

Conf-9409107--3

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LEU ⁹⁹Mo TARGET FABRICATION AND TESTING:
OVERVIEW, STATUS, AND PLANS*

by

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September 1994

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For presentation at 17th International Meeting on Reduced Enrichment for Research and Test Reactors (RERTR), September 18-22, 1994, Williamsburg, VA, U.S.A.

*Work supported by the U. S. Department of Energy, Office of International Affairs and Energy Emergencies, under Contract No. W-31-109-ENG-38

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ABSTRACT

As part of the RERTR program, the development of technology to use low-enriched uranium (LEU) for production of the fission product ⁹⁹Mo has continued. Progress in fabrication development and out-of-pile thermal testing of targets based on uranium metal foils is summarized. Uranium foil of 125 μm (0.005 in.) thickness has been fabricated. Heat treatments have been developed to provide a random crystal structure, which is required for satisfactory irradiation properties. Two target designs, a tapered thermal expansion type and a split-outer-tube type, are presented. After anionic coating and thermal treatment of prototypical targets, no diffusional or dimensional changes were observed. A formal agreement for cooperation between Argonne National Laboratory (ANL) and the Indonesian National Atomic Energy Agency (BATAN) for irradiation testing is in the final stage of negotiation. The first irradiation tests of targets fabricated by ANL are scheduled to begin during the first half of 1995.

INTRODUCTION

As reported last year,¹ the RERTR Program has reactivated its effort to develop use of low-enriched uranium (LEU) to produce the fission product ⁹⁹Mo. This work comprises both target and chemical processing development and demonstration. Two major target systems are now being used to produce ⁹⁹Mo 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 UO₂. An LEU replacement for the former is feasible from the target-performance standpoint using a U₃Si₂-aluminum dispersion, whose outstanding irradiation performance properties have already been demonstrated. Considerable work is needed, however, to develop and demonstrate a chemical process for extracting the ⁹⁹Mo. Progress in this area will be reported in another paper at this meeting.² Both a target and its associated chemical processing must be developed to replace the deposited UO₂ target. This paper summarizes progress in fabrication development and in out-of-pile thermal testing of targets based on uranium metal foils. Chemical processing of foil targets will be discussed in a later paper at this meeting.³

Before discussing our progress in target development, we will summarize our overall development plans. Today, Indonesia is the only active user of deposited UO₂ targets. For that reason and because the U.S. no longer has suitable irradiation facilities for testing ⁹⁹Mo targets, we will work with the Indonesian National Atomic Energy Agency (BATAN) to test the uranium metal foil targets and their chemical processing. A formal agreement for cooperation is in the final stage of negotiation. BATAN is already working on

the safety analysis for the irradiation, and we plan to perform the initial irradiation tests during the first half of 1995. The first test targets will be fabricated at ANL, but future targets may be fabricated in Indonesia. Chemical processing studies are under way at ANL and at the University of Illinois. Several tests on irradiated targets are anticipated during the next year in Indonesia. We are in the early stages of negotiating a similar cooperation agreement with the Argentine National Atomic Energy Commission (CNEA) for the testing of U_3Si_2 -aluminum dispersion targets. Because target irradiation performance is not an issue here, we will concentrate on chemical processing. Work is also underway in this area at ANL and the University of Illinois-Urbana-Champaign.

EQUIPMENT AND EXPERIMENTAL PROCEDURES

Because all of the proposed irradiation target designs for the production of ^{99}Mo using low-enriched uranium (LEU) are based on use of a clad uranium metal foil, techniques to produce uranium foils were required. A range of 25-125 μm (0.001-0.005 in.) was the goal for the final foil thickness. The decision to produce the foils at ANL was made after an unsuccessful search for a cost-effective commercial source of LEU foil, and also to have in-house control of the processing parameters. After an extensive literature search, a flowchart of current common fabrication methods was produced (Figure 1). Due to safety and time constraints, canning was chosen as the fabrication method to produce foil from cast uranium ingots. Initial rolling experiments were performed with 50 x 50 x 6 mm (2 x 2 x 0.25 in.) depleted uranium plate stock, which was previously cast and hot-worked. This material was canned in plain carbon steel and rolled on a 12 x 20 cm (5 x 8 in.) two-high Stanat mill in the high alpha uranium range ($\approx 625^\circ C$). After hot-rolling, the material was cold-rolled on a 15 x 30 cm (6 x 12 in.) four-high Bliss mill.

