

DEVELOPMENT OF ANNULAR TARGETS FOR ⁹⁹MO PRODUCTION-1999*

C. Conner, E. F. Lewandowski, J. L. Snelgrove,
M. W. Liberatore, D. E. Walker, T. C. Wiencek,
D. J. McGann, G. L. Hofman, and G. F. Vandegrift

Argonne National Laboratory
Argonne, Illinois U.S.A.

RECEIVED
JAN 18 2000
OSTI

To be Presented at the
1999 International Meeting on
Reduced Enrichment for Research and Test Reactors

October 3-8, 1999

Budapest, Hungary

The submitted manuscript has been created by the University of Chicago as Operator of Argonne National Laboratory ("Argonne") under Contract No. W-31-109-ENG-38 with the U.S. Department of Energy. The U.S. Government retains for itself, and others acting on its behalf, a paid-up, nonexclusive, irrevocable worldwide license in said article to reproduce, prepare derivative works, distribute copies to the public, and perform publicly and display publicly, by or on behalf of the Government.

*Work supported by the U.S. Department of Energy,
Office of Nonproliferation and National Security,
Under Contract No. W-31-109-38-ENG.

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, make 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.

the heat to be dissipated to the coolant, the uranium foil must be in good thermal contact with the target structural materials (i.e., the target tubes). If there is poor thermal contact between the uranium foil and the target tubes, the temperature in the uranium foil will increase until the point where uranium will melt and/or react with the fission-recoil barrier (see below) or the target structural materials, a condition that must be avoided.

The original target for irradiating the uranium foils encapsulated the foil between tapered inner and outer tubes. When the inner tube was pushed into the outer tube, the taper would push the foil against the outer wall of the target tube, thus providing good thermal contact of the foil with the target wall [15, 16]. After irradiation, the ends of the target were cut off, the inner tube was pushed out, and the uranium foil was retrieved for subsequent recovery of ^{99}Mo . Fission-recoil barriers were added to the uranium foil to prevent it from bonding to the target tubes [17, 18]. Unfortunately, this target was expensive to make (due to the fabrication of the tapered tubes). Also, without serious modifications, this target allows coolant to flow only over the outer target tube. However, much was learned from this target. We were able to complete several irradiations using this style of target and prove that using LEU metal foil to produce ^{99}Mo was feasible. A new annular-style target was prepared for an August 1999 irradiation in Indonesia.

The design of the new annular target significantly reduces the cost of target fabrication by using target tubes that are not tapered. Also, because the tubes need not be tapered and the inner tube does not need a substantial end surface to accommodate disassembly by pushing, both inner and outer tubes can be made thinner, thus reducing neutron absorption in the reactor and reducing the amount irradiated waste for disposal. In addition, the annular design will allow coolant to flow over the outside and through the inside of the target, effectively doubling the heat transfer surface. In general, the annular target is fabricated by expanding an inside tube into an outside tube (a schematic of the annular target is shown in Fig. 1). As follows:

- A piece of uranium foil is wrapped around the inner tube. A small "undercut" on the inner tube ensures that the uranium foil is positioned correctly within the target.
- The inner tube with the uranium foil wrapped around it is slid into the outer tube.
- The inner and outer tubes with the uranium foil sandwiched between them are slid into a draw die (see Fig. 2).
- The inner tube is then expanded by pulling a draw plug through the inside of the inner tube. The diameters of the inner and outer tubes and of the draw plug are chosen such that the inner tube is expanded plastically and the outer tube is expanded elastically. This creates a target in which the uranium foil is held tightly between the two tubes, effecting good heat transfer within and out of the target.
- After removal from the draw die, the two ends of the target are welded shut.

The target can then be inserted into the reactor using an insertion rig that balances the flow of coolant between the inside and outside of the target. A sketch of a target insertion rig is shown in Fig. 3.

