PROGRESS OF U-Mo DISPERSION ROD FUEL QUALIFICATION PROGRAM IN KOREA

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ABSTRACT

In the irradiation test performed in HANARO since June in 2001 relating to a qualification program of atomized U-Mo dispersion rod fuel for HANARO, a failure happened from the cleavage of cladding under the influence of the fuel meat swelling. An examination on the cross sections showed that the Al matrix of fuel meat at high power region had almost interacted with fuel particles. The depletion of the Al matrix deteriorated the thermal conductivity of fuel meat and resulted in the very high temperature around the center region of fuel meat. Swelling calculations using the Al matrix fractions of fuel meats showed a relatively good agreement with the measured swellings by immersion densities. The swelling in the failure region of about 15% does not seem to be enough to affect the rupture of the pure aluminum cladding. A failure reason could be attributed to the fabrication defects of the cladding. The next irradiation test for U-Mo dispersion rod fuels was planned and the fuel fabrication is under way. The loading density was changed from 6.0 g-U/cc to 4.5 g-U/cc and the smaller fuel meat diameter of 5.49 mm was chosen in the viewpoint of the centerline temperature of fuel meat to be lower than 200 °C. The next irradiation will include the various tests for the following rods; 1) a fuel rod containing U-Mo hollow cylinder, 2) two different particle size fuel rods to investigate the particle size effect, 3) a fuel rod coated with Ni to prevent corrosion, 3) a fuel rod added with poison material of Er_2O_3 relating to fuel life extension, 4) a fuel rod dispersed with U-9wt.Mo particles, 5) A U_3Si dispersion fuel rod for a reference. It is scheduled that the loading in HAHARO will be done around the end of November.

1. INTRODUCTION

The irradiation tests performed by the RERTR program showed that U-Mo dispersion fuel with high uranium density of 8.0 g-U/cc was acceptable for high performance research reactors[1,2]. In case of using high uranium density fuel with U-Mo dispersion in HANARO in Korea, it is assumed that some beneficial effects could be obtained such as fuel life extension, higher neutron flux, and availability to use a few of the driving fuel sites for irradiation holes. Relating to the spent fuel disposal after expiration of the FRR SNF program as well as the beneficial effects of using high uranium density fuel, a qualification program of atomized U-Mo dispersion rod fuel for HANARO has been carried out since 1999[3]. A fuel assembly consisting of fuel rods with 3.15 g-U/cc and 6.0 g-U/cc had been fabricated successfully using atomized U-7wt%Mo and U-9wt%Mo powder. The fuel assembly containing 10 fuel rods had been irradiated in HANARO since June 26, 2001 and was discharged on August 27, 2001 due to a failure occurrence of a fuel rod[4].

In order to find out the failure cause the irradiated fuel rods have been examined from the beginning of January 2002. Swellings of fuel meats were obtained by dimension measurement and immersion density measurement. A calculation on swelling of fuel meat was done using the remaining fraction of Al matrix on the cross section in fuel meat. Temperatures of fuel meat under irradiation were calculated using estimated thermal conductivities from the volume fraction of interacted (U-Mo)Al_x phase on fuel meat and the correlation with thermal conductivity. In planning the next irradiation efforts have been made to prevent any failure on fuels.

In this paper, in connection with the qualification program for U-Mo rod type fuel in KAERI some results obtained from the PIE on the failed irradiation fuels, the next irradiation plan, and the preparation for the irradiation fuels are reported.

2. IRRADIATION TEST OF U-Mo DISPERSION FUEL RODS AND THE PIE

4 different types of fuel rods were selected for the irradiation test as shown in Table 1. Two different Mo contents of 7 wt.% and 9 wt.% and two different uranium loadings of 3.4 and 6 g-U/cc were applied. 3.4 g-U/cc of U-Mo dispersion fuel is equivalent to the uranium density of current HANARO fuel of U_3Si dispersion, the density of which is 3.15 g-U/cc. In order to reduce the center temperature of fuel meat form a safety point of view the smaller diameter fuel of 5.49 mm than that of standard diameter of 6.35 mm was designed.

