Proceedings of the Korean Nuclear Society Autumn Meeting
Yongpyong, Korea, October 2003

Casting of Wide Polycrystalline Uranium Foils by A Single Cooling Roll

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ABSTRACT

As the uranium foils for a Mo-99 irradiation target, which are charged into a reactor can be conventionally fabricated at a laboratory scale, but not yet at a commercialized scale by the hot rolling method due to some problems in foil quality, productivity and economic efficiency, attention has shifted to the development of new improved technology. Under these circumstances, an alternative fabrication method for wide U polycrystalline foils has been investigated using the cooling roll casting method, in order to produce a fission isotope ^{99}Mo, the parent nuclide of ^{99m}Tc.

Continuous polycrystalline uranium foils with a thickness range of 100 to 150㎛ and a width of about 50 mm were fabricated, by adjusting the process parameters of the cooling-roll casting apparatus. The uranium foils had a good roughness on the surface, with a few impurities. The cooling-rolling casting process produced a uranium foil with a high purity and a high productivity. The uranium foils had fine and uniform polycrystalline grains below about 10 microns in size with the α-U phase, irrespective of the process parameters.

1. Introduction

Generally, the conventional fabrication method for uranium foil for the Mo-99 irradiation target [1-2] has the disadvantages of complicated processes as follows: casting the uranium; cutting the resulting ingot to a suitable size for hot rolling; rolling a thick piece of the ingot through many passes to gradually thin it to fabricate a uranium foil of 100-150 µm thickness; and finally heat-treatment at ~800 °C and quenching the fabricated uranium foil to produce the required gain size and orientation.

In the conventional method, the uranium must be heated and rolled under a vacuum or in an inert atmosphere because it is a reactive material. The hot rolling is repeated several times to obtain a suitable thickness of the uranium foil. As the hot-rolling process takes a long time, productivity is relatively low. A washing/drying process must be done to remove surface impurities after hot rolling. In order to obtain the fine polycrystalline structure which has a more stable behavior during irradiation, heat-treatment and quenching must be performed. The high hardness and low ductility of uranium make it difficult to roll the foil. The foil is liable to crack owing to residual stress during the process, resulting in a low yield.

Meanwhile, a uranium foil having excessive residual stress from hot rolling may be deformed or damaged by thermal cycling during irradiation. Furthermore, deformed areas or cracks generated during thermal cycling may act as penetration paths through which there can be an interdiffusion reaction of the uranium with a coating layer, such as Al or Ni, which serves as a protector against the reaction of the uranium with a fixed tube in an irradiation target.

In the present study, an alternative method using rapid cooling solidification of a uranium melt [3] was applied to obtain wide polycrystalline uranium foils. The uranium foils were directly prepared from a melt, not through vacuum melting & casting, ingot cutting, hot-rolling and heat-treatment process, but through a cooling-roll casting process, and characterized for application as a Mo-99 irradiation target.

2. Experimental procedure

Depleted uranium lumps (99.9 % pure), aluminum wire (99.9%), and iron (99.8%) were charged and induction-melted in a high-temperature-resistant ceramic nozzle. The superheated molten U metal was discharged through a small slot in the nozzle onto a rotating cooling roll under the condition that the slot was located close to the cooling roll. The U foil was rapidly cooled by contact with the rotating roll driven by an electric motor in an inert atmosphere so that the fine crystalline granules of the uranium foil with an irregular orientation are formed. The rapidly solidified foil was collected in a container. Fig. 1 shows the experimental apparatus for the cooling-roll casting (a) and the schematic diagram of the melt puddle area (b).

The thickness of the foils was measured at several positions along each foil using a micrometer. The foils were polished to 0.3μm in diamond paste, and the metallographic observation was performed for the longitudinal and the transverse sections of the foils, using a scanning electron microscope (SEM). X-ray diffractometer (XRD) using Cu K_α radiation and a Ni filter were used to determine the phase and the preferred orientation for both the surfaces of the foils.

