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In-Reactor Behaviour of Centrifugally Atomized U_3Si Dispersion Fuel Irradiated at High Temperature in HANARO

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ABSTRACT

The irradiation test on full-size U_3Si dispersion fuel elements, prepared by centrifugal atomization and conventional comminution method, has been performed up to about 77 at.% U-235 in maximum burn-up at CT hole position having the highest power condition in the HANARO reactor, in order to examine the irradiation performance of the atomized U_3Si for the driver fuels of HANARO. The in-reactor interaction of the atomized U_3Si dispersion fuel meats is generally assumed to be acceptable with the range of 5~15 μm in average thickness. The atomized spherical particles have more uniform and thinner reaction layer than the comminuted irregular particles. The U_3Si particles have relatively fine and uniform size distribution of fission gas bubbles, irrespective of the powdering method. The bubble population in the atomized particles appears to be finer and more homogeneous with the characteristics of narrower bubble size distribution than that of the comminuted fuel. The atomized U_3Si dispersion fuel elements exhibit sound swelling behaviours of 5 % in $\triangle V/V_m$ even at ~77 at.% U-235 burn-up, which meets with the safety criterion of the fuel rod, 20vol.% for HANARO. The atomized U_3Si dispersion fuel elements show smaller swelling than the comminuted fuel elements.

1. Introduction

U_3Si fuel dispersed in aluminium matrix was developed over the world from 1970s, and has been used as nuclear fuel for research reactors with verification of the in-reactor performance. The rod-type U_3Si fuel assembly has also been applied in HANARO as a driver fuel after confirmation of fuel safety in AECL [1]. KAERI has performed the localization development of the U_3Si driver fuel since 1987.

During the development process of the nuclear fuel for HANARO, new fuel fabrication technology was applied, preparing the spherical fuel powders directly from the alloy melt by centrifugal atomization method instead of conventional comminution method, was applied and eventually registered as an invention patent in USA, Canada, Germany and Korea. The centrifugal atomization technology, without the homogenization treatment and the mechanical comminution of an as-cast uranium alloy billet, has brought the advantages of the process simplification, the minimization of the fabrication space, the improvement of the production yield, the fuel productivity, the powder purity, the fuel formability and the thermal conductivity along the transverse direction, and the decrease of the as-fabricated porosity and the thermal swelling (Fig. 1) [2-4].

In this study, the U_3Si dispersion fuel elements with a fuel loading of 24vol.% were irradiated to characterize the in-reactor behaviours of atomized U_3Si dispersion fuels up to about 77at.% U-235 in maximum burn-up at CT hole position showing the highest power in the HANORO reactor. Thereafter, the in-reactor performance of the atomized U_3Si dispersion fuels was examined primarily using an optical microscope, a scanning electron microscope, an electron micro-probe analyser and an immersion densimeter, compared with that of the comminuted fuels.

2. Experimental procedure

The U_3Si fuel powders were prepared by rotating-disk centrifugal atomization and conventional mechanical comminution, using low enriched uranium lumps (99.9wt.% pure) and silicon chips (99.9999wt.% pure) [5]. The full-size fuel cores being ϕ6.35mm in diameter and L700mm in length were dispersed by the atomized or the comminuted U_3Si particles of 24vol.% in pure aluminum matrix having a uranium loading of 3.15 g-U/cm^3. The type and the dimension of the fuel assembly for the irradiation test were the same as that of the 36 rod type driver fuel assembly for HANARO reactor. The test rod was composed of an

extruded U_3Si-Al dispersion fuel meat, an aluminum cladding having eight cooling fins, and two aluminum end plugs. The fuel assembly for the irradiation test was fabricated with six test fuel elements (three atomized fuel elements and three comminuted fuel elements) and thirty dummy fuel elements.

The full-size elements with the atomized U_3Si fuel and the comminuted fuel, were loaded at CT hole position showing the highest power in the HANARO reactor and irradiated for 173.7 F-P-Day residence time at about 75 % operating capacity from June 1999 to July 2000. Table 1 summarizes typical irradiation conditions for fuel elements in the HANARO reactor. The maximum linear power was evaluated to be about 122 kW/m, in which the peak temperature of fuel elements was calculated to have been 320℃ during irradiation. The average burn-up, the maximum burn-up, and the fission density in the fuel meats after irradiation test were estimated to be about 63 at.% U-235, about 77 at.% U-235, and 9.2 x 10^{20} fissions/cm^3, respectively.

