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A Study on the Characteristics of U-(5.4~10 wt%)Mo Alloy Powders Prepared by Centrifugal Atomization

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Abstract

The characterization on powders of U-(5.4~10 wt%) Mo alloy depending on Mo content prepared by rotating disk centrifugal atomization process was carried out in order to investigate the phase stability of the atomized U-Mo alloy system. All of atomized U-(5.4~10 wt%)Mo alloy particles exhibits, irrespective of Mo composition, a Mo micro-segregation at grain boundaries as well as fine polycrystalline structures with γ-U grains below 5 μm in size. It revealed from the EDS and neutron scattering analyses that the Mo content at the cell boundaries is about 2~3 at% lower than that in the cell, which corresponds to the existence of two γ-U phases with both having Im3m bcc space groups. As Mo concentration increases, the lattice parameters of both γ_1-U and γ_2-U phases become decreasing with increasing the fraction of γ_2-U phase. The decomposition of γ-U phase at 500℃ take place by the cellular mechanism, nucleated at γ cell boundaries due to its somewhat lowered Mo content. Then most γ-U cells were separated into α-U and γ-U platelets by a cellular reaction as the annealing time increases.

1. Introduction

Recently, increased attention has been paid to the development of aluminum matrix dispersion fuels with increased fissile uranium density to make possible the conversion of research reactors from the use of high-enriched uranium (HEU) to

low-enriched uranium (LEU)[1-2]. Although U_3Si_2-Al dispersion fuel possesses very stable irradiation behavior, new dispersion fuels with loading up to 8~9 g U/cm^3 are required for the conversion of several high performance reactors[3]. Because there is a practical limit of 55% for the volume loading of the dispersed fuel phase in a commercial process, fuel dispersants with uranium densities higher than about 15 g U/cm^3 are required to reach this goal[3]. Since earlier studies have shown the γ-uranium phase to behave rather stably under irradiation[3], uranium alloys which retain the γ phase are preferred. Until now, the most promising candidate is proven to be the U-Mo alloys, which have a relatively large range of γ phase that is retainable in a metastable state at a temperature at which the fuel is fabricated and irradiated. Recently, It has also been found by RERTR irradiation test that the U-Mo(U-10Mo, U-8Mo, U-6Mo) and U-6Mo-X(X=0.6Ru,1Pt) alloys appeared to be acceptably with relatively small fuel-matrix interaction layers and stable bubble growth, which is similar to the irradiation behavior of the U_3Si_2 fuel.[4] The U-10Mo exhibited generally stable irradiation performance compared to U-8Mo, U-6Mo, and U-6Mo-X alloys. In addition, the atomized U-10Mo alloy showed the smallest-sized and lowest density of fission gas bubbles forming almost exclusively on grain boundaries. Kim et. al.[5] characterized the atomized powder of U-10Mo alloy, in which the atomized U-10Mo alloy exhibited the cell structure due to a Mo micro-segregation at grain boundaries and has a high γ-U phase stability. The stable irradiation behavior of atomized U-10Mo can be closely related with the metastable phase arised from rapid solidification.

In this study, The characterization on powders of U-(5.4~10 wt%) Mo alloy depending on Mo content prepared by rotating disk centrifugal atomization process was carried out in order to investigate the phase stability of the atomized U-Mo alloy system.

2. Experimental Procedures

Atomized U-(5.4, 6, 7, 8, 9 and 10 wt%)Mo alloy powders were atomized using depleted uranium lump(99.9 wt%) and molybdenum lump(99.7 wt%). The alloying elements were induction melted in a graphite crucible coated with high temperature resistant ceramics, yttria stabilized zirconia. In order to obtain a suitable size distribution of U-(5.4~10 wt%)Mo alloy powders, atomization parameters such as the super-heating and rotating speed of the turn disk were adjusted.

