

USE OF A CENTRIFUGAL ATOMIZATION PROCESS IN THE DEVELOPMENT OF RESEARCH REACTOR FUEL

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Received September 27, 2007

A centrifugal atomization process for uranium fuel was developed in order to fabricate high uranium density dispersion fuel for advanced research reactors. Spherical powders of U_3Si and U-Mo were successfully fabricated and dispersed in aluminum matrices. Thermal and mechanical properties of dispersion fuel meat were characterized. Irradiation tests at the research reactor HANARO confirm the excellent performance of high uranium density dispersion fuel.

KEYWORDS : Centrifugal Atomization, Research Reactor, Dispersion Fuel, U_3Si , U-Mo, Spherical Powder, Rotating Disk

1. INTRODUCTION

Research reactors are used for research and training, materials testing, neutron activation analysis, and the production of radioisotopes for medicine and industry. The fuel plates used in the fuel elements for most research and test reactors consist of a fuel core in aluminum alloy cladding [1]. The question of enrichment has been a major focus, especially in relation to the security concerns of the early 1970s. It was concluded that in order to guard against weapons proliferation from HEU(High Enriched Uranium) fuels enrichment should be reduced to no more than 20% U-235. The program for Reduced Enrichment for Research and Test Reactors (RERTR) was launched by the USA in 1978 [2]. The RERTR program has led to the development and qualification of new high uranium density and LEU(Low Enriched Uranium) fuels. Enrichment can be lowered by increasing the uranium density. The original uranium density of U-Al fuel types was about 1.3 g/cm³ to 1.7 g/cm³. The RERTR program chose to pursue the use of high-density U-Si alloys instead of UAl_x and U_3O_8 in conventional aluminum matrix dispersion fuel to take advantage of the large commercial base of available equipment and experience with respect to the fabrication of such fuels. One uranium silicide compound, U_3Si_2 , has been found to perform extremely well under irradiation and can provide a uranium density of at least 4.8 g U/cc. There were successful tests with denser U_3Si -Al dispersion fuel up to 6.1 g U/cc [1-4].

The RERTR program concentrates on reactors over 1 MW, which have significant fuel requirements. In 2007, the program has set a target of 129 reactors out of the 207

reactors that use HEU fuel. To early 2007, 48 research reactors (13 in the USA) either had been or were being converted to LEU silicide fuel, and another 53 are convertible using present fuels and 28 more, mostly Russian designs, need higher-density fuels not yet available. The goal is to convert 105 reactors by 2013 [5].

This development work has been undertaken to provide fuels that can extend the use of LEU fuel to those reactors requiring higher densities than available in silicide dispersions and to provide a fuel that can be more easily reprocessed than the silicide type. The US RERTR program at the Argonne National Laboratory recommenced its R&D activities in 1996 with the application of gamma-phase (such as U-Mo) alloy dispersion fuels. The French U-Mo Group (CEA, CERCA, COGEMA, Framatome-ANP and Technicatome) has been involved in U-Mo R&D activities since 1999 and the Argentine Atomic Energy Commission (CNEA) has been involved since 2000. An international effort is underway to develop, qualify and license a high-density fuel based on U-Mo alloy dispersed in aluminum, with a U-density of 6 g/cm³ to 8 g/cm³. Approval of this fuel was expected in 2006 but tests since 2003 have failed to confirm performance due to unstable swelling under high flux irradiation condition, and the target is now 2010 [5].

In a further stage of U-Mo fuel development, which has now become the main priority, the Argonne National Laboratory, CEA and CNEA are testing U-Mo fuel in a monolithic form, instead of the dispersion of U-Mo in aluminum. The uranium density is 15.6 g/cm³ and this would enable every research reactor in the world to convert from HEU to LEU fuel without loss of performance [6].

This paper focuses on the use of a centrifugal atomization process in fuel development activities.

2. USE OF A ROTATING DISK CENTRIFUGAL PROCESS IN THE DEVELOPMENT OF TECHNOLOGY FOR THE FABRICATION OF FUEL POWDER

The technology for developing U₃Si dispersion rod-type fuel was launched for the localization of fuel at the High-Flux Advanced Neutron Application Reactor (HANARO). Conventionally, U₃Si powder is produced by comminuting a U₃Si billet. However, because U₃Si has a hardness of 200 VHN and elongation of about 1%, the powdering work is tedious and laborious. An alternative way of fabricating the powder is to use an atomization process from the melt [7]. Furthermore, in the case of U-Mo powder, U-7wt%Mo is relatively tough with an ultimate tensile strength of more than 90 kg/mm² and elongation of about 5%. The task of making U-Mo powder by a mechanical method of breaking or machining is difficult. We chose the rotating disk atomization process because it enables the melt to be handled simply and it has a relatively narrow particle size distribution [8]. As shown in Fig. 1, the rotating disk atomization system involves the tasks of melting, atomizing, and collecting. The melting chamber contains a crucible with a nozzle, a heat-generating ring by induction, an insulator, and an induction coil. The atomizing chamber has a rotating disk and a driving motor. The diameter of the atomizing chamber is 2.5 m, which allows sufficient solidification before the atomized melt droplets reach the chamber wall. Through the successful development of our rotating disk atomization technology, we have simplified the process, enabled mass production, improved the yield, and obtained a purer product.

