Development of Atomized Powder Fabrication Technology for High Uranium Density LEU Dispersion Targets

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1. Introduction

Tc-99m can be widely used in the radiopharmaceutical field owing to its short half-life and gamma radiation of 140 keV; also, it is possible to obtain Tc-99m from the decay of Mo-99[1]. Uranium aluminide(UAl_x) - aluminum matrix dispersion targets have been used to produce Mo-99 radioisotope in a research reactor for medical diagnostic purposes[2]. However, major producers of Mo-99 still use high enriched uranium(HEU) targets, which is not recommended for use under the nuclear non-proliferation policy[3]. It is necessary, therefore, to develop high-uranium-density targets with low enriched uranium (LEU) to replace HEU targets[4].

The Korea Atomic Energy Research Institute (KAERI) has been developing high-density LEU targets using atomized U-Al alloy powder. Atomization is a key technology for achieving high-uranium-density because atomized powder is able to have various U-Al compositions and a high uranium content. Fig. 1 shows the atomization technology that can fabricate spherical powder; its process is much simpler than that of conventional comminuted technology. The atomized powder has high purity with fewer defects, excellent irradiation performance, and high production yield rate. In this study, U-xAl (x = 0, 5, 10, 15, 20, and 25 wt.%) powders for high-uranium-density targets were fabricated using centrifugal atomization.

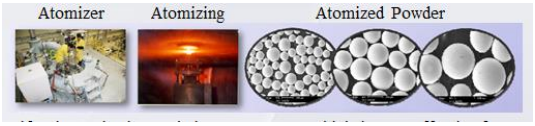

Fig. 1. KAERI centrifugal atomization technology

2. Experimental Procedures

Fig. 2 provides a schematic of the centrifugal atomization process for the fabrication of the U-Al alloy powder. The first step was the arc melting process with U and Al as raw materials. Various compositions of U-xAl alloys (x = 5, 10, 15, 20, and 25 wt.%) were fabricated from arc melting process, which is necessary to reduce thermal shock during atomization.

The mother alloy obtained from the arc melting process was used for the atomization. The atomization used a high frequency induction heating system; a carbon crucible set was used as a susceptor. Before feeding molten alloy onto the rotating disk, which reached a speed of up to 40,000 RPM, superheating was necessary, which required a temperature 200℃ higher than the melting point of U-xAl, to increase the fluidity of the molten metal. Ceramic crucible was made of yttria-stabilized zirconia(YSZ). Molten alloy metal droplets were spread from the rotating disk toward the chamber wall and cooled rapidly under Ar atmosphere. Sieving processes followed to obtain a U-Al powder with diameter under 150 ㎛. The particle surface was observed through scanning electron microscopy, and the fractions of U, UAl_2, and UAl_3 in each U-xAl powder were identified using X-ray diffraction.

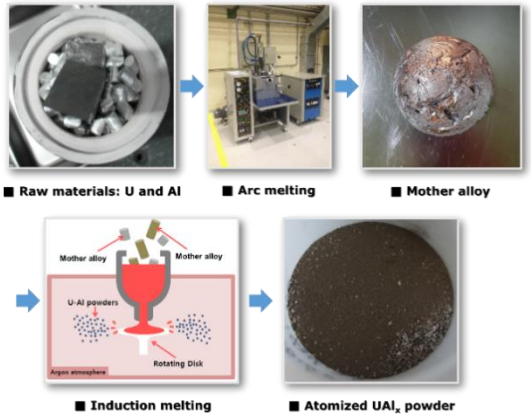

Fig. 2. Schematic of centrifugal atomization process for fabrication of U-Al alloy powder[5]

3. Results and Discussion

3.1 Powder Fabrication

The pouring temperature and yield rates for each U-xAl (x= 0, 5, 10, 15, 20, and 25 wt.%) are presented in Table I. The pouring temperature had to be set based on the phase diagram of U-Al system, as shown in Fig. 3. Higher Al content led to lower yield rate because UAl_2 phase alloy, which has a melting point of 1620℃ (the highest among the U-Al intermetallic compounds), solidified more on the disk surface and in the crucible during the rapid cooling in the atomization chamber.

