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Comparison of Irradiation Behavior Between Atomized and Comminuted U_3Si_2/Al Mini-Plate Fuels

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Abstract

The irradiation test on both the atomized and comminuted U_3Si_2 dispersion mini-plate fuels, with connection to international research co-operation under the RERTR program, has been performed in the HANARO reactor in order to understand fuel irradiation performance of two different types of fuel powder. The mini-plates, irradiated until 70 at%U_{235} burn-up at OR6 hole in HANARO were subjected to IMEF for post-irradiation analysis. Visual inspection, gamma scan, dimension measurement, microstructure analysis including EPMA have been conducted. From the PIE results, it is of note that both the atomized and comminuted U_3Si_2 dispersion mini-plate fuels irradiated at HANARO exhibit sound swelling behaviors. The thickness of the aluminide layer formed in all samples ranges from 1.73 ㎛ to 2.13 ㎛, which implies fuel meat swellings of ~3.3 $\triangle V/V_m$. However a bubble distribution with slightly smaller mean diameters was found in the atomized fuel than in the comminuted sample, and the bubble population in atomized fuel appears to be more homogeneous than that in the comminuted fuel.

Introduction

The development of low enriched uranium nuclear fuel has been the center attraction for research reactors in the world to retard the nuclear proliferation [1]. Until now, high enriched uranium (HEU) has been converted into low enriched uranium (LEU) with a uranium density of 4.8 gU/cm^3 using uranium silicide alloys (U_3Si, U_3Si_2) in about 90% of research reactors [1-2]. Therefore the U_3Si-Al dispersion fuel is now being used at HANARO reactor of KAERI (Korea Atomic

Energy Research Institute) as a driver fuel.

KAERI has collaborated with ANL(Argonne National Laboratory, USA) since 1992 in applying a centrifugal atomizing technology developed by KAERI to a proliferation-resistant nuclear fuel for research reactors. KAERI made a agreement on a collaboration program with ANL at December in 1996. The atomized U_3Si_2 powder of 2 kg was offered to ANL and used for the fabrication of ANS nuclear fuel at BWXT and the results were published at the 19th RERTR international conference [3]. Recently, the irradiation test of micro-plate using this atomized powder carried out at ATR reactor of INEEL in USA and the results showed the performance of atomized powder was superior to that of comminuted powder [4]. The irradiation test for U_3Si_2 dispersed fuels at HANARO reactor has been carried out in order to compare the in-pile performances of between the two types of U_3Si_2 fuels, prepared by both the atomization and comminution processes. KAERI has also conducted all safety-related works such as the design and the fabrication of irradiation rig, the analysis of irradiation behavior, thermo-hydraulic characteristics, thermal and mechanical analyses for both irradiation rig and fuel plate.

In this study, the fabrication process, the post-irradiation works including the safety analysis and irradiation history of the U_3Si_2 dispersed mini-plate fuels tested in the HANARO reactor are comprehensively investigated in order to compare the in-pile performances of the two types of U_3Si_2 fuels, prepared by both the atomization and comminution processes.

Fuel Fabrication

In order to fabricate U_3Si_2 powder by atomization method, low-enriched uranium lumps and silicon chips with a slightly hyper-stoichiometric composition were charged and induction-melted in heat-resistant ceramic crucible[5]. The molten metal was heated to approximately 200℃ higher than the melting point and was fed through a small nozzle onto a rapidly rotating graphite disk on a vertical axis. Liquid alloy droplets were then spread from the disk by a centrifugal force and cooled in an argon atmosphere. The atomized powder was collected in a container at the bottom of the funnel-shaped chamber. BWXT Co. fabricated the mini-plates with a uranium density of 4.8 gU/cc, four mini-plates from the atomized U_3Si_2 powder and two from the comminuted fuel powder, respectively. The mini-plates fabricated were inspected in accordance with the specification of the inspection standard and procedure for plate-type fuel[6]. Then mini-plates were provided to KAERI for irradiation test. Figures 1 and 2 show the dimension and general feature of an atomized mini-plate fabricated in BWXT, respectively.

Safety Evaluation

The requirements which should be satisfied for the safe irradiation test is a test rig flow rate at a pressure drop of 200 kPa less than 12.7 kg/s, margin of onset of nucleate boiling greater than 12.7℃, critical heat flux ratio greater than 2.50, fuel centerline temperature less than 350℃, temperature rise across the fuel surface oxidation layer less than 114℃, and fuel swelling less than 20 vol%. These should be fulfilled in order to prevent fuel failure mainly due to blistering or spallation.

The reactivity change due to the insertion of a mini-plate test rig was 0.5 mk which is far less than the operational limits and conditions of 12.5 mk. The fuel power including uncertainty and control rods position effect was evaluated as 25.5 kW when four of all fuel plates are summed up. The thermal margin was estimated large enough to perform the safe irradiation test.

Irradiation Rig

The irradiation test vehicle, named as the mini-plate rig, was designed and fabricated according to the safety-related requirements for HANARO[7]. In the housing part of mini-plate rig, as shown in Fig. 3, four mini-plates can be loaded independently. The most parts consisting of the rig, except fuel loading section and outer protection tube, have a similar structure with HANARO driver fuel to maintain a sufficient strength during irradiation test in the HANARO.