When the molten metal is heated to a temperature that is approximately 200 K higher than the melting point, it is fed through a small nozzle onto a rotating disk in an argon atmosphere. The alloy melt droplets, which formed from the melt by centrifugal force, are spread from the disk toward the atomizing chamber wall by the flowing argon gas during the flight of the droplets. In general, the atomized particle size is influenced as follows by process parameters such as the revolution speed, the surface tension, the melt density, the melt feeding rate and the disk diameter [9]:

$$d = 3.65 \cdot 10^6 \cdot \frac{1}{\omega^{0.96}} \cdot \left(\frac{\gamma}{\rho} \right)^{0.43} \cdot \frac{Q^{0.12}}{D^{0.64}}, \quad (1)$$

ω = disk revolution

γ = surface tension

ρ = melt density

Q = melt feed rate

D = disk diameter.

The atomized particle size tends to decrease when the disk revolution speed, the melt density, and the disk diameter decrease; in contrast, the atomized particle size tends to increase when the surface tension of melt and the melt-feeding rate increase. In general, atomized particles have a rapidly solidified microstructure, a spherical shape, and a relatively narrow particle size distribution. The melt that leaves the tip of the disk forms droplets by means of the uniform spreading force, yielding particles with a relatively narrow particle size distribution. The powders, whose particles vary in diameter from tens of microns to hundreds of microns, are easily available for the rotating disk atomization process. The spherical shape is influenced by the surface tension of the droplet. The large specific area of fine melt droplets enables the melt droplets to cool rapidly.

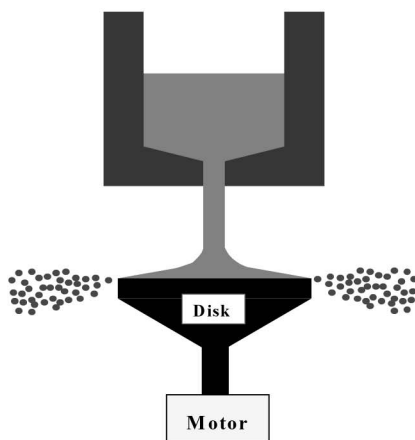


Fig. 1. (a) A Schematic Illustration and (b) a Photograph of the Centrifugal Atomization Process

2.1. The U_3Si Fuel Powder Produced by Rotating Disk Atomization

The particle size of U_3Si powder for HANARO fuel must be less than $150\text{ }\mu\text{m}$ in diameter. The rotating disk atomization process can meet this requirement. Normally the as-cast structure of U_3Si alloy exists as a U_3Si_2 precipitated phase in the eutectic phase of U and U_3Si_2 [7]. Heat treatment is required to transform this alloy to U_3Si . A cast U_3Si alloy billet usually takes about 72 h due to the large U_3Si_2 precipitates. The atomized U_3Si alloy powder can be easily heat-treated in 6 h because the U_3Si_2 precipitate is 10 times smaller than that in the as-cast U_3Si alloy billet [7].

2.2. The U-Mo Powder Produced by Rotating Disk Atomization

Figure 2 shows that the U-Mo particles produced by rotating disk atomization are spherical with a smooth surface [8]. The densities of the atomized powders are

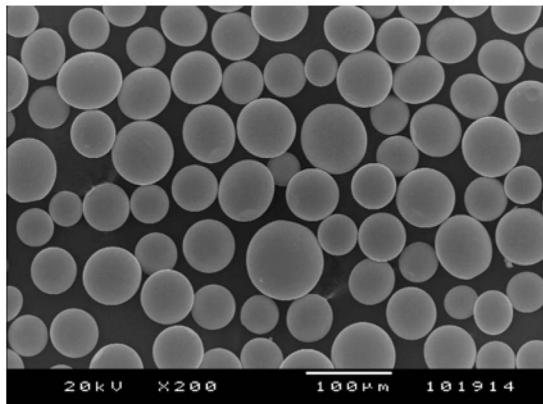


Fig. 2. Morphology of the Atomized U-Mo Alloy Powder

lower than the densities of powders from cast alloy due to the internal porosities of some particles. The internal porosity is known to form as the cooling gas is captured during the flight of droplet through the cooling chamber.

Figure 3 shows a typical cross-sectional SEM micrograph of the U-Mo atomized powders. A cellular structure appears in the atomized U-Mo particle but the grain boundaries are difficult to be distinguishable. The cell structure is thought to occur as a result of the Mo segregation during the solidification, which is caused by the substantial liquid-solidus gap of the U-Mo alloy [8]. Compositional analysis by means of EDS reveals that, irrespective of the alloy composition, the Mo content at the cell boundaries is about 2 at% to 3 at% lower than that inside the cell. The cell size is less than $5\text{ }\mu\text{m}$. In general, the cell size decreases as the particle size becomes smaller because a higher cooling rate of the smaller droplets tends to suppress Mo segregation during solidification. The cell morphology varies somewhat as the Mo concentration varies. As the Mo concentration increases from 5.4 wt% to 10 wt%, the thickness of the cell boundary increases from about $0.3\text{ }\mu\text{m}$ to about $0.6\text{ }\mu\text{m}$ and the size of the cell remains the same [10]. X-ray diffraction analysis for the atomized U-7Mo particles indicates that a meta-stable gamma structure is formed as a result of the rapid cooling effect.

