Development Status of Very High-density LEU Dispersion Targets Using Atomized Uranium-Aluminum Alloy-Powders

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1. Introduction

One of the representative medical radioisotopes, technetium-99m (^{99m}Tc), is mostly used in nuclear medical diagnostic fields. ^{99m}Tc is one of the decay products of molybdenum-99 (^{99}Mo). In many countries, ^{99}Mo has been produced by irradiating highly-enriched uranium (HEU > 93% U-235) targets in a commercial scale. However, worldwide intentions have been focused on replacing HEU targets with low-enriched uranium (LEU < 20% U-235) targets to reduce proliferation risks. The decrease of uranium enrichment involves a performance degradation in ^{99}Mo production and an increase of radioactive waste [1].

As a remedy, the use of high-density LEU targets has been proposed using the atomized uranium alloy powders [1]. The atomized powder is favorable for high uranium loadings and it showed better irradiation performances compared to pulverized powder. In our previous works, we successfully made out 3.2 g-U/cm^3 targets using atomized U-Al powders [2]. As a next step, very high-density targets in which uranium density is more than 4.0 g-U/cm^3 has been developed. It is expected that very high-density targets can dramatically increase ^{99}Mo production efficiency with a reduced radioactive waste amount.

The target fabrication includes an annealing process to convert α-U and UAl_2 phases to UAl_3 and UAl_4 phases. This phase transformation accompanies a considerable volume expansion, which makes a fabrication failure. This volume expansion is magnified as the uranium density increases. It seems that this is one of the keys in the development of very high-density dispersion targets. In this study, the overall development status of the very high-density target is described focusing on the optimization of the annealing conditions.

2. Methods and Results

2.1 Fabrication of very high-density targets

Very high-density dispersion target plates were fabricated using atomized U-xAl (x = 5, 10, and 15wt%) powders. The target plates were fabricated by adopting a typical fabrication procedure of a research reactor plate fuel including mixing and blending, compaction, heat-treatment, assembling, and hot-rolling. **Fig. 1** shows a typical schematic of the hot rolling process for the fabrication of the dispersion target plates.

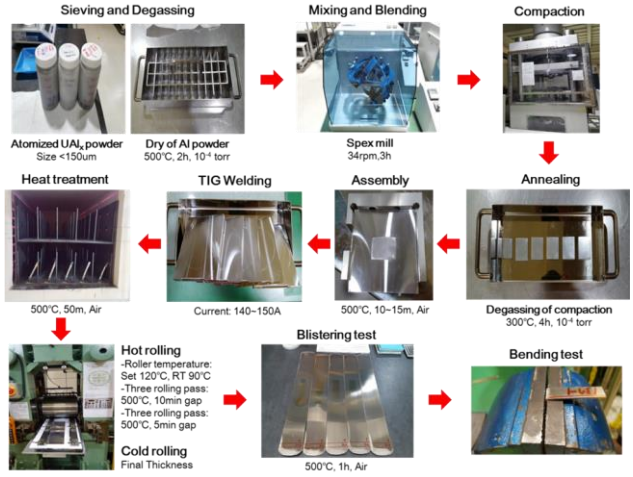


Fig. 1. Schematic of the centrifugal atomization process for the fabrication of the U-Al alloy powder [3].

In this flow, U-xAl (x = 5, 10, and 15 wt%) targets were fabricated as seen in **Fig. 2** (a). The surface condition was fine without any defects. The targets were subsequently subjected to the annealing process. The targets were annealed at 570 °C for 12 hours to transform the U and UAl_2 phases to UAl_3 and UAl_4 phases. **Fig. 2** (b) shows the annealed targets where the cladding burst out due to the excessive volume expansion.

(a) as-fabricated targets (b) annealed targets

Fig. 2. Images of very high-density targets before and after the annealing process.

