Improvement and Optimization of Atomization Technology for U_3Si_2 Powder Fabrication

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1. Introduction

KAERI has been developing atomization technology, which is a key technology for achieving high-density low-enriched uranium (LEU) fuel. With the atomization technology presented in Fig. 1, KAERI can fabricate a spherical type of powder via a process much simpler than that used to make the conventional comminuted type. Atomized powder is known for its high purity with fewer defects, excellent irradiation performance, and high production yield rates.

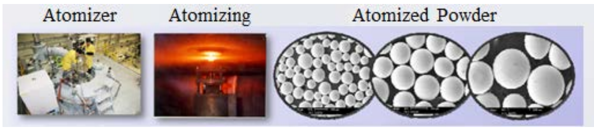

Fig. 1. KAERI centrifugal atomization technology [1]

The melting point of U_3Si_2 is 1665℃, meaning that U_3Si_2 molten alloy should be heated to more than 1800℃ before it can be poured molten alloy onto a rotating disk. U_3Si_2 droplets spread from edge of the rotating disk only for a few minutes and then solidify rapidly when flying in the atomization chamber.

During the atomization process, the parts made of carbon, i.e., the disk, must be coated to protect them from reacting chemically with the molten U-metal alloys. The conventional coating method with yttria-stabilized zirconia (YSZ) powder involves sprayed this powder onto the carbon surfaces of the parts through a plasma spray-coating process. The YSZ-coated surface can also be coated additionally by applying a releasing agent if needed.

The U_3Si_2 atomization process with its severe molten alloy temperatures causes a chemical reaction between the U_3Si_2 and parts made of carbon due to the lack of durability of the coatings on the surfaces of these parts. Although the atomization process at KAERI was designed to fabricate more than 3 kg of powder per batch, fabrication amounts were currently limited to less than 1.2 kg per batch to alleviate this problem.

2. Methods and Results

2.1 U_3Si_2 atomization experiments

To improve the U_3Si_2 atomization process, two goals were set: soundness of the coating on parts made of carbon to be maintained until the end of atomization and an increase in powder productivity from 1.2 kg to more than 3 kg per batch. Depleted uranium (DU) was used for three steps of the atomization experiments, which involved eight batches in total. The results of the U_3Si_2 (U-7.6wt.%Si) atomization improvement experiments are presented in Table I.

Table I: The Results of U_3Si_2 Atomization Experiments

Batch	Total Loading (g)	Powder (150 ㎛↓) (g)	Yield Rate (%)	Coating Soundness of Parts
C2208	2,267.13	1,329.58	58.65	X
C2211	2,207.92	2,100.85	95.15	O
C2212	3,288.66	0.00	0.00	O
C2213	3,260.80	2,249.72	68.99	X
First Subtotal	11,024.51	5,680.15	51.52	-
C2214	3,303.79	3,102.32	93.90	X
C2215	2,287.03	2,215.29	96.86	X
Second Subtotal	5,590.82	5,317.61	95.11	-
C2217	3,337.69	3,190.02	95.58	O
C2218	3,290.57	3,151.46	95.77	O
Third Subtotal	6,628.26	6,341.48	95.67	-
Overall total	23,243.59	17.339.24	74.60	-

For the first four experiment batches of C2208 to C2213, the results showed a total yield rate of 51.52%, and coating problems occurred in C2208 and C2213. New process parameters during U_3Si_2 atomization were tested with these four batches. These non-optimized parameters caused both a low yield and coating soundness problems. The damaged disk noted for the C2208 batch, related to the coating problems including a chemical reaction between U_3Si_2 and the disk is shown in Fig. 2.

To improve the yield rate and durability of the coating, we optimized the process parameters, specifically temperature control of the molten U_3Si_2 alloy. The total yield rate from C2214 to C2215 increased to 95.11%. Except for the disks, the coating soundness of the other carbon parts showed an improvement in both batches. However, chemical reactions between the center of the disk and the U_3Si_2 alloy, as presented in Fig. 3, were found to have occurred in both batches. Increasing the loading of each batch to approximately 3.3kg caused a longer droplet spreading time from the disk such that the chemical reactions thus occurred. The coating soundness of the disks was needed to be improved.

Fig. 2. Damaged disk from the C2208 Batch

Fig. 3. Chemical reaction effects between the center of the disk and the U_3Si_2 alloy in the C2214 batch

Before starting the third experiment with the batches of C2217 to C2218, the coating thickness and coating powder size on the disks were optimized to improve the durability and adhesion of the disk coating. Two coating conditions were applied to the disks. In the first condition for the C2217 batch, the coating thickness was increased to 167%, with the same coating powder size used previously. The second condition for the C2218 batch was one in which the coating powder was decreased by 65% with the aforementioned 167% increase in the coating thickness. These two disk-coating conditions are presented in Table II.

Table II: Disk Coating Conditions for the C2217 and C2218 Batches

Batch	Disk Coating Thickness	Disk Coating Powder Size
C2217	167% Increase	Same as Previous
C2218	167% Increase	65% decrease

After the C2217 and C2218 batches, results showed a total yield rate of 95.67%, with the coating soundness of all carbon parts improved. There were no chemical reactions between molten U_3Si_2 alloy and disks. The disks applied to the C2217 and C2218 batches showed that both the increase in the disk coating thickness and the decrease in the disk coating powder size had a positive effect.

2.2 Particle Size Analysis

The C2211, C2214, C2215, C2217 and C2218 batches were chosen for a particle size analysis because the yield rates of these batches were approximately 95% and no abnormalities arose during the atomization process, except for the chemical reactions between U_3Si_2 alloy and disks. The particle size analysis was conducted based on ASTM B214-15 and B215-15, which specify both the standard sieve analysis method and the sampling of the metal powders.