Prior to the irradiation test, the endurance test for mini-plate rig was conducted to verify the integrity of the rig under the HANARO operating condition[7]. The flow velocity across the fuel rig corresponding to the pressure drop of 200 kPa, was measured to be about 6.78 kg/sec. Vibration frequency for the mini-plate rig ranged from 14 to 19 Hz. RMS(Root Mean Square) displacement for the fuel rig was less than 7 ㎛, and the maximum displacement was less than 20 ㎛. Based on the endurance test results, the appreciable fretting wear for the mini-plate rig was not observed. It was confirmed from the above results that the mini-plate rig is designed and fabricated to satisfy the safety-related requirement for the HANARO.

Irradiation Conditions

The mini-plates, three mini-plates from atomized U_3Si_2 fuel and one from comminuted fuel, were loaded at OR6 hole in the HANARO and irradiated for 110.7 F-P-Day residence time at about 70 % operating capacity from July 1999 to Match 2000. Table 1 summarizes typical irradiation conditions for mini-plates in the HANARO. Fig. 4 shows the linear power history of mini-plate as well as reactor power during irradiation test. The maximum linear power was evaluated to be about 82 kW/m, in which the peak temperature of mini-plate was estimated to have been 88.3℃ during irradiation. The average burn-up and fission density after the irradiation test was calculated to be 70.5 at.%U_{235} and 1.58×10^{21} fissions/㎤, respectively.

Post Irradiation Examinations

1. Visual Examinations

The mini-plates irradiated in the HANARO were subjected to post-irradiation after the irradiation following suitable periods, for about eight months, of cooling. Visual examination showed that the mini-plates have been irradiated without any defects and wear formed on the surface, indicating no abnormal swelling. Only a small amount of fretting wear due to a fast coolant flowing in a fuel test zone has been taken place in the upper cap part of the irradiation rig that acts as a role of fixing the mini-plates. White-colored oxide in the fuel meat region of every plate which can be easily distinguished from the cladding zone in mini-plate was also observed.

2. Gamma Scanning

Gamma scanning for four mini-plates has been carried out after visual examination in M1 hot cell. The gamma ray from mini-plate 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. Fig. 5 shows the typical gamma scan result from mini-plate. It was observed from all samples that, although gamma scan can only give qualitative data for evaluating the burn-up of irradiated fuel, there seems to be no apparent position dependence of burn-up in fuel meat region due to the relatively short size of the mini-plate itself.

3. Dimension measurement

The dimension of mini-plate was measured from several positions in the fuel meat region. Table 2 represents the result, in which the thickness was in the ranged from 1.372~1.379 mm. This implies the uniform growth of the plate. Considering the initial of mini-plate(1.29~1.30 mm) before irradiation, it is noted that the increase of plate is mainly originated from fuel swelling as well as oxide layer growth. The fuel swelling can be calculated by the increase of thickness in the fuel meat zone by removing the oxide layer formed at the plate surface. The thickness increase from the fuel meat zone was estimated to be about 44 microns, which implies fuel meat swellings of ~3.3 $\triangle V/V_m$. Therefore the mini-plate tested the in HANARO after 70 at.% burn-up showed a sound swelling behavior.

4. Cladding Oxide Layer Thickness

The oxide layer for each atomized and comminuted mini-plates was investigated from metallographic sample in order to examine whether the spallation of cladding took place during irradiation. The measurement was taken at the several positions, at the sample location for metallography, from the fuel meat center to the edge region in the middle section of the mini-plate. Fig. 6 shows typical microstructure representing oxide layers formed at the most center region in atomized mini-plate. As listed in Table 3, the oxide layer, irrespective of the kind of mini-plate, was measured to range from 8.6 ㎛ to 17.46 ㎛ in average value. The maximum layer was 28.01 ㎛ in atomized sample. It is of certain that the difference of oxide layer thickness depending on fuel meat position seems to be minimal due to its small fuel meat size.

5. Fuel Microstructure(Optical)

In order to prepare the sample for microstructural examination, the mini-plates were cut down into four pieces. Then the optical observation was focused at the sample positions from the fuel meat center to the edge region in the middle section of mini-plate. Figures 7 shows the optical microstructures of each atomized and comminuted U_3Si_2 mini-plates after 70 at.% burn-ups. Irrespective of the difference of fuel shape, a large amount of as-fabricated porosity remains but with being a rounded inner surface to decrease the surface energy, and no fission gas bubbles are visible in the fuel particles. Moreover, the uranium silicide particles reacted slightly with the aluminum matrix around fuel particle. As listed in table 4, where the value was averaged from the thickness measured at each fifteen fuel particles, a reaction layer thickness ranges from 1.73 ㎛ to 2.13 ㎛ in all samples. The interfacial layer formed between the fuel particle and the Al matrix becomes slightly thicker as the particle locates from the meat-edge side to the center, but there was no discernable difference depending on the fuel particle type.

