INDONESIA'S CURRENT STATUS FOR CONVERSION OF Mo-99 PRODUCTION TO LEU FISSION

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ABSTRACT

Indonesia has a conversion program from HEU to LEU for producing Mo-99 from LEU foil target. Limited and restricted row material of HEU is the basic reasons to have conversion program in producing Mo-99 from LEU fission. The substitution of low-enriched uranium (LEU) metal foils for the HEU UO2 used in current target designs will be applied for production of Mo-99 Indonesia commercially. Batan has a joint research project with ANL to develop LEU-metal-foil target fabrication. Presented here is the current status of the experiment of foil fabrication using depleted uranium metal. The program of HEU will be reviewed as a background of the conversion program, and discussion will be focused on experiment results of foil target fabrication. The results show that foil targets resulted from the rolling has good characteristics. The surface of the foil is very smooth, and the grain has random orientation after heat treatment and quenching. Foil targets are fabricated from Ni foil-wrapped-DU foil inserted in between two concentric aluminum tubes, and the ends of the tubes are welded. Testing is conducted to assure that there is no leakage in the welded tubes.

1. Introduction

Since 1996 Indonesia has produce Mo-99 from HEU fission product mainly for domestic consumption. Due to limited and restricted row material of HEU, Indonesia has a conversion program from HEU to LEU for producing Mo-99 from LEU foil target. The substitution of low-enriched uranium (LEU) metal foils for the HEU UO2 used in current target designs will be applied for production of Mo-99 commercially. Batan has a joint research project with ANL to develop LEU-metal-foil target fabrication since 1992. Many achievements have been resulted from the experiments. ANL has developed several of LEU target design and fabrication, and has been demonstrated in Radio Metallurgy Installation hot cell in Batan for disassembly process to take out LEU foil from the target after being irradiated in reactor RSG-GAS BATAN. Chemical processing were conducted in Isotope Production Center hot cell for producing Mo-99 which will be use to produce Tc-99m Generator. The experiment was terminated temporary in 2004 due to September eleven accident. In November 2005, ANL provided training LEU target assembly to BATAN by simulation of Cu-foil as LEU foil. And then BATAN personnel demonstrated re-assembly of 2 ANL LEU targets to replace Zn and Al foil barrier with Ni foil barrier in January 2006. Good coordination work had been conducted by BATAN, ANL and IAEA for CRP workshop on LEU foil target at Serpong in March 2006. All of these activities are for preparing Indonesia to change the production of Mo-99 from HEU fission product to LEU fission product. Presented here is the current status of the experiment of foil fabrication using depleted uranium metal. The purpose of Batan experiment using depleted uranium is for better understanding of foil target fabrication steps.

2. Program of HEU

Since 1996 Indonesia has producing Mo-99 from HEU fission product to produce Tc-99m Generator mainly for domestic consumption and some for Bangladesh and Malaysia. Rate of production since 1996 is up and down as seen in Table1.

Table-1. Mo-99 production

Year	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006
Mo-99, Ci	300	1775	2002	1746	783	140	420	806	1550	1575	1786

HEU, as row material, currently Indonesia very difficult to buy HEU from abroad, while the stock of HEU is very limited. This condition led Indonesia to faster the conversion program from HEU to LEU for producing Mo-99. As a plan, next year Mo-99 commercial production by using LEU should be started. To prepare the conversion program, experiment of depleted uranium foil target fabrication has been conducted.

3. DU foils target fabrication

3.1. Foil rolling

Actually the experiment will be done by using LEU but because of no stock of LEU, it has been used depleted uranium to conduct the experiment. ANL will supply Indonesia LEU this year for foil target experiment. So, in this paper will be presented depleted uranium foil target fabrication. Material of depleted uranium (DU) is also supplied by ANL. The form of the material was a plate with dimension of 50 x 50 x 3 mm.

Fig.1. DU plate, frame and cover of Carbon Steel

Hot rolling and cold rolling were done to make foil, and hot rolling was done first before cold rolling. For hot rolling, frame and cover from carbon steel were used to make composite assembly of DU plate, as shown in Figure 1. Plate of DU ingot was put into the frame and cover of carbon steel plates which were coated by paste from mixed of 50 % dextrine $(C_6H_{10}O_5)n$. x H_2O and 50 % alcohol. DU in the frame and cover was welded by SMAW using Cu chill block to protect heat conduction from weld to DU plate. Every sides of assembly were welded. Figure 2 shows the assembly of cover, frame, and DU plate which is ready for welding. The result of welding is shown in Figure 3. The detailed of procedure is refer to RERTR ^{99}Mo Uranium Foil Rolling[1].