The full-size fuel elements irradiated in the HANARO reactor were subjected to post-irradiation after the irradiation following suitable periods, for about sixteen months, of cooling. The gamma scanning for the fuel elements has been carried out after visual examination. The gamma ray from the fuel element itself was transferred to the detector through a lead-filtered collimator, in which the dimension is 3 mm X 30 mm X 150 mm. The detected signal was analysed by using a multi-channel analyser. The specimens for density measurements and microscopy observations were prepared in locations of 87mm(: top part), 212mm (: middle part), 492mm (: bottom part) from the top should of the end plug, referring to total counts showing the relative axial burn-up distribution of the fuel element. Thereafter, the post-irradiation examinations of the fuel elements were performed, primarily using an optical microscope, a scanning electron microscope, an electron micro-probe analyser and an immersion densimeter.

3. Experimental Results and Discussion

Figs. 2 and 3 shows the typical optical micrographs of polished surfaces in the atomized and the comminuted U_3Si dispersion fuel elements at about 63 at.% burn-up. The fuel-matrix interaction layer of the atomized spherical particles is relatively uniform and generally acceptable in the range of 5~15 μm in average thickness. Irrespective of the powdering method, the U_3Si dispersion fuel elements have a considerable thickness of reaction layer between U_3Si fuel and aluminium matrix; however, the comminuted irregular particles have

less uniform and even thicker reaction layer relative to the atomized particles all over burn-ups (Fig. 4). The possible reason is supposed as follows. The atomized particles do not have a prominent deformation damage formed during the powdering process. Whereas, the comminuted particles have severe deformation damage with a high dislocation density formed during the powdering process. The local deformation zone formed during the comminution process increases the diffusion rate of aluminium atoms to the irregular particles, which leads to less uniform and thicker fuel/Al reaction layer relative to the atomized particles.

The typical scanning electron micrographs of fractured surface and the apparent fission gas bubble size distribution in the atomized and the comminuted U_3Si dispersion fuel elements at about 60 at.% burn-up are shown in Figs. 5 and 6. There are no fission gas bubble-free zones in the U_3Si particles, in contrast to the atomized U-Mo particles. There are apparent similarities in comparing the bubble formation as well as bubble distribution between the atomized and the comminuted fuel samples. The bubbles formed to be visible have covered entirely the fuel particles, nucleating and growing at the whole regions. The U_3Si particles have relatively fine and uniform bubble size distribution, and relatively thin and uniform fuel/Al layer thickness, irrespective of the powdering method. The bubble population in the atomized fuel appears to be finer and more homogeneous with the characteristics of narrower bubble size distribution than that in the comminuted fuel. The maximum and the average bubble diameter of the U_3Si fuel elements are approximately 1.4 μm and 0.36 μm. The U_3Si dispersion fuels show no indication of breakaway swelling, similar to that of U_3Si_2 and U-Mo [6-8]. There is no evidence of interlinking as the relatively uniformly and randomly distributed fission gas bubbles in places in particles. The atomized U_3Si dispersion fuels do not show any indication of breakaway swelling, but stable irradiation behaviour, similar to that of U_3Si_2 and U-Mo. The fuel elements have swollen by about 0.7 vol.% per 10 atomic percent burn-up to maximum burn-up of about 77 atomic percent, which meets with the safety criterion of the fuel rod, 20vol.% for the HANARO.

According to the previous studies, the stoichiometry of the interaction layer formed in uranium silicide dispersion fuel during irradiation, has been known to be $U(Si_3,Al)_3$ [8]. In this study, the more quantitative analysis for compositional changes was carried out across the fuel/aluminum interface by using Electron Prove Micro Analysis (EPMA). Fig. 7 shows the typical compositional changes near the fuel particle surface in U_3Si sample. In an unreacted fuel particle region, the result is fairly consistent with fuel composition of U_3Si. Moreover, it is also observed that the compositions uranium, silicon and aluminum in the inter-diffusion

layer changes gradually and continuously across the reacted layer, not being in unique composition such as $U(Si_3,Al)_3$, indicating that the formation of interfacial layer in U_3Si/Al during irradiation is a diffusion controlled reaction.