The as-cold-rolled foils were encapsulated inside Vycor tubes filled with argon gas (99.95%) at 15 in.-Hg partial pressure. Small zirconium chips were sealed inside the tubes to prevent high-temperature oxidation of uranium during the β -treatment. Two Vycor tubes were heated to $690^\circ C$ at $4^\circ C/min$, held for 3 min, and then quenched in water and in air. Another tube was air-cooled after a 10 min hold at $690^\circ C$. Texture analysis of the uranium foils before and after the β -treatment was made by X-ray diffraction, using a Philips XRG-3100 X-ray diffractometer with a copper target. Voltage and current used were 40 kV and 15 mA, respectively. The scanning range of 2θ was between 25° and 65° , and the collecting division and time were 0.05° and 2 sec, respectively.

Prototype one-third-scale targets of the tapered plug/thermal expansion design and split-outer-tube design were assembled and thermally tested from $300-500^\circ C$ for up to one week. Details of these designs were previously presented.¹

RESULTS AND DISCUSSION

Foil Texture

The first uranium foil produced was 125 μm (0.005 in.) thick. The resulting sheet had a very good surface finish, and no pinholes were observed.

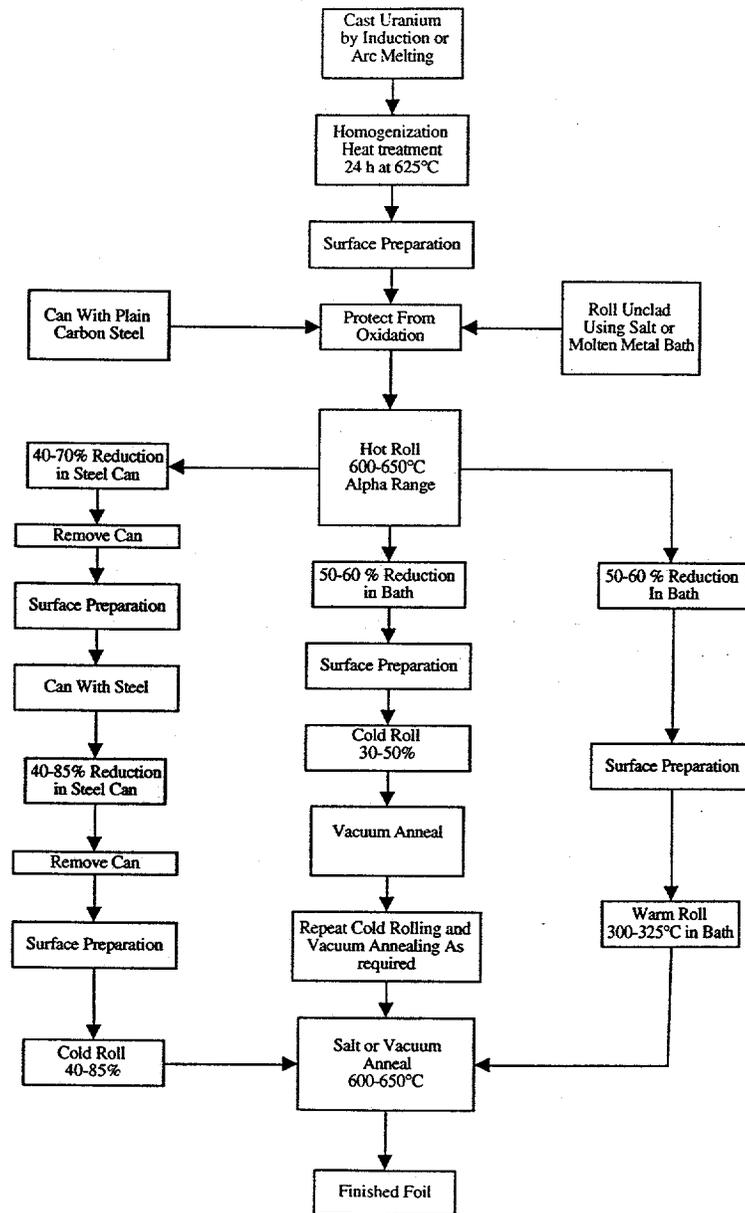


Figure 1. Production of Foil from Cast Uranium

A highly textured structure is inevitably formed after the rolling process,⁴ which causes undesirable anisotropic growth that leads to undesirable surface effects such as swelling during irradiation. Paine et al.⁵ showed that uranium single crystals elongate in the [010] direction and contract in the [100] direction with no appreciable change along the [001] direction. Buckley⁶ explained this phenomenon as mass transport from {110} planes to {010} planes, based on the observation that fission-generated interstitials and vacancies form loops on {010} planes and {100} planes, respectively, as the result of anisotropic thermal expansion induced in uranium by thermal spikes in displacement cascade. In general, randomly oriented and moderately fine-grained uranium is necessary for successful irradiation behavior. β -treatment after rolling is essential to eliminate the foil texture.