First, it was considered that thick Al cladding can prevent the bursting out. Hence, the very high-density targets were annealed in the middle of the hot-rolling process instead of annealing at the last step. After the three passes of hot-rolling, the U-15Al targets were annealed at 550 ℃ for 20 hours and the U-5Al and U-10Al targets were annealed 570 ℃ for 12, 15, and 18 hours. **Fig. 3** shows the three-time passed targets after the annealing process. There were similar results. All the targets were burst out.

(a) U-5wt.%Al (b) U-10wt.%Al (c) U-15wt.%Al

Fig. 3. Images of three step passed targets after the annealing process.

2.2 Optimization of the annealing process

The annealing process was applied dividedly after each hot-rolling pass to control the excessive volume expansion and prevent the burst of cladding. It was expected that the short annealing leads a less expansion, which can be hot-rolled again.

First, a condition that the annealing time was evenly distributed was considered. U-5Al and U-10Al targets were annealed at 570 ℃, which followed the annealing condition 1 in Table 1 so that the total annealing time was to be 12 hours. U-15Al targets were annealed at 550 ℃, which followed the annealing condition 6 in Table 2 so that the total annealing time was to be 10 hours. shows the target images during the fabrication. At the early pass, there was very low volume expansion due to the thick cladding thickness. However, as the targets were hot-rolled, the volume expansion became relatively remarkable. The expanded cladding was folded during the hot-rolling process.

In order to avoid the cladding folding, optimization of the annealing conditions was conducted. Long-time annealing was applied after the first and second hot-rolling pass as seen in Table 1 and Table 2. As a result, the folding of cladding was avoided.

Table 1. Annealing conditions of U-5Al and U-10Al.

#	Annealing Temp	Hot-rolling pass					Total time
		1	2	3	4	5	
1	570℃	2h	2h	2h	3h	3h	12h
2	570℃	5h	4h	2h	1h	0h	12h
3	570℃	5h	4h	1h	1h	1h	12h
4	570℃	5h	4h	3h	1h	1h	14h
5	570℃	4h	3h	1h	1h	1h	10h

Table 2. Annealing conditions of U-15Al.

#	Annealing Temp	Hot-rolling pass					Total time
		1	2	3	4	5	
6	550℃	2h	2h	2h	2h	2h	10h
7	550℃	4h	3h	2h	1h	0h	10h
8	550℃	5h	4h	3h	1h	1h	14h

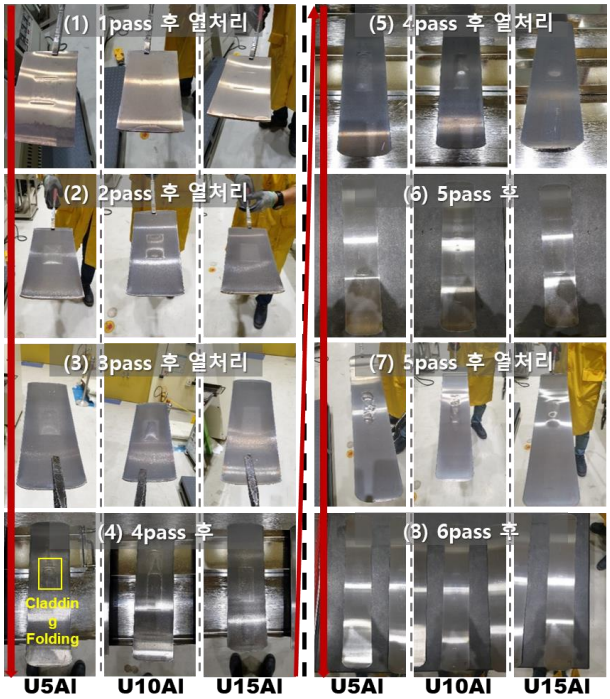
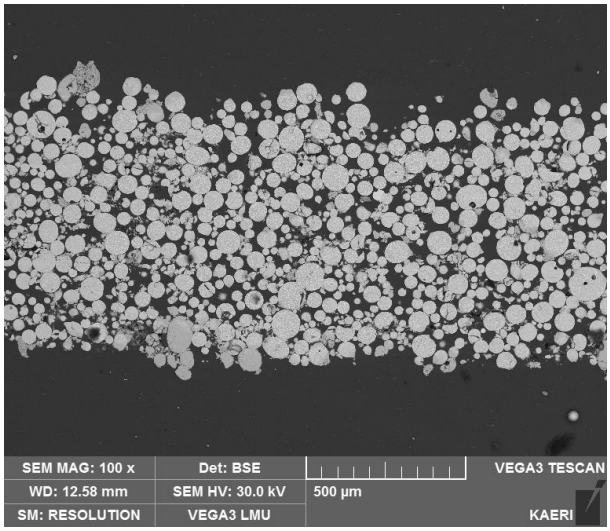