6. Fuel Microstructure(EPMA)

In order to prepare SEM sample from irradiated mini-plate, a special punching tool was designed to make samples for SEM observation. Two small pieces of irradiated samples(atomized and comminuted) with having a diameter of 1.57 mm by using a punching jig in the hot cell was taken from the fuel meat center region. Then the fuel meat in the punched sample was cut down by hand in a glove box to observe the fractured surface of the irradiated fuel particle. SEM observation on the polished fuel samples were also carried out to investigate the bubble size distribution.

Figures 8 and 9 show each fractographs and polished surfaces of the U_3Si_2 particles in mini-plate after 70 at.% burn-ups, which reveal the extent of fission gas bubble formation as well as the depth of the fuel-Al inter-diffusion layer at the particle surface. There are apparent similarities in comparing the bubble formation as well as bubble distribution between the atomized and comminuted fuel samples. The fission gas bubbles have formed, in general, on small grain boundaries. It seems that both fuel samples irradiated in the HANARO are at the beginning stage of bubble formation. The bubbles which are visible have not covered entirely the fuel particle, nucleating and growing at the preferred regions having a maximum bubble size of under 1 micron. However, a bubble distribution with slightly smaller mean diameters was found in the atomized fuel than in the comminuted sample, and the bubble population in atomized fuel appears to be more homogeneous than that in the comminuted fuel. This observation indicates that the bubble morphology in the atomized U_3Si_2 fuel results from more fine grain structure formed during atomization, compared to the comminuted fuel.

Fig. 10 represents the comparison of fission gas bubble distribution formed in each type of fuel. The bubbles in the atomized fuel, about 0.25 ㎛ in the mean size and 0.6 ㎛ of the maximum, have a characteristics of narrower bubble size distribution than in the comminuted fuel where the bubbles have the mean size of ~0.35 ㎛.

According to the previous studies, the composition of the interaction layer formed in uranium silicide dispersion fuel during irradiation has been known to be $U(Si_3Al)_3$.[4] In this study, the more quantitative analysis for the compositional changes were carried out across the fuel/aluminum interface by using Electron Prove Micro Analysis(EPMA). Fig. 11 shows the typical compositional changes at near the fuel particle surface in atomized U_3Si_2 sample. In an un-reacted fuel particle region, the result is fairly consistent with the fuel composition of U_3Si_2. 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 $U(Si_3Al)_3$, indicating that the formation of interfacial layer in U_3Si_2/aluminum during irradiation is a diffusion controlled reaction.

When comparing the bubble morphology from the fuels irradiated in the HANARO with that from the RERTR-1&2 irradiation(see Fig. 12) as well as the previous result[8], it is of note that there are some discrepancies in bubble population, in which the bubble formation from the HANARO irradiation, although the fuels have been irradiated with an estimated burn-ups of 70 at% U_{235}, are apparently similar to, or possibly at burn-ups between 40 at% U_{235} and 70 at% U_{235}, the RERTR-1 microstructures. The main reason for such a difference of bubble population is still not clear although it can possibly be caused as a result of irradiation conditions such as fission rate and so on. Therefore uranium and plutonium isotopic analysis for mini-plate samples are being performed to provide an absolute burn-up information.

Summary

The irradiation test on both the atomized and comminuted U_3Si_2 dispersion mini-plate fuels has been performed in the HANARO in order to demonstrate a better irradiation performance of the atomized U_3Si_2 fuel powder as well as to compare in-reactor behaviors. The mini-plates, irradiated until 70 at% U_{235} burn-ups at OR6 hole in the HANARO were subjected to IMEF for post-irradiation analysis. Visual inspection, gamma scanning, dimension measurement, microstructure analysis including

EPMA have been conducted. From the PIE results, it is of note that both the atomized and comminuted U_3Si_2 dispersion mini-plate fuels exhibit sound swelling behaviors. The aluminide layer formed in all samples ranges from 1.73 ㎛ to 2.13 ㎛ in thickness, which implies fuel meat swellings of ~3.3 $\triangle V/V_m$. However a bubble distribution with slightly smaller mean diameters was found in the atomized fuel than in comminuted sample, and the bubble population in atomized fuel appears to be more homogeneous than that in the comminuted fuel.

Acknowledgements

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Table 1. Irradiation Conditions in the HANARO Reactor.

Thermal Neutron Flux	1.7X10^{14} n/cm^{2}
Coolant Inlet Temperature Outlet Temperature Velocity pH	 35℃ 36.9℃ 18.9 m/sec 5.5-6.5
Max. Centerline Temperature	88.3℃
Max. Thermal Flux	2.64 MW/㎡
Pressure Drop on mini-plate	137 kPa
$\triangle T_{ONB}$	39.5℃
CHFR	9.2
Fission Density	1.58X10^{21} fissions/㎤
Irradiation Time	110.7 FPD at 20 MW

Table 2. Thickness of mini-plates after irradiation.

Sample I.D.	Thickness(mm)	
	before irr.	after irr.
(A)129-0016-02	1.29～1.30	1.372±0.011
(A)129-0016-03	1.29	1.352±0.012
(A)129-0016-04	1.29～1.30	1.378±0.008
(C)129-130-20	1.29	1.379±0.009

※ (A) : atomized U_3Si_2 mini-plate
(C) : comminuted U_3Si_2 mini-plate

Table 3. The averaged oxide layer thickness of mini-plates.