Figure 2 shows the relative ratios of the characteristic peaks, (110), (002), (111), (112), and (131), to the (021) peak (the strongest of the random sample) in each sample in the cold-rolled and various β -treatment conditions. Apparently, strong texture was generated after cold-rolling because substantially higher ratios of (110), (111), (112), and (131) peaks than those of the random sample are observed. However, the as-rolled texture was different from that reported by Adam and Stephenson,⁷ since most of the (010) planes were perpendicular to the rolling direction after cold-rolling. Relatively high ratios of (111), (112), and (131) peaks were found for the sample that was air-cooled after 10 min at 690°C. This may be attributed to the preferred growth of these planes along the rolling direction due to prolonged exposure at elevated temperature. The sample that was water-quenched after a 3-min hold at 690°C showed higher (110)/(021) and (111)/(021) ratios than those of the random sample. This implies that a certain amount of cold-rolled texture still exists, and that 3 min at 690°C followed by a water quench may not be adequate for completion of the α - β transformation. Comparably, the specimen that was air-cooled after 3 min at 690°C exhibited more random microstructural characteristic than the other two specimens. In summary, we suggest that β treatment consist of an isothermal soaking (3 min at \approx 690°C) and air-cooling to minimize undesired texture of cold-rolled uranium foils.

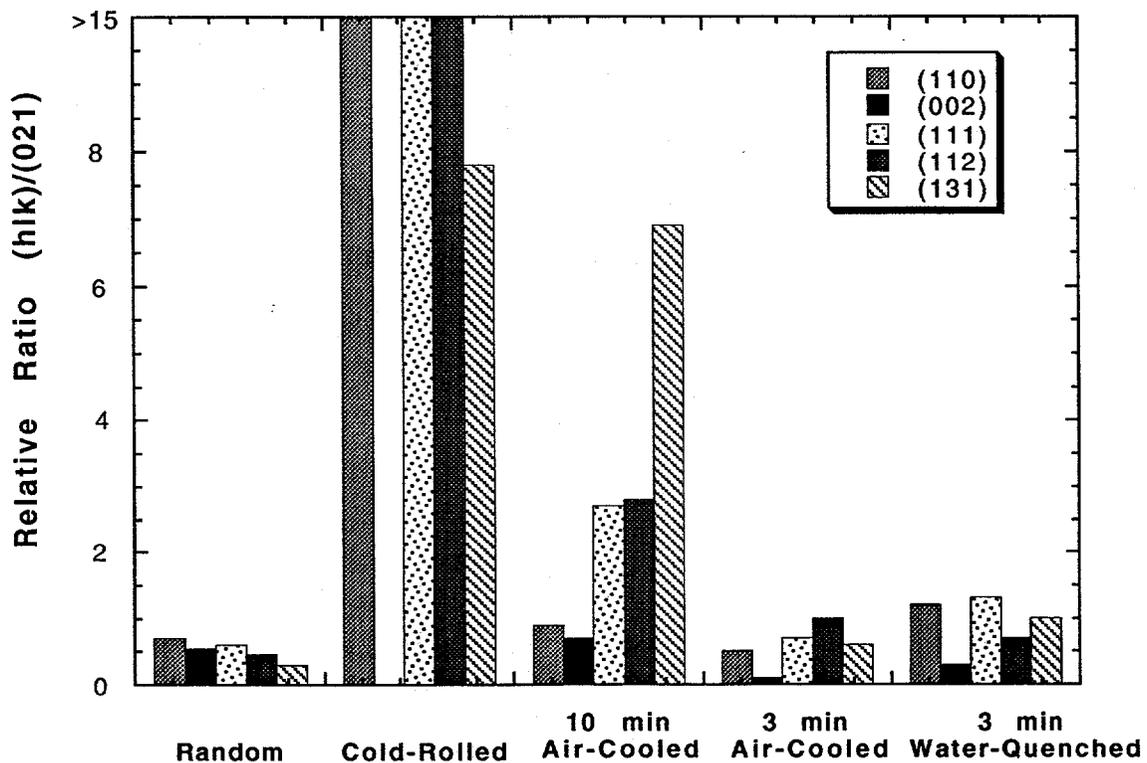


Figure 2. Relative Ratios of (h1k) Peak to (021) Peak for Various Metal-Working and β -Treatment Conditions