3. Results and Discussion

Fig. 1 shows the typical appearance of a uniform foil of 50mm in width with good surface quality and high flexibility, fabricated by the cooling-roll casting apparatus. The uranium foils with a thickness ranging from 100 to 150μm were cast continuously exceeding 5m in length for one batch. The width of the foil was almost the same with the slot width under the stable process condition. Since the melt of the uranium is rapidly cooled to directly fabricate the uranium foil, the uranium foil, being difficult to roll due to its high toughness, may be easily fabricated. The fabrication process of uranium foil by cooling roll is greatly simplified compared to the conventional fabrication method, which includes a vacuum induction melting process, a repetitive hot-rolling process, a washing/drying process to remove the impurities, such as surface oxides, and a refinement heat-treatment process to obtain fine grains. The melt may be cast at once to fabricate the uranium foils of 1~2 kg in a few seconds by the cooling-roll casting method, thereby having high productivity, which leads to high economical benefits in producing the uranium foil. A long time period is required to conduct the repetitive troublesome conventional hot-rolling process to adjust the thickness of a uranium ingot. However, as the uranium has a low thermal conductivity, it is not easy to collect the uranium foils soundly without wrinkles and cracks.

In addition, Figs. 2~3 show the scanning electron micrographs having various magnifications of the free surface (Fig. 2) and the wheel-contact surface (Fig. 3) of the obtained foils. The wheel-contact surface has a smooth surface state like the roll surface; however, the free surface exhibits a somewhat rough surface state, with some defects. As the feeding temperature of the melt and the gap distance between the nozzle slot and the roll increase, the free surface shows a rougher state. Figs. 4~5 show the scanning electron micrograph, and the X-ray diffraction pattern of the obtained foils. The uranium foils have homogeneous and fine grains below 5 microns in size, so it is expected that it will prevent the uranium foils from excessively growing by the an-isotropic growth behavior during irradiation. All the phases of the rapidly solidified foil are found to be the α-U (orthorhombic) phase. Hence, it is not necessary to heat-treat the hot-rolled foil and quench it from about 800 °C to form fine grains, as an uranium foil having very fine grains is directly obtained by the rapid solidification effect.

In addition, because the uranium is lacking in ductility, the uranium foil may be damaged and cracked owing to an induced stress during the hot-rolling process, which leads to a low yield and a reduced economic efficiency. Furthermore, the uranium foil fabricated by cooling roll has a smaller stress than the foil obtained through the repetitive hot rolling of a uranium plate. Accordingly, deformation or cracking of the foil generated by thermal cycling during the irradiation process can be prevented. Defects in the deformation areas or cracks can act as penetration paths for the elements in the coating layer of the target. The interaction between the coating layer and the target will be enhanced by the defects or cracks. However, the uranium foil fabricated by cooling roll does not have such paths. Commonly, the uranium foil undergoes a large anisotropic growth during irradiation in a reactor. However, the uranium foil fabricated by cooling roll has homogeneous and fine grains with a random orientation so as to prevent the uranium foil from excessively growing during irradiation.

4. Conclusions

1) Continuous polycrystalline uranium foils with a thickness range of 100 to 150㎛ and a width of about 50 mm were fabricated, by adjusting the process parameters of the cooling-roll casting apparatus.
2) The uranium foils had a good roughness on the surface, with a few impurities. The cooling-rolling casting process produced uranium foils with a high purity and a high productivity.
3) The uranium foils had fine and uniform polycrystalline grains below about 10 microns in size with the α-U phase, irrespective of the process parameters. It is expected to be able to prevent the uranium foils from excessive swelling by the an-isotropic growth behavior during irradiation.

Knowledgements

The authors would like to express their appreciation to the Ministry of Science and Technology (MOST) of the Republic of Korea for its support of this work through the mid- and long-term nuclear R&D Project.

Reference

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[3] K. H. Kim, Hee-Jun Kwon, Jong-Man Park, Yoon-Sang Lee, and Chang-Kyu Kim, Journal of Korean Nuclear Society, Vol. 33, No. 4, pp. 365-374, Aug. 2001.