Type	Mo contents (wt.%)	Targeted uranium density (g-U/cc)	Fuel meat diameter (mm)	% to theoretical density
1	7.0	3.4	6.35	99.4
				99.1
				99.8
2	7.0	6.0	5.49	98.3
				97.6
				96.9
3	9.0	3.4	6.35	99.9
4	9.0	6.0	5.49	97.7
				95.9
				95.5

Table 1. Fuel rods loaded in OR hole for irradiation

The fuel assembly containing 10 U-Mo dispersion rods were loaded in the OR-5 hole in HANARO on June 26, 2001 and irradiated under the operating power of 20 MW. The maximum linear power was estimated to be about 80 kW/m. The U-Mo fuel assembly was discharged from the HANARO core to the cooling pool on August 27, 2001 due to the high cooling water radioactivity representing a leakage of fuel. It was calculated that the U-Mo fuel had been irradiated for 26.6 full power days.

On the cladding of the fuel rod with high uranium loading of 6.0g-U/cc under the highest power an axial cleavage occurred from 300 mm to 600 mm at the 700 mm fuel rod. A calculation on linear power of the failed fuel region showed that the maximum power would be 107.1 kW/m, which was much higher than the expected value of 80 kW/m. This phenomenon happened due to the influence of the loaded fresh fuels in the core fuel sites situated near the irradiating fuel. The peak burnup of the failed fuel rod was calculated as 12.9 at.%.

Micrographs taken on the cross sections of the failed fuel rod are shown in figure 1. It seems that cladding was cleaved along the weak boned area during the co-extrusion process because the cleavage surface is very smooth. If the cleavage happened in sound pure aluminum cladding, the ductile rupture pattern would be shown. Al matrix in the central region of severely cleaved fuel position was interacted with fuel particles completely. Some dark area looks like the formation of void. On the periphery region of fuel meat much of Al matrix remained due to the low temperature. The fuel meat cross section of middle position initiating a cleavage appeared that the interaction of Al matrix was almost completed except the periphery region. However, the cross section of the top position of fuel rod revealed a little interaction due to the low temperature from low power rate. The observation by SEM on a cut surface of a fuel particle was obtained as shown in figure 2. It seems that a few fission gas bubbles appeared along the grain boundaries. The bubble size is very small with submicron. This morphology is similar with the results of RERTR tests[5].