Fig.2. Assembly of cover, frame, and ingot DU; ready for welding.

Fig.3. Welding result of assembly

After welding, the assembly was wrapped in SS foil envelope and sealed to protect oxidation during annealing. Annealing was done at 625 C for 24 hours and cooled naturally in furnace. Before hot rolling, assembly was heat treated at 625 C for 1 hr in argon environment. Hot rolling was conducted gradually to reduce the thickness of assembly (sample). Sample entered to roller machine is always with the same front position, but the surface of the sample should be reversed to 180 degree. Heat treatment at 625 C for 10 minute was conducted to the sample before entered to the next step of rolling. Figure 4 is shown the process of hot rolling. The result of hot rolling for the assembly is shown in Figure 5. After hot rolling, the foil was annealed at 625 C for 30 minutes.

Fig.4. Assembly going into the hot roll.

Fig.5. Assembly condition after hot rolling.

DU foil resulted from hot rolling was cleaned with perchloretylene ($C_2H_4Cl_4$) to remove dextrin. Pickling was also conducted with solution of 50 % vol. HNO_3 and 50 % vol. water and cleaned with water and alcohol. The thickness foil resulted from hot rolling was 0.67 mm with the length and the width were 240 mm and 52.7 mm. It was cut became 3 pieces equally long each 80 mm for cold rolling. Stainless steel plate was used to cover the foil during cold rolling. Stainless steel plate thickness was 0.6 – 0.7 mm with the length of 2 x 152 mm folded equally long, and the width was 133 mm. While the length and width of the foil was 80 mm and 52.7 mm. Inner surfaces of SS plate were de-greased with alcohol. Cold rolling was conducted step by step with thickness reduction of 0.5 - 0.1 mm as shown in Figure 6.

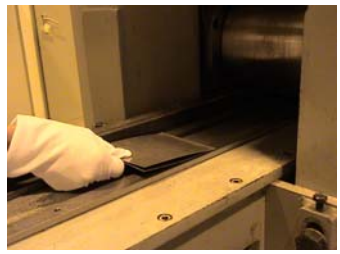

Fig. 6. Cold rolling

3.2. Heat treatment and quenching

Stainless steel foil envelope was used to protect the foil from oxidation during heat treatment and quenching. DU foil with dimension of 44 x 76 x 0.124 mm was inserted to the stainless steel foil envelope, evacuated and sealed for heat treatment and quenching as shown in Figure 7. Heat treatment temperature was 720 C for 20 minutes, and quenching in water for 15 minutes. It was been done 2 times, reheating and quenching [2]. Figure 8 shows the process of heat treatment and quenching.

Fig.7. DU foil wrapped by SS foil and oxygen evacuation with rotary pump.

Fig.8. Heat treatment and quenching

3.3. Foil target assembly

Foil resulted from heat treatment and quenching was used to assemble foil target. DU foil dimension was 44 x 76 x 0.124 mm. Before assembling, the foil was wrapped with nickel foil

of 0.015 mm thick as shown in Figure 9. The wrapped foil was inserted between two concentric aluminum tubes as shown in Figure 10. To prevent foil slip during insertion, a grove was made in the inner tube. A longitudinal line was scribed on the outer tube for the foil gap indication. To make rigid the position of foil in the concentric tubes, draw plug was used to pull the inner tube as shown in Figure 11. The ends of the target were welded by TIG welding. The detailed procedure was written in the ANL procedure for target assembly [3]. Dimension of aluminum tube for target is as follows: Outer tube has ID 28.27 mm and OD 30 mm. While, inner tube has ID 26.23 mm and OD 27.978 mm.

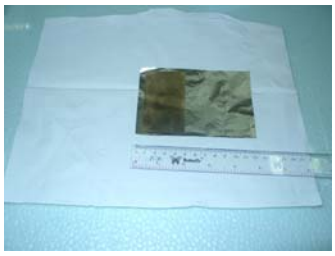

Fig. 9. DU foil will be wrapped with Ni foil.

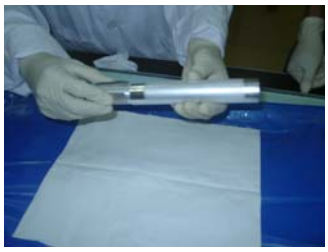

Figure 10. Insertion a foil in Al tubes

Fig. 11. Assembled draw die with target

4. Result and discussion

4.1. Foil

Foil resulted from hot roll was dark color as shown in Figure 12a, perhaps oxidation was occurred during annealing before rolling. It could be come from the leak of carbon steel cover for depleted uranium plate. To prevent the air contact to DU foil, it needs to improve welding practice for the carbon steel assembly. Pickling has been conducted to clean the color, and the result is as shown in Figure 12b. Pickling solution is from mixture of 50 % HNO3 and 50 % water. Cold roll has a good surface performance that is flat and smooth as shown in Figure 13 and 14. Thickness of the resulted foil is 0.124 mm.