Analysis of the neutron diffraction patterns for U-(5.4-10wt%)Mo alloy powders shows that two kinds of γ -U phases, namely γ_1 -U inside the cell and γ_2 -U at the periphery of the cell [10]. Two phases of γ_1 -U and γ_2 -U appeared to have a crystal structure having Im3m bcc space group. However, the peaks of the two phases could be distinguishable, mostly at high diffraction angles, with a higher instrumental resolution. In the case of the U-10wt%Mo, the lattice constants were 0.34116 nm for the γ_1 phase and 0.34261 nm for the γ_2 phase, respectively. As the Mo concentration increases, the lattice parameters

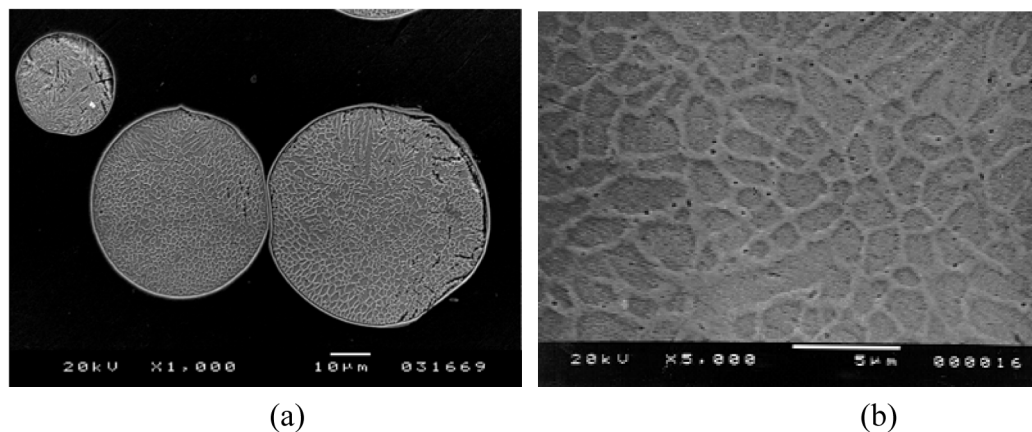


Fig. 3. Cross-sectional SEM Images of the Atomized U-Mo Powders

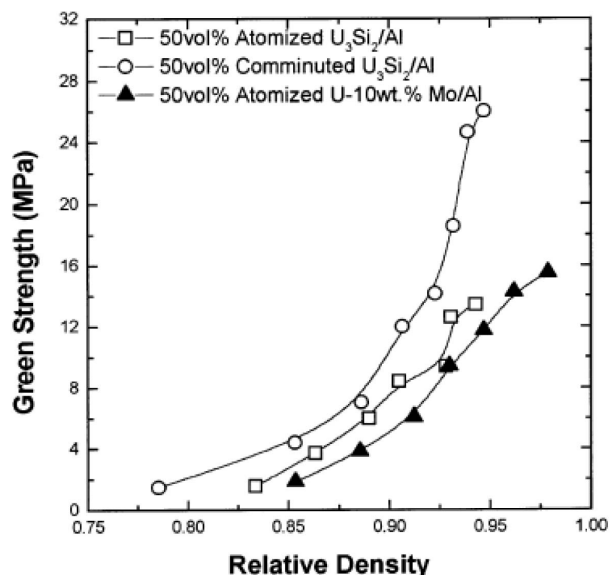


Fig. 4. The Green Strength of the Compacts of 50 vol% Atomized U-10Mo, 50 vol% Atomized U_3Si_2 and 50 vol% Comminuted U_3Si_2 with a Varying Relative Density

of both the γ -U phase and the β -U phase generally increase linearly with an increased fraction of the β -U phase.

3. IMPROVEMENT OF DISPERSION FUEL MEAT THROUGH THE IMPLEMENTATION OF ATOMIZED FUEL POWDER

As shown in Fig. 1, the green strengths of the fuel meat compacts of comminuted fuel powder are much larger than those of the compacts of atomized powder [11,12]. The green strength of a powder compact is mainly due to the bonding force of the interparticle metallic contact during the compacting process, and the bonding force of the interparticle metallic contact is enhanced as the plastic deformation of particles increases. The bonding force between Al particles is much larger than that between Al and fuel particles; thus, the green strength of the compact can be controlled by the contact area between the Al particles and the fuel particles. The contact area between the Al particles and the comminuted fuel particles is larger than that between Al particles and atomized fuel particles. Therefore, the green strength of the comminuted fuel-Al compact is much greater than that of the atomized fuel-Al compact. The shape of the particles clearly plays a major role in the green strength of the fuel-Al compacts.

The thermal diffusivities in transverse and longitudinal directions for atomized and comminuted U_3Si -Al fuel meat rods were measured to investigate the effect of particle shape on thermal conductivity [13,14]. The diffusivities of atomized fuel meats appear to be slightly lower along

the longitudinal direction than those of the comminuted fuel meats. Along the transverse direction, however, the diffusivities of the atomized fuel meats are higher. This tendency becomes more apparent as the U_3Si content increases. The comminuted angular particles are aligned along the extrusion direction, whereas the atomized spherical particles are randomly distributed. The effective thermal diffusivity is generally improved through the use of atomized spherical U_3Si powder, probably due to the reduction of porosity as well as the isotropic shape of the particles [13]. The thermal conductivity of U-Mo/Al fuel meats, in general, appears to have a linear dependence with the U-loadings and to be similar with the thermal conductivity of the U_3Si/Al fuel meats (Fig. 5).