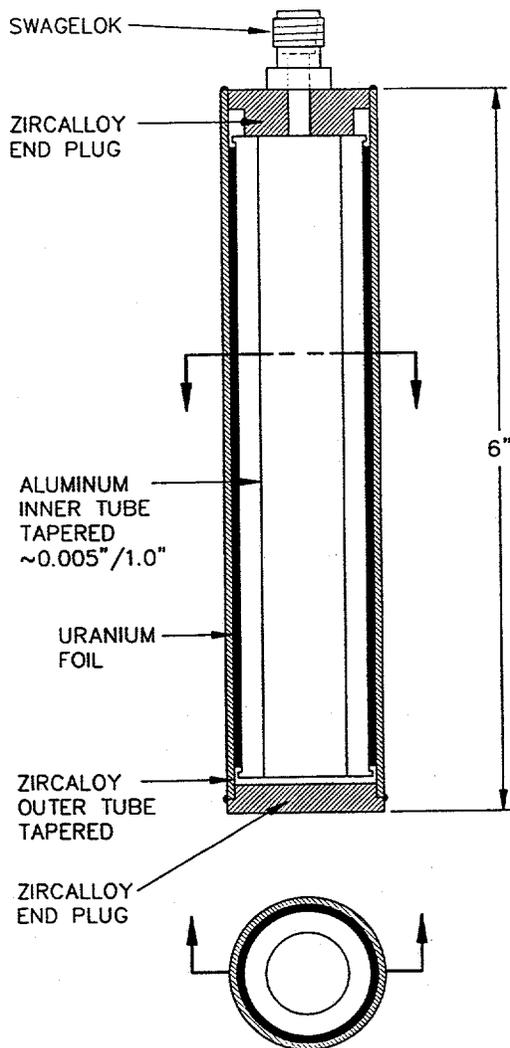


Figure 3. Differential Thermal Expansion (DTE) Target

Other design improvements are being considered. If a tighter fit between the cladding and the foil is required, the outer cladding could be fabricated from an aluminum alloy such as 2219 Al, which has a lower coefficient of thermal expansion than the 6061 Al inner tube. The effect would be similar to that of the DTE target but on a lesser scale. To eliminate the need for a gas-removal fitting and to simplify the production and cost of each target, a device may be designed to seal onto a target, pierce the outer cladding and allow for gas removal.

SUMMARY AND CONCLUSIONS

Two designs of irradiation targets have been developed for the production of ^{99}Mo using LEU. Both concepts, a tapered thermal expansion design and a split-outer-tube target design, are based on cladding over an LEU foil. Out-of-pile testing of LEU targets for the production of ^{99}Mo has progressed to the stage where preparation for irradiation testing has begun. After satisfactory irradiation of the prototypes is demonstrated, scale-up to full-size targets is planned.

Differential Thermal Expansion Target

The Differential Thermal Expansion (DTE) target design is shown in Figure 3. The principal feature of this design is the use of two tubes of materials with different thermal expansion coefficients, between which is situated the U metal target. By installing Zr, which has the lower coefficient of thermal expansion, on the outside, good contact and thus good heat transfer between the U foil and both tubes is ensured when the target temperature is raised during irradiation. The outer tube is Al. Assembly and disassembly of the target is facilitated by machining a fitting taper on both tubes. The outer Zr tube is sealed by welding a blind bottom plug and a top plug equipped with a swagelock fitting to allow removal of fission gas from the target after irradiation. The target is evacuated and filled with He prior to sealing.

To prevent interdiffusion of U with either Al or Zr during irradiation, the Al inner tube was black-anodized and the Zr outer tube was air-oxidized at 300°C to a dark blue color.