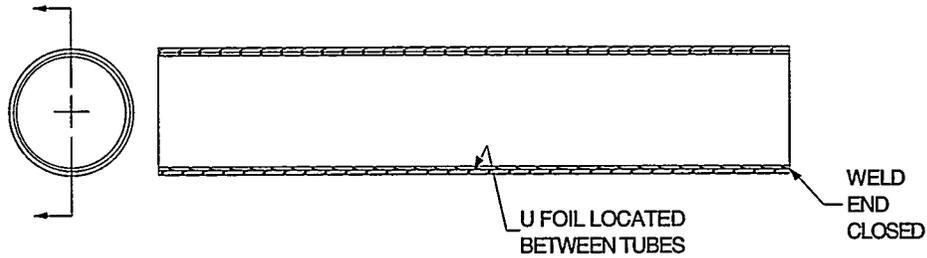


Fig. 1. Annular Target

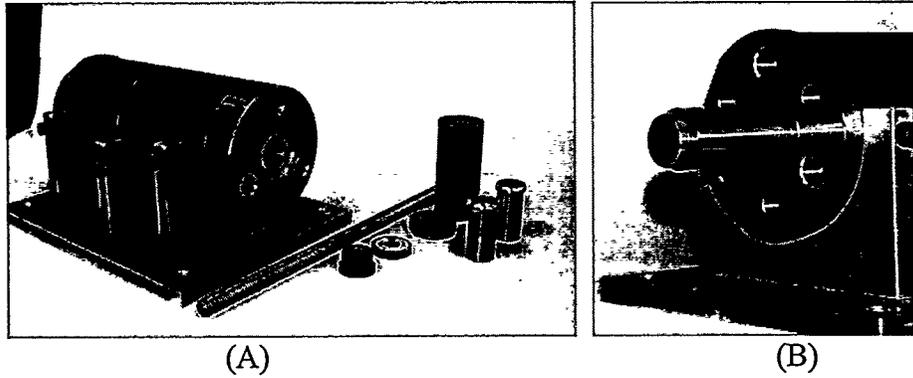


Fig. 2. (A) Draw Die Components, (B) Draw Die with Draw Plug and Target Tubes Exposed

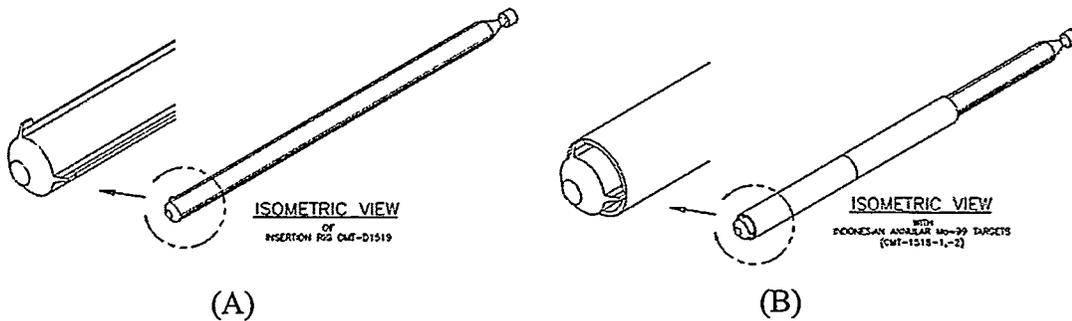


Fig. 3. (A) Target Insertion Rig, (B) Target Insertion Rig with Target Showing Cooling Channel on Inside of Target

THERMAL CYCLING TESTS AND FAILURE ANALYSIS

To verify that the new annular target tubes are in good contact with the uranium foil, several prototype targets were fabricated. Table 1 shows a matrix of the test samples that were prepared. These samples were prepared using Type 3003 aluminum. Most of these samples underwent thermal cycling to simulate the mechanical and thermal performance of the targets during irradiation. Thermal cycling simulates irradiation conditions by "stress relieving" the target tubes. The contact of the tubes with the uranium foil relies on the fact that the outer tube squeezes down on the inner tube, owing to the residual hoop stress remaining from its elastic deformation during drawing. Irradiation in a reactor can relieve stress; if all of the hoop stress in the outer tube is relieved during irradiation, then good contact between the target tubes and uranium may

not be maintained. Even though two targets for the August 1999 irradiation were made from type 702 zirconium, only aluminum samples were tested since zirconium does not stress relieve as readily as aluminum.