The extrusion pressure of the mixture of atomized U_3Si powders is lower than that of the mixture of comminuted U_3Si powder [14]. The pressure difference increases as the volume fraction of the U_3Si powders increases. The spherical atomized particles with a smooth surface are thought to allow the Al matrix to flow more easily than the comminuted particles with a rough angular surface.

Figure 6 shows that the tensile strength of the atomized fuel meats of U_3Si and U-10wt%Mo is generally lower than that of comminuted fuel meats because comminuted particles are aligned along the extrusion direction. Fuel particles with a certain aspect ratio rotate and lie in the extrusion direction. Comminuted fuel particles are reoriented in the extrusion direction of the fuel meat. In contrast, atomized particles are isotropically orientated in the fuel meat. The atomized particles never affect the aspect ratio because they are mostly spherical. However, the elongation of the atomized fuel meats is much higher than that of the comminuted fuel meats. Observations of the fractured surface show that the aluminum matrix is mostly bonded with the atomized particles, whereas some of the comminuted

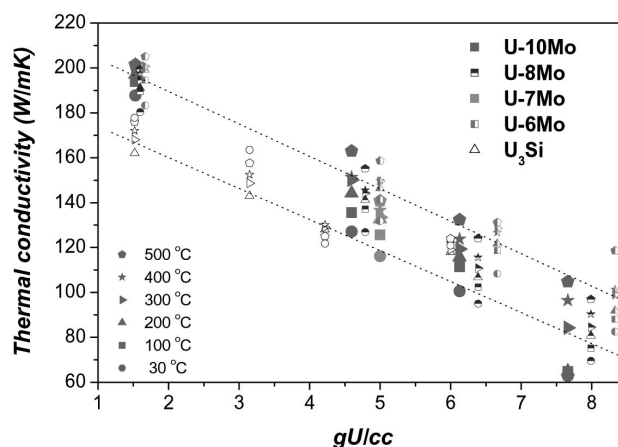


Fig. 5. Comparison of Thermal Conductivities of Both U-Mo/Al and U_3Si/Al Fuel Meats

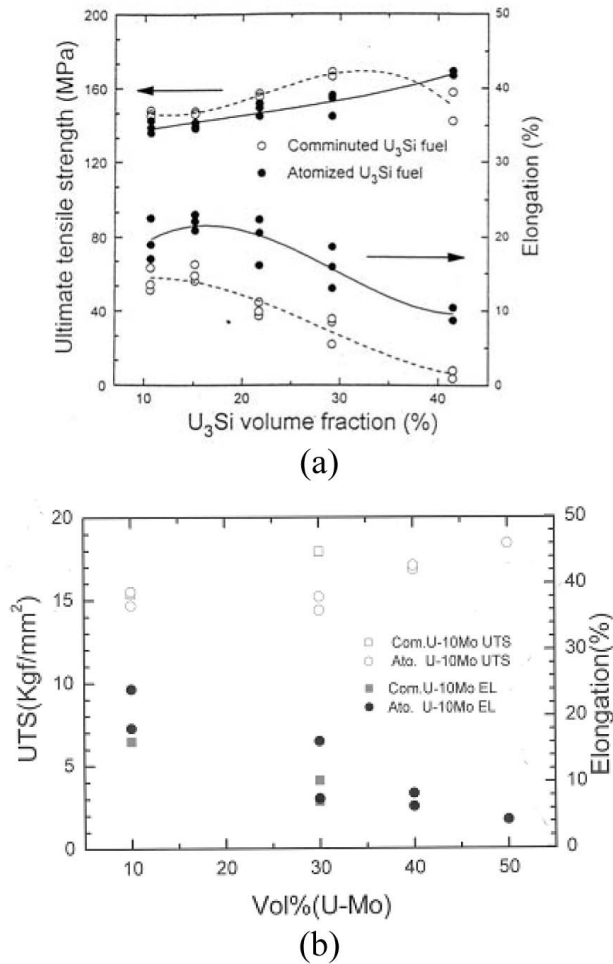


Fig. 6. (a) Tensile Properties at Room Temperature of Atomized and Comminuted U_3Si/Al Fuel Meats; (b) Tensile Properties at Room Temperature of Atomized and Comminuted $U-10Mo/Al$ Fuel Meats

particles are pulled out or fractured in a transgranular manner [14].

Thermal swelling occurs as a result of the reaction between dispersed fuel particles and the aluminum matrix in the dispersion fuels. The volume expansion during thermal annealing is known to be caused by the formation of uranium aluminide phases that have a low density due to the chemical reaction between fuel particles and aluminum matrix. Figure 7 shows that, at any test temperature, the atomized U_3Si/Al fuel meats show fewer volume changes than the comminuted powder fuel meats [15]. The growth values of the atomized U_3Si/Al fuel meats correspond to about 70% of that of comminuted fuel meats at 350°C or 400°C. This phenomenon is likely due to the fact that the specific surface area of the atomized powder is smaller than that of the comminuted irregular powder.

