1] -- 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_3. As will be detailed in the following section, several modifications led to a successful design in which thin (10-μm) non-fissile foils were installed between the U 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	$U^{(1)}$	inextractable U-Al interaction
2	Zr	air oxidized	Al	Zr coated	$U^{(1)}$	extractable U-AlZr bonding
	Zr	air oxidized	Mg	as-machined	$U^{(1)}$	extractable U-Mg bonding
	Zr	air oxidized	Zr	air oxidized	$U^{(1)}$	inextractable U-Zr bonding
3	Zr	air oxidized	Al	as-machined	Ni-$U^{(2)}$-Ni	extractable Al-Ni bonding
	Zr	air oxidized	304SST	as-machined	$U^{(2)}$	inextractable
	Zr	air oxidized	304SST	as-machined	$U^{(2)}$-Ni	extractable U-SST bonding
	Zr	air oxidized	304SST	as-machined	Cu-$U^{(2)}$-Cu	extractable foil easily removed
(1) Pure uranium, β treated						
(2) Adjusted uranium, β treated						

TEST NUMBER ONE

A single target was irradiated in this test. This target consisted of an anodized Al inner tube, an air oxidized Zr outer tube, and a β-treated pure U foil.

Out-of-reactor heating tests had shown that during six days at 400°C no interdiffusion between either Al or Zr and U 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_2 to precipitate the gas, after which the swaglok 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.

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 Zr outer tube with a plumbers 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 2, there was substantial interaction between the U foil and the Al 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 U and Al results in the formation of, primarily, UAl_3, an intermetallic compound with a much lower density than U. However, there appears to be no indication of interaction having occurred between the U foil and the outer Zr tube.

TEST NUMBER TWO

Three targets were irradiated in this test, each one with a different inner tube. In one target the Al inner tube was retained but it was coated with Zr by means of flame spraying. This choice was based on the apparent non-bonding of U foil and outer Zr tube in the first test. A second target contained a Zr inner tube for the same reason; however, this combination lacked the thermal expansion difference. The third target contained a Mg inner tube. Mg has all the desirable properties of the Al but does not form compounds with U. The Al (Zr) and Mg inner tubes could both be extracted from the outer tube without difficulty. The U foils, however, were firmly bonded to both, (see for example Fig. 3) and could not be removed. The Zr inner tube could not be extracted from the all-Zr 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 U and Mg, or U and the Zr coating on Al (see Fig. 4 and 5). Intimate contact and an intense flux of fission recoil atoms at the U surface apparently results in sufficient atomic mixing at the interface to affect a strong metallurgic 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 U foil-tube interface.

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

Four metals were thus selected as barrier candidates: Cu, Ni, Fe, and Zn. The first three are suitable for the acid dissolution process while Zn can be used for a base process as well. For the current test Cu and Ni were selected. Since the recoil range in Cu and Ni is approximately 7 μm, a 10-μm-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 U 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 U 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 Zr, satisfying that criterion as well.

Another concern was raised during the examination of the second test. The U 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 U-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 U foils. It was decided to use so-called "adjusted" U for the third test. This required addition of small amounts of Al and Fe to the U, which drastically reduced the grain size in the finished U foils.

The third test was thus comprised of the following four targets. Three targets with a type 304 stainless steel inner tube, of which one had no recoil barriers (serving as a control sample), a second had a 10-μm foil at the U-outer Zr interface (this target represents the option of immersing the inner tube together with the U foil), and a third had Cu foils on both inner and outer foil-tube interface (a Cu-U-Cu sandwich).

To round out the test, an Al inner tube with Ni foils at both interfaces (a Ni-U-Ni sandwich) was added. Time constraints did not permit anodizing the Al 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 Ni foil at the U-Zr outer tube interface was easily extracted. There was no evidence of bonding between the Ni foil and the outer Zr tube. The foil (U-Ni combination) was firmly locked to the SST inner tube as

expected. The target with Cu on both sides of the U foil was also easily extracted and the Cu-U-Cu foil (sandwich) could be easily slid off the inner SST tube (see Fig. 6 and Fig. 7). Small samples were cut from both targets for chemical dissolution studies. The target with a SST inner tube, without recoil barriers on either side could not be extracted. The target with an Al inner tube and (Ni-U-Ni) foil combination was easily extracted; however, the Ni inner foil appeared bonded to the Al 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 Ni-Al 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 Al inner tube surface will prevent this interdiffusion.