The size distribution of U-(5.4~10 wt%)Mo alloy powders were measured by

sieve analysis. The morphology and microstructure of U-(5.4~10 wt%)Mo alloy powders were characterized with a scanning electron microscope(SEM). Compositional analysis was done by energetic dispersive spectroscopy(EDS) with SEM. The powder diffraction data on as-atomized powders were obtained by using X-ray diffraction analysis with Cu $K\alpha$ radiation as well as neutron diffraction analysis from the 32-detector HRPD of KAERI, using 1.8339Å neutron from Ge(331), and 1.2193Å from Ge(335) monochromator, both with a 90° take-off angle.

In order to investigate the high temperature phase stability of U-(5.4~10 wt%)Mo alloy powders, each powders were vacuum-sealed in quartz tubes and were annealed at 500℃ with increasing time, where the microstructural changes and phase transformation behaviors of the alloy powders were determined using X-ray diffraction analysis and scanning electron microscopy(SEM).

3. Results and discussion

3.1 Characterization of powders

Most of U-(5.4~10 wt%)Mo powders are nearly closed to the desired composition. The atomized particles shown in Fig. 1, representing the typical SEM microstructure of atomized powder, appeared to be spherical in shape, having smooth surface.

The cross-sectional SEM micrographs of atomized U-Mo alloy particles depending on Mo content are illustrated in Fig. 2. It can be seen that all of the atomized U-(5.4~10 wt%)Mo alloy particles exhibit, irrespective of Mo composition, fine polycrystalline structures with many non-dendritic γ-U grains below 5 ㎛ in size. The grain size becomes smaller as the particle size becomes finer, which indicates that the higher cooling rate of the finer droplet, even though considering the very high cooling rate of about 10^4 K/sec during atomizing process, decreases the time available for solidification and thus enhances finer polycrystalline structure. It was also revealed that the atomized U-(5.4~10 wt%)Mo alloys, at all Mo concentration, exhibit Mo micro-segregation at grain boundaries due to the rapid solidification, named as a cell structure, which is the characteristic of an alloy with a substantial liquidus-solidus gap, such as U-Mo alloy. Compositional analysis by EDS in atomized U-(5.4~10 wt%)Mo alloy powders revealed that the Mo content at the cell boundaries is, irrespective of alloy composition, about 2~3 at% lower than that in the cell, which represents the micro-segregation of Mo in the atomized U-Mo alloys system. At the same time, there exhibit somewhat different cell morphology depending on Mo concentration. As the Mo concentration increases in U-Mo from 5.4

wt% to 10 wt%, the cell boundary becomes thick from about 0.3 ㎛ to about 0.6 ㎛ with maintaining similar grain size. This observation indicates that the fraction of cell boundary, i.e. Mo depleted zone, increases as the Mo content increases.

3.2 X-ray and Neutron Diffraction Analysis

In order to identify the as-quenched phase in atomized U-(5.4~10 wt%)Mo alloy powders, X-ray and neutron diffraction analyses were carried out. Irrespective of alloy composition, all X-ray diffraction data on U-(5.4~10 wt%)Mo alloy powder showed single γ-U phase. However, when analysing by using neutron diffraction method, there existed two γ-U phases in U-(5.4~10 wt%)Mo alloy powders. Fig. 3 shows the typical neutron diffraction pattern for atomized U-10 wt% Mo powder. Since the two primary phases (γ_1-U and γ_2-U) with both having Im3m bcc space groups were so similar in unit cell sizes, the peaks of the two phases could be distinguished mostly at higher scattering angles with higher instrumental resolution. The different lattice dimensions may be caused by different Mo concentration but it is impossible to determine Mo content in each phase by diffraction method alone. Using the information from an earlier study by A. E. Dwight [6] that the lattice parameter of phase in the U-Mo system is related to the Mo concentration by the equation α(Å) = 3.4808-0.00314(at% Mo). Based on these assumption, the atomic concentration of Mo atoms in the U-10 wt% Mo solid solutions was calculated to be 22 at% with a lattice parameter of 3.4116 Å for γ_1-U phase and 17.5 at% with a lattice parameter of 3.4261 Å for γ_2-U phase. This result is in accord with the SEM/EDS micrograph observation, in which there exists cell structure with Mo micro-segregation. Therefore it revealed from the above results that γ_1-U and γ_2-U phases are related to the major phase and cell boundary phase, respectively.

