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In-Reactor Behaviour of Atomized U-Mo Dispersion Rod Type Fuel Irradiated at High Temperature in HANARO

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ABSTRACT

In order to examine the in-reactor behaviour of atomized U-Mo dispersion rod type fuel, U-Mo fuel element has been irradiated to ~9at.% burn-up at high temperature at HANARO. There are few fission gas bubbles in the U-Mo particles even at high temperature, irrespective of the linear power of the dispersion fuel. The thickness of the fuel-matrix interaction layer in the atomized U-Mo dispersion fuel is very sensitive to linear power. The elevation effect between interaction layer and fuel temperature results in the formation of thick uranium-aluminide layer and the extensive cavity in the center, and leads to a significant contribution to large swelling. The cladding, in the region having a linear power of higher than about 80 KW/m, shows a long crack like an axial cleavage, initiated at the outer periphery of the cladding. The reasons of the cladding failure could be attributed to the weak bonding of aluminium plastic flows formed in the thinnest part during co-extrusion process from the viewpoint of breakage pattern and the swelling induced from severe interaction between Al and U-Mo particles.

1. Introduction

The conversion of research reactors from high enriched uranium (HEU) to low enriched uranium (LEU) fuel requires a large increase of uranium per unit volume to compensate for the reduction in enrichment. Dispersion fuel loading must be increased from the current NRC approved maximum of 4.8 g-U cm^{-3} to 8~9 g-U cm^{-3}. The relatively high-density compound U_3Si_2, with a uranium density of 11.6 g-U cm^{-3}, has been found to possess stable irradiation behaviour; however, fabricability limits do not allow fuel element loadings higher than 6 g-U cm^{-3} [1-5]. Uranium alloys with small amounts of alloying elements have the highest possible uranium density [6-7]. U-Mo alloys in particular are known to stabilize the cubic γ-U phase at modest alloy content [8]. Early irradiation experiments with uranium alloys showed the promise of acceptable irradiation behaviour, if these alloys could be maintained in their cubic γ-U crystal structure [9].

Commercial dispersion fuel fabrication is based on powder metallurgical techniques, and requires that the fuel phase be prepared in the powder form. The fuel phase for research and test reactors has, in the past, been brittle materials such as UAl_x and U_3Si_2 prepared by the comminution of ingots. Uranium alloys are ductile, and do not lend themselves to such processing methods. In order to simplify the preparation process and improve fuel performance, a rotating-disk centrifugal atomization method was applied to produce the powder used in this study [10]. This process, in which the powder is prepared by a centrifugal force, has the characteristics to suggest that the powder has a rapidly solidified microstructure, a relatively narrow particle size distribution, and a spherical shape [11-12].

U-Mo alloy with about 16.8 g-U/cc has been assessed to be acceptable for the above fuel from the stable in-reactor behaviour as shown in the ATR irradiation tests [13]. In connection with the end of the FRRSNF Acceptance Program of spent research reactor fuel in May 2006, an alternative available disposal was requested [14]. Uranium silicide dispersion fuel was investigated to be difficult for the reprocessing process, while U-Mo fuel is reprocessable [15]. In order to replace the current fuel with the reprocessable fuel, an accelerated program to qualify a U-Mo fuel was initiated by the RERTR program in 1999. In case of using high uranium density fuel of U-Mo dispersion in HANARO in Korea, it is assumed that some beneficial effects can be obtained such as versatility in spent fuel management, fuel life extension, higher neutron flux, and availability to use a few of the driving fuel sites for irradiation holes. In this paper, the in-reactor behaviour of atomized U-Mo dispersion rod type fuel irradiated at high temperature in HANARO has been examined.

