논 문

Fabrication and Characterization of Wide Uranium Foils by Planar Flow Casting Method

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Abstract

원자로에 장전되는 ^{99}Mo 조사표적을 제조하기 위한 우라늄박판은, 박판 품질, 생산성, 경제성 문제로 인해, 기존의 열간압연방법에 의해 실험실 규모로는 제조가 가능하나, 상용 규모로는 제조되기 어려운 실정이므로, 새로운 제조방법의 개발이 요구되고 있다. 이와 같은 상황에서, ^{99m}Tc 의 모핵종인 방사선 동위원소^{99}Mo생산하기 위하여 planar flow casting (PFC) 법에 의해 다결정질 우라늄박판에 대한 새로운 제조방법이 연구되었다. 100~150 μm 의 두께 및 너비 약 50 mm 의 연속적인 다결정질 우라늄박판이 하나의 batch 에서 5 m 이상의 길이로 제조되었다. 우라늄박판은 불순물이 거의 없었으며 양호한 표면조도를 가지고 있었다. 우라늄박판의 냉각롤 접촉표면은 자유표면 보다 매끈한 자유표면을 가지고 있었다. 우라늄박판은 제조공정변수와는 상관없이 α-U 상을 가진 약 10 μm 이하의 미세한 다결정립을 가지고 있었다.

Key words : PFC, Uranium foil, Continuous Polycrystalline Foil, Medical isotope

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1. Introduction

The Reduced Enrichment for Research and Test Reactors (RERTR) program was established in 1978 by the Department of Energy (DOE) in USA. As a part of a nonproliferation effort, the RERTR program has investigated the production of the fission isotope ^{99}Mo using low-enrichment uranium (LEU) instead of high-enrichment uranium (HEU) since 1993, a parent nuclide of ^{99m}Tc, which is a major isotope for a medical diagnosis [1~2]. Fissile part of the Mo-99 irradiation target using LEU is a thin (100~150 mm thick) pure uranium foil sandwiched between inner and outer Al tubes.

The conventional fabrication method of a uranium foil for a Mo-99 irradiation target generally has the disadvantages of complicated processes such as the following [3~4]: casting the uranium; cutting the resulting ingot to a suitable size for a hot rolling; rolling a thick piece of the ingot through many passes to gradually thin it to fabricate a uranium foil with a thickness of 100~150 μm; and finally a heat-treatment at ~1073°K and quenching the fabricated uranium foil to produce fine gain size and random orientation.

As uranium foils have been fabricated by the conventional method on a laboratory scale by a repetitive hot-rolling method with significant problems in foil quality, productivity and economic efficiency, attention has shifted to the planar flow casting (PFC) method. PFC is a variation of a free jet melt spinning (FJMS), which has attracted lots of interest as an efficient and continuous process for producing relatively wide foils [5~6]. Under these circumstances, an alternative fabrication method of uranium foils has been investigated using the PFC method, in order to produce a fission product ^{99}Mo. In the present study, the PFC method was applied to obtain wide polycrystalline 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 PFC process, and characterized for an application as a Mo-99 irradiation target.

2. Experimental

Uranium lumps (99.9 % pure) were charged into a 60-mm diameter quartz tube, and a vacuum was formed within the PFC caster by the operation of an exhaust pump. The U lumps were induction-melted and then discharged through a rectangular slot with an argon gas over-pressure on to a 300-mm diameter polished copper wheel, rotating with a tangential velocity of 450-800 rpm (9.4-16.7 ms^{-1}). The wide U foils were formed with a rapid cooling by contacting then with a rotating cooling-roll under a condition where the slot was located close to the cooling roll. The slot/wheel gap was carefully set to 0.5 mm by using a thickness

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gauge just before an ejection. The rapidly solidified foils were collected in a long container.

The uranium foils were measured for their thickness and widths at several positions, and then average values were obtained. The morphology and the microstructure of the foils were observed by using a scanning electron microscope (SEM). X-ray diffractometer (XRD) by using a Cu K_α radiation and a Ni filter was used to determine the phases present in the obtained foils.

3. Results and Discussion

3.1 *Fabrication of wide U foils by a planar flow casting*

Fig. 1 shows the typical appearance of a uranium foil of 50mm in width with smooth surface and fine edges, fabricated by the PFC method. Since the foil is directly fabricated from a melt by a rapid solidification, it was possible to fabricate a wide uranium foil continuously with the PFC method. The wide U foil with a thickness ranging from 100 to 150 µm was cast continuously, exceeding 5m in length for one batch. The width of the foil was almost the same as the slot width under a stable process condition.

Fig. 2 shows the scanning electron micrographs with various magnifications of the free surface and the wheel-contacted surface of the obtained uranium foil. The wheel-contacted surface has a smooth surface state, like the cooling-roll surface; however, the free surface exhibits a somewhat rough surface state. The roughness of the uranium foil also revealed that the obtained foils exhibited a rougher state on the free surface and a smoother state on the wheel-contacted surface.