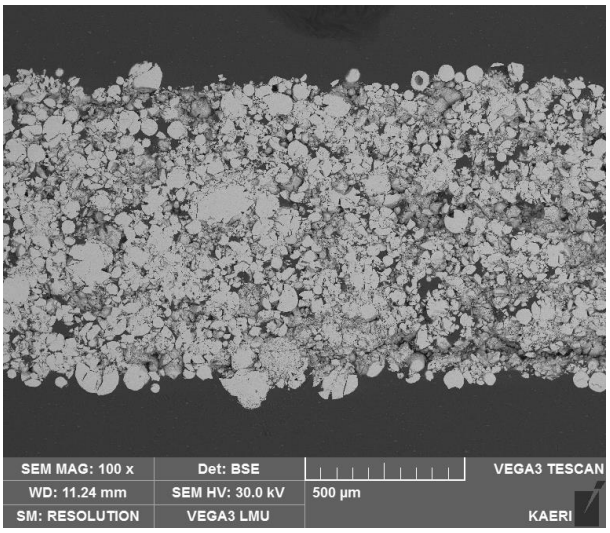
Fig. 4. Images of very high-density targets after each pass and annealing process; annealing condition 1 for U-5Al and U-10Al, annealing condition 6 for U-15Al.

2.3 Microstructure analysis

The microstructures and constituent phases of annealed very high-density targets were identified using scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), and X-ray diffraction (XRD). Microstructure images of unannealed and annealed very high-density U-10Al targets are shown in Fig. 5. The unannealed targets have a very uniform distribution of particles retaining its spherical shape. On the other hands, the particles of the annealed targets swelled and cracked. It seems that most of the Al matrix were consumed for the phase transformations.



