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Fabrication of Continuous Polycrystalline Uranium Foil Using the Cooling-roll Casting Method

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ABSTRACT

As the uranium foils for Mo-99 irradiation target charged into a reactor can be fabricated at a laboratory scale, but not 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 technology. Under these circumstances, the alternative fabrication method of uranium foil was developed in KAERI using cooling roll, in order to produce the fission isotope ^{99}Mo, the parent nuclide of ^{99m}Tc. The continuous and uniform uranium foil has been rapidly solidified, directly from a melt through the cooling-roll casting method. In order to develop the fabrication technology of the wide foils with reliability, the fabrication and characterization of the uranium foils using the cooling-roll casting method were carried out in this study. The polycrystalline uranium ribbons with a thickness ranging from 100 to 200ì m were cast continuously exceeding 10m in length for one batch procedure. Major advantages of the cooling-roll casting process were obtained as follows: 1) a simplified process without the hot-rolling process and heat-treatment process, 2) an improvement in productivity and yield in foil fabrication, and 3) a high purity and a high quality of foil, 4) a very fine polycrystalline structure.

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 through many passes a thick piece of the ingot to gradually thin it to fabricate a uranium foil of 100-200 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 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 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 the 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 in obtaining uranium foils. The uranium foils were directly prepared from a melt, not through a vacuum melting & casting, ingot cutting, hot-rolling and heat-treatment process, but through a cooling-roll casting process, and characterized by geometry and microstructure for application as a Mo-99 irradiation target.

2. Experimental procedure

Depleted uranium lumps (99.9 % pure) were charged and induction-melted in a high-temperature-resistant ceramic nozzle. The superheated molten U metal was fed through a small orifice onto a rotating cooling roll on a vertical axis. The liquid metal was then rapidly solidified with the rotating roll driven by an electric motor in an inert atmosphere. The rapidly solidified foil was collected in a container. Fig. 1 shows the experimental apparatus for cooling-roll casting (a) and the schematic diagram of the melt puddle area (b). The foils were polished to 0.3ìm in diamond paste, and metallographic observation was performed on the wheel-side and gas-side surfaces and the longitudinal and the transverse sections of the foils, using an optical microscope (OM) and a scanning electron microscope (SEM). X-ray diffractometer (XRD) using Cu K_{α} radiation and a Ni filter was used to determine the phase and the preferred orientation on both surfaces of the foils.

3. Results and Discussion

Fig. 2 shows the typical appearance of continuous and uniform foils with a good surface quality and high flexibility, fabricated by the cooling-roll casting apparatus. The uranium foils with a thickness ranging from 100 to 200ì m were cast continuously exceeding 10m in length for one batch procedure. The width of the foil was almost the same with the slot width under a stable process condition. Since the melt of 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 of uranium foil process by the cooling roll is greatly simplified compared with the conventional fabrication method, which includes a vacuum induction melting process, a repetitive hot-rolling process, a washing/drying process for removing impurities, such as surface oxides, and a heat-treatment process to obtain fine grains. The melt may be cast at once to fabricate large amounts of the foil in a few seconds by the cooling-roll casting method, thereby having a higher productivity and yield, which leads to a greater economy in producing uranium foil. In contrast, a long time period is required to conduct the repetitive troublesome conventional hot-rolling process to adjust the thickness of a uranium ingot. In addition, because the uranium is lacking in ductility, the uranium foil fabricated by the conventional method 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.