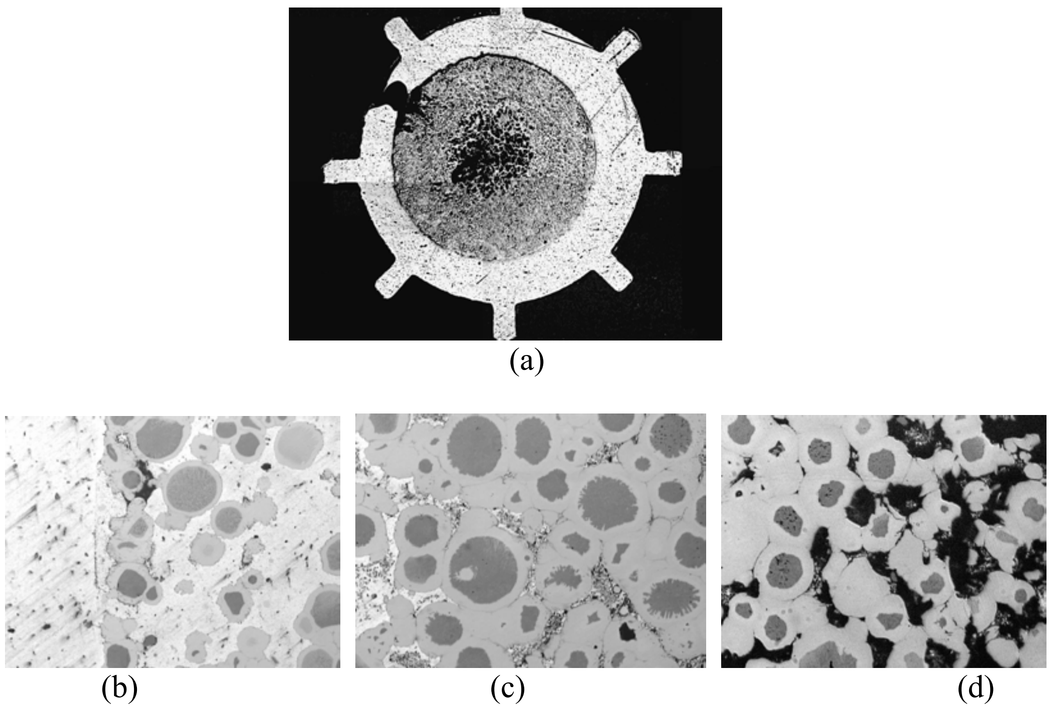

Fig. 1. Cross sections of fuel meat at the failure position; (a) whole cross section, (b) periphery, (c) middle, (d) center.

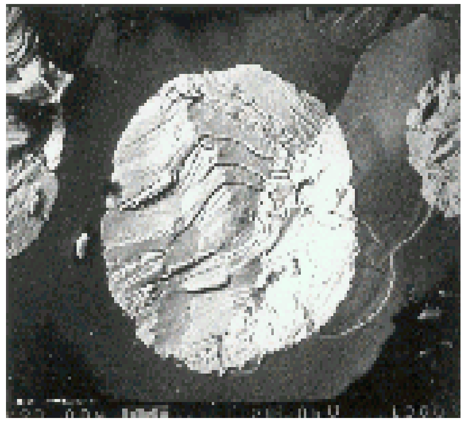

Fig. 2. The morphology of the U-Mo particle in the bottom part of the irradiated fuel rod.

3. CALCULATION FOR INTERACTION SWELLING OF FUEL MEAT

It is considered that swelling of fuel meat occurs by interaction of Al matrix with fuel particles, bubble formation of gas elements of the fission product and lattice swelling of solid elements. Presumably the volume expansion of solid elements can be estimated from the burnup of fuel meat. The maximum burnup of irradiated U-Mo rod fuel was calculated as 13.6 at.%. The above burnup is corresponded to the increasing fraction of atom number in fuel meat of about 4.0%. Presumably the bubble swelling is negligible because of the small size and a little population of bubbles. The interaction swelling occurs mainly from the formation of the interacted product having low density. In order to calculate the interaction swelling a reaction relation is supposed as below.

$$\text{U-Mo} + \text{Al} \rightarrow \text{U-Mo} + \text{U(Al, Mo)}_4 + \text{Al} \qquad (1)$$

From the viewpoint of the conservation the product was considered as $U(Al, Mo)_4$ instead of (U-Mo) $Al_{4.4}$. It is supposed that the densities of U-Mo and $U(Al,Mo)_4$ are 17.5 g/cc and 5.7 g/cc, respectively. Equations regarding the mass balance of U and Al can be written as below.

$$V^1_{(U\text{-}Mo)} \times \delta_{U\text{-}Mo} \times W_{U(U\text{-}Mo)} = V^2_{(U\text{-}Mo)} \times \delta_{U\text{-}Mo} \times W_{U(UMo)} + V^2_{(U\text{-}Mo\text{-}Al)} \times \delta_{U\text{-}Mo\text{-}Al} \times W_{U(U\text{-}Mo\text{-}Al)} \qquad (2)$$

$$V^1_{(Al)} \times \delta_{Al} = V^2_{(U\text{-}Mo\text{-}Al)} \times \delta_{U\text{-}Mo\text{-}Al} \times W_{Al(U\text{-}Mo\text{-}Al)} + V^2_{(Al)} \times \delta_{Al} \qquad (3)$$