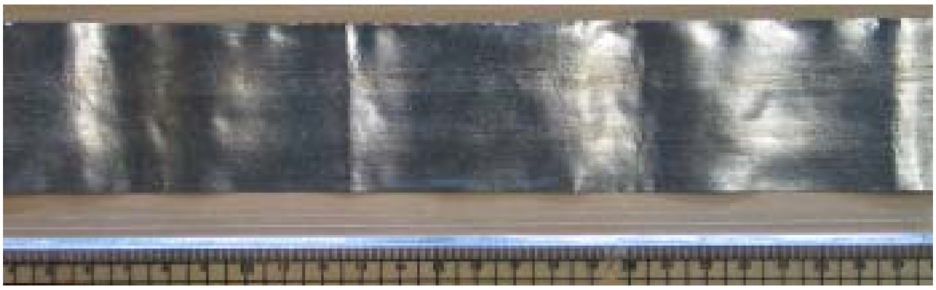

Fig. 1. Uanium foils produced by cooling-roll casting apparatus.

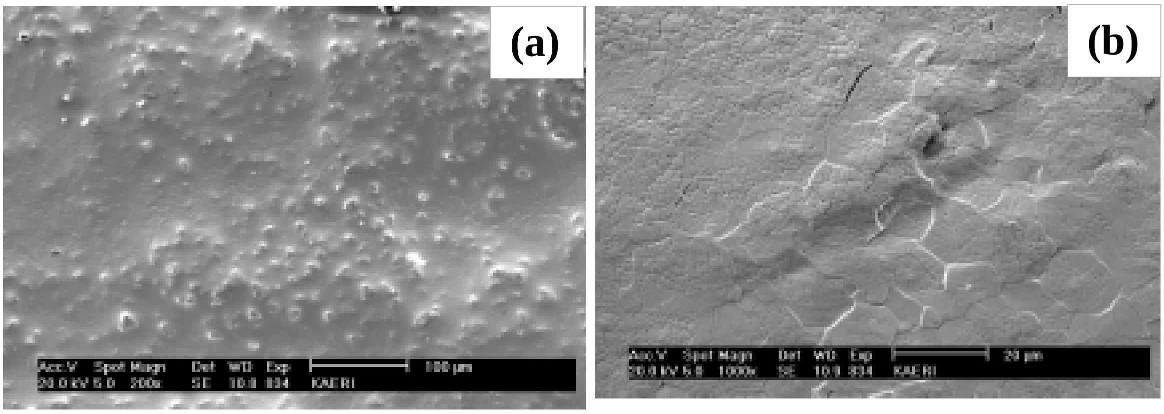

Fig. 2. Scanning electron micrographs of the free side surface of obtained foils with various magnifications; (a) x200, (b) x2000.

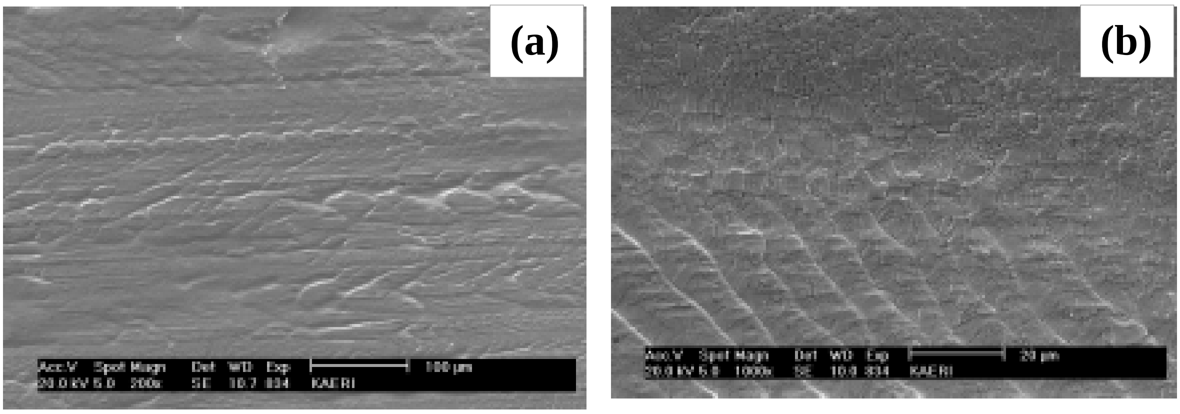

Fig. 3. Scanning electron micrographs of the wheel-side surface of obtained uranium foils with various magnifications; (a) x200, (b) x2000.