In the planar flow casting, the foil thickness is generally dependent upon the rotating wheel velocity. The rotating wheel velocity was the most dominant process parameter influencing the foil thickness. As the rotating wheel velocity increases, the foil thickness continuously reduces, as can be seen Fig. 3. Uranium foils were fabricated under the optimum conditions of 100°K superheat, 0.5 mm slot/wheel gap, 160 kPa ejection pressure, and 660 rpm revolution speed of the rotating wheel, and the average foil thickness was found to be approximately 130 µm. It has been reported extensively that a foil thickness decreases as the rotating wheel velocity increases due to a decrease of the contact length of the melt puddle with the rotating wheel [7~8].

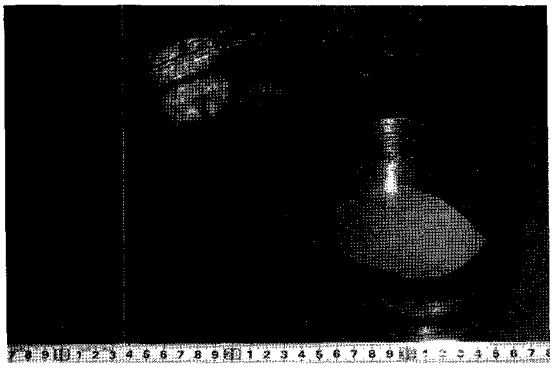

Fig. 1. Typical appearance of the obtained foil of 50 mm in width, continuously fabricated by the PFC method.

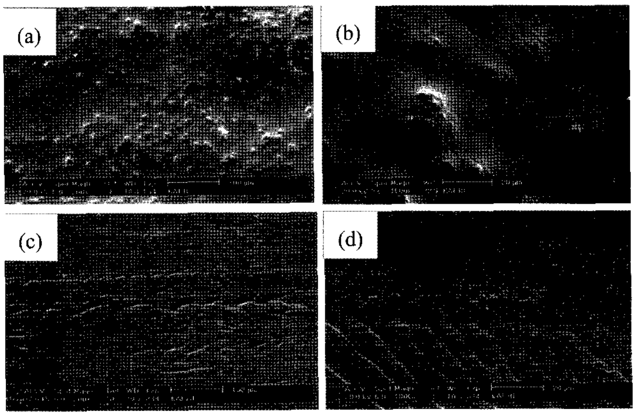

Fig. 2. Scanning electron micrographs of the free surface (a-b) and the wheel-side surface (c-d) of the uranium foils with various magnifications.

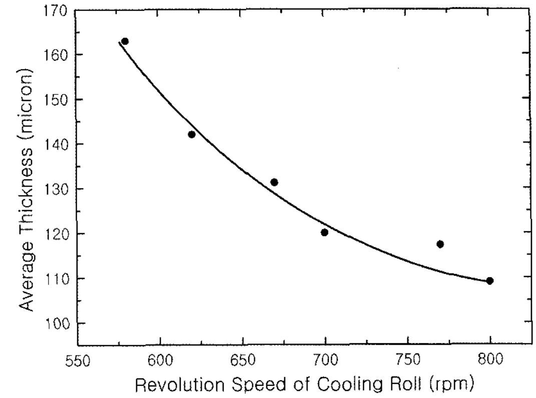

Fig. 3. The effect of the revolution speed of the cooling roll on the average thickness of the uranium foils.

3.2 *Characterization of wide U foils by a planar flow casting*

Fig. 4 shows a typical SEM microstructure transition from a wheel-side surface to free surface of the uranium foil, fabricated under the optimum conditions of 100°K superheat, 0.5 mm slot/wheel gap, 140 kPa ejection pressure, and 500 rpm revolution speed of the rotating wheel. The uranium foil generally had a three-layed structure consisting

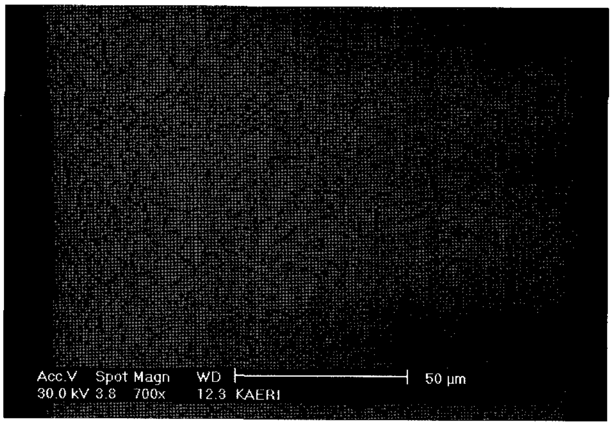

Fig. 4. Typical SEM microstructure transition from the wheel-side surface to free surface of the uranium foil, with revolution speed of wheel of 500 rpm (10.4 m/sec).