CONCLUSIONS

Low enriched uranium metal foils can be conveniently removed from tubular target assemblies after irradiated. Recoil-atom absorbing barriers are necessary between U foil and 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 ^{99}Mo Target Fabrication and Testing," Proceedings of 1994 International Meeting on Reduced Enrichment for Research and Test Reactors, Williamsburg, VA USA (1994).

PRESSED + WELDED

ZIRCALLOY
END PLUG

ALUMINUM
INNER TUBE
TAPERED
~0.005"/1.0"

URANIUM
FOIL

6"

ZIRCALOY
OUTER TUBE
TAPERED

ZIRCALLOY
END PLUG
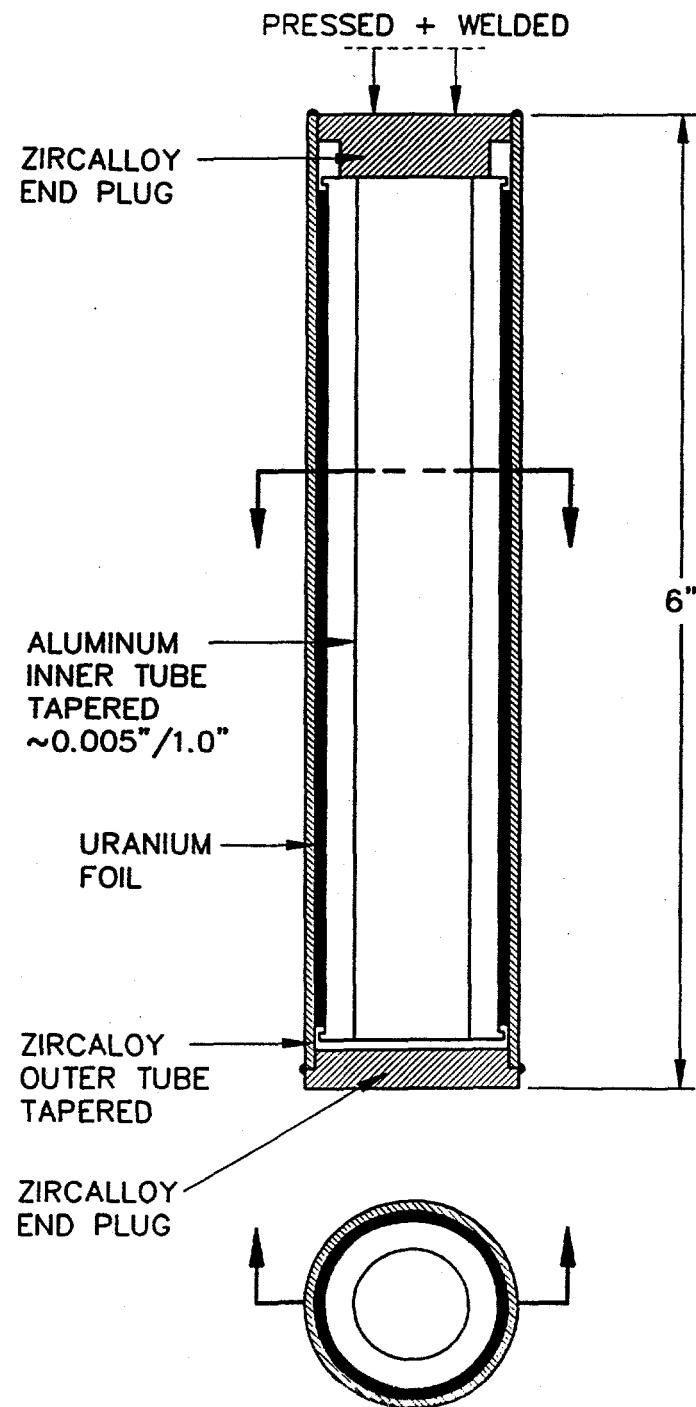
Figure 1. Duifferential Thermal Expansion (DTE) Target