2. Experimental procedure

The U-Mo fuel powder was prepared by rotating-disk centrifugal atomization, using low enriched uranium lumps (99.9wt.% pure) and molybdenum buttons (99.7wt.% pure) [16]. The reduced fuel core having ϕ5.49mm in diameter and L700mm in length was dispersed by the atomized U-Mo particles of 38vol.% in pure aluminium matrix having uranium loading of 6.0 g-U/cm^3. The porosity of fuel meat was measured to be less than about 4% in volume fraction. The type and the dimension of the fuel assembly for the irradiation test were the same as that of the 18 rod type fuel assembly for HANARO reactor. The test rod was composed of an extruded (U-Mo)-Al dispersion fuel meat, an aluminium cladding having eight cooling fins, and two aluminium end plugs.

The fuel element from the atomized U-Mo fuel, was loaded at OR-5 hole position in the HANARO reactor and irradiated for 26 F-P-Day residence time at about 70 % operating capacity from on June 26, 2001 to August 27, 2001. Table 1 summarizes typical irradiation conditions for a fuel element in the HANARO reactor. Fig. 1 shows the linear power distribution according to distance from the fuel meat top in the in the atomized U-Mo fuel element. The maximum linear power of the fuel rod was evaluated to be about 107 KW/m, in which the peak temperature of fuel element was calculated to have been 276℃ during irradiation. The peak burn-up, the average burn-up and the fission density in the fuel rod after irradiation test were estimated to be about 12.9 at.%, about 9.0 at.% U-235 and 1.47 x 10^{21} fissions/cm^3, respectively. Thereafter, in order to prepare the samples for optical observation from irradiated fuel elements, the fuel elements were cut down into three pieces according to linear power. The optical observation was focused at the sample positions from the center to the edge region in the fuel meat. In order to prepare SEM sample, a special punching tool was used to make the samples for SEM observation. Two small pieces of irradiated samples with having a diameter of 1.57 mm using punching jig in hot cell was taken from the center region of fuel meat. Then the fuel meat in the punched sample was cut down by hand in a glove box to observe the fractured surface of irradiated fuel particles. SEM observation on the polished fuel samples was also carried out to investigate the bubble size distribution and the fuel/Al reaction layer.

3. Experimental Results

Fig. 2 shows the optical micrographs of polished surfaces in the atomized U-Mo fuel meat, having the linear power of 47KW/m after ~9 at.% U-235 burn-up. Most particles show a round "kernel-like" unreacted island surrounded with reacted intermetallic compound. The

fuel-matrix interaction layer of the spherical particles is relatively uniform and generally thin in the range of 2~4 μm in thickness. The U-Mo fuel meat does not have a prominent reaction layer between U-Mo fuel and aluminium matrix, irrespective of the specimen part of the fuel meat. Figs. 3~4 show the optical micrographs of as-polished cross sections in the atomized U-Mo fuel specimen, having the linear power of 80KW/m (Fig. 3) and 107KW/m (Fig. 4). Most particles in the atomized U-Mo fuel specimen having the linear power of 80KW/m show a round "kernel-like" unreacted island surrounded with reacted intermetallic compound; however, a considerable amount of fuel particles in the atomized U-Mo fuel specimen, having the linear power of 107KW/m show several small unreacted islands surrounded with reacted intermetallic compound. The fuel-matrix interaction layer of the spherical particles is not uniform and generally thick in the range of 9~17 μm in thickness. The fuel-Al interaction thickness, caused by inter-diffusion of the matrix Al and the dispersed fuel alloy, significantly increases, as the specimen part of the fuel meat changes from the periphery region to the center region, in other words, as the fuel meat temperature increases. Most particles in the center region of the atomized U-Mo fuel specimen come in contact among fuel particles due to volume increase by extensive reaction between fuel and aluminium matrix. A considerable amount of small particles exhibit almost complete reaction. In addition, even some coarse fuel particles show almost complete reaction, in the center part of the atomized U-Mo fuel specimen having the linear power of 107KW/m. The center part of the atomized U-Mo fuel specimen, having the linear power of 107KW/m, shows extensive cavity region. It is assumed that the cavities resulted from the agglomeration of many voids, due to severe reaction between fuel and aluminium matrix at centerline temperature much higher than 276℃ during irradiation.