The final phase composition and structural parameters on atomized U-(5.4~10 wt%)Mo alloys analysed by the neutron diffraction method are listed in Table 1. In the case of U-5.4 wt% Mo alloy, As Mo concentration increases, the lattice parameters of both γ_1-U and γ_2-U phases become decreasing with increasing the fraction of γ_2-U phase.

3.3 Annealing Test on Alloy Powders

The γ-U phase of U-Mo alloy annealed below the eutectoid temperature(560℃) has a tendency to be decomposed as the thermodynamically stable lamellar structure including α-U and γ'-U_2Mo phases[7]. The SEM micrographs of atomized U-(5.4~

10 wt%)Mo alloy powders after isothermal annealing at 500℃ for 1 hour are shown in Fig. 4. The majority γ-U phase(γ_2 phase) at cell boundary region of the U-5.4 wt% Mo alloy powder, was decomposed to two (α-U and γ'-U_2Mo) phase structure by a cellular reaction, with remaining γ_1 phase in the cell. As the Mo concentration increase up to 8 wt%, U-Mo powder generally retained a γ-U cell structure except for finely decomposed structure formed primarily around the cell boundary and the extent of decomposed phase becomes smaller. When compared with the X-ray diffraction results that are listed in Table 1, with Fig. 4, U-7 wt% Mo and U-8 wt% Mo powders illustrate no distinct indication of phase decomposition by X-ray diffraction data even though there already exist small amount of finely decomposed α-U and γ'-U_2Mo at cell boundaries. However the γ-U phase of U-9 wt% Mo and U-10 wt% Mo powder was not affected during 1 hour heat treatment at 500℃.

As the heat treatment time increases to 5 hours, it revealed from the X-ray result that the γ-U phases in the U-5.4 wt% Mo powder were almost decomposed into two (α-U and γ'-U_2Mo) phases. In addition, there were also detectable indications of phase decomposition by X-ray diffraction pattern from γ-U phase to the α-U and γ'-U_2Mo phases up to the alloy composition of 9 wt% Mo.

Fig. 6 illustrates the scanning electron micrographs of atomized U-(5.4~10wt%)Mo powders after annealing at 500℃ for 100 hours. The micrograph of the atomized powder after annealing at 500℃ for 100 hours showed most γ-U cells were separated into α-U and γ-U platelets by a cellular reaction. However, as the Mo content increases, the fraction of retained γ-U phase increases with maintaining a cell morphology. The X-ray diffraction pattern of the atomized powder after annealing at 500℃ for 100 hours showed that, irrespective of Mo content, some γ-U phases of U-Mo powder were transformed into α-U and γ'-U_2Mo phases.

From the above results, it is of certain that the decomposition of γ-U phase at 500℃ take place by the cellular mechanism, nucleated at γ cell boundaries due to its somewhat lowered Mo content. In addition, alpha uranium platelets also precipitated in a poorly defined Widmanstetten form (Fig. 6). It seems that precipitation of α uranium platelet competes with the cellular reaction at 500℃. It is thought that below the eutectoid temperature, the cells consist initially of α-U and enriched γ-U rather than α-U and γ'-U_2Mo as would be expected for a normal eutectoid reaction[8,9]. Then the transformation occurs continuously involving formation of an ordered intermediate phase (γ'-U_2Mo), with α-U presumably forming from a solute depleted matrix around the cell boundary.

4. Conclusion

The characterization on powders of U-(5.4~10 wt%) Mo alloy depending on Mo content prepared by rotating disk centrifugal atomization process was carried out in order to investigate the phase stability of the atomized U-Mo alloy system. All of atomized U-(5.4~10 wt%)Mo alloy particles exhibits, irrespective of Mo composition, a Mo micro-segregation at grain boundaries as well as fine polycrystalline structures with γ-U grains below 5 μm in size. It revealed from the EDS and neutron scattering analyses that the Mo content at the cell boundaries is about 2~3 at% lower than that in the cell, which corresponds to the existence of two γ-U phases with both having Im3m bcc space groups. As Mo concentration increases, the lattice parameters of both γ_1-U and γ_2-U phases become decreasing with increasing the fraction of γ_2-U phase. The decomposition of γ-U phase at 500℃ take place by the cellular mechanism, nucleated at γ cell boundaries due to its somewhat lowered Mo content. Then most γ-U cells were separated into α-U and γ-U platelets by a cellular reaction as the annealing time increases.