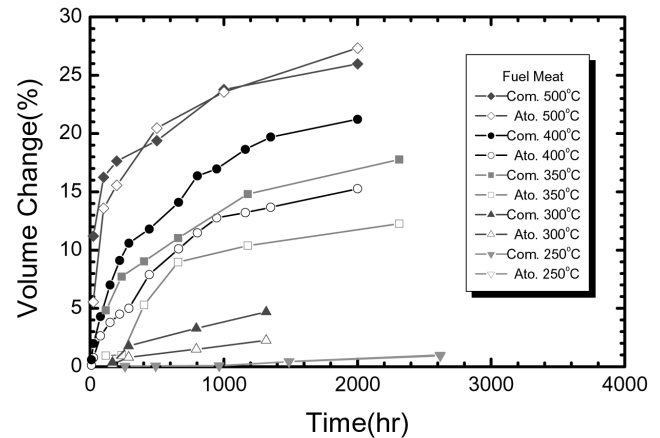


Fig. 7. The Dependency of Temperature and Time on the Thermal Swelling Behavior of Atomized and Comminuted U_3Si-Al Fuel Meats

4. PERFORMANCE COMPARISON OF FUELS PROCESSED BY ATOMIZATION AND COMMINATION

4.1 Irradiation Behavior of Plate-Type Atomized U_3Si_2/Al Dispersion Fuel

An irradiation test was carried out on both the atomized and comminuted U_3Si_2 dispersion mini-plate fuels at HANARO in order to compare the irradiation performance [16]. The mini-plates, which were irradiated until 50 at% to 54 at% U_{235} burn-up at HANARO were subjected to IMEF for post-irradiation analysis. The PIE results confirm that both the atomized and comminuted U_3Si_2 mini-plate fuels exhibit sound swelling behaviors. The aluminide layer that formed in all fuel particles ranges from 1.73 μm to 2.13 μm in thickness, implying a fuel meat swelling of approximately 3.3 vol%. However, the atomized fuel has a bubble distribution with slightly smaller mean diameters than the comminuted sample, and the bubble population in the atomized fuel appears to be more homogeneous than that in the comminuted fuel (Fig. 8).

4.2 Comparison of Irradiation Behaviors of Both Atomized and Comminuted U_3Si/Al Dispersion Fuel

Two irradiation tests were conducted on U_3Si dispersion fuel elements, which were prepared by centrifugal atomization and a conventional comminution method; the tests were performed in connection with the localization of HANARO fuel [17,18]. The first irradiation test was performed at a lower power than normal and with a higher burn-up, 70 at%, than normal

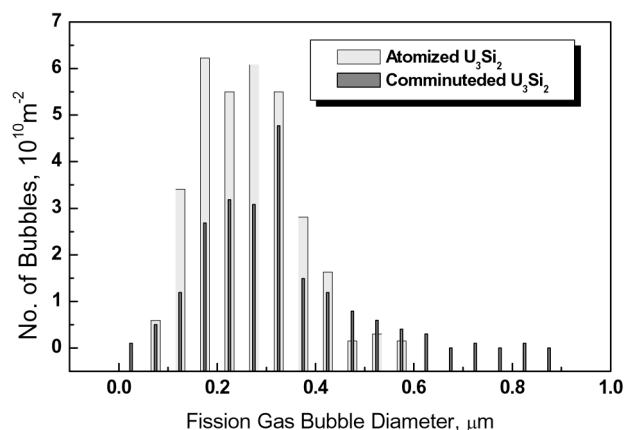


Fig. 8. Bubble Size Distribution of Atomized and Comminuted Fuels

for mini-elements with a shorter length than the standard fuel; the second test was done at the irradiation conditions of normal power and burn-up of 60 at% for full size fuels.

Figure 9 shows typical SEM micrographs of the atomized and comminuted U_3Si fuels after the first irradiation test. The fuel-matrix interaction layer in the atomized particle is relatively uniform and has a generally acceptable average thickness in the range of approximately 9 μm . Irrespective of the powdering method, the U_3Si mini-elements do not have a prominent reaction layer between the U_3Si fuel and aluminum matrix; however, the comminuted particles showed less uniform and even thicker reaction layer than the atomized particles. SEM observation on the cross section of particles reveals a relatively fine and uniform bubble size distribution as well as a relatively thin and uniform

fuel-Al layer, irrespective of the powdering method. Compared with the comminuted fuel, the atomized fuel appears to have a more homogenous bubble population and a narrower bubble size distribution.

In general, the U_3Si particles form a relatively fine and uniform bubble size distribution, irrespective of the powdering method. The bubble population in the atomized fuel particles appears to be finer and more homogeneous than that of the comminuted fuel particles; the atomized fuel particles also have a narrower bubble size distribution. The irradiation behavior of the atomized U_3Si dispersion fuels gives no indication of breakaway swelling and the final swelling of 5 vol% is acceptable.

4.3 Irradiation Behavior of Plate-Type U-Mo/Al Dispersion Fuel

The irradiation tests, which were performed in accordance with the RERTR program, show the irradiation performance of plate-type U-Mo/Al dispersion fuels with U-10wt%Mo powders processed by both the atomization and grinding methods [19,20]. The fuel zone is elliptical with major and minor axes of approximately 51 mm and 9.5 mm, respectively; the thickness of the fuel zone is nominally 0.5 mm. The U-10wt%Mo micro-plates were independently irradiated with burn-ups of 40 at% and 70 at% at an average fuel central temperature of 65°C.

Scanning electron micrographs of fractured surfaces of U-10wt%Mo irradiated up to a 70 at% burn-up show that the bubble size and the population density increase greatly as the burn-up of the micro-plates increases from 40 at% to 70 at%. The maximum bubble diameters of both atomized and ground fuel particles are approximately 0.7 μm and 1.0 μm , respectively, and the average bubble diameters are approximately 0.2 μm and 0.3 μm , respectively.