Table 1. Prototype Targets Fabricated for Thermal Cycling Tests

Prototype No.	Inner Tube Expansion*	Foil Size and Material
A**	+0.0085" (0.22 mm)	1.75"x3" (44 mm x 76 mm) Cu
B	+0.0095" (0.24 mm)	1.75"x3" (44 mm x 76 mm) Cu
C	+0.0095" (0.24 mm)	1.75"x3" (44 mm x 76 mm) Ni Plated Cu
D	+0.0095" (0.24 mm)	1.75"x3" (44 mm x 76 mm) Zn Plated Cu
E	+0.0115" (0.29 mm)	1.75"x3" (44 mm x 76 mm) Zn Plated Cu
F	+0.0095" (0.24 mm)	1.75"x3" (44 mm x 76 mm) Al Foil Wrapped Cu
G	+0.0095" (0.24 mm)	1.75"x3" (44 mm x 76 mm) DU

*The difference between the ID of the outer tube and the OD of the inner tube was 0.009" (0.23 mm). The number in this column indicates the amount of expansion given to the inner tube.

**Not Tested

The thermal cycling procedure for prototypes B-F was as follows: The targets were placed in a 200°C ($\pm 10^\circ\text{C}$) furnace and intermittently removed over a period of 7 days. To effect a thermal cycle, the targets, after at least 4 hours and up to 54 hours at temperature, were removed from the furnace and allowed to air cool to room temperature (~1 hour). After the targets cooled they were examined, measured, and then returned to the furnace. A minimum of five cycles was performed on each target. Examination of the targets during the tests showed little change. The deviation in the outer diameter of the targets did not exceed 0.001" (25 μm).

After completion of the thermal cycling, the targets were cut into seven small sections. The two end sections were ~1/2" (13 mm) long and the five middle sections were each ~1" (25 mm) long. From left to right the sections were numbered 1-7 (see Fig. 4). Section 4 from prototypes B, C, D, E, and F were used to measure the residual hoop stress after thermal cycling. Hoop stress was measured as follows:

- First the target section was clamped in a circular jig.
- The outer tube was slit using a slitting-saw of known thickness [0.012" (0.30 mm)]. The gap the slitting saw produced, which is typically slightly larger than the slitting-saw blade, was measured using feeler gauges [typically 0.014" (0.36 mm)].
- Then the clamp retaining the outer tube of the target was released, which caused the slit in the outer tube to widen owing to the release of the hoop stress in the tube. The width of the gap was measured again using feeler gauges [typically 0.030"-0.035" (0.76 mm-0.89 mm)].

The difference between the width of the gap before and after releasing the clamp can be used to calculate the residual hoop stress in the outer tube. Values of the hoop stress

ranged from 370 to 530 psi (2.5 to 3.7 MPa); with an average of 440 psi (3.0 MPa). These values indicate that the outer tube is squeezing the inner tube, and, hence, the uranium foil should maintain good thermal contact during irradiation.