Table I: Yield rate of the U-xAl (x= 0, 5, 10, 15, 20, and 25 wt.%) powder

U-xAl	Pouring Temp. (℃)	Total Loading (g)	Fabricated Powder (g)	Yield Rate (%)
0Al	1,415	3,230	3,135	97.1
5Al	1,658	1,044	1,006	96.3
10Al	1,725	1,045	969	92.7
15Al	1,750	1,048	957	91.4
20Al	1,800	1,080	940	87.0
25Al	1,800	954	815	85.5

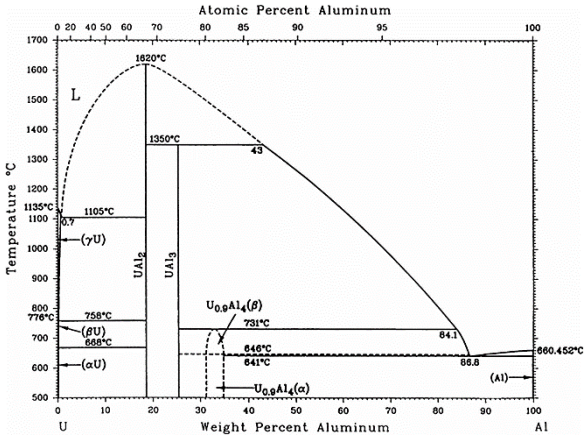

Fig. 3. Phase diagram of U-Al system[6]

Table II: Sieving data of the U-xAl (x= 0, 5, 10, 15, and 20 wt.%) powder for diameters under and over 150 ㎛

U-xAl	under 150 ㎛ (wt.%)	over 150 ㎛ (wt.%)
0Al	99.6	0.4
5Al	98.2	1.8
10Al	95.4	4.6
15Al	93.7	6.3
20Al	89.1	10.9

The sieving results with the 150 ㎛ sieve are presented in Table II. The under 150 ㎛ powder proportion decreased as Al content increased. Higher Al content contributed to lighter weight of droplets, so that the density of the molten alloy became decreased.

Champagne and Angers developed an empirical model for determining the diameter of droplets[7]. The diameter was given as:

$$d = 4.63 \times 10^{6} \frac{\gamma^{0.43} Q^{0.12}}{\rho^{0.43} \omega^{0.98} D^{0.64}}$$

where d is the diameter of droplets in ㎛, γ is the interfacial energy in J/m^2, Q is the liquid supply rate in m^3/s, ρ is the density in kg/m^3, ω is the angular velocity in rad/s, and D is the rotating disk diameter in m. According to the formula above, the density(ρ) and the diameter(d) were in inverse proportion to each other. More over-150 ㎛ particles, therefore, were fabricated as Al content increased.

3.2 Microstructure and Phase Fraction of U, UAl_2, and UAl_3

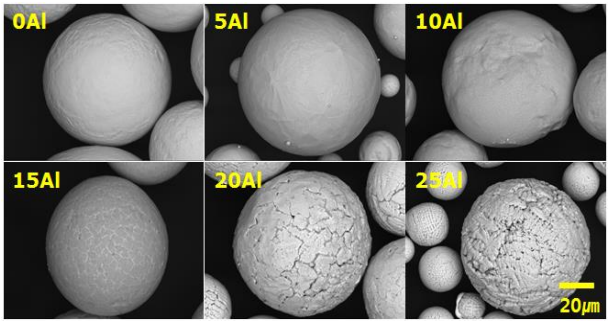

Fig. 4. SEM images collected on U-xAl (x= 0, 5, 10, 15, 20, 25 wt.%) particle surface[5]

U-Al_x powder was observed that spherical particles with average diameter of 70 ㎛ were obtained, as shown in Fig. 4. When Al content was more than 20 wt.%, remarkable cracks occurred on the surfaces of the particles owing to solidification shrinkage of the liquid phase during cooling in the atomization chamber with Ar atmosphere[5].

Table III: XRD quantitative analysis for U, UAl_2, and UAl_3 fraction of U-xAl (x= 0, 5, 10, 15, 20, and 25 wt.%) powder

U-xAl	U (wt.%)	UAl_2 (wt.%)	UAl_3 (wt.%)
0Al	100	0	0
5Al	68	32	0
10Al	27	73	0
15Al	13	87	0
20Al	0	80	20
25Al	0	25	75

The data from the quantitative analysis for UAl_x by XRD is presented in Table III. The table shows the fractions of U, UAl_2, and UAl_3 in each U-xAl powder. UAl_2 increased from 32 wt.% in U-5Al to 87 wt.% in U-15Al and then deceased. UAl_3 increased from 20 wt.% in U-20Al to 75 wt.% in U-25Al. This is explained by Fig. 3, which shows that UAl_2 formed from 18wt.% Al and UAl_3 formed from 25wt.% Al, respectively. $U_{0.9}Al_4$ was not found because it formed from 31 wt.% Al.

4. Conclusions

Considering the analysis results, we can conclude that KAERI has successfully developed a fabrication method for U- xAl (x= 0, 5, 10, 15, 20, and 25 wt.%) powder using atomization technology. The phase fraction table, determined using XRD quantitative analysis, can help in follow-up processes such as mixing, compaction, and

heat treatment, which mainly require UAl_3 phase. This study can be used as important evidence for the fabrication process of high-density LEU dispersion targets.

Acknowledgements

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