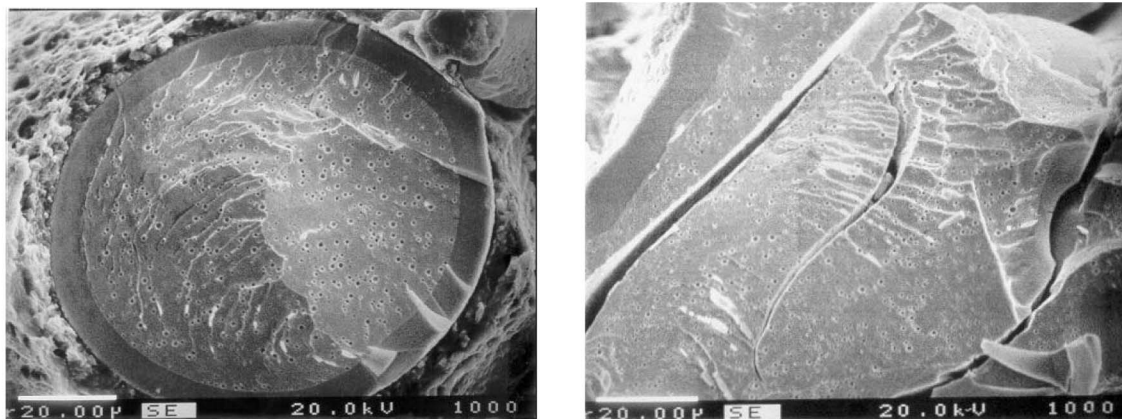


Fig. 9. Scanning Electron Micrographs of Fractured Surfaces in (a) the Atomized and (b) the Comminuted U_3Si Mini-Elements with a Burn-Up of about 69 at% (the First Irradiation Test)

The bubble population density for the atomized fuel particles is about $1.4 \times 10^{12} \text{ m}^{-2}$, while the ground particles have a prominent bimodal bubble size distribution and a population density of about $2.6 \times 10^{12} \text{ m}^{-2}$, which is twice as high as the population density of the atomized powder (Fig. 10).

4.4 Irradiation Behavior of Rod-Type U-Mo/Al Dispersion Fuel at HANARO

A qualification program of atomized U-Mo dispersion rod fuel for HANARO has been conducted since 1999 with some beneficial effects, such as fuel life extension, higher neutron flux, availability to use a few of the driving fuel sites for irradiation holes, and disposition flexibility of the spent fuel after expiration of the FRR SNF program [21-27]. In the first step, we targeted high U density fuel of 6.0 g-U/cc by using

atomized U-7wt%Mo and U-9wt%Mo powder [21,22]. A fuel assembly with 10 test fuel rods commenced irradiation, named as the KOMO-1 test, at HANARO on June 26, 2001, and was discharged on August 27, 2001, due to the failure of a fuel rod during irradiation test.

The linear power of the failed fuel region reached a maximum of 107.1 kW/m, which is much greater than the expected value of 80 kW/m. The calculated burn-up of the failed fuel rod was 12.9 at%. Figure 10 shows the cross sections of the failed fuel rod. The cladding was apparently cleaved along the weak bonded area during the co-extrusion process because the cleavage surface is very smooth. However, in the central region of the fuel meat, all aluminum matrix was consumed to form the U-aluminide phase as a result of the high temperature. However, the peripheral region shows that, the interaction between the fuel particles and the matrix, as

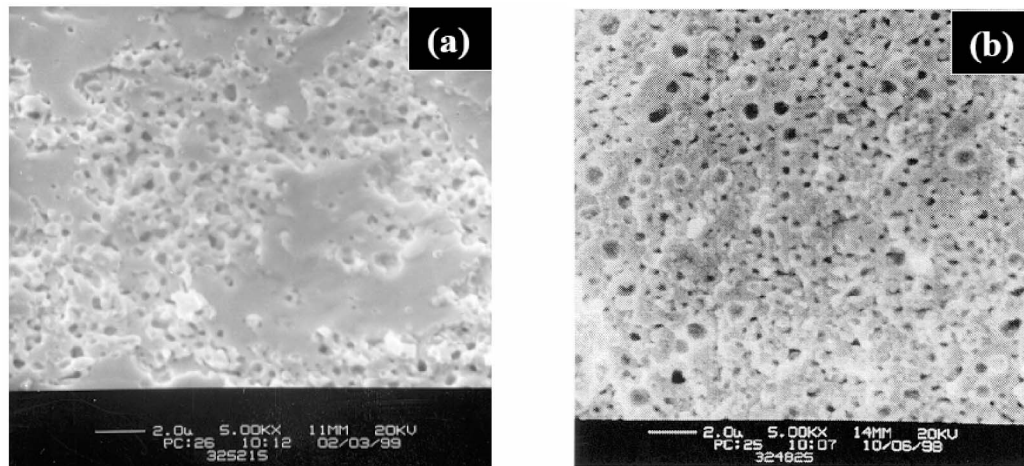


Fig. 10. Scanning Electron Micrographs for the U-10Mo/Al Micro-Plate at a Burn-Up of 70 at% for (a) Atomized Fuel Powder and (b) Ground Fuel Powder