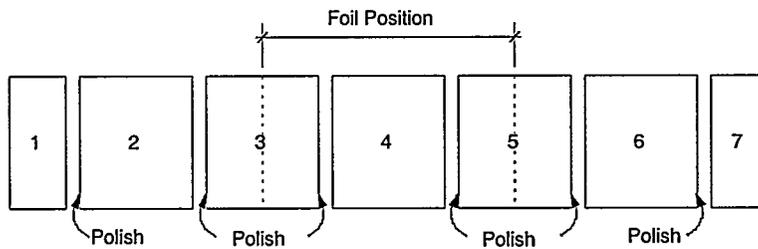


Fig. 4. Sectioning Diagram for Prototype Targets

Sections 2, 3, 5 and 6 were polished for microscopic examination. The faces that were polished are shown in Fig. 4. The right edge of section 3 and the left edge of section 5 were examined to determine the contact of the foil with the target tubes. The other sections were examined to observe the interface between the inner and outer target tubes. This interface is not as critical as the interface between the foils and the tubes, but it does give an indication of the general quality of the target. Microscopic examination of the interface between the target tubes showed that the tubes were tight. In general, a thin line visibly delineates the interface between the inner and outer tubes. In some cases the contact between the two tubes was so good that this line was not visible. Even when the interface was not visible, the outer tube could easily be separated from the inner tube after being slit. (This will be important during disassembly.) Example photomicrographs of target tube interfaces are shown in Figs. 5-7. Microscopic examination of the critical interface between the foil and the target tubes showed excellent contact of the foil with the target tubes. This indicates that the foil is in good thermal contact with the target tubes and that the targets should perform well during irradiation. Sample photomicrographs of the target-tube/foil interface are shown in Figs. 8-12. Figures 13 and 14 show the end of the foil and how the target tubes configure around the foil ends. As seen in Fig. 13, the target tubes draw down tightly around the end of the foil (i.e., the gap at the end of foil gets progressively narrower farther from the foil edge). In Fig. 14, the target tubes have actually cold flowed around the end of the foil to completely encapsulate it. Either configuration is acceptable, because little to no heat transfer is expected to occur out the end of the foil.

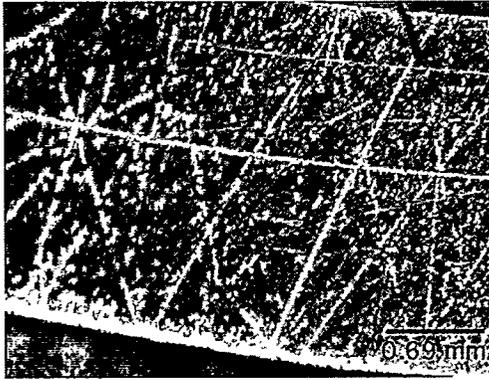


Fig. 5. Section Part B2 – The interface between inner and outer tubes is seen as a thin white line.

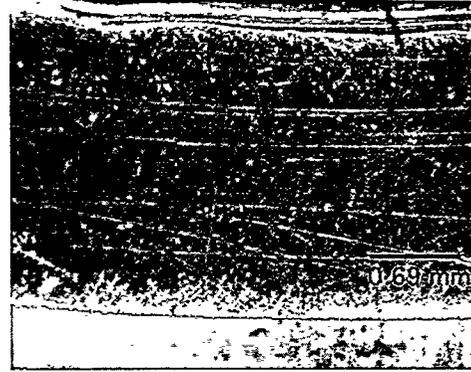


Fig. 6. Section Part D5 – The interface between inner and outer tubes cannot be seen in photomicrograph.

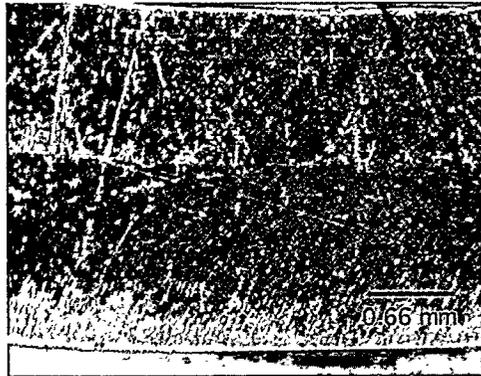


Fig. 7. Section Part E5 – The interface between inner and outer tubes can barely be seen in photomicrograph.



Fig. 8. Section Part B3 – No gaps are apparent in interface between inner and outer tubes and foil.

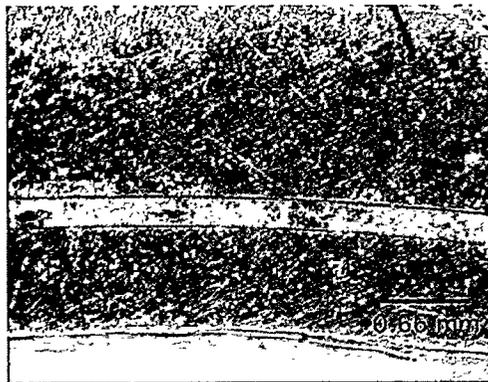


Fig. 9. Section Part C5 – No gaps are apparent in interface between inner and outer tubes and foil.