Fig. 4. Multi-level slot sampler

Fig. 5. Vibration action sieve shaker

KAERI used the multi-level slot sampler presented in Fig. 4 and sampled 100±10 g of U_3Si_2 powders for each batch which had been sieved through a 100-mesh sieve (150 μm) in advance. To conduct the sieve analysis of the U_3Si_2 powders, KAERI used a vibration action sieve shaker using sieves 203 mm in diameter, as presented in Fig. 5. Here, 140-mesh (106 μm), 200-mesh (75 μm), 230-mesh (63 μm), and 325 mesh (45 μm) sieves were used. These were assembled with the coarsest sieve at the top, and the setup was operated for 15 minutes after pouring the sampled U_3Si_2 powders onto the coarsest sieve at the top. After sieving, the procedures of cleaning the sieves and collecting the powders proceeded. The sum of the masses of all fractions had to be more than 99 wt.% of the initial mass for credibility of the analysis [2].

The particle size analysis results are presented in Table III and Fig. 6. Except for the C2211 batch, the C2214, C2215, C2217 and C2218 batches had results showing two peaks associated with the -140+200 mesh and the -230+325 mesh. The C2211 batch had two peaks associated with the -325 mesh and the -230+325 mesh, indicating that the C2211 batch had finer powders than the other batches. This particularity of the C2211 batch was attributed to the relatively high U_3Si_2 molten alloy temperature when pouring the molten alloy onto the rotating disk. The first four experimental batches, including the C2211 batch, were tested at a higher U_3Si_2 molten alloy temperature than the last four batches. The higher molten alloy temperature improved the alloy's flowability, which resulted in low viscosity.

Champagne and Angers developed an empirical model for determining the diameter of droplets[3]. In this method, it is explained that the viscosity (η) and particle diameter (d) are directly proportional to each other. The particle size analysis results were in good agreement with Champagne and Angers' findings.

Table III: Particle Size Analysis Results

	C2211 (wt.%)	C2214 (wt.%)	C2215 (wt.%)	C2217 (wt.%)	C2218 (wt.%)
-325 mesh (≦45 ㎛)	28.45	17.12	17.47	16.02	12.98
-230+325 mesh (≦63>45 ㎛)	31.36	30.81	36.19	30.66	28.20
-200+230 mesh (≦75>63 ㎛)	14.37	14.02	17.27	18.02	17.03
-140+200 mesh (≦106>75 ㎛)	24.53	34.67	27.68	32.52	37.88
-100+140 mesh (≦150>106 ㎛)	1.29	3.38	1.40	2.78	3.90

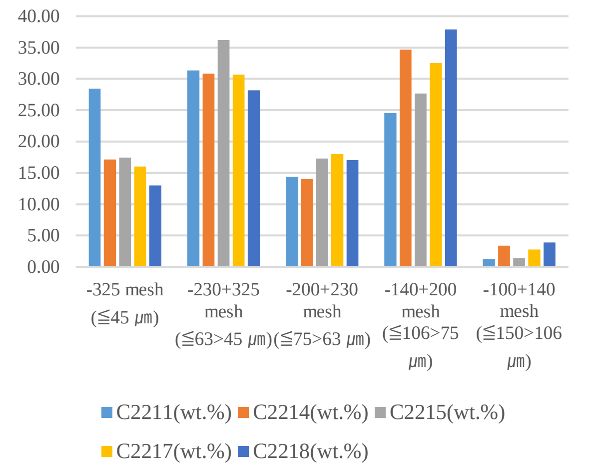

Fig. 6. Particle size distribution graph

2.3 Chemical Analysis

Chemical analyses of the C2211, C2214, C2215, C2217 and C2218 batches were also conducted. These results are presented in Table IV.

Except for Fe and Al impurities stemming from DU, all batches met the specification requirements for impurity levels. The C2214 and C2215 batches, for which chemical reactions occurred between the disks and the U_3Si_2 alloy, showed carbon impurity outcomes of 870 and 610 µg/g, respectively. These carbon impurity results for the C2214 and C2215 batches were slightly higher or similar to those of the C2211, C2217 and C2218 batches.

Table IV: Chemical Analysis Result

Unit : µg/g *wt%

	C2211	C2214	C2215	C2217	C2218
Si*	7.67	7.59	7.75	7.55	7.58
C	540	870	610	640	600
H	<1	14	3	8	8
O	1780	1600	1460	1420	1510
N	<5	<5	<5	<5	40
Zr	1200	1000	900	1100	1100
Y	<50	<50	<50	<50	<50
Fe	1600	800	800	800	800
Al	600	800	800	800	800
Cu	<50	100	100	100	100
Ni	<50	<50	<50	<50	<50
Zn	<50	<50	<50	<50	<50
Li	<5	<5	<5	<5	<5
Co	<5	<5	<5	<5	<5
Cd	<5	<5	<5	<5	<5
B	<5	<5	<5	<5	<5

3. Conclusion

Atomized U_3Si_2 powders were produced at less than 1.2 kg per batch with a low yield rate due to severe process conditions, including a required U_3Si_2 molten alloy temperature of more than 1800℃. To improve KAERI's atomization technology for U_3Si_2 powder, the coating conditions on parts made of carbon and temperature control process parameters required optimization.

Considering the results of the analyses conducted here, new coating conditions and process parameters set for U_3Si_2 have been proven and qualified. The results of this study can be used to design and establish effective manufacturing procedures for atomized LEU_3Si_2 powder with a 95% yield rate and 3kg powder per batch.

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