4. Conclusions

In order to localize the driver fuels for HANARO reactor, centrifugally atomized U_3Si dispersion fuel elements, have been fabricated and irradiated up to about 77 at.% U-235 in maximum burn-up having the highest power condition at CT hole position of HANARO reactor, compared with mechanically comminuted fuel elements. The fuel-matrix interaction layer of the atomized spherical particles is relatively uniform and generally acceptable in the range of 5~15 μm in average thickness; however, the comminuted irregular particles have less uniform and even thicker reaction layer relative to the atomized particles. The U_3Si particles have relatively fine and uniform bubble size distribution, irrespective of the powdering method. The bubble population in the atomized fuels appears to be finer and more homogeneous with the characteristics of narrower bubble size distribution than that of the comminuted fuels. The atomized U_3Si dispersion fuels do not show any indication of breakaway swelling, but stable irradiation behaviour with the irradiation swelling of 5 % in $\triangle V/V_m$ even at ~77at.% U-235 burn-up, similar to that of U_3Si_2 and U-Mo, which meets with the safety criterion of the fuel rod, 20vol.% for the HANARO.

Acknowledgements

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Fig.1. The fabrication process flow sheet of for HANARO fuel.

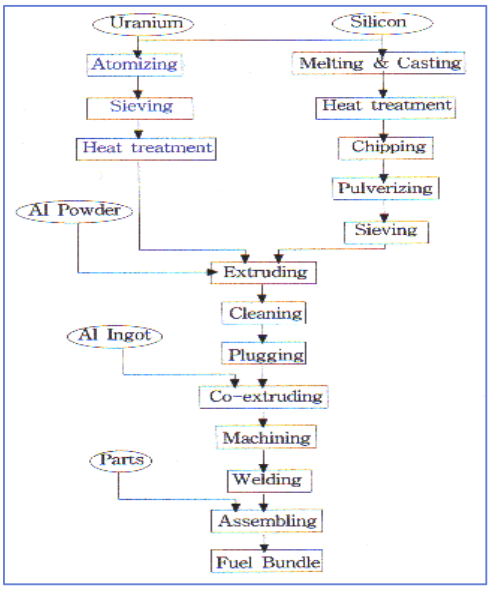

Table 1. Irradiation Conditions in the HANARO Reactor.

Irradiation time	174 Full Power Days at ~23 MW
Average (maximum) linear power	83 (122) KW/m
Centerline temp.	320°C (at 122 KW/m)
Fission density	9.2×10^{20} n/cm^2
Average (maximum) Burn-up	63 (77) at.% U-235

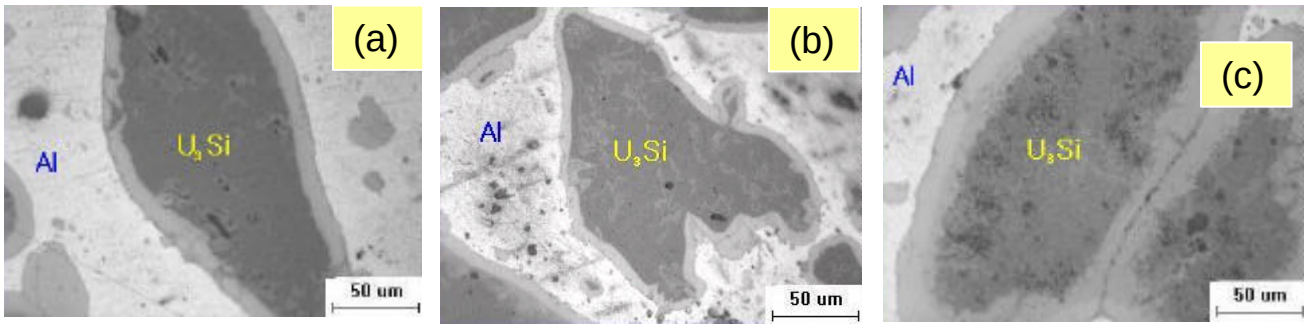

Fig. 2. The optical micrographs of polished surfaces in the comminuted U_3Si fuel element; (a) 39 at.% burn-up, (b) 60 at.% burn-up, (c) 76 at.% burn-up.