		Atomized Mini-Plate 129-0016-02 [㎛]		Comminuted Mini-Plate 129-130-20 [㎛]	
		Average	Max.	Average	Max.
LEFT	1(center)	13.39	22.56	12.93	21.79
	2	13.04	17.12	13.60	21.40
	3	12.37	16.73	10.83	17.12
	4	13.99	28.01	11.40	18.28
	5	9.92	19.07	17.46	25.29
	6(edge)	14.41	20.23	10.76	17.90
LIGHT	1(center)	10.22	17.90	13.52	18.68
	2	10.38	17.12	10.14	21.79
	3	8.60	14.40	9.39	13.62
	4	8.61	14.79	11.12	14.79
	5	10.99	17.12	12.68	21.01
	6(edge)	7.97	13.62	10.71	15.17

Table 4. The measured thickness of interfacial layer.

	Atomized U_3Si_2 129-0016-02 [㎛]	Comminuted U_3Si_2 129-130-20 [㎛]
TOP	2.08	2.13
MIDDLE	1.79	1.83
BOTTOM	1.73	1.75

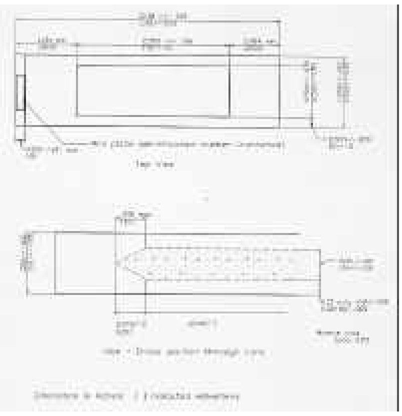


Fig. 1. Dimension of mini-plate.

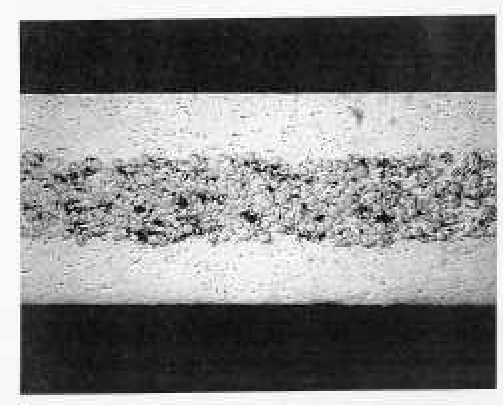


Fig. 2. Microstructure of the atomized U_3Si_2 mini-plate.

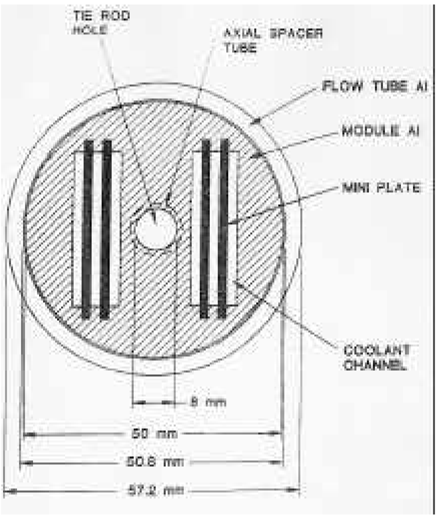


Fig. 3. Cross sectional view of fuel loading section.

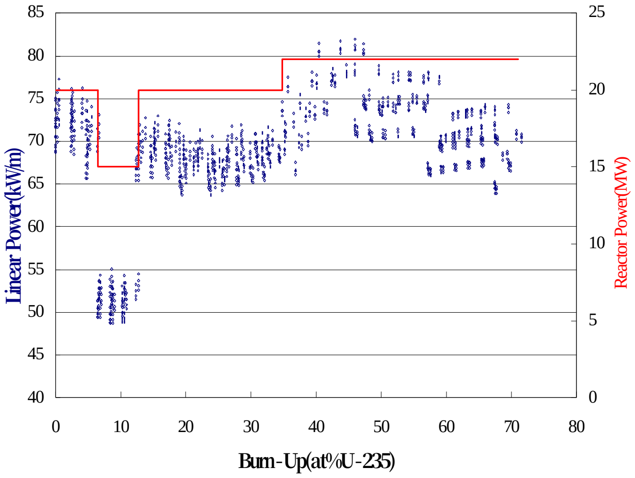


Fig. 4. The linear power history of mini-plate.