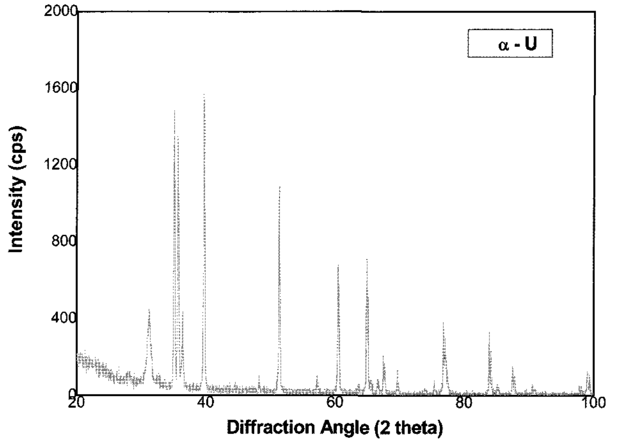

Fig. 5. Typical X-ray diffraction pattern of the obtained uranium foil, with revolution speed of wheel of 500 rpm (10.4 m/sec).

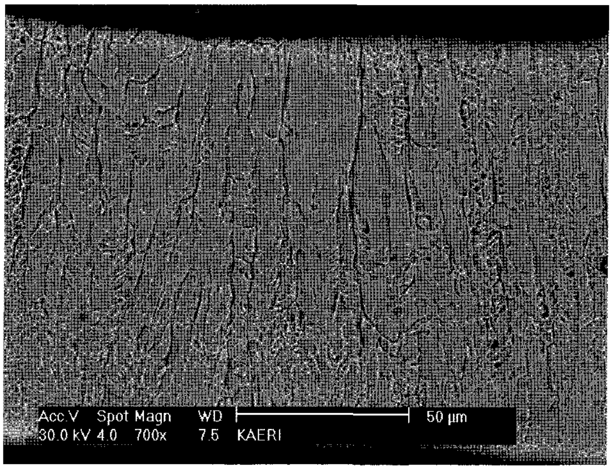

Fig. 6. Scanning electron micrograph of the uranium foil, with revolution speed of wheel of 660 rpm (13.8 m/sec).

of an equi-axed layer near the wheel surface followed by a columnar layer and, then a cellular layer in the free surface part of the foil. It can be seen from Fig. 4 that the columnar grains grew from the wheel side surface towards the middle of the foil, showing a microstructural transition from a planar morphology to a cellular morphology. This transition can be explained by the absolute stability criterion which has been described previously [9-10]. The existence and the relative thickness ratio of each layer (to the entire foil thickness) depended on the process variables which affect the solidification process [11-15]. In general, the microstructures, fabricated with a low revolution speed of the rotating wheel below about 600 rpm (12.5 m/sec), were dominant with a cellular zone of about 5 μm, affected by a low solidification velocity. Fig. 5 shows a typical X-ray diffraction pattern of the uranium foil. 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 1073°K to form fine grains, as a uranium foil with fine grains is directly obtained by a rapid solidification effect. As usual, the uranium foil undergoes a large anisotropic growth during an irradiation in a reactor [16~17]. However, the uranium foil fabricated by the PFC method has fine grains with a random orientation so as to prevent the uranium foil from growing excessively during an irradiation.

Fig. 6 shows the scanning electron micrograph of the uranium foil, fabricated under the optimum conditions of 100°K superheat, 0.5 mm slot/wheel gap, 160 kPa ejection pressure, and 800 rpm revolution speed of the rotating wheel. In general, the microstructure of the uranium foil fabricated with a high revolution speed of the rotating wheel above about 600 rpm (12.5 m/sec), mainly consisted of a columnar zone, affected by a high solidification velocity.

A columnar layer has been reported to consist of a partition-less solidification structure owing to a very rapid heat extraction during a foil formation [18-19]. It has been found to be composed of thermal dendrites which are formed in a higher undercooled region near the wheel surface [20]. However, as the columnar grains grow, the degree of undercooling in the liquid, ahead of the growing dendrites, decreases due to a release of latent heat from a freezing. In the lower undercooling region, a solutable diffusion is predominant to a thermal diffusion, so solutal dendrites with a low growth velocity are formed. When the wheel speed increases, the length of the columnar zone in the foils increases. This is considered to be caused by the fact that the higher the wheel speed, the larger the heat transfer coefficient at the melt/wheel interface. This results in a high undercooling due to an increase of the cooling rate.

4. Conclusions

(1) Continuous polycrystalline uranium foils with a thickness range of 100 to 150 μm and a width of 50 mm were fabricated, which exceeded 5 m in length for one batch by the PFC method.

(2) The uranium foils had a relatively good roughness on the surface, with few impurities. The wheel-side surface of the uranium foils was rather smoother than the free surface of the uranium foils.

(3) The uranium foils had fine polycrystalline grains below about 10 μm in size with a α-U phase, due to a rapid solidification effect from a melt.

(4) The fabrication of uranium foils by the PFC method can be considered as an alternative process for a fabrication of the uranium foils for a Mo-99 irradiation target.

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