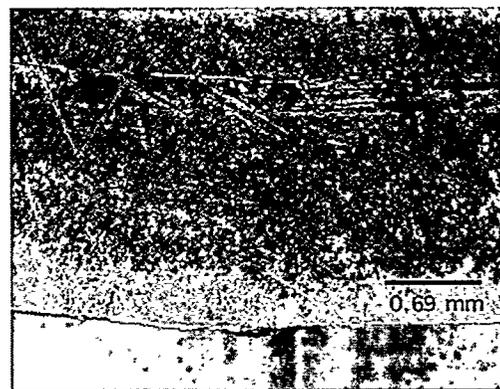


Fig. 10. Section Part D3 – No gaps are apparent in interface between inner and outer tubes and foil.



Fig. 11. Section Part E3 – No gaps are apparent in interface between inner and outer tubes and foil.

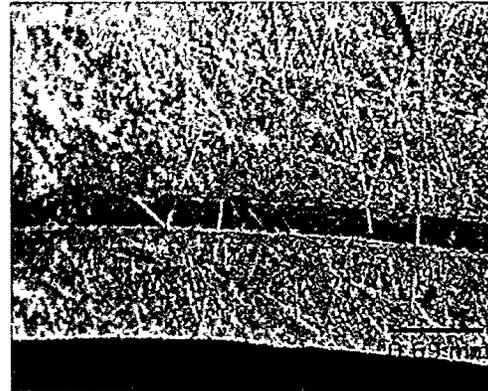


Fig. 12. Section Part F5 – No are gaps apparent in interface between inner and outer tubes and foil.

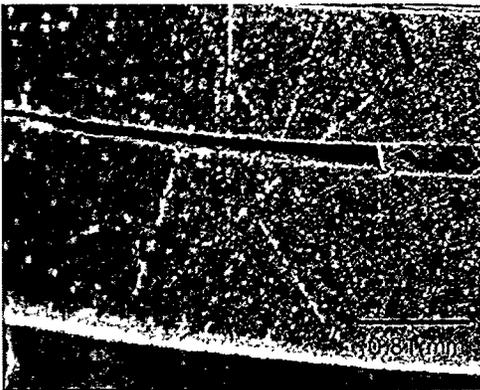


Fig. 13. Section Part B3 – Shows end of foil where target tubes draw down around end of foil leaving small gap.

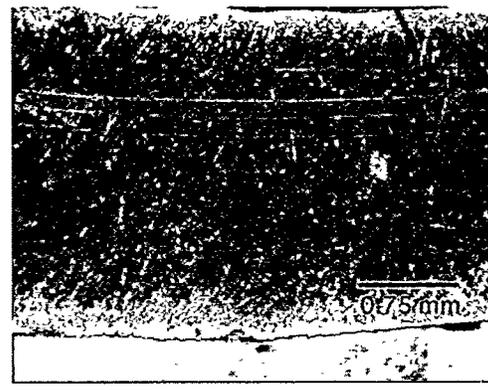


Fig. 14. Section Part D5 – Shows end of foil where target tubes draw down around end of foil completely encapsulating the end of foil.

During past irradiations we have seen that the uranium foil can grow in thickness up to twice the original thickness. An analysis was performed to determine if the resulting pressure on the aluminum inner tube could cause the tube to buckle, thereby disrupting the heat-transfer path from the uranium foil to the inner tube, and to determine if the aluminum outer tube could fail in tension, exposing the foil to the coolant. The strain resulting from the combination of fabrication and uranium growth during irradiation is ~1%. Because of the high ductility of Type 3003 aluminum, the inner tube can accommodate more than ten times the strain resulting from the growth of the aluminum before buckling. For the same reason, the outer tube cannot fail in tension. Since zirconium is so much stronger than aluminum, there is no concern about mechanical failure of zirconium targets.