In addition, Figs. 3~4 show the scanning electron micrographs having various magnification of the free surface (Fig. 3) and the wheel contact surface (Fig. 4) 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 nozzle slot and roll increase, the free surface shows a rougher state. Figs. 5~6 show the optical and the scanning electron micrographs, and the X-ray diffraction pattern of the obtained foil. 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 due to an-isotropic growth behavior during irradiation. All phases of the rapidly solidified foil are found to be the á-U (orthorhombic) phase. Hence, it is not necessary to heat-treat and quench the hot-rolled foil at about 800 °C to form fine grains, as the uranium foil having very fine grains is directly obtained by such a 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 the cooling roll process has a smaller stress level than foil obtained through repetitive hot rolling of the uranium plate. Accordingly, deformation or cracking of the foil generated by thermal cycling during the irradiation process can be prevented. Defects in deformation areas or cracks can act as penetration paths for elements in the coating layer of the target. The interaction between coating layer and target will be enhanced by the defects or cracks. However, the uranium foil fabricated by the cooling roll process does not have such paths. Commonly, the uranium foil undergoes large an-isotropic growth during irradiation in a reactor. However, the uranium foil fabricated by the cooling roll process 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 and uniform polycrystalline uranium ribbons with a thickness ranging from 100 to 200ì m were produced exceeding 10m in length for one batch procedure, by adjusting the process parameters of the cooling-roll casting apparatus. The cooling-roll casting process exhibited very high yield and productivity.
2) The uranium foils have a good roughness. The wheel-contact surface has a smooth surface state like the roll surface, with a few gas pockets; however, the free surface exhibits a rougher surface state.
3) The uranium foils have homogeneous and fine grains below 5 microns in size with the á-U phase.

Knowledgements

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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.

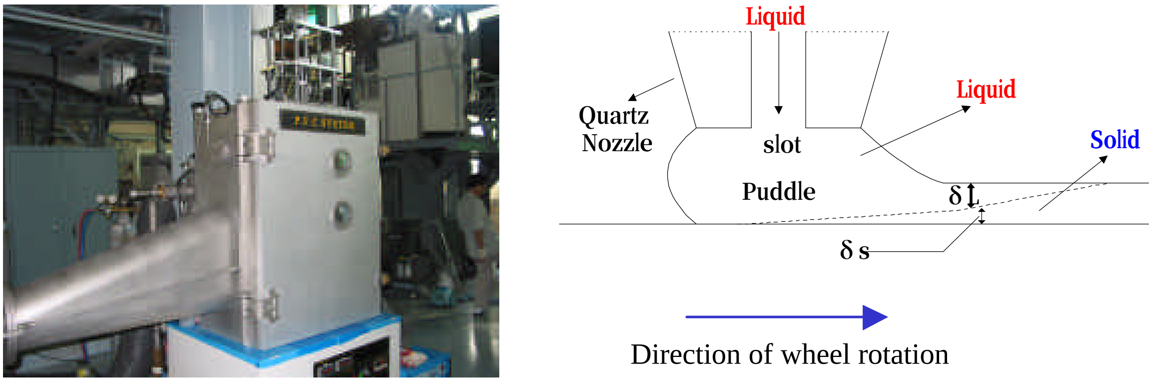

Fig. 1. Experimental apparatus for cooling-roll casting (a) and schematic diagram of melt puddle area (b).

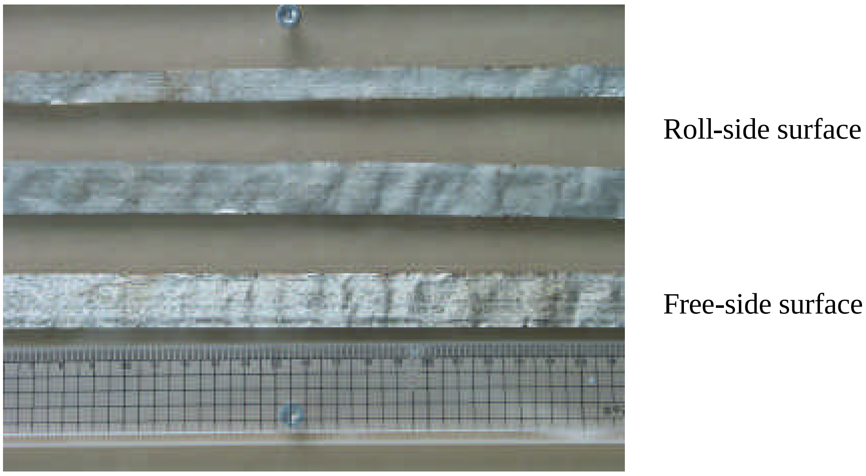

Fig. 2. Continuous uanium foils produced by cooling-roll casting apparatus.