The scanning electron micrographs of the fractured surface in the atomized U-Mo fuel element at about 9 at.% burn-up are shown in Fig. 6. The crystalline grains of the irradiated U-Mo alloy even at ~9at.% burn-up has homogeneized grains of 15~25 μm in grain size, much larger than the cell size of the as-atomized U-Mo fuel of 2~3 μm. There are few fission gas bubbles in the U-Mo particles, irrespective of the linear power of the fuel specimens. The extent of the reaction product formation in the fuel meat having the linear power region of 47KW/m does not exceeds approximately half of the particle cross-section due to the lower center-line temperature, which leads to a volume change of less than 1%; the extent of the reaction product formation in the fuel meat having the linear power region of 107KW/m

exceed half of the particle cross-section due to the higher center-line temperature, which leads to a volume change of less than 10%.

Fig. 5 shows the macrographs of polished cross-sections in the atomized U-Mo fuel element after ~9 at.% U-235 burn-up. It is observed that the U-Mo fuel meat and the cladding do not exhibit a concentric circle, and the difference of the cladding thickness reaches about 40% in maximum. The cladding of the fuel rod, having a linear power of lower than 80 KW/m, shows a sound state without any damage; however, the cladding, in the region having a linear power of higher than 80 KW/m, shows an substantial crack like an axial cleavage with extensive cavities of larger than 1mm in diameter, initiated at the outer periphery of the cladding (Fig. 5). Some black spots and longish fabrication defects, shown in the region having the linear power of 80 KW/m, are also observed on the cladding surfaces of the fuel rod. It is assumed that the spots were formed from pitting Al cladding by cooling water and the longish defects from co-extrusion during fabrication process of the fuel rod.

The fuel element has swollen by about 1vol.% in the linear power region of 47KW/m, about 10vol.% in the linear power region of 80KW/m, and about 15 vol.% in the linear power region of 107KW/m to a final burn-up of about 9 atomic percent. It is thought that large swelling in the atomized U-Mo fuel specimen having a linear power of higher than 80KW/m results from severe interaction between U-Mo particles and Al matrix at center-line temperature higher than 276℃.

4. Discussion

It is supposed that the atomized U-Mo fuel maintains γ-U phase with homogeneized grain size of 15~25 μm. The atomized U-Mo fuel having γ-U phase shows a nucleation stage of fission gas bubble at grain boundaries due to low fission density irradiated in about 9 at.% burn-up. It results in few fission gas bubbles in the U-Mo particles irrespective of the linear power of the fuel specimens. The U-Mo dispersion fuel does not show large fission gas bubbles even at high temperature, similar to low temperature [16]. The fission gas bubble terms in the atomized U-Mo fuel do not make little contribution to the irradiation swelling, irrespective of irradiation temperature.

During irradiation of the atomized U-Mo fuel element, the interaction layer between fuel particle and aluminium matrix greatly degrades the thermal conductivity of the fuel meat. The corrosion layer of the cladding by cooling water deteriorates the thermal conductivity of the fuel cladding. The interaction layer and the corrosion layer also lead to increase overall fuel

temperature. The elevated temperature accelerates the fuel-matrix interaction and the cladding corrosion, as the fuel-matrix interaction rate and the corrosion rate are especially sensitive to the fuel temperature. Hence, the atomized U-Mo fuel specimen, having a linear power of higher than 80KW/m has a thick reaction layer in the range of 9~17 μm in thickness, greatly depending on the specimen part of the fuel meat, in other words, the fuel temperature. This results in volume increase due to the formation of uranium-aluminide having lower density. The cavity region in the center part of the atomized U-Mo fuel specimen, having the linear power of 107KW/m, also makes a significant contribution to the irradiation swelling. However, such swelling does not go beyond 15 vol.%, that is, a diametrical increase of about 5% in radial direction, even in the linear power region of 107KW/m.