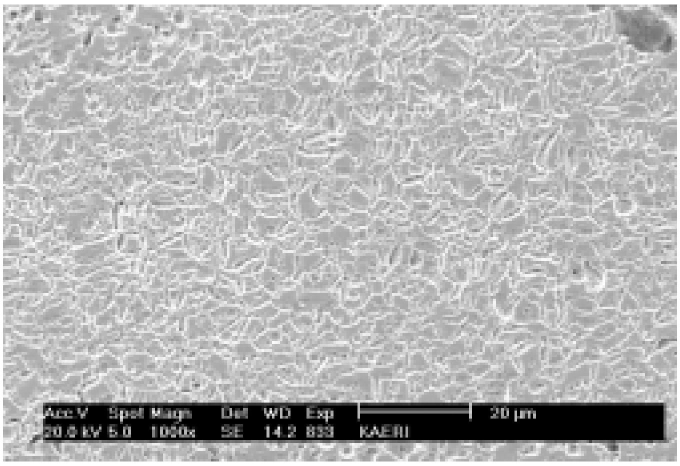

Fig. 4. Scanning electron micrograph of the polished uranium foil.

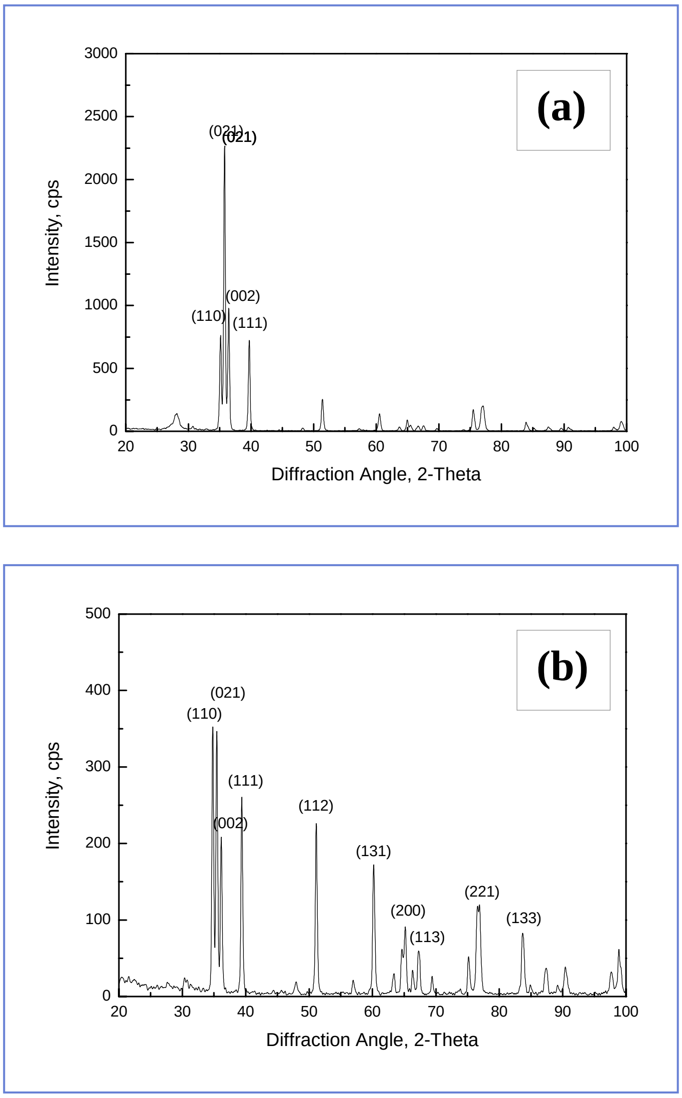

Fig. 5. The X-ray diffraction pattern of obtained uranium foil; (a) wheel-contact surface, (b) free surface.