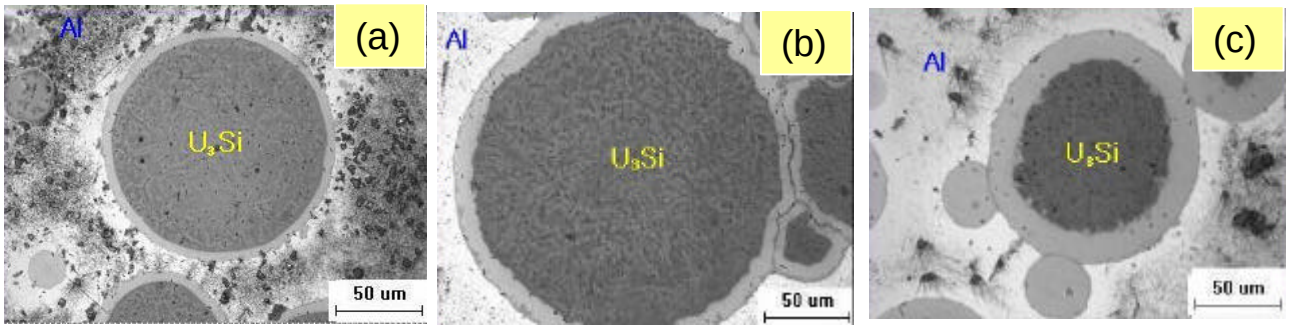

Fig. 3. The optical micrographs of polished surfaces in the atomized U_3Si fuel element; (a) 39 at.% burn-up, (b) 60 at.% burn-up, (c) 76 at.% burn-up.

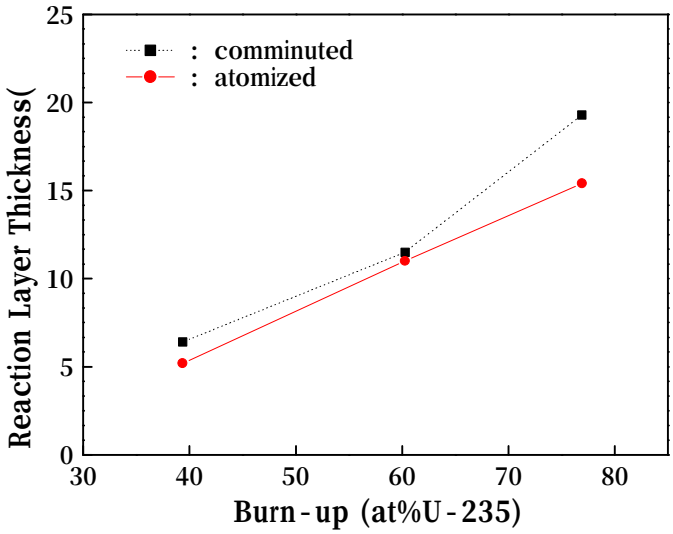

Fig. 4. The fuel/Al reaction layer thickness of the atomized and the comminuted U_3Si fuel element according to burn-up.

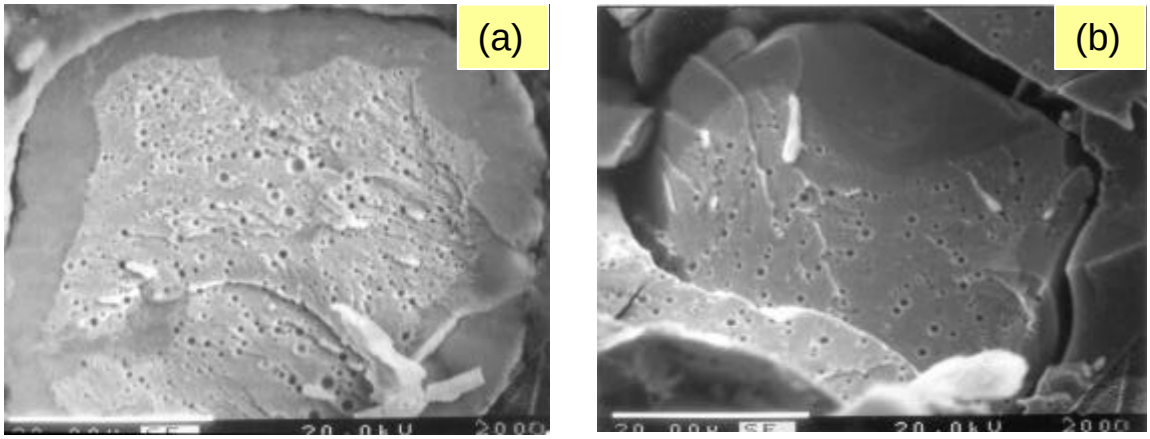

Fig. 5. The scanning electron micrographs of fractured surface in the atomized (a) and the comminuted (b) U_3Si fuel elements at 60 at.% burn-up.