It is not acceptable to explain the cladding failure only by such irradiation swelling, as it is supposed that the tensile strength and the elongation of the cladding is enough to sustain a diametrical increase of about 5%. The cladding of the rod type dispersion fuel is made by forming aluminium ingot at 500℃. When aluminium metal flows with covering from one side surface of fuel meat to other side surface, two metal flows contact each other at the other side and should be bonded each other. The contacted interface would have mechanically weak bonding with remaining a defect as misfits of atoms, as the cladding of the rod type dispersion fuel was made at a temperature of lower than normal co-extrusion temperature. In addition, the U-Mo fuel meat and the cladding do not exhibit a concentric circle, and the difference of the cladding thickness reaches even about 40% in maximum. It is thought that the thinnest part of the cladding would have the lowest strength of the cladding with the weak mechanical bonding of aluminium. As the interaction layer grows rapidly with irradiation time, the swelling of the fuel meat acts a tensile stress at the cladding. It is assumed that the cracking would be initiated at the outer periphery in the thinnest part of the cladding having the weakest bonding strength, with the swelling of the fuel element. Accordingly, the reasons of the cladding failure could be attributed to the weak bonding of aluminium plastic flows in the thinnest part from the viewpoint of breakage pattern and the swelling induced from severe interaction between Al and U-Mo particles.

5. Conclusions

1) There are few fission gas bubbles in the U-Mo particles even at high temperature at low burn-up, irrespective of the linear power of the fuel element.

2) The thickness of the fuel-matrix interaction layer in the atomized U-Mo dispersion fuel is very sensitive to linear power. The atomized U-Mo dispersion rod type fuel with a diameter of 5.49mm and an uranium loading of 6 g-U/cm^3 has a prominent reaction layer in case of having a linear power of higher than about 80KW/m, especially with extensive cavity region in the center part of the atomized U-Mo dispersion fuel with the linear power of about 107KW/m.

3) During irradiation, the interaction layer and the corrosion layer with low thermal conductivity lead to increase overall fuel temperature. The elevated temperature accelerates the fuel-matrix interaction and the cladding corrosion. This elevation effect results in the formation of thick uranium-aluminide layer and the extensive cavity, and leads to a significant contribution to large swelling of the fuel element.

4) The cladding, in the region having a linear power of higher than about 80 KW/m, shows a long crack like an axial cleavage, initiated at the outer periphery of the cladding. It is not acceptable to explain such cladding failure only with a swelling of less than 10 vol.% in the linear power region of about 80KW/m. The reasons of the cladding failure could be attributed to the weak bonding of aluminium plastic flows formed in the thinnest part during co-extrusion process from the viewpoint of breakage pattern and the swelling induced from severe interaction between Al and U-Mo particles.

Acknowledgements

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Table 1. Irradiation conditions in the HANARO reactor.

Thermal neutron flux Fuel element No. 15 average	1.24×10^{14} n/cm^2
Average coolant temperature pH	37.0℃ 6.0
Irradiation time	26.6 Full Power Days at 24 MW
Average linear power Fuel element No. 15 average	75.1 KW/m
Maximum linear power For fuel element No. 15	107.1 KW/m

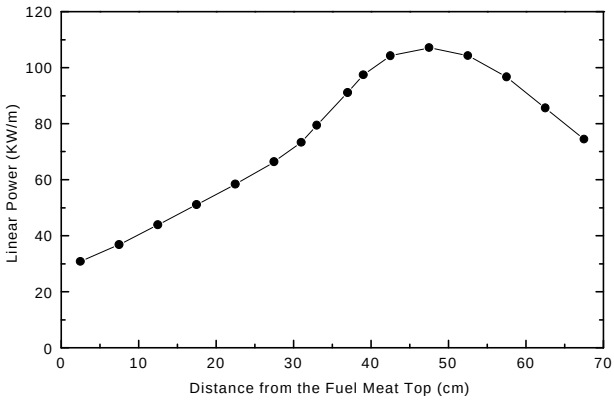

Fig. 1. Linear power distribution according to distance from the fuel meat top.