Using the above equations interaction swelling can be calculated if the remaining Al matrix fraction is measured. Accordingly area fractions of phases on the micrographs of the fuel cross section were measured using image analysis and shown in Table 2. Then calculations for swellings were performed. For the cross section containing the void the total swelling was calibrated by adding void volume to the interacted volume. The calculated swellings are shown in Table 3. Separately swellings of fuel meats were measured by the immersion weighing method and shown in Table 4. Fuel meat samples for immersion weighing were taken by removing cladding from fuel rod by machining. So a little of the periphery part of the fuel meat was cut away in machining. Presumably the measured density of fuel meat seems skewed to a little less density than the true density. Generally it is assumed that the calculated densities agree with the measured densities by the immersion method.

Table 2. Area fractions of phases on micrographs of cross sections of fuel rod

Position	Phase	Center	Inner middle	Outer middle	Periphery
Top (Low power)	U-Mo	47.9%	42.9%	42.9%	40.6%
	(U-Mo)Alx	Negligible	Negligible	Negligible	Negligible
	Al matrix	52.1%	57.1%	57.1%	59.4%
	Void	None	None	None	None
Middle (Medium power, cleavage initiation)	U-Mo	23.0%	21.6%	28.2%	34.8%
	(U-Mo)Alx	72.9%	69.4%	59.7%	23.8%
	Al matrix	3.7%	8.9%	12.1%	41.4%
	Void	0.38%	0.12%	None	None
Bottom (High power, severe cleavage)	U-Mo	25.2%	31.6%	24.1%	27.2%
	(U-Mo)Alx	50.1%	55.4%	66.2%	29.4%
	Al matrix	None	4.9%	9.4%	40.7%
	Void	24.7%	8.1%	0.29%	2.7%

Table 3. Calculated swellings for irradiated U-Mo fuel rod.

	Position	Center	Inner middle	Outer middle	Periphery	Average
Excluding Void	Top	2.3%	1.2%	1.2%	0.7%	1.1%
	Middle	13.5%	12.2%	11.4%	4.5%	8.7%
	Bottom	14.5%	13.1%	12.1%	4.5%	9.1%
Including Void	Top	2.3%	1.2%	1.2%	0.7%	1.1%
	Middle	14.0%	12.3%	11.4%	4.5%	8.7%
	Bottom	52.0%	23.1%	12.4%	7.4%	14.7%

Table 4. Swellings of fuel meats measured by immersion method.

Position		Top	Middle	Bottom
Density (g/cc)	As-fabricated	7.92	7.92	7.92
	After irradiation	7.84	7.16	6.84
Swelling		1.0 %	10.6%	15.8%

In order to know the temperature of fuel, meat thermal conductivities for 4 divided parts were estimated from the correlation between the volume fraction of the interaction phase and thermal conductivity. The correlation had been obtained from thermal conductivities measured for compatibility test specimens. The corrosion layer on the fuel cladding was measured about 30 μm. Temperature at the failure position of fuel meat could be calculated as shown in figure 3. The centerline temperature seems to be higher than 700 °C. It is considered that the void at the center region would form from gas evolution at the very high temperature.