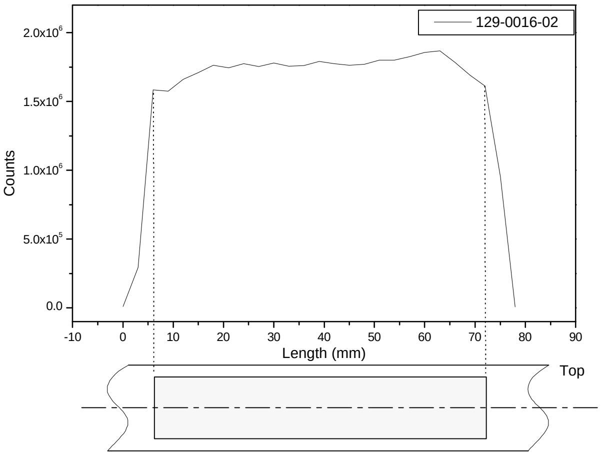


Fig. 5. Gamma scanning spectrum of the atomized mini-plate.(129-0016-02)

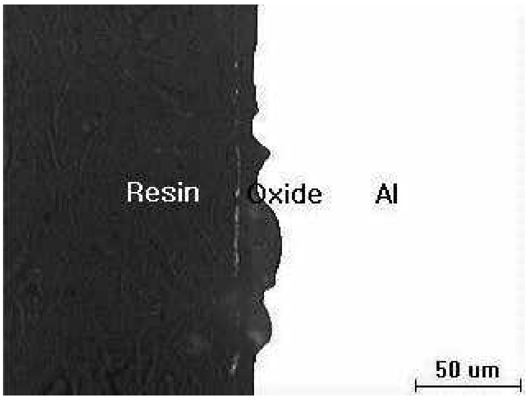
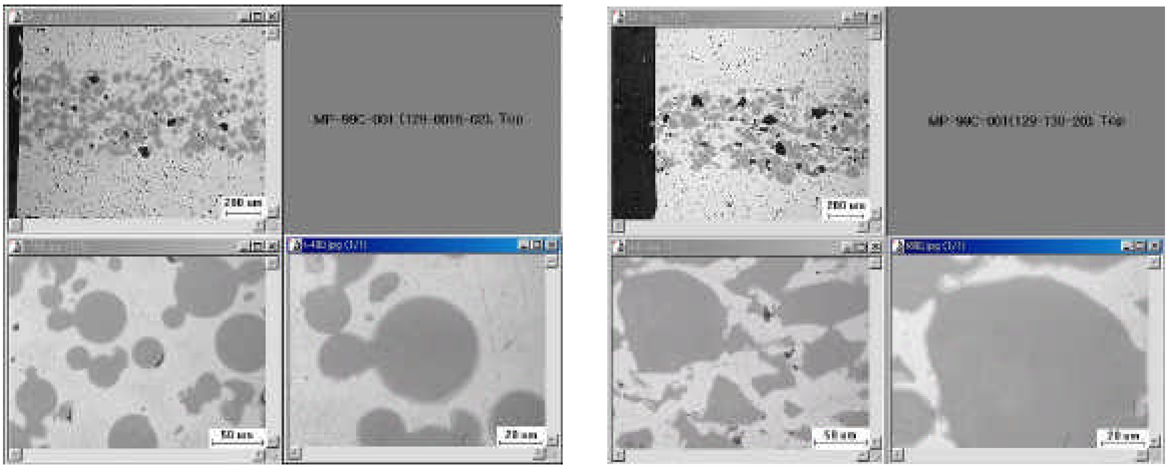


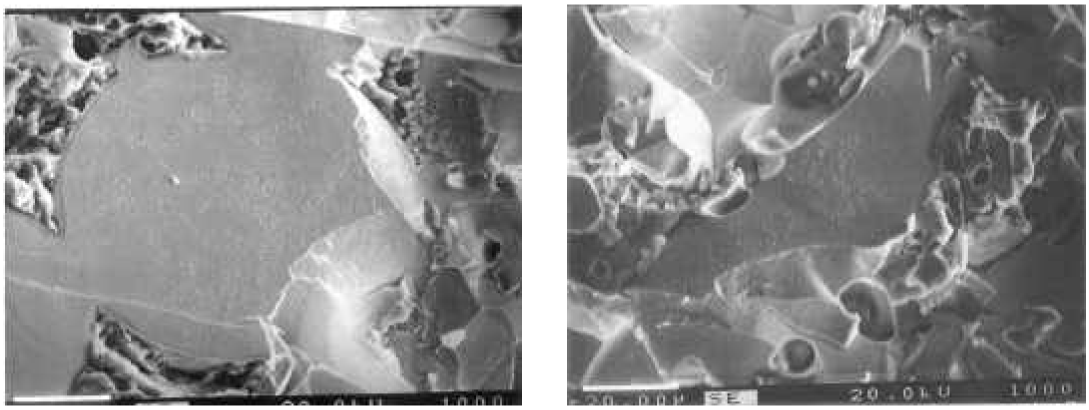
Fig. 6. Typical oxide layer on mini-plate 129-0016-02.