IRRADIATIONS IN INDONESIA

The thermal-cycling tests confirmed that the new annular targets should perform well during irradiation. Therefore, seven targets of the new annular design were fabricated containing LEU foils. The uranium foil in the targets was adjusted uranium (specified to be approximately 450 ppm iron, 1000 ppm aluminum) and had been heat-treated to produce a fine, random grain structure [18]. Various fission-recoil barriers were added to the uranium foil [17,18]. Table 2 shows the details of the fabricated targets. These targets were then irradiated in the Indonesian RSG-GAS reactor at a reactor power of 15 MW for approximately 120 hours. For insertion into the reactor, the targets were slid onto the reusable irradiation rig, shown in Fig. 3. Each irradiation rig could accommodate up to two annular targets. The overall irradiation performance of the targets was good. There was no evidence of heat-transfer problems during irradiation. All targets were easily removed from the irradiation rigs after irradiation, indicating no significant mechanical distortions during irradiation.

Table 2. Test Matrix for August Irradiations

Target ID	Tube Material	Barrier Material	U Foil Size	Notes
99-9	Zr	Electroplated Ni	1.75" x 3"	Foil designated for processing to ⁹⁹ Mo recovery
99-11	Al	Wrapped with Al Foil	3" x 3.5"	Contact surfaces on Al tube was anodized ^a
99-12	Al	Wrapped with Al Foil	3" x 3.5"	Contact surfaces on Al tube was NOT anodized
99-13	Zr	Electroplated Zn	1.75" x 3"	Foil designated for processing to ⁹⁹ Mo recovery
99-14	Al	Electroplated Zn	1.75" x 3"	Contact surfaces on Al tube was anodized ^a
99-15	Al	Electroplated Ni	3" x 3.5"	Contact surfaces on Al tube was NOT anodized
99-16	Al	Electroplated Zn	3" x 3.5"	Contact surfaces on Al tube was NOT anodized

^aBlack sulfuric acid anodization following MIL A 8625 F Type II Class 2 specifications. Only the inner surface of the outer tube and the outer face of the inner tube were anodized. The inner and outer tubes were masked at either end so a clean, oxide-free surface remained for welding.

After irradiation the targets were transported to a target disassembly hot cell at the Radiometallurgy Installation. Disassembly was accomplished by cutting off the ends of the target and then making a longitudinal cut in the outer tube. After cutting the outer tube, our procedure called for the outer tube to be pried off the inner tube and for the uranium foil recovered. Results of the disassembly are shown in Table 3. In general we found the following:

- Both aluminum and zirconium target tubes work well.
- Nickel fission-recoil barriers perform the best.

- Zinc fission-recoil barriers showed some bonding of the foils to the target tubes.
- Aluminum fission-recoil barriers show promise with anodized aluminum target tubes.
- The zirconium tubes were significantly harder to cut than the aluminum tubes, and it was harder to pry off the outer tube. However, the target tube thickness was the same as that of the aluminum target tubes; owing to the strength of zirconium, the thickness could be considerably reduced, making disassembly easier.

During assembly of two of the targets to be tested in Indonesia, we found that there was insufficient clearance between the target tubes to accommodate a uranium foil wrapped in the desired 25- μm -thick aluminum-foil fission-recoil barrier, so we substituted 15- μm -thick aluminum foil. However, the range of fission-recoil fragments in aluminum is $\sim 14 \mu\text{m}$, so some fission-recoil fragments probably penetrated the thinner aluminum barrier and caused some localized bonding of the uranium foil to the target tubes. Using thicker aluminum foil should improve the disassembly of the targets with an aluminum fission-recoil barrier.

In addition, all the foils used in this irradiation showed more brittleness than has been observed in the past. We are investigating the cause of the brittleness; we suspect that there was some problem with the composition of the LEU foil used in the targets.

Metallographic examination of the disassembled targets (either separated foils or foils stuck to aluminum target tubes) is underway in Indonesia. Once the results of these examinations and other examinations of the unirradiated foils are available, we intend to define a new test matrix to address the issues raised during the present tests, such as thickness of the fission-recoil barriers and the composition of the uranium foils.