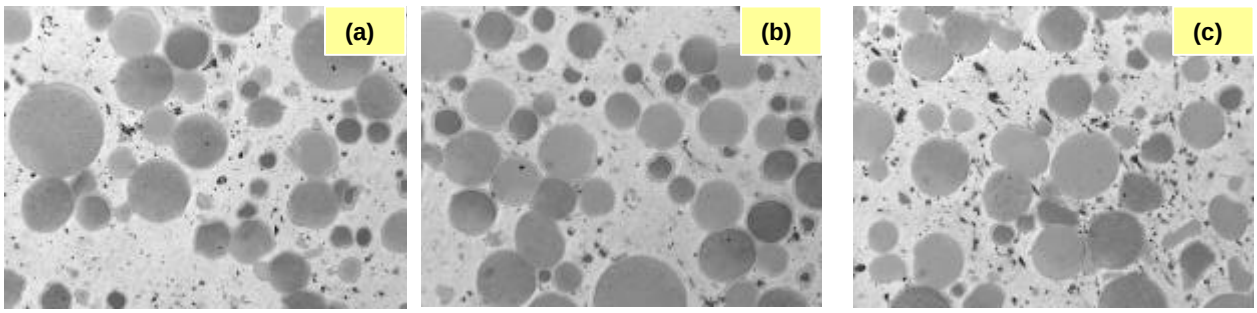

Fig. 2. The optical micrographs of polished surfaces in the atomized U-Mo fuel meat with the linear power of 47KW/m (x200); (a) periphery region, (b) middle region, (c) center region.

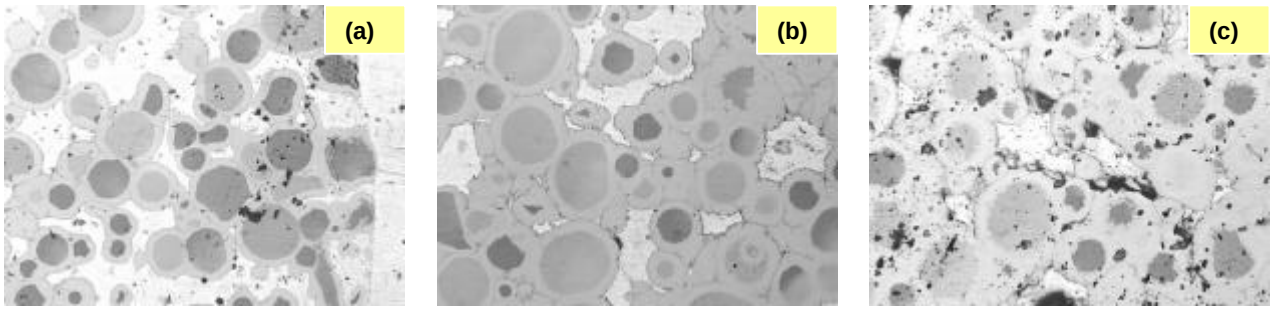

Fig. 3. The optical micrographs of polished surfaces in the atomized U-Mo fuel meat with the linear power of 80KW/m(x200); (a) periphery region, (b) middle region, (c) center region.

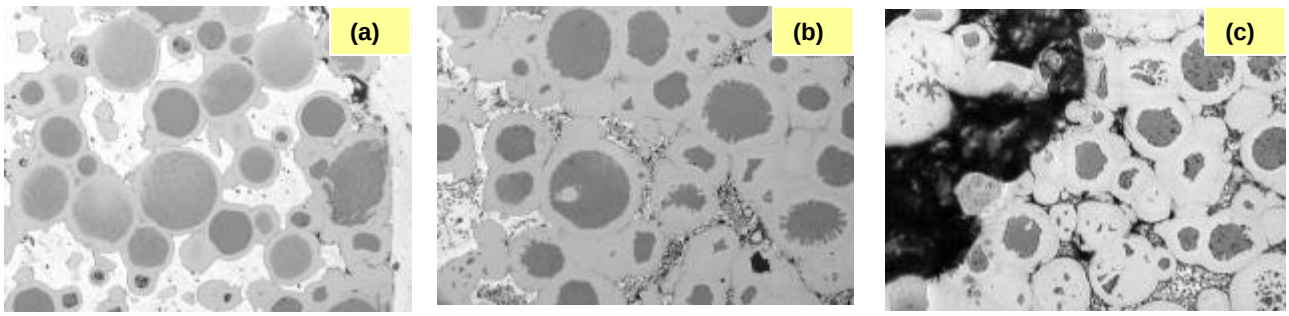

Fig. 4. The optical micrographs of polished surfaces in the atomized U-Mo fuel meat with the linear power of 107KW/m(x200); (a) periphery region, (b) middle region, (c) center region.