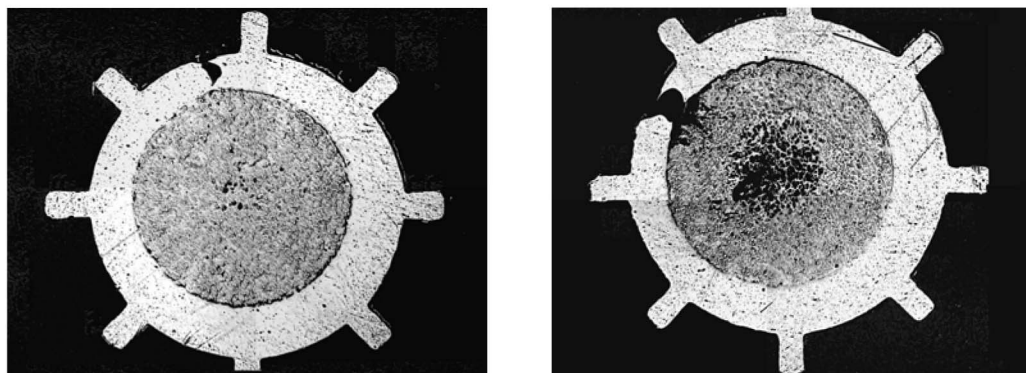


Fig. 11. Cross Sections of the Failed U-Mo/Al Fuel Rod of the KOMO-1 Test

well as the thickness of the interaction layer, increased from the periphery to the central region.

The second irradiation test (KOMO-2) was carried out to find a stable irradiation condition with a lower U density because such a condition can reduce the fuel temperature [23-26]. Atomized U-7%Mo/Al dispersion fuels with U-loadings of 4.5 g-U/cc and 4.0 g-U/cc were fabricated with the meat diameters of 5.49 mm and 6.35 mm, respectively. The fuel assembly for the irradiation test was loaded at HANARO on January 9, 2003, and discharged on March 7, 2003. The average and local maximum burn-ups were estimated to be 60.8 at% and 71.2 at%, respectively.

All U-Mo/Al fuel rods had swelling values greater than 10 vol% at burn-ups higher than 60 at% [24,25]. However, fuel rods with large U-Mo particles showed a small swelling value of about 8.5 vol%.

Figure 12 shows optical micrographs of a cross section of the 4 g-U/cc U-7Mo dispersion fuel rod with local burnups of 50%, 62% and 68% [24]. When the local burn-up is higher than 62%, almost all the U-Mo fuel particles reacted with aluminum matrix and several pore-

like defects were observed at the central area of the fuel rod. However, in the peripheral area of the fuel rod, no pore-like defects were evident even after a burn-up of 68%.

5. R&D ACTIVITIES FOR ATOMIZED U-MO DISPERSION FUEL

The KOMO-3 irradiation test is under way in order to improve the irradiation performance of atomized U-Mo fuel through the use of larger U-Mo particles [26-28]. U-Mo powder with larger particle size can be produced successfully by centrifugal atomization [28]. To obtain larger particles, we adjusted the atomizing parameters by adopting a smaller diameter and lower revolution speed of the rotating disk. Additionally, we used helium instead of argon as the cooling gas. As a result, we obtained U-Mo powders with various larger-sized distributions. As shown in Fig. 13, we chose a large U-Mo particle powder with a diameter range of 212 μm to 300 μm as a reference for the KOMO-3 test. To investigate the effect of particle size on irradiation behavior, the irradiation test