← Zr

← U-foil

← Al

(500X)
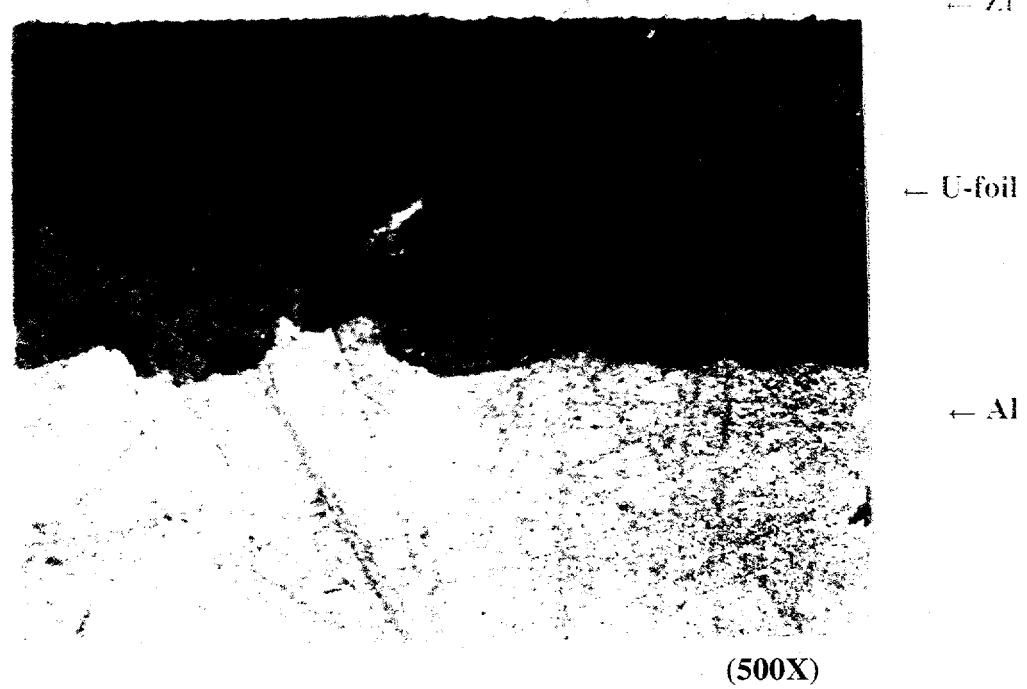
**Figure 2. Cross Section of First Target Showing Interaction of U-foil and Al Inner Tube**
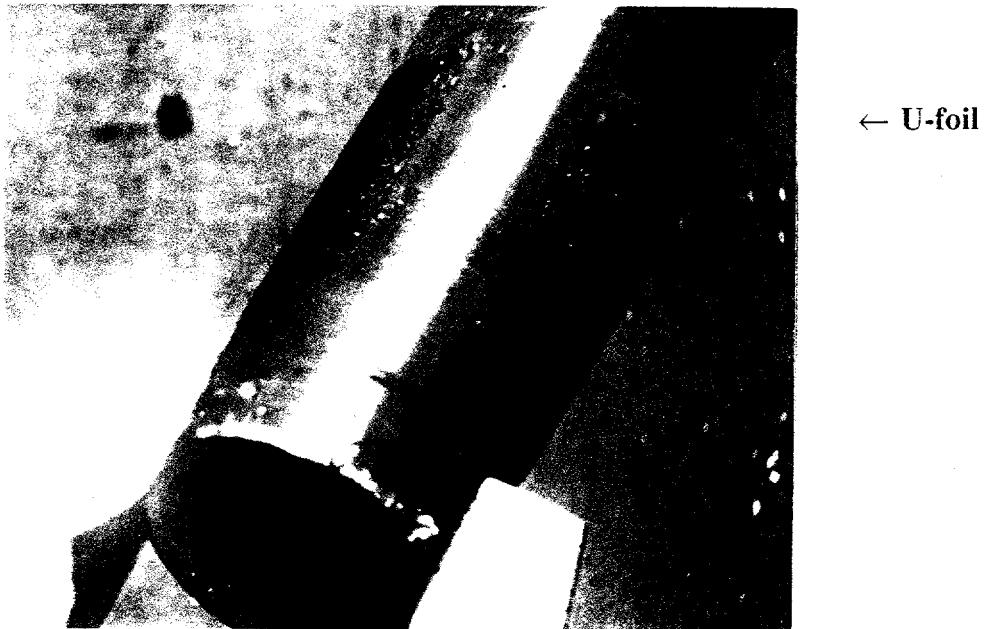
← U-foil