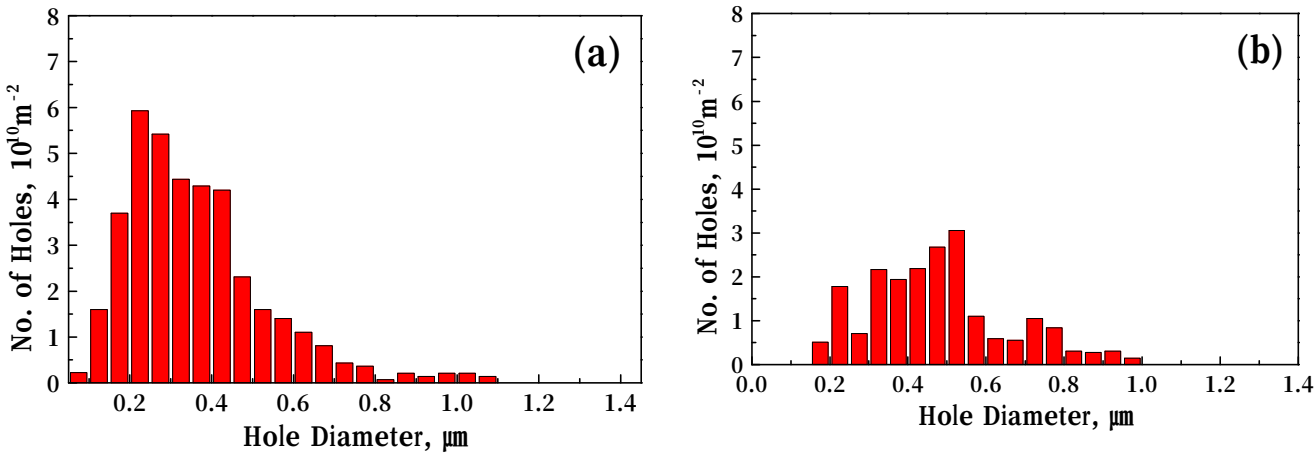

Fig. 6. The apparent fission gas bubble size distribution in the atomized (a) and the comminuted (b) U_3Si fuel elements at 60 at.% burn-up.

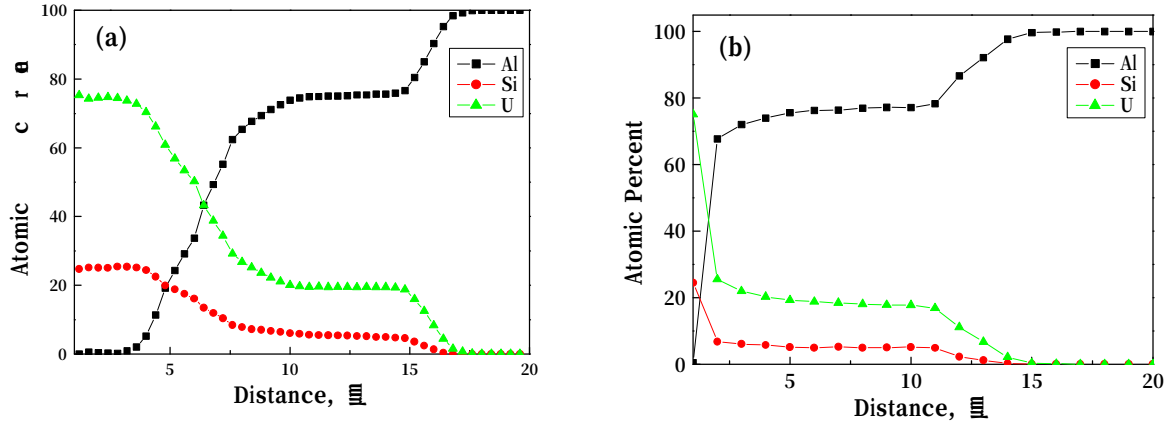

Fig. 7. The typical compositional changes at near the fuel particle surface in the atomized(a) and the comminuted (b) U_3Si fuel elements at 60 at.% burn-up.