Table 3. Performance Results of Targets Irradiated in August in Indonesia

Target	Disassembly Performance	Notes
99-9	Good	When making the longitudinal cut, the outer tube was not cut completely, which made prying it off the inner tube difficult. However, the foil was not bonded to the target and was recovered in 3 large pieces. The foil was broken when trying to pry off the outer tube.
99-11	Good	In general, the foil was not bonded to the target tubes except for a little ring at the top and bottom of the foil. The little bit of foil that bonded was where a small amount of the aluminum foil barrier used to wrap the uranium foil was pinned between the target tubes in an unanodized region. Extending the anodizing slightly past the undercut for the foil should alleviate this problem. Even though the aluminum foil barrier was slightly thinner than it should have been, the anodizing on the target tubes prevented bonding of the foil to the target tubes.
99-12	Poor	Some bonding of the foil to the target tubes (approximately half to the outer tube and half to the inner tube) was noted in this target. A thicker barrier might prevent bonding.
99-13	Good	When making the longitudinal cut, the outer tube was not cut completely, which made prying it off the inner tube difficult. The foil was not bonded to the target, but it was brittle and broke into many pieces when trying to pry the outer tube off the inner tube.
99-14	Moderate	The foil did not bond to the inner tube, but it was brittle and somewhat bonded to the outer tube.
99-15	Good	The foil was not bonded to the target tubes, but was somewhat brittle and broke into 3 large pieces during disassembly.
99-16	Poor	The foil showed some bonding to the target tubes.

CONCLUSION

The new annular target performed well during irradiation. The target is inexpensive and provides good heat transfer during irradiation. Based on these and previous tests, we conclude that targets with zirconium tubes and either nickel-plated or zinc-plated foils work well. We proved that we could use aluminum target tubes, which are much cheaper and easier to work with than the zirconium tubes. In aluminum target tubes nickel-plated fission-recoil barriers work well and prevent bonding of the foil to the new target tubes during irradiation. Also, zinc-plated and aluminum-foil barriers appear promising in anodized aluminum tubes. Additional tests are anticipated to address such issues as fission-recoil barrier thickness and uranium foil composition. Overall, however,

the target was successful and will provide an inexpensive, efficient way to irradiate LEU metal foil for the production of ^{99}Mo .

ACKNOWLEDGMENTS

The authors gratefully acknowledge the contributions of R. S. Smith and D. R. Henley of Argonne National Laboratory to the buckling analysis discussed herein and the assistance of D. L. Amin and A. Basiran of the Indonesian National Nuclear Energy Agency in performing the target disassembly.

REFERENCES

- [1] J. Konrad, "Facilities for the Irradiation of ^{235}U for the Production of ^{99}Mo at the HFR Petten," Irradiation Technology, Proc. of the Int. Topical Mtg., Grenoble, France (1982) 677-683.
- [2] J. Salacz, "Production of Fission Mo-99, I-131 and Xe-133," Revue IRE Tijdschrift, Vol. 9, No. 3 (1985).
- [3] C. J. Fallais, A. Morel De Westgaver, L. Heeren, J. M. Baugnet, J. M. Gandolfo, and W. Boeykens, "Production of Radioisotopes with BR2 Facilities," BR2 Reactor Mtg., Mol, Belgium, INIS MF 4426, pp. IX-1 to -11 (1978).
- [4] A. A. Sameh and H. J. Ache, "Production Techniques of Fission Molybdenum-99," Radiochimica Acta 41 (1987) 65-72.
- [5] R. O. Marques, P. R. Cristini, H. Fernandez, and D. Marziale, "Operation and Installation for Fission ^{99}Mo Production in Argentina," Fission Molybdenum for Medical Use, Proc. of Technical Committee Mtg. Organized by the International Atomic Energy Agency, Karlsruhe, October 13-16, 1987, IAEA-TECDOC-515 (1989) 23-33.
- [6] A. A. Sameh and H. J. Ache, "Production Techniques of Fission ^{99}Mo ," Fission Molybdenum for Medical Use, Proc. of Technical Committee Mtg. Organized by the International Atomic Energy Agency, Karlsruhe, October 13-16, 1987, IAEA-TECDOC-515 (1989) 47-64.
- [7] J. Salacz, "Processing of Irradiated ^{235}U for the Production of ^{99}Mo , ^{131}I , and ^{133}Xe Radioisotopes," Fission Molybdenum for Medical Use, Proc. of Technical Committee Mtg. Organized by the International Atomic Energy Agency, Karlsruhe, October 13-16, 1987, IAEA-TECDOC-515 (1989) 149-154.
- [8] A. A. Sameh and A. Bertram-Berg, "HEU and LEU MTR Fuel Elements as Target Materials for the Production of Fission Molybdenum," Proc. of the 1992 International Meeting on Reduced Enrichment for Research and Test Reactors, Roskilde, Denmark, September 27-October 1, 1992, Argonne National Laboratory Report, ANL/RERTR/TM19, CONF-9209266 (1993) 313-333.