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

—U-foil

← Zr

← Al

(200X)
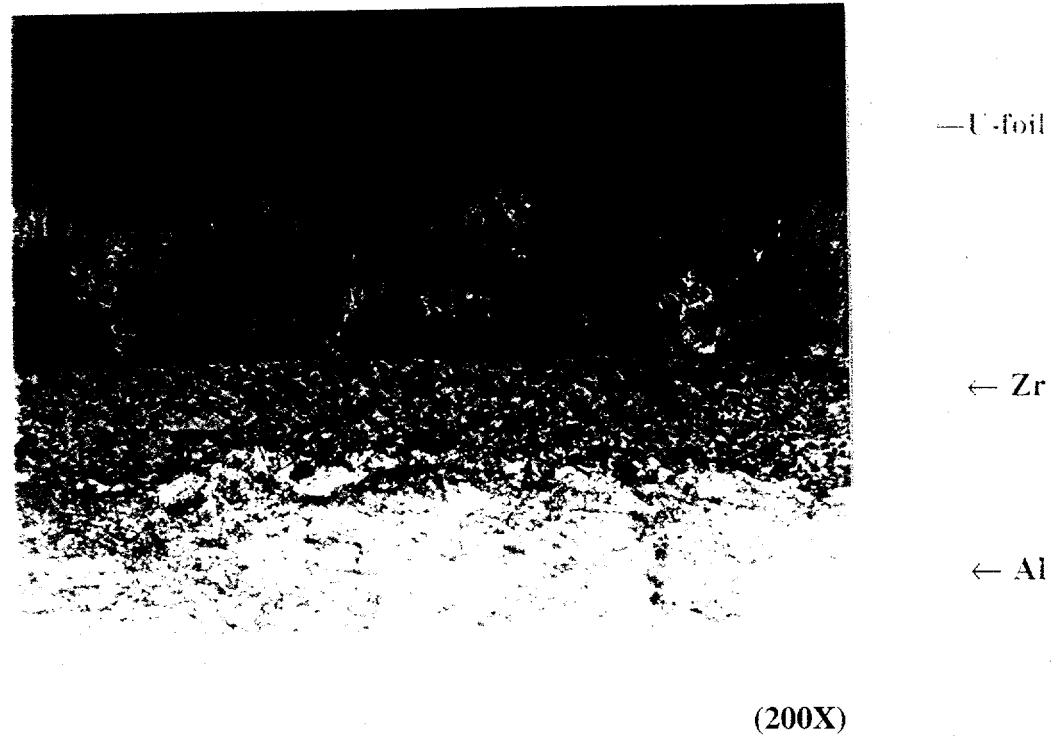
Figure 4. Cross Section of Extracted Al-Zr Coated
With U-foil Bonded to Zr Coating