5. Acknowledgements

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6. References

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Table 1. Neutron and X-ray diffraction results for U-(5.4~10 wt%)Mo alloy powders.

			5.4 Mo	6 Mo	7 Mo	8 Mo	9 Mo	10 Mo
Neutron Diffraction Result	Space Group		Im3m	-	-	Im3m	Im3m	Im3m
	a_o (Å)	γ_1	3.4431	-	-	3.4244	3.4164	3.4112
		γ_2	3.4493	-	-	3.4365	3.4272	3.4261
	F (%)	γ_1	86	-	-	78	57	64
		γ_2	14	-	-	22	43	36
Neutron Diffraction Result	as-atomized		single γ	single γ	single γ	single γ	single γ	single γ
	After Heat Treat-ment	500℃ 1 hr	$\alpha+\gamma'$ cell boundary	$\alpha+\gamma'$ cell boundary	single γ	single γ	single γ	single γ
		500℃ 5 hr	$\alpha+\gamma'$ at cell boundary	$\alpha+\gamma'$ at cell boundary	$\alpha+\gamma'$ at cell boundary	$\alpha+\gamma'$ at cell boundary	single γ	single γ
		500℃ 100 hr	$\alpha+\gamma'$	$\alpha+\gamma'$	$\alpha+\gamma'$	$\alpha+\gamma'$	$\alpha+\gamma'$	$\alpha+\gamma'$

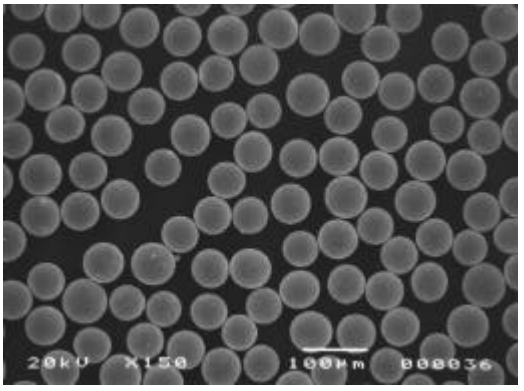
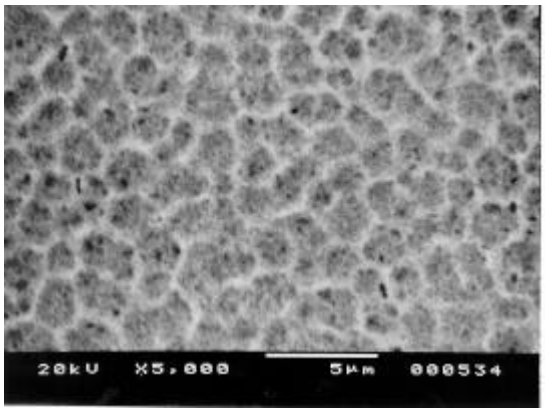
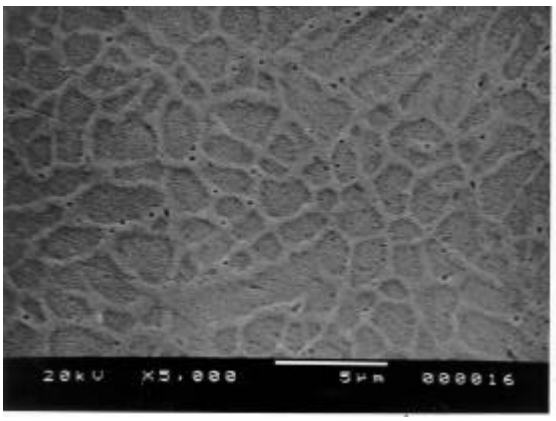