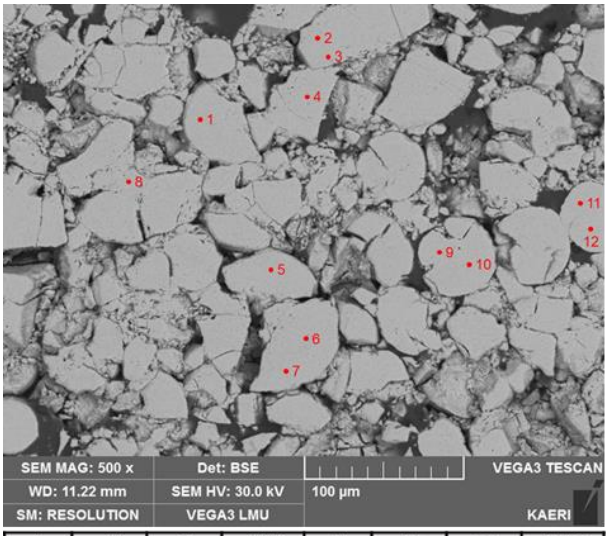
(a) without annealing



(b) with annealing

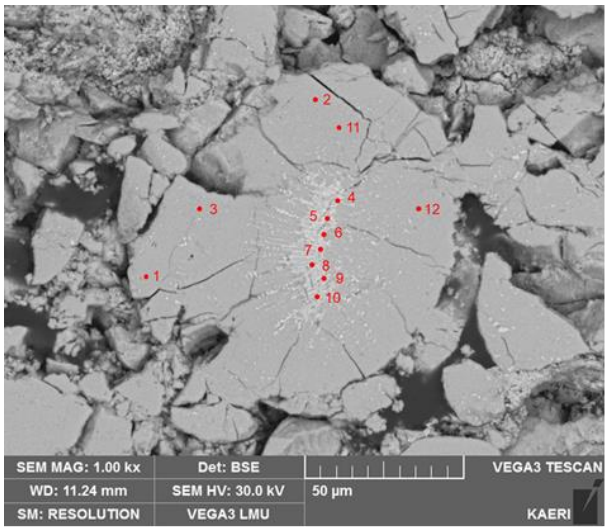
Fig. 5. SEM images of very high-density U-10wt.%Al target plates after 12hours annealing at 570 ℃.

The compositions of U-Al particles were identified as shown in Fig. 5. As seen in Fig. 5 (a), most U-Al phases were successfully changed to UAl_3. Meanwhile, untransformed phases were sparsely observed especially at the sites near the center of particles. The phases of them were expected to be U and UAl_2 phases.
In order to investigate the exact phase, it is required to conduct more phase analyses using XRD. This work is ongoing and the results will be obtained in the near future.



#	Al	U	Al/U	#	Al	U	Al/U
1	75.4	24.6	3.1	7	73.6	26.4	2.8
2	74.7	25.3	3.0	8	72.3	27.7	2.6
3	70.7	29.3	2.4	9	73.5	26.5	2.8
4	75.2	24.8	3.0	10	76.2	23.8	3.2
5	71.5	28.5	2.5	11	70.6	29.4	2.4
6	75.9	24.1	3.1	12	71.2	28.8	2.5

(a) overall region



#	Al	U	Al/U	#	Al	U	Al/U
1	75.0	25.0	3.0	7	55.1	44.9	1.2
2	73.3	26.7	2.7	8	0.0	100	0.0
3	52.3	47.7	1.1	9	38.9	61.1	0.6
4	50.6	49.4	1.0	10	72.0	28.0	2.6
5	53.0	47.0	1.1	11	72.7	27.3	2.7
6	55.0	45.0	1.2	12	73.4	26.6	2.8

(b) at the center of particles

Fig. 6. SEM and EDS results of U-10Al targets annealed at 570 ℃ for 12 hours.

3. Conclusions

For the first time, very high-density LEU targets with a uranium density higher than 4.0 gU/cm^3 were successfully fabricated using atomized uranium-aluminide powders at KAERI. There were many difficulties in the target fabrication, especially on the annealing process. The excessive volume expansion from phase transformation caused bursting out of cladding. Hence, the annealing processes were conducted in the middle of the hot-rolling process. It enables the swollen regions were hot-rolled and prevents the bursting. Annealing of U-5Al and U10Al targets at 570 ℃ for 12 hours led enough phase transformation. Most phases were identified as UAl_3 phase while U and UAl_2 phases were observed at the center of particles. Further optimizations of the target fabrication process are in progress.

REFERENCES

[1] H.J. Ryu, C.K. Kim, M.S. Sim, J.M. Park, J.H. Lee, Development of high density U/Al dispersion plates for 99Mo production using atomized uranium powder. Nucl Eng Technol 45:979–986, (2013)
[2] T.W. Cho, K.N. Kim, S.C. Park, Y.J. Jeong, K.H. Lee, S.H. Kim, J.M. Park, Preliminary Study on High-density LEU Dispersion Targets Using Atomized Uranium-Aluminum Alloy Powder, *Transaction of KNS spring meeting*, Jeju, Korea, (2019)