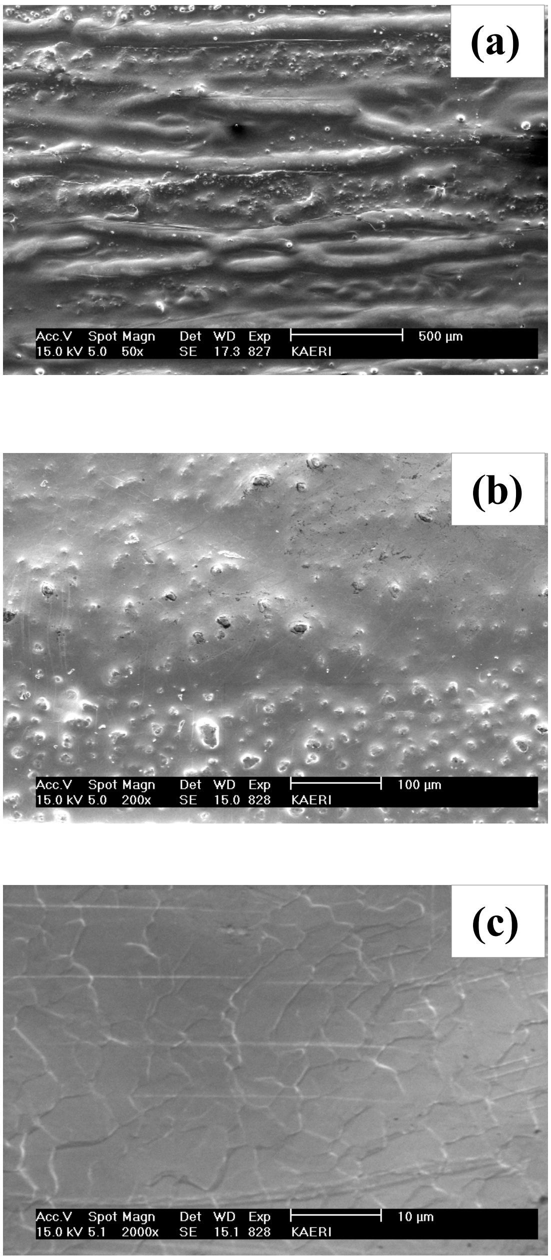

Fig. 3. Scanning electron micrographs of the free side surface of the obtained foils with various magnifications; (a) x50, (b) x200, (c) x2000.