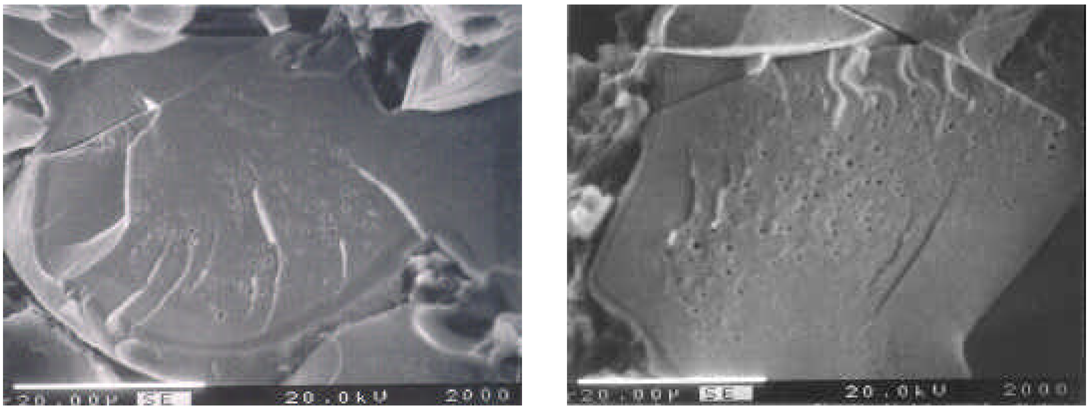
(a) atomized

(b) comminuted

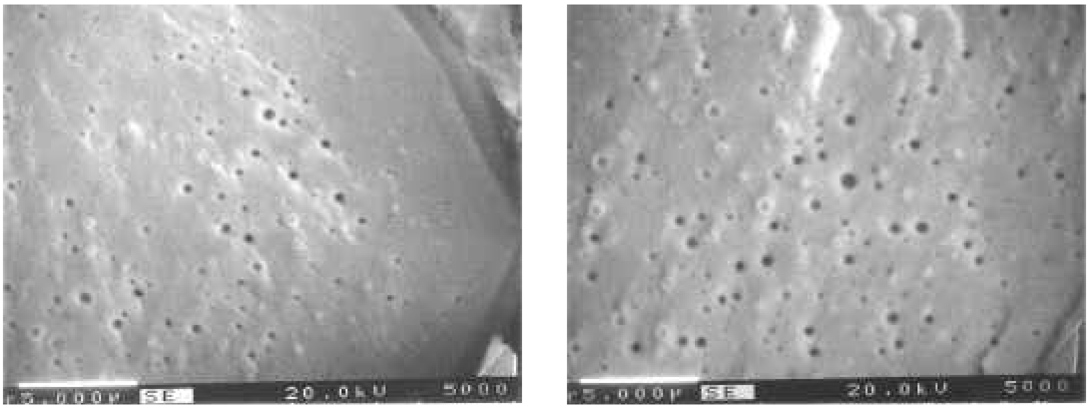
Fig. 7. Optical microstructures from atomized and comminuted mini-plates.



(a) Atomized(X1000)　　　　　(d) Comminuted(X1000)

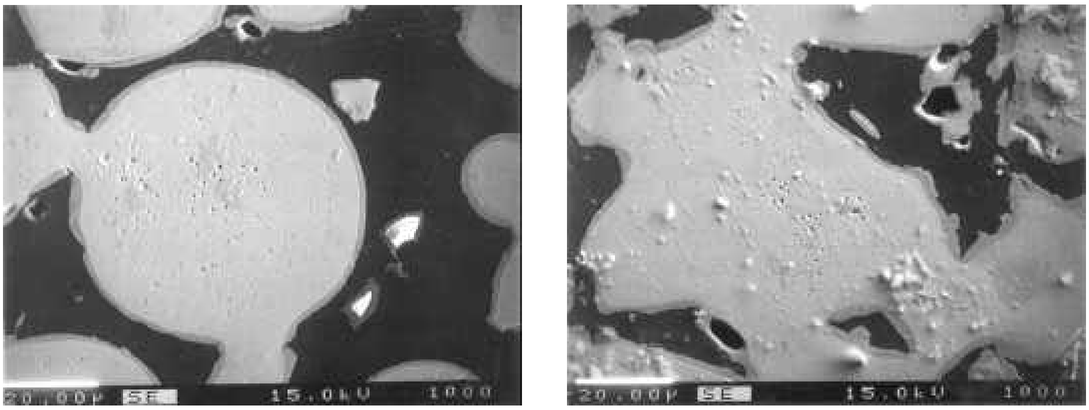


(b) Atomized(X2000)　　　　　(e) Comminuted(X2000)

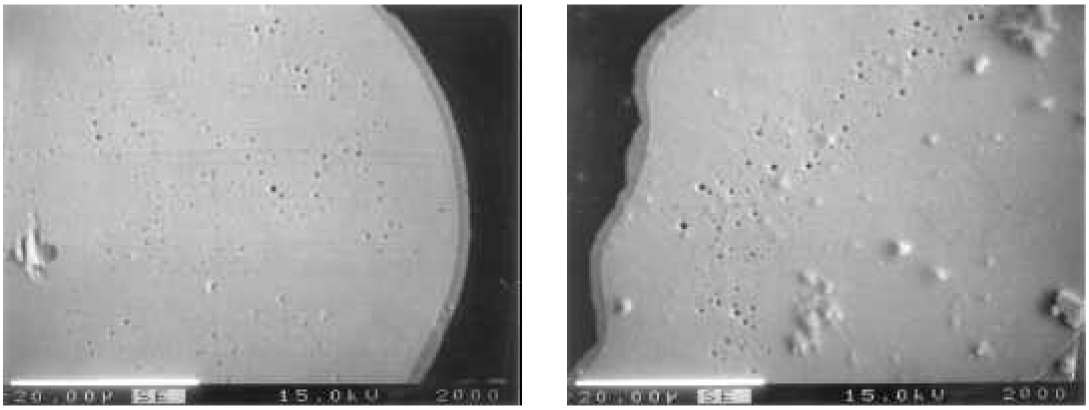


(c) Atomized(X4000)　　　　　(f) Comminuted(X4000)