Several 15 cm (6 in.) targets were fabricated and heat-treated for approximately one week, after which they were disassembled (the U foil removed) and metallographically examined; test results are summarized in Table 1. The test temperatures were deliberately chosen to be well above the expected irradiation temperature in order to establish whether interdiffusion was likely to occur. Only the non-anodized-oxidized target, which was heat-treated at 400°C, could not be disassembled due to interdiffusion of U and Al (Figure 4). In contrast, no interdiffusion was evident in the other targets (Figure 5). An example of a disassembled target is shown in Figure 6.

The results of the assembly, heat treatment, and disassembly experiments with these DTE targets give us confidence to proceed with the further design and fabrication of in-reactor test targets. Application of oxide diffusion barriers on both the Al and Zr surfaces appear to provide ample protection against diffusion bonding between the U foil and target tubes.

Split-Outer-Tube Target

The latest design of the split-outer-tube target design is shown in Figure 7. A port has been added to allow removal of the irradiation gases before the irradiated target is disassembled. Fittings identical to those of the current production design will be used to facilitate compatibility with existing equipment.

As detailed in the previous section, an anodized coating on the aluminum prevented diffusion during thermal heat treatments. Because it was assumed that the split-outer-tube target would behave in the same manner; no thermal diffusion tests on this design were performed. A single sample of a scaled-up 15 cm (6 in.) version of the initial 7 cm (3 in.) design was assembled and thermal-cycled four times at 400°C over a 168-hr period. No changes in dimensions were detected and destructive examination of the target after the test showed no gap or diffusion.

Table 1. Testing Conditions of Differential Thermal Expansion (DTE) Targets

Capsule	U Foil	Glovebox Atmosphere	Temperature (°C)	Time (h)	Swelling on Center	Remarks
1	4	Ar-He	Not Heat Treated	-	-	Easily assembled.
2	4	Ar-He	300	144	<1 mil (<0.025 mm)	Foil and Al came out as one piece. Foil removed from Al with a little prying.
3	4	Ar-He	300	156	<1 mil (<0.025 mm)	Consists of 4 cool-down cycles. Cycling did not affect the interface.
4 Anodized	5	He	400	168	4 mil (0.10 mm)	After sectioning ends, foil remained tight around centered Al rod.
5 Anodized	5	He	500	168	1 mil (0.025 mm)	Easy fit. Foil removed very easily.
6	5	He	400	168	12 mil (0.30 mm)	Appeared to have a U-Al reaction.



Figure 4. Unanodized Capsule #6 after Heat-Treatment for One Week at 400°C Showing Interaction Between U and Al. ≈25x

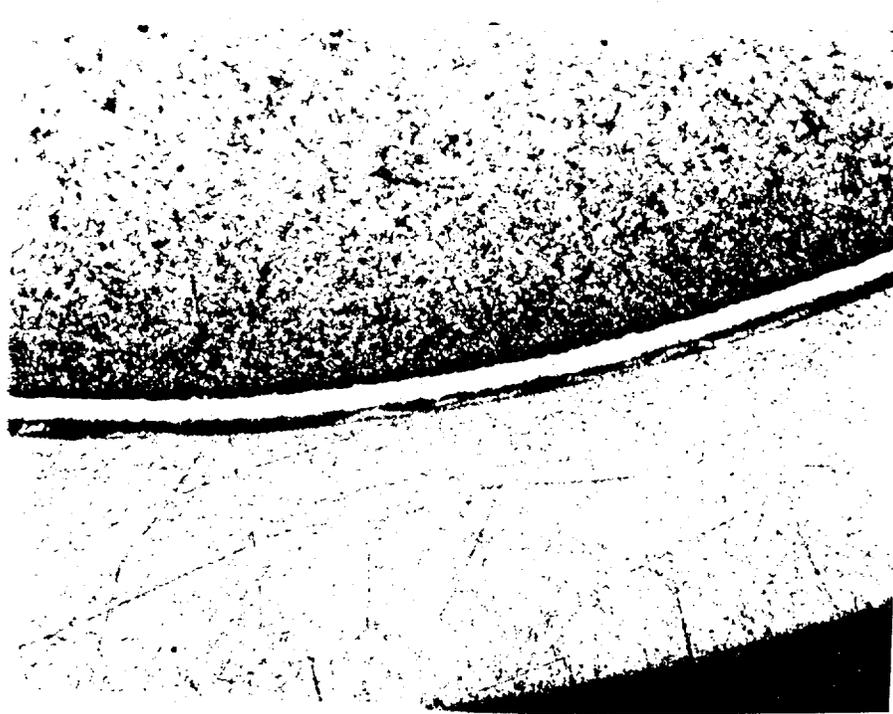


Figure 5. Anodized Capsule #4 after Heat-Treatment for One Week at 400°C Showing No Interaction Between U and Al. $\approx 25\times$