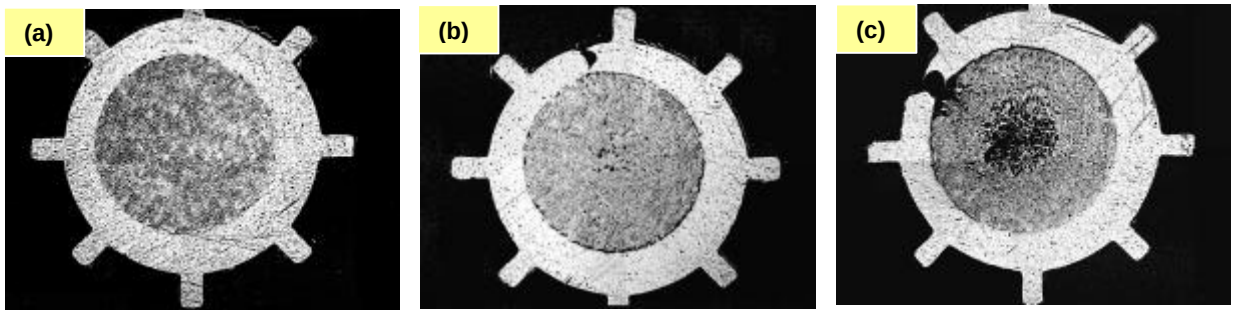

Fig. 5. Macrographs of U-Mo fuel element cross-sections after irradiation; (a) 47 KW/m, (b) 80 KW/m, (c) 107 KW/m.

(a) (b)

Fig. 6. Photograph showing the longish cleavage-like defect on cladding in the region fuel rod having the linear power of 80 KW/m(a) and 107 KW/m (b).

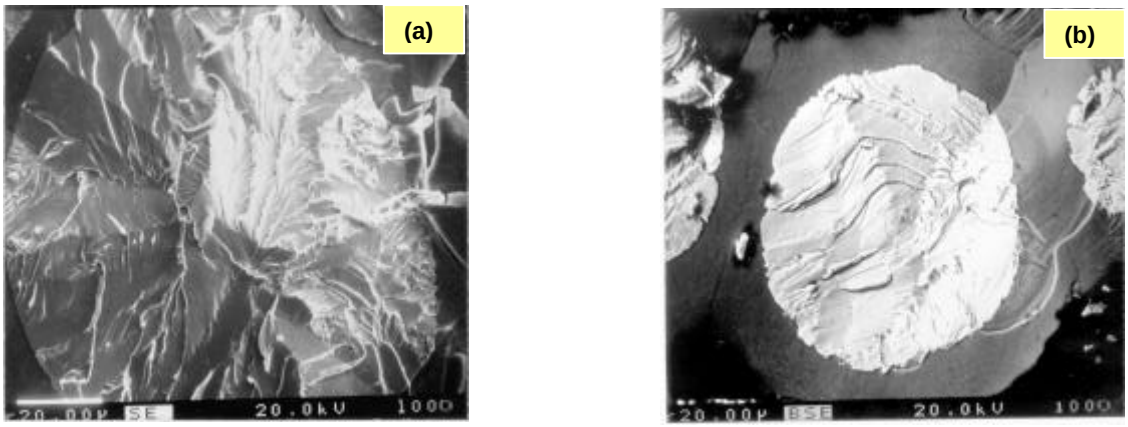

Fig. 7. The scanning electron micrographs of fractured surface in the atomized U-Mo fuel meat; (a) 80KW/m, (b) 107KW/m.