Preliminary Study of High-density LEU Dispersion Targets using an Atomized Uranium-Aluminum Alloy Powder

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1. Introduction

Technetium-99m (^{99m}Tc) is one of the medical radioisotopes typically used in nuclear medical diagnostic fields. The demand for ^{99m}Tc had increased recently [1]. ^{99m}Tc is mainly obtained from the radioactive decay of molybdenum-99 (^{99}Mo), with a half-life of 67 hours. ^{99}Mo is commercially supplied by irradiating highly enriched uranium (HEU > 93) targets; approximately 6.1% of the fission of U-235 produces ^{99}Mo.

However, international efforts have been made to replace HEU targets with low-enriched uranium (LEU < 20) targets in accordance with non-proliferation policies. The conversion of HEU to LEU involves a decrease in the U-235 content in the targets, which leads a loss of Mo-99 production efficiency [2]. In order to compensate for this, a possible alternative is to increase the uranium density in the targets, thus making high-density uranium targets. KAERI developed a centrifugal atomization technology to produce various uranium alloy powders. An atomized powder is favorable for high uranium loadings, and it showed better irradiation performance outcomes compared to those of a pulverized powder. However, atomized powders have different microstructure characteristics from pulverized powders, as they are cooled rapidly, a fact that was not clearly revealed. In this work, preliminary studies of the development of high-density targets using atomized uranium-aluminide powders were conducted to evaluate the feasibility of such a process.

2. Experimental Methods

2.1 Preparation of the atomized U-Al powder

U-xAl (where x=0, 5, 10, 15, 20, and 25 wt.%) alloy powders were fabricated using a centrifugal atomization technique at KAERI. Fig. 1 presents a schematic of the centrifugal atomization procedure for the fabrication of the U-Al alloy powder. First, U and Al ingots were prepared and were arc-melted together in an Ar atmosphere to avoid a thermal shock. Next, U-Al mother alloys were inserted into a ZrO_2 crucible, which was heated to a temperature 300℃ higher than the melting point of the intermetallic compounds to increase the fluidity of the molten metals. Finally, the molten metals were fed onto a rotating graphite disk. The rotating disk produced numerous tiny droplets. The droplets were scattered toward the chamber and cooled very rapidly. The microstructures and constituent phases of the powders were identified using SEM/EDS and XRD.

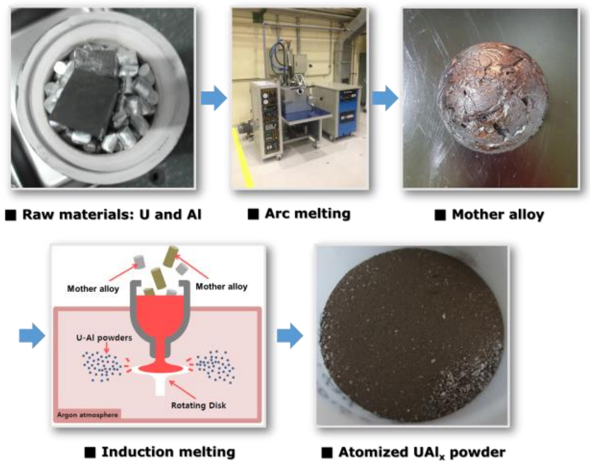

Fig. 1. A schematic of the centrifugal atomization process for the fabrication of the U-Al alloy powder [3]

2.2 Fabrication of high-density dispersion targets

High-density dispersion targets with a uranium density of 3.2 gU/cm^3 were fabricated using atomized U-xAl (x=0, 5, 10, 15, and 20 wt.%) powders. U-25Al was excluded because the U-Al powder volume percent in the targets was limited to 50 vol.%. The targets were fabricated using typical plate fuel fabrication procedures, including mixing and blending, compaction, heat-treatment, assembling, and hot-rolling. The targets were annealed at 550 °C to transform the U and UAl_2 phases to the UAl_3 and UAl_4 phases. The constituent phases were identified using XRD.

3. Results and Discussion

2.1 Fabrication of the atomized U-Al powders

The atomized U-Al powders were successfully fabricated. Fig. 2 shows the microstructures of these powders. The addition of Al formed precipitates with a dendrite structure. The dendrite structure became more distinct as the Al composition increased. As shown in

Fig. 2, remarkable cracks appeared along the grain boundaries when the composition of Al exceeded 20 wt.%. In this range, the α-U phase was consumed entirely and only the UAl_2 and UAl_3 phases existed, which may have led to cracks due to the differences in the volume shrinkage between the uranium and aluminum phases.