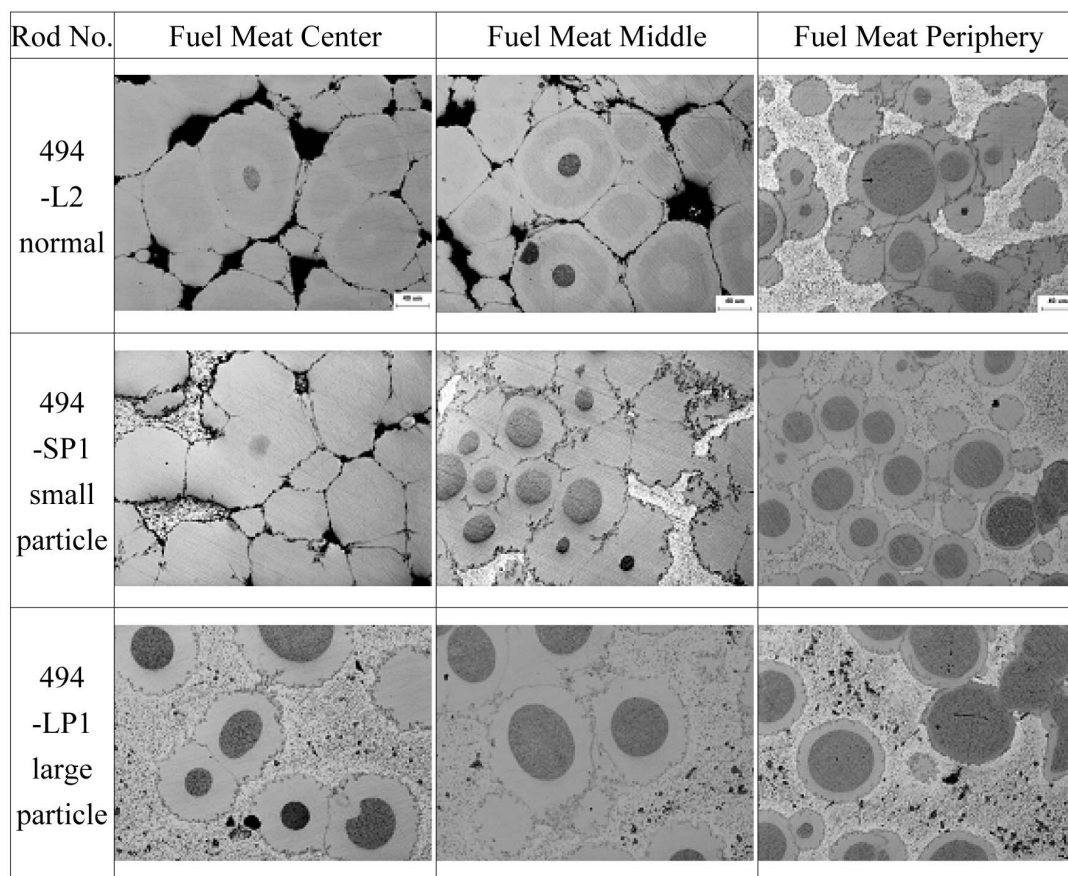


Fig. 12. Optical Micrographs of 4.0 g-U/cc U-7Mo Dispersion Fuel Rods with Respect to the Fuel Particle Size (62 at% BU)

fuels were fabricated by using the powder with smaller particles ranging from 100 μm to 212 μm in diameter as well as the powder with larger particles ranging from 300 μm to 425 μm in diameter.

When the fuel particles in fuel meat become larger, the interparticle space in the dispersion fuel meat increases and the volume fraction of the interacted product decreases due to a smaller specific interface area. The centerline temperature and the meat swelling decrease as the average diameter of the fuel particles increases because the volume fraction of a smaller conducting interaction layer increases more slowly. Another proposal features plate-type dispersion fuel with large uniform spherical particles of a U-Mo arranged regularly on an Al cladding in a single layer or in two layers. We can fabricate high-density plate-type dispersion fuel by hot pressing the arranged U-Mo particles and Al powder within a picture frame of Al cladding.

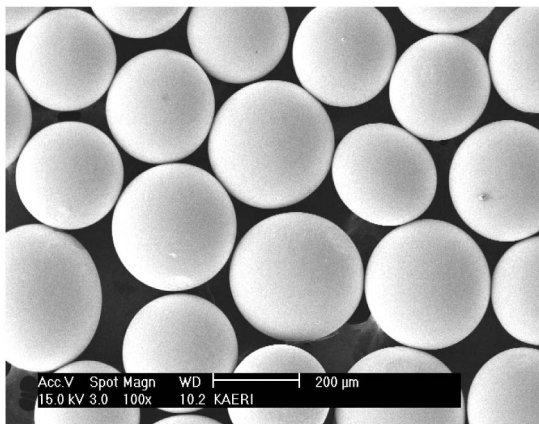


Fig. 13. Morphology of Large U-Mo Particles

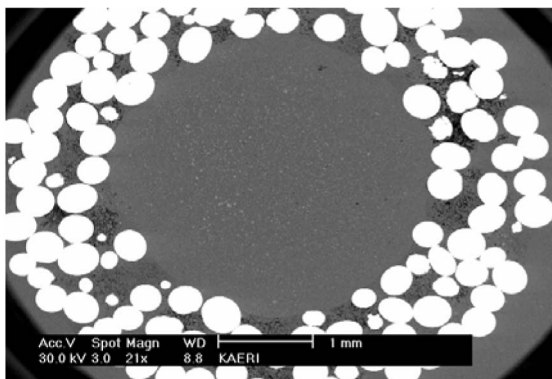


Fig. 14. A SEM Micrograph of Annular Fuel Meat Processes with Atomized U-Mo Powder

As an alternative for avoiding the interaction problem between U-Mo fuel particles and an Al matrix, we considered the annular-shaped fuel meat. In rod-type dispersion fuel, the swelling occurs mainly in the central region because of the longer heat diffusion distance. First, we prepared an aluminum tube with an annular space by fixing an Al rod at the bottom center. We then put relatively coarse U-Mo powder into the annular space and then vibro-packed fine aluminum powder into the interparticle space. To make the annular fuel meat more dense, we performed the swaging work for the fuel rod several times. Finally, we consolidated the work by sintering at 400°C for 10 h. When we conducted a test in this way, we produced a volume fraction of packed U-Mo and Al powders of about 75 vol% to 89 vol%, which represents a U-loadings of 9.3 g-U/cc to 10.2 g-U/cc. Furthermore, when the thickness of the annular region is half of the inside radius of the Al tube, we can achieve an apparent uranium density of 6.9 g-U/cc to 7.6 g-U/cc. As shown in Fig. 14, the swaged and sintered annular fuel meat shows fairly dense consolidation.

6.CONCLUSION

We developed spherical powders of U_3Si and U-Mo by means of a centrifugal atomization process in order to develop high-density dispersion fuel for advanced research reactors. Dispersion fuel with atomized fuel powder showed better thermal and mechanical properties. Compared with the comminuted fuel, the atomized fuel showed good irradiation stability such as a smaller fission gas bubble size distribution. The atomized U-Mo/Al fuel meats with large particles and annular-shaped fuel meat were fabricated to reduce the detrimental interaction between U-Mo and Al.

ACKNOWLEDGMENT

This work was supported by the National Nuclear R&D Program of the Ministry of Science and Technology in the Republic of Korea.

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