- [9] H. Cols, P. R. Cristini, and R. O. Marques, "Preliminary Investigations on the Use of Uranium Silicide Targets for Fission Mo-99 Production," Proc. of 1994 International Meeting on Reduced Enrichment for Research and Test Reactors, Williamsburg, Virginia, September 18-23, 1994, Argonne National Laboratory Report, ANL/RERTR/TM-20 (in press).
- [10] R. T. Jones, "AEC-2 Experiments in Support of ⁹⁹Mo Production in NRU," Atomic Energy of Canada Limited, AECL-7335 (1982).
- [11] K. A. Burrell and R. J. Harrison, "Development of the ⁹⁹Mo Process at CRNL," Fission Molybdenum for Medical Use, Proc. of Technical Committee Mtg. Organized by the International Atomic Energy Agency, Karlsruhe, October 13-16, 1987, IAEA-TECDOC-515 (1989) 35-46.
- [12] H. Arino, H. H. Kramer, J. J. McGovern, and A. K. Thornton, "Production of High Purity Fission Product Molybdenum-99," U.S. Patent 3,799,883 (1974).
- [13] H. Arino, F. J. Cosolito, K. D. George, and A. K. Thornton, "Preparation of a Primary Target for the Production of Fission Products in a Nuclear Reactor," U.S. Patent 3,940,318 (1976).
- [14] C. D. Massey, D. L. Miller, S. D. Carson, T. A. Wheeler, S. W. Longley, R. L. Coats, E. J. Parma, M. McDonald, M. E. Vernon, S. C. Bourcier, S. G. Mills, A. J. Trennel, and K. R. Boldt, "Feasibility Study of Medical Isotope Production at Sandia National Laboratories," SAND95-2703 Rev. O, Sandia National Laboratories (1995).
- [15] G. L. Hofman, T. C. Wiencek, E. L. Wood, J. L. Snelgrove, A. Suropto, H. Nasution, D. Lufti-Amin, and A. Gogo, "Irradiation Tests of ⁹⁹Mo Isotope Production Targets Employing Uranium Metal Foils", Proc. of the XIXth International Meeting on Reduced Enrichment for Research and Test Reactors, October 7-10, 1996.
- [16] J. L. Snelgrove, G. F. Vandegrift, C. Conner, T. Wiencek, and G. Hofman, "Progress in Converting ⁹⁹Mo Production from High- to Low-Enriched-1999", this conference.
- [17] J. A. Smaga, J. Sedlet, C. Conner, M. W. Liberatore, D. E. Walker, D. G. Wygmans, and G. F. Vandegrift, "Electroplating Fission-Recoil Barriers onto LEU-Metal Foils for ⁹⁹Mo Production Targets" Proc. of the XXth International Meeting on Reduced Enrichment for Research and Test Reactors, October 5-10, 1997, Jackson Hole, Wyoming, U.S.A., in press.
- [18] C. Conner, M. W. Liberatore, A. Mutalib, J. Sedlet, D. E. Walker, and G. F. Vandegrift, "Progress in Developing Processes for Converting ⁹⁹Mo Production from High- to Low-Enriched Uranium-1998" Proc. of the XXIth International Meeting on Reduced Enrichment for Research and Test Reactors, October 17-23, 1998, Sao Paulo, Brazil, in press.