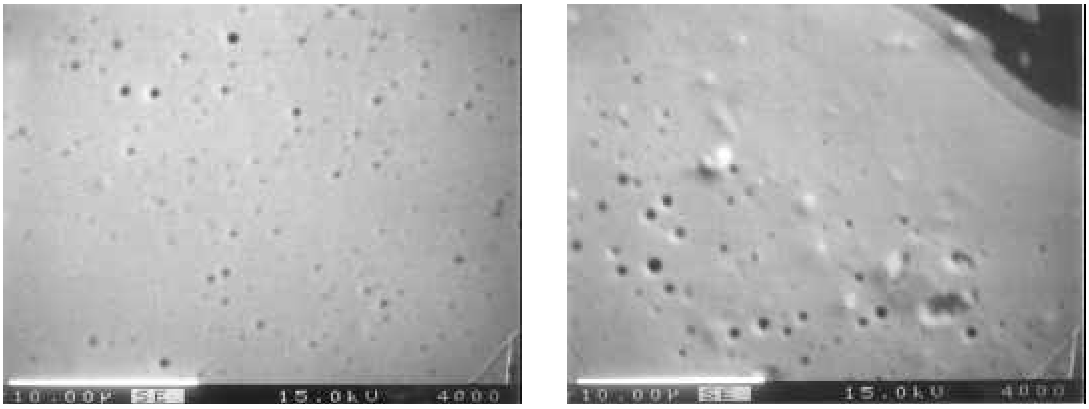
Fig. 8. SEM microstructures of fractured mini-plate fuels.



(a) Atomized(X1000)　　(d) Comminuted(X1000)



(b) Atomized(X2000)　　(e) Comminuted(X2000)



(c) Atomized(X4000)　　(f) Comminuted(X4000)

Fig. 9. SEM microstructures of polished mini-plate fuels.

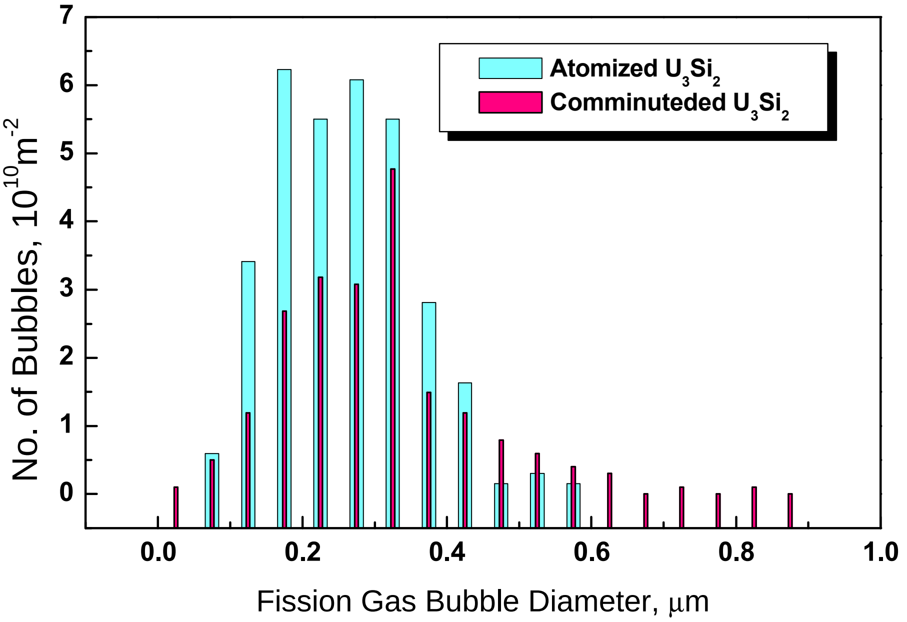


Fig. 10. Bubble size distribution of atomized and comminuted fuel.