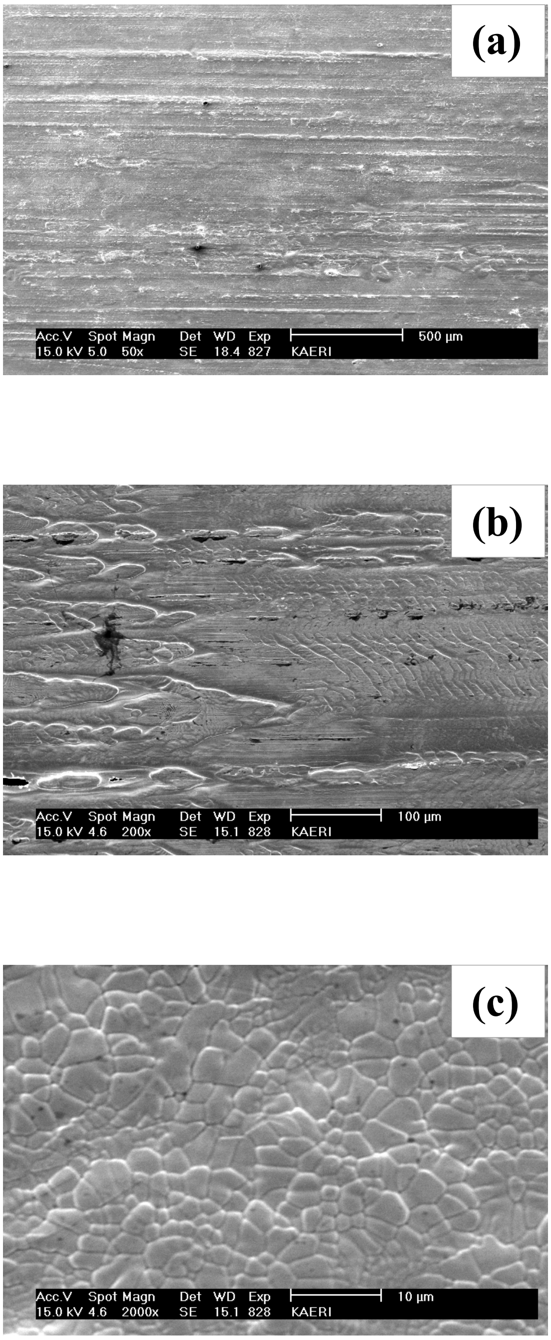

Fig. 4. Scanning electron micrographs of the wheel-side surface of obtained uranium foils with various magnifications; (a) x50, (b) x200, (c) x2000.

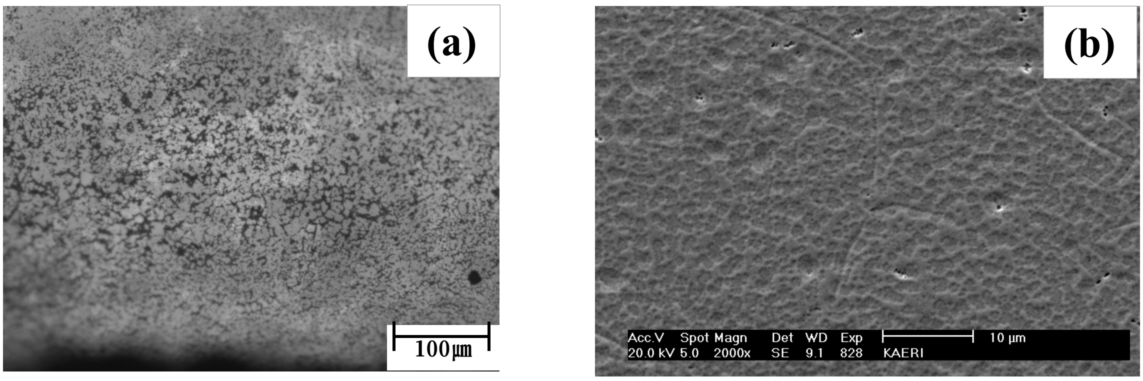

Fig. 5. Optical microscope (a) and scanning electron micrograph (b) of the polished uranium foil.

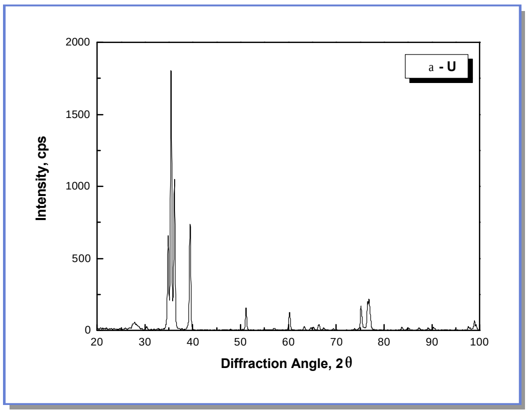

Fig. 6. The X-ray diffraction pattern of the obtained uranium foil.