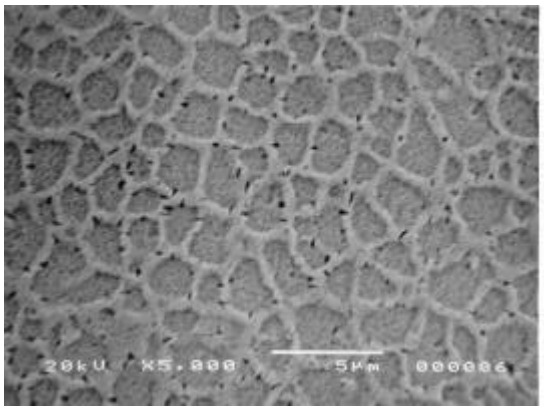
Fig. 1. Scanning electron micrograph of atomized U-10 wt% Mo powder.



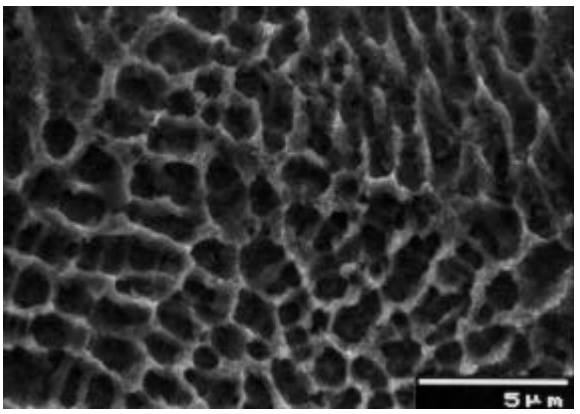
(a) U-5.4 wt% Mo



(b) U-7 wt% Mo



(c) U-8 wt% Mo



(d) U-10 wt% Mo

Fig. 2. Cross-sectional SEM images of the atomized U-(5.4~10 wt%) Mo powders.

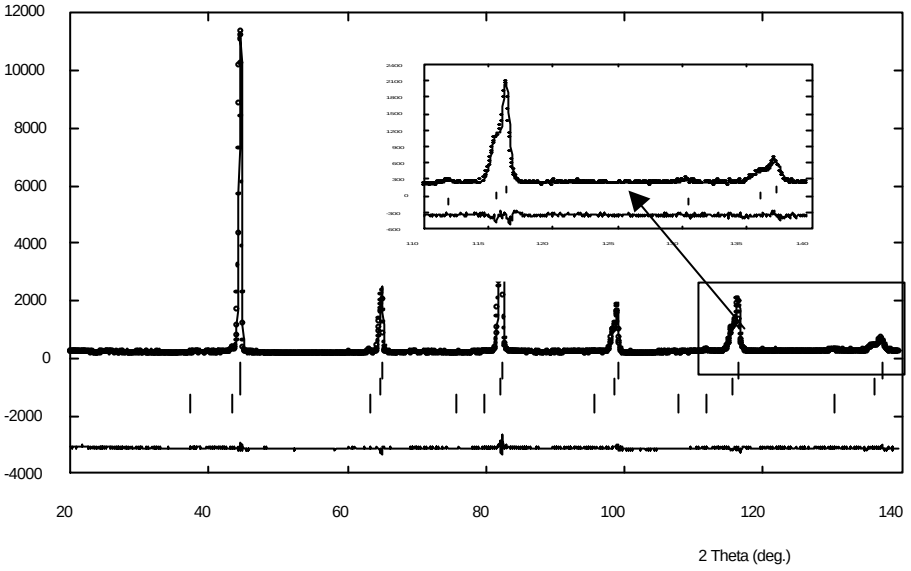
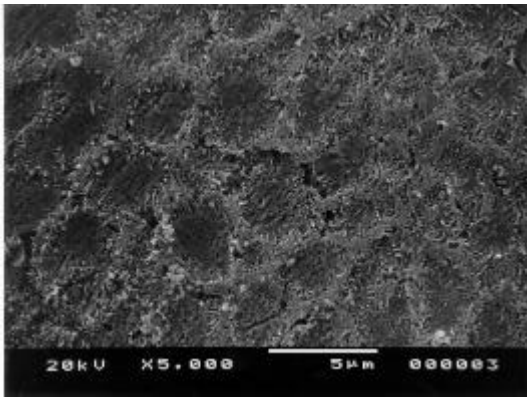
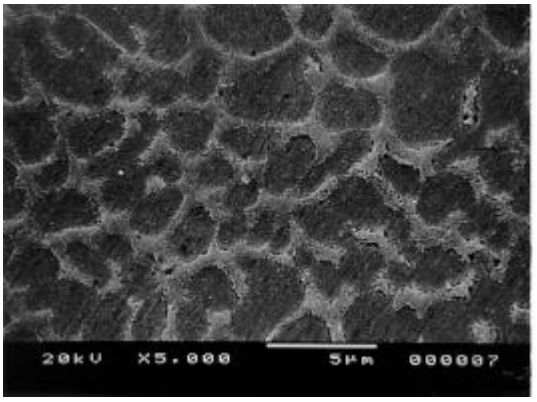