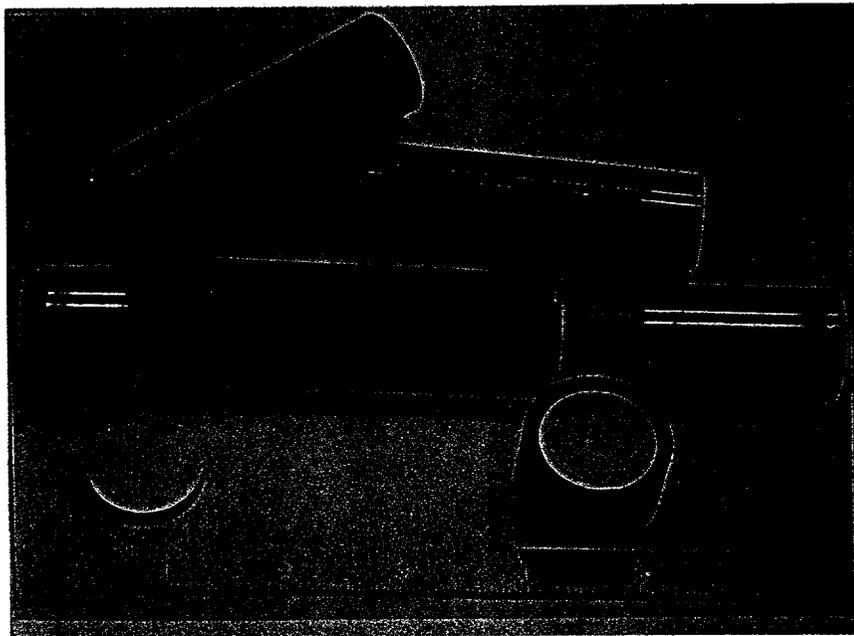


Figure 6. Disassembled DTE Target after Heat-Treating Showing Uranium Foil Removed (center)

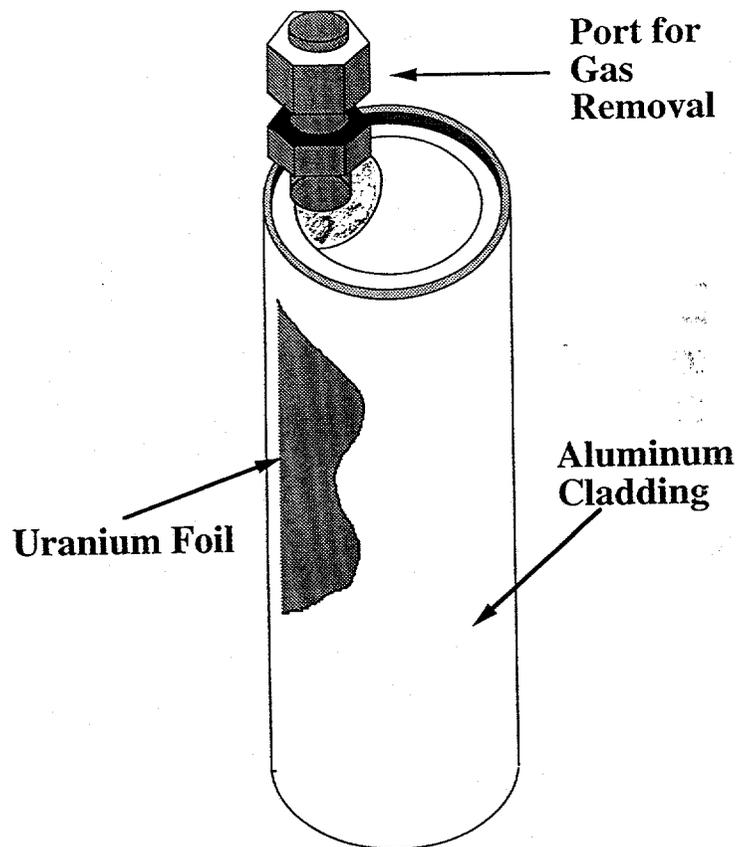


Figure 7. Split-Outer-Tube Target

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