← U-foil

← Zr
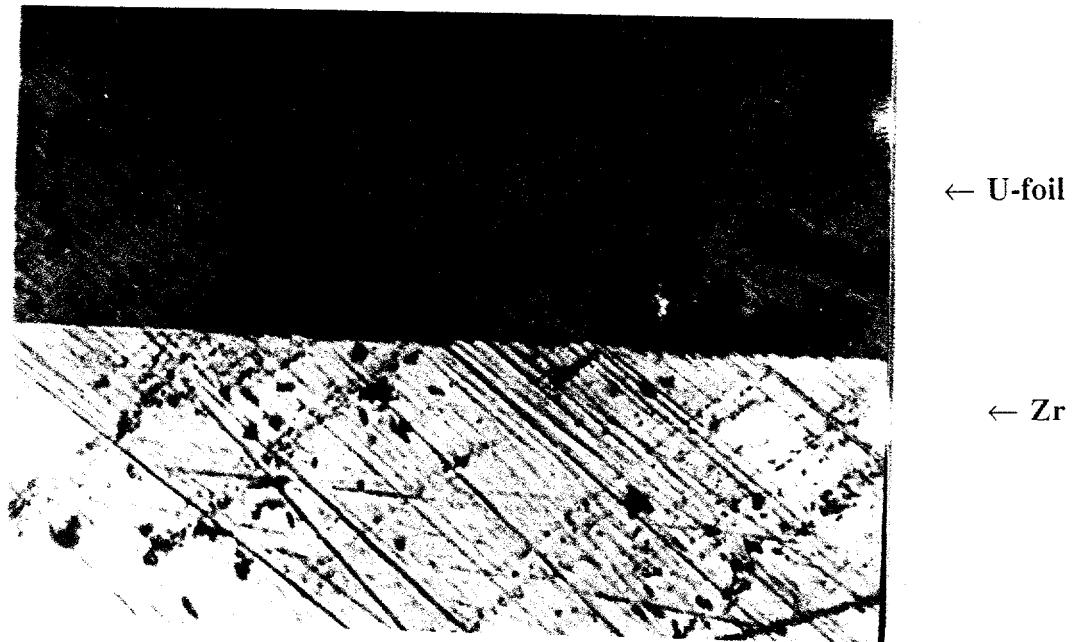
Figure 5. Cross Section of Zr-U Interface in all Zr Target
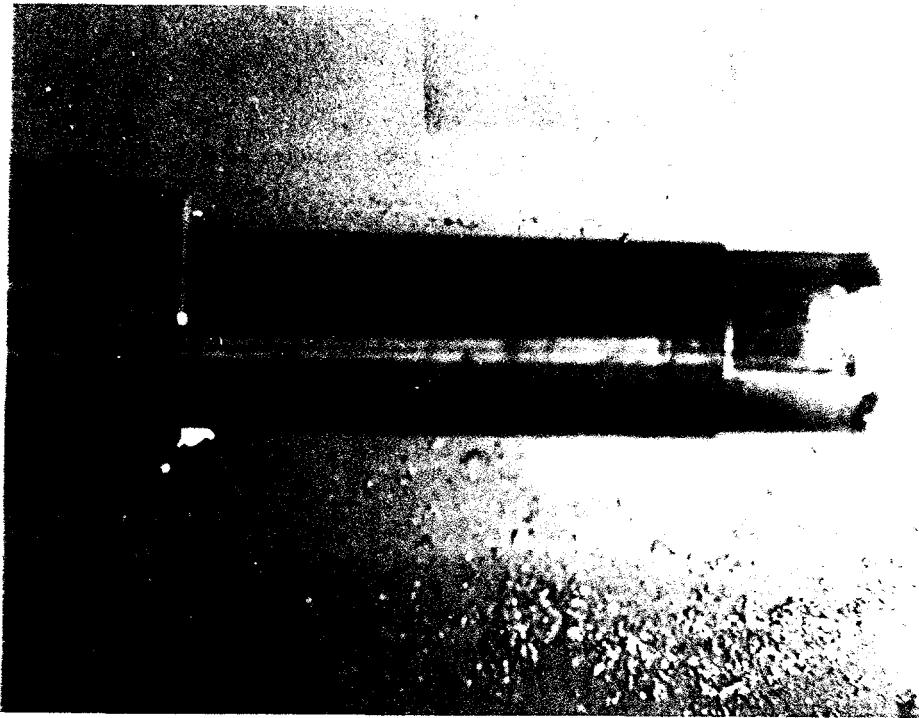
Figure 6. Cu-U-Cu Foil Partially Extracted
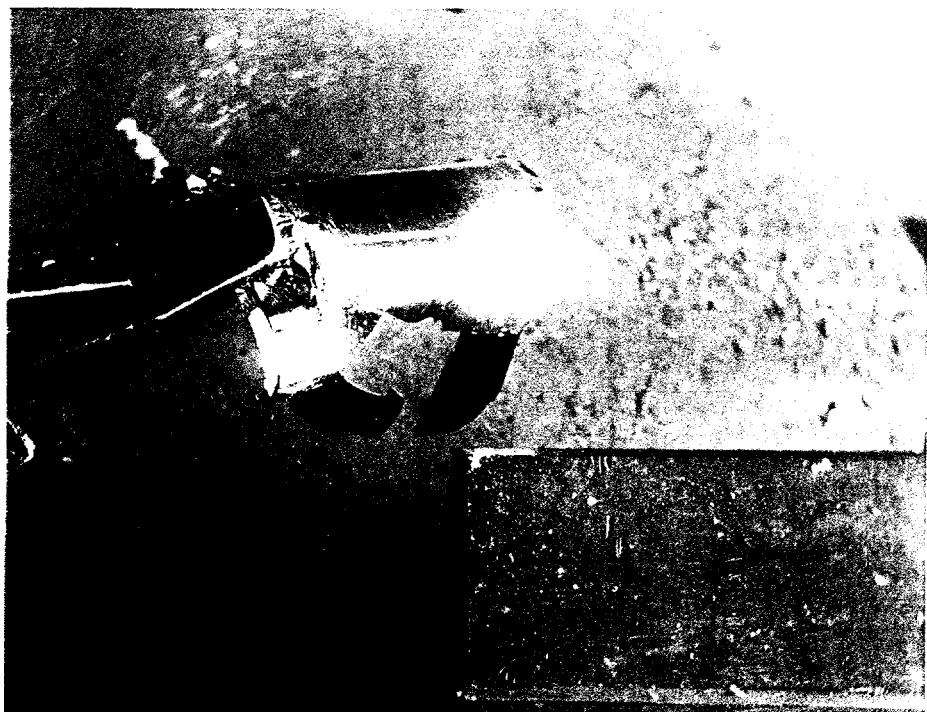
Figure 7. Cu-U-Cu Foil Removed from Target Tube