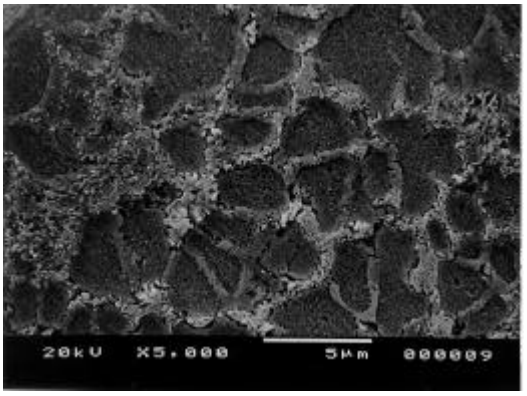
Fig. 3. Neutron diffraction pattern of the atomized U-10 wt% Mo alloy powder.



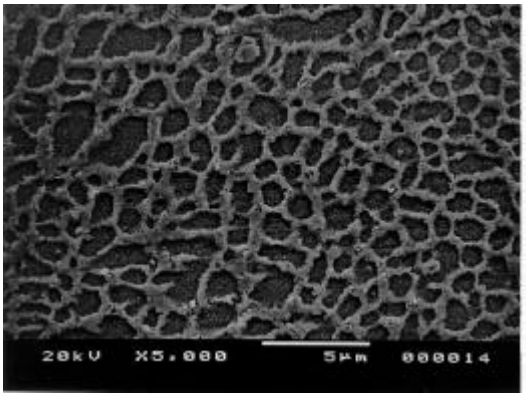
(a) U-5.4 wt% Mo



(b) U-7 wt% Mo

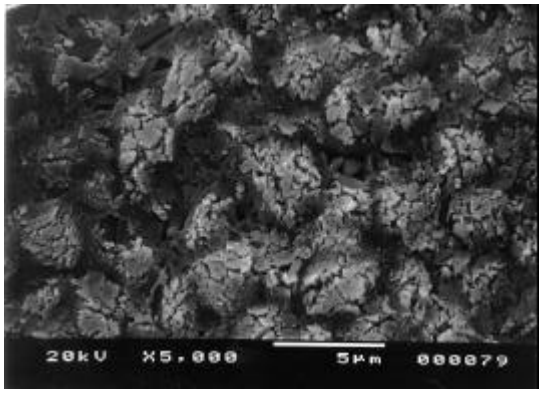


(c) U-8 wt% Mo

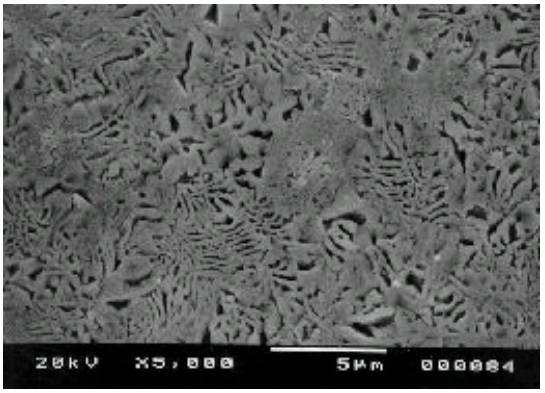


(d) U-10 wt% Mo