a b

Fig.12. DU foil resulted from hot rolling

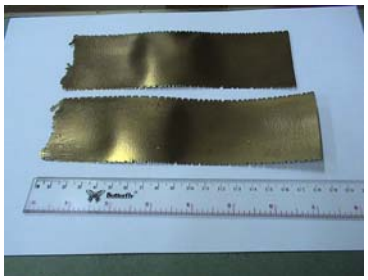

Fig.13. Foil DU resulted from cold rolling

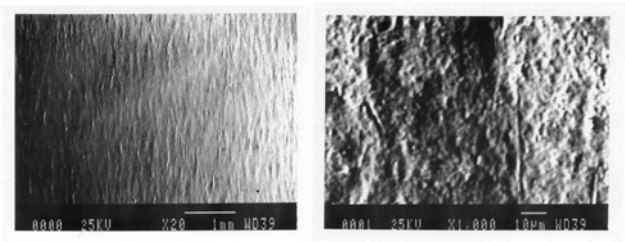

Fig. 14. SEM photograph of DU foil

4.2. Heat treatment foil

Hot and cold rolling of DU foil can produce large grains and oriented in the direction of rolling. For safe irradiation, the grains must be fine and randomly oriented. For that purpose, the foil should be heat treated and quench. DU foil resulted from heat treatment and quenching is shown in Figure 15 and microstructure of the foil is shown in Figure 16. From the picture, it shows that the grain size is small and the orientation is random.

Figure 15. DU foil resulted from heat treatment and quenching

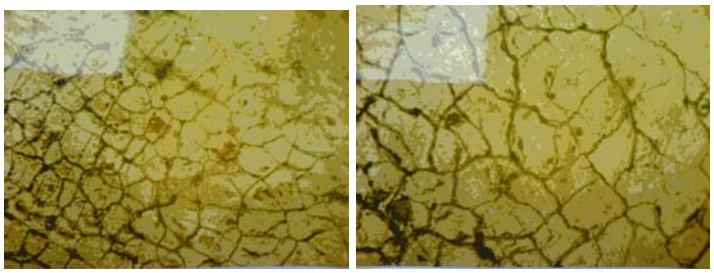

Fig. 16. Microstructure of DU foil heat treated at 670 C, 20 men (left) and 720 C, 20 men (right), each was quenched in water and repeated 2 times. Magnification 200 x. Grain size is almost the same, left is slightly smaller.

β phase was resulted from the heat treatment of DU foil at temperature 670 °C and 720 °C and quench in water, but α phase is also still exist and typical of the XRD graph is shown in Figure 17. It could be improve by repeated the heat treatment condition.

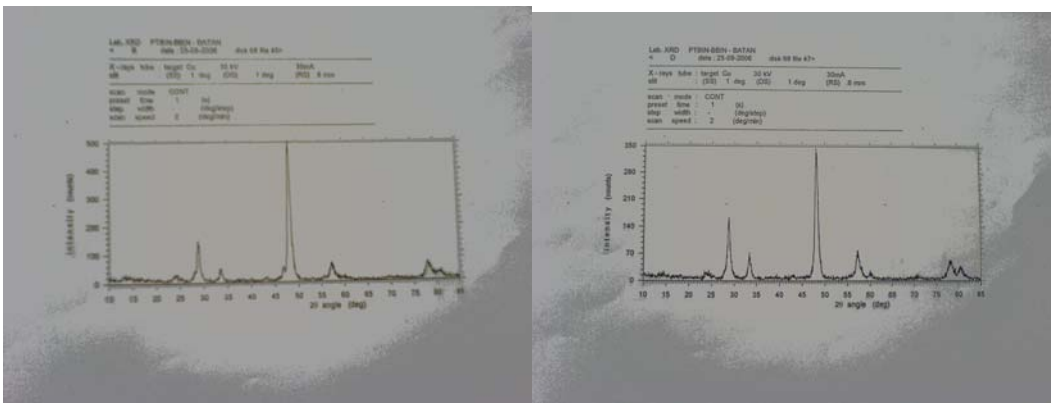

Fig. 17. Typical of XRD graphs of DU foil after heat treatment at 670 C (left) and 720 C (right).

4.3. Foil target assembly

The groove depth of the inside tube has been varied to find the optimum condition of the procedure. It was used Cu foil target to make variation of groove depth, that was 40 µm and 13 µm less than the thickness of CU foil wrapped in Ni foil, and the same as the thickness of CU foil wrapped in Ni foil. SEM was used to analyze the result of the different of groove depth used in the foil assembly as shown in Figure 18.