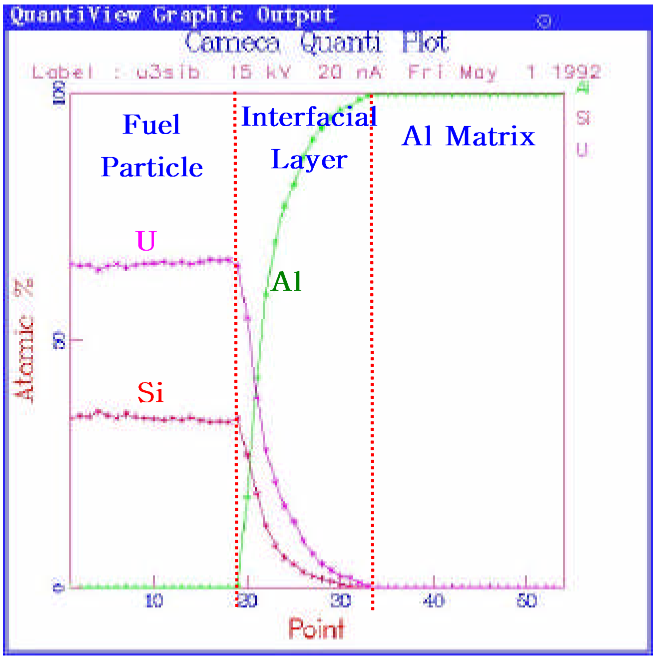
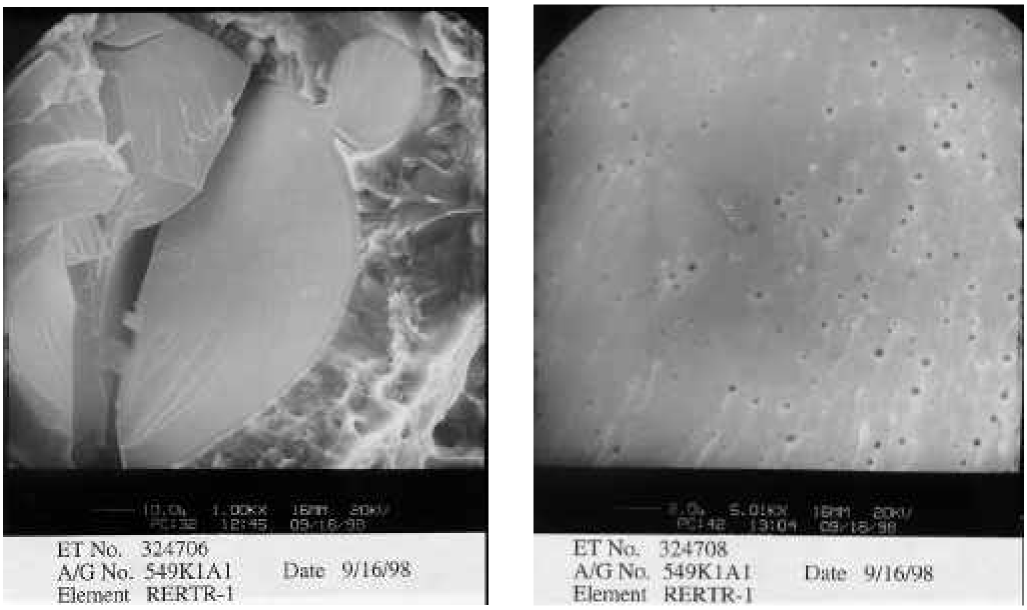
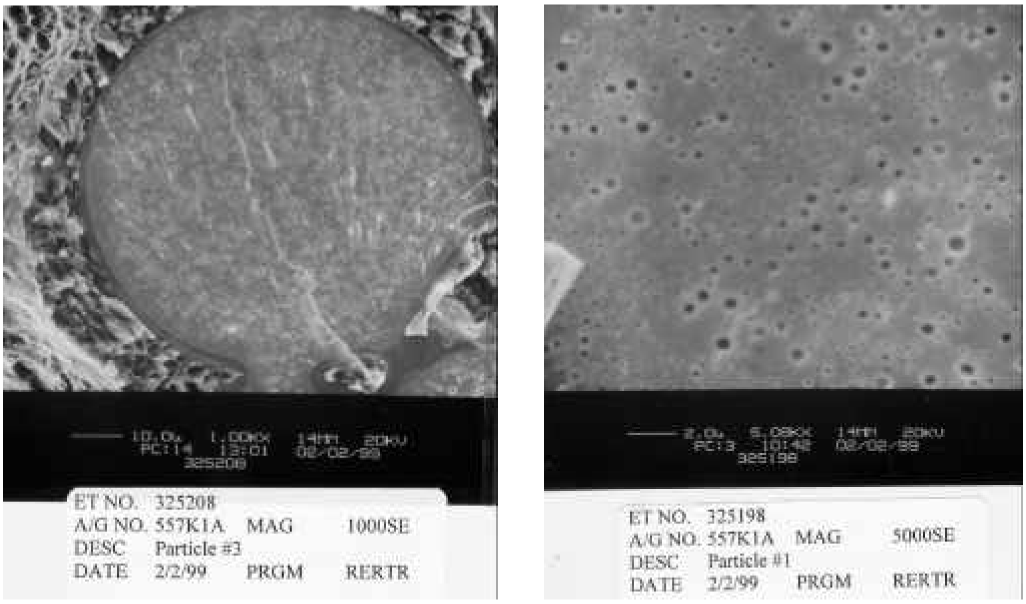


Fig. 11. Compositional changes of across the interfacial layer in U_3Si_2 fuel meat.



(a) after 40 at.% BU(X1000) (b) after 40 at.% BU(X5000)



(c) after 70 at.% BU(X1000) (d) after 70 at.% BU(X5000)

Fig. 12. RERTR-1&2 irradiation data of atomized U_3Si_2 fuel.