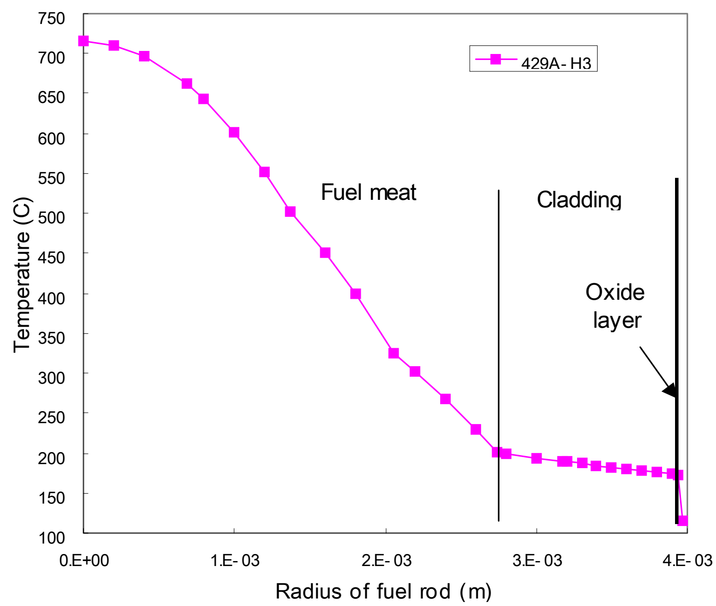

Fig. 3. Profile of calculated temperatures

The swelling value of fuel meat such as 15 % is not large enough to affect cladding failure because of pure aluminum with ductile property. In order to examine the cladding ductility some enlarging tests for fuel claddings were performed by inserting a wedge. Elongations of cladding appeared with large variations of 2.5% to 45.8% in various fuels. An observation on the cladding with very low elongation of 2.5% showed that some defects exist in cladding as shown in figure 4. It seems that the defects are formed from the inclusion of lubricant used in the co-extrusion and incomplete bonding of plastic deforming aluminum in the co-extrusion process. The reason for cladding rupture under irradiation could be attributed to the defect in cladding formed in the fabrication process.

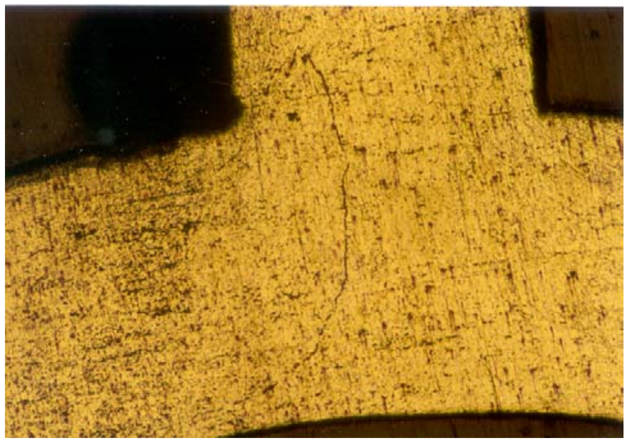

Fig. 4. Photograph showing defect existence in fuel cladding.

4. NEXT IRRADIATION TEST

In planning the next irradiation test for U-Mo dispersion rod fuels every effort was made to avoid the swelling problem induced from interaction. Loading density and diameter of fuel meat were decided with the viewpoint of the centerline temperature of fuel meat to be lower than 200 °C. The loading density was changed from 6.0 g-U/cc to 4.5 g-U/cc and the smaller fuel meat diameter of 5.49 mm was chosen. The interaction rate is dependent on the specific surface area of fuel powder. Two fuel rods with different particle size will be loaded to investigate the particle size effect on the interaction swelling.

A consideration has been given for solving the interaction problem in eliminating U-Mo/Al interface of dispersion fuel meat using monolithic U-Mo hollow cylinders with the thin thickness of less than 1 mm. The hollow cylinder fuel has only one interface between the outer surface of U-Mo hollow cylinder and cladding. The interface temperature is governed by the thickness of cladding and the total linear power. The interface temperature is considered not to rise severely. A gap is put between the inner surface of the U-Mo hollow cylinder and the aluminum rod inserted at the center of the hollow. A fuel rod containing one hollow cylinder of U-7wt%Mo is under fabrication and will be loaded for irradiation.