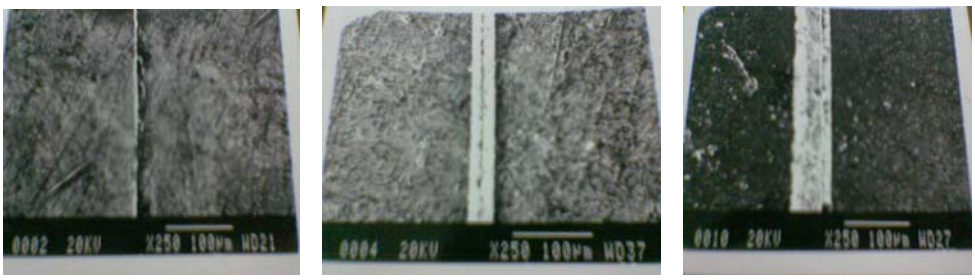

Fig. 18. SEM photograph of Cu foil target from different grooving depth, from the left: 20 µm, 47 µm and 60 µm.

From the figure could be understood that high deformation was occurred in the foil for shallow grooving. To see integrity of Cu foil in the target, dye-penetrant test was conducted on the sample of Cu target and the result is as shown in Figure 19. Grooving depth of 20 μm is very rigid, but it was very difficult to assemble the foil wrapped in the inner tube to slide into the outer tube. With this condition and data from SEM photograph in Fig. 18, it has been chosen that the optimum groove depth is 47 μm. It means that the groove depth is 13 μm less than the thickness of DU foil wrapped in Ni foil.

Figure 19. Dye-penetrant test result

Effect of extension on the change of diameter of the inner and outer tube has been analyzed, and data is shown in Table 2. Before extension, average inside diameter of inner tube is 26.210 mm and average outside diameter of outer tube is 30.000 mm. After extension, average inside diameter of inner tube is 26.5433 mm and average outside diameter of outer tube is 30.0807 mm. So, plastic deformation of outer tube and inner tube are 0.269 % and 1.194 % respectively.

Table 2. Data of outside and inside diameter of foil target assembly.

DU TARGET	After extension, mm				Before extension, mm			
	1	2	3	Average	1	2	3	Average
OD outer tube	30.030	30.086	30.126	30.0807	30.000	30.000	30.000	30.000
ID inner tube	26.550	26.550	26.530	26.5433	26.230	26.230	26.230	26.230

Welding of foil target was conducted with different parameter of welding to find the best condition. We have tried to vary the DC current (40 ampere, 50 ampere and 60 ampere) and angle chamfer (90°, 60°, 45° and flat) on the outside diameter and inside diameter surfaces of the aluminum tubes as shown in Figure 20. DC current with 60 ampere and 45° welding angle has a good result, and it was used for welding the DU foil target. The result of DU foil target is shown in Figure 21. To check the leakage, bubble test was conducted for DU foil target and the result is no leakage.

Fig. 20. Results of welding with different parameter of ampere and angle

Fig. 21. Result of DU foil target

5. Conclusion

Indonesia has preparing to convert from HEU to LEU for Mo-99 production. Batan progress is working to develop foil target fabrication. Batan and ANL have good cooperation to develop LEU foil target. Batan demonstrated all steps of foil target fabrication use depleted uranium. Some important result could be concluded from DU foil target fabrication practice are:

1. To prevent dark color of DU foil resulted from hot roll, it needs improvement in welding practice for the assembly of DU plate and carbon steel plate.
2. Resulted DU foil from the hot and cold roll has flat surface with thickness of 0.124 mm.
3. Grains resulted from heat treatment is small and randomly oriented.
4. Phase β and α were resulted in the DU foil after heat treatment and quench. It still needs improvement for heat treatment practice.
5. The optimum groove depth of inner tube is 13 μm less than the thickness of DU foil wrapped in Ni foil.
6. Plastic deformation of outer tube and inner tube resulted are 0.269 % and 1.194 % respectively.
7. The optimum welding parameter is DC current with 60 ampere and 45° chamfer on the outside and inside diameter surfaces of the end aluminum tubes.

6. Reference

1. ANONIM, RERTR ^{99}Mo Uranium Foil Rolling, IAEA CRP Workshop, March 2006.
2. ANONIM, Heat Treating (Beta-Quench), IAEA CRP Workshop, March 2006.
3. ANONIM, Draft Instruction for Assembling 8 gr or 16 gr Ni Foil Wrapped LEU Metal Foil Annular Targets, IAEA CRP Workshop, March 2006.