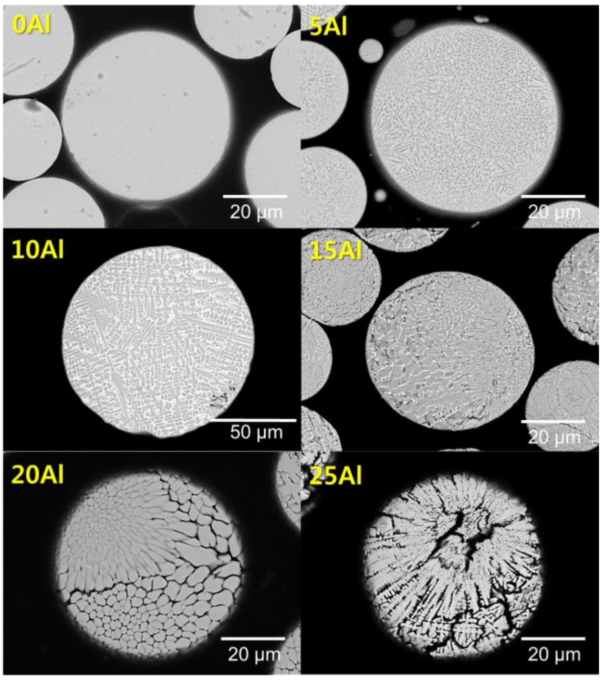

Fig. 2 SEM images of atomized U-xAl (x x=0, 5, 10, 15, 20, and 25 wt.%) powders

Previous work revealed that UAl_x powders are composed of complex intermetallic phases [2] [3]. Fig. 3 shows the XRD results of the atomized U-Al powders. In the atomized powders, the three phases of α-U, UAl_2, and UAl_3 were observed. The addition of Al formed UAl_2 precipitates in the α-U matrix with Al contents of less than 15 wt.%. In the range of 20 to 25 wt.% of Al, the UAl_3 phase was observed instead of α-U. Other characteristics of the atomized aluminide powders were analyzed in our previous studies [3].

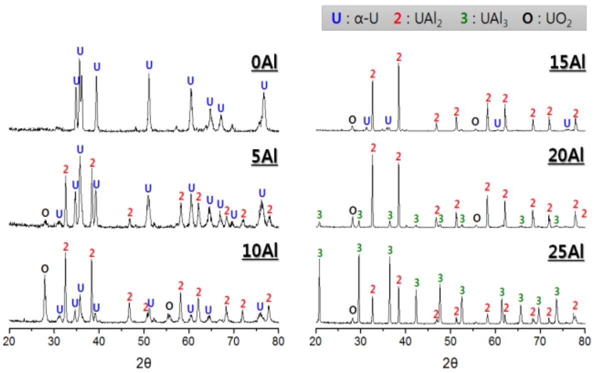

Fig. 3. XRD results for atomized U-xAl (x x=0, 5, 10, 15, 20, and 25 wt.%) powders [3]

2.2 Fabrication of the hot-rolled targets

Fig. 4 shows a conventional target annealed after a hot-rolling process. The phase transformation caused a considerable volume expansion in the targets by means of a large amount of deformation. In order to prevent this type of deformation, the annealing process was embedded between the hot-rolling processes. Table 1 shows the modified annealing conditions. Hence, the targets were successfully fabricated without any deformations, as shown in Fig. 5

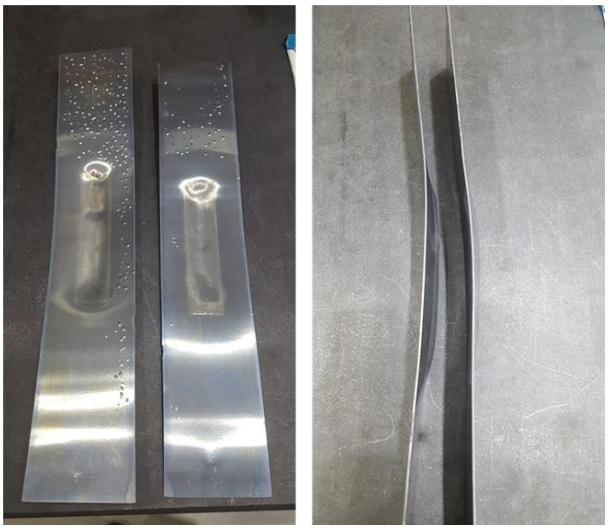

Fig. 4. An image of a hot-rolled target after thermal annealing

Table 1. Annealing conditions for phase transformations with U-15Al at 550℃ for five hours