In the previous irradiation test the fuel cladding surface was corroded by cooling water and a corrosion layer known as boehmite formed with the thickness of about 30 micrometers[6]. The temperature increase at the corrosion layer was calculated as about 60 °C. It is considered that thin Ni coating on the cladding surface with negligible thermal conductivity degradation will improve the corrosion resistance of fuel cladding. A corrosion test for a fuel rod coated thinly with Ni (less than 10 micrometers) and a fuel rod without any coating was performed at the elevated temperature of 350°C under high pressure water at two different waters of pH 5.5 and pH 6.5. The cladding without coating was corroded up to the layer of more than 70 micrometers in both pH conditions. However, the cladding coated with Ni showed no corrosion. A fuel rod coated with Ni is under preparation for irradiation test.

The replacing of HANARO fuel with higher uranium loading density would allow for extended fuel life. In this case, an addition of poison material to fuel meat is required. A fuel rod added with poison material of Er_2O_3 is loaded. In the preliminary fabrication and an investigation on the physical properties of poison added fuel meats, any differences that would affect the irradiation test were not shown.

Generally as Mo content in U-Mo particles increases, the irradiation performance has been shown to improve. A fuel rod dispersed with U-9wt.Mo particles has been fabricated and will be irradiated. A U_3Si dispersion fuel rod will be loaded in the irradiation fuel assembly as a reference. A comparison of U-Mo fuel with U_3Si fuel will be conducted after the 2nd irradiation test. Even though fuel meat swelling is large, the cladding with good ductility could accommodate the swelling. In order to improve the cladding ductility co-extrusion was done at the higher temperature under careful conditions to avoid any unwanted inclusions. This irradiation test assembly consisting of 9 fuel rods as shown in Table 5 is scheduled to be loaded in HAHARO through a safety evaluation by a committee organized by HANARO around the end of November.

Table 5. Fuel rods to be loaded for the next irradiation test

Serial No.	Fuel Material	Loading Density (g-U/cc)	Diameter (mm)	Length (mm)	Number of fuel rod	Remarks
1	U-7Mo	4.5	5.49	360	1	High density
2	U-9Mo	4.5	5.49	360	1	High Mo
3	U-7Mo	4.0	6.35	360	1	Low density
4	U-7Mo	4.0	6.35	110	1	Larger particles
5	U-7Mo	4.0	6.35	110	1	Smaller Particles
6	U3Si	4.0	6.35	210	1	Reference
7	U-7Mo	4.5	5.49	210	1	Poison
8	U-7Mo	Pure	$^{OD}6.35\times^{t}0.7$	5	1	Hollow cylinder
9	U-7Mo	4.0	6.35	360	1	Ni coating

5. SUMMARY

The failure of the 1st U-Mo dispersion rod fuel irradiation was observed to have occurred through the cleavage of cladding. The very smooth cleavage surface implies that the defects in cladding would act as a dominant role in failure. The swelling calculations performed using the measured densities by the immersion method and the Al matrix fraction on micrographs of fuel meat cross section revealed a fairly good agreement. The maximum swelling is assumed about 15 Vol. %, which is not large for affecting the cladding failure because of pure aluminum with ductile property. The estimated centerline temperature seems to be higher than 700 °C. It is considered that the void at the center region would form from gas evolution at the very high temperature.

In planning the next irradiation test for U-Mo dispersion rod fuels, loading density was changed from 6.0 g-U/cc to 4.5 g-U/cc and the smaller fuel meat diameter of 5.49 mm was chosen with the viewpoint of the centerline temperature of fuel meat to be lower than 200 °C. The next irradiation will include the various fuel rods; 1) a fuel rod containing U-Mo hollow cylinder, 2) two different particle size fuel rods to investigate the particle size effect, 3) a fuel rod coated with Ni to prevent corrosion, 4) a fuel rod added with poison material of Er_2O_3 relating to fuel life extension, 5) a fuel rod dispersed with U-9wt.Mo particles, 6) A U_3Si dispersion fuel rod for a reference.

Acknowledgement

This work has been performed under the nuclear R&D support by the Ministry of Science and Technology, Republic of Korea.

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