Annealing condition	Hot-rolling passes					
	1	2	3	4	5	6
1	5h					
2			5h			
3	2h		2h		1h	
4	1h	1h	1h	1h	1h	

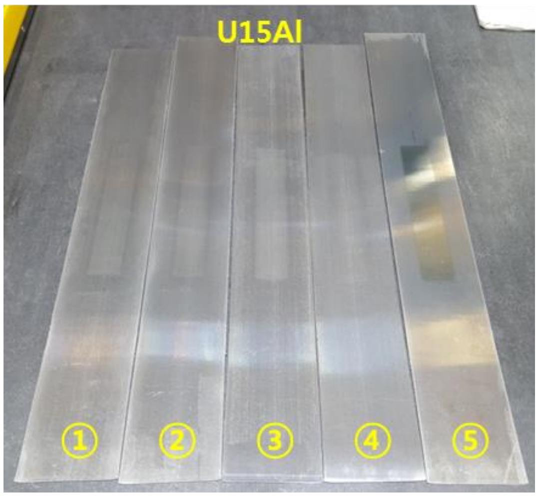

Fig. 5. An image of hot-rolled targets after modification of the annealing conditions (5: conventional method)

Fig. 6 shows the XRD results for the targets annealed under the modified conditions. They were compared to the data of the as-fabricated target and the annealed target after the fabrication process was completed, indicating that annealing condition 3 (two hours at 1, 3, and 5 passes) and 4 (one hour at 1-5 passes) in Table 1 resulted in a desirable phase transformation.

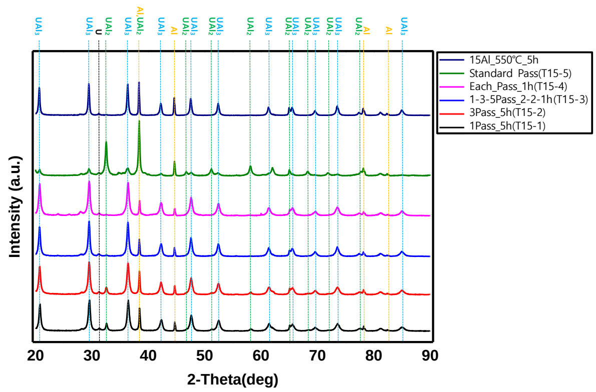

Fig. 6. XRD data for hot-rolled targets annealed under different conditions

As shown in Fig. 6, annealing for five hours at 550 ℃ was not enough to transform all of the UAl_2 phase. The annealing time was adjusted to find a proper annealing time. Fig. 7 shows the XRD data of targets annealed after seven, ten and twenty hours, respectively. There were no peaks for UAl_2 in any of the target data. Therefore, it appears that the minimum annealing time is approximately seven hours at 550 ℃.

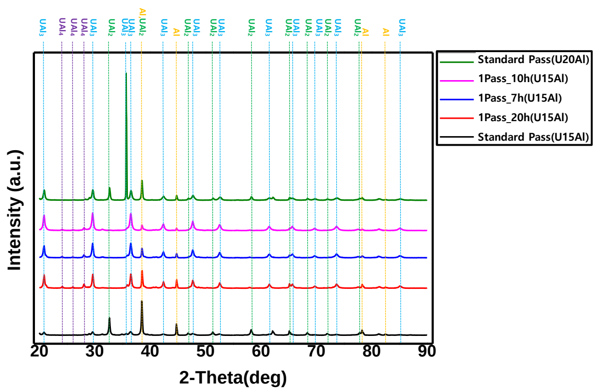

Fig. 7. XRD results for targets annealed at 550 ℃

3. Conclusions

High-density LEU targets with a uranium density of 3.2 gU/cm^3 were fabricated using atomized uranium-aluminide powders at KAERI. The characteristics of the atomized powders depend on Al contents: α-U and UAl_2 phases with an Al content of less than 15 wt.%, and UAl_2 and UAl_3 phases for an Al content in the range of 20 – 25 wt.%. Annealing of the targets was conducted to eliminate the UAl_2 phase. The annealing processes were conducted during a hot-rolling process to avoid deformation of the targets. It was found that annealing for seven hours at 550 ℃ is required for the entire phase transformation of the UAl_2 phase. Further optimizations of the target fabrication process are in progress.

Acknowledgement

This research was supported by a grant from the Nuclear Energy and Safety Program of the National Research Foundation of Korea (NRF) funded by the Korean government (MSIT) (No. NRF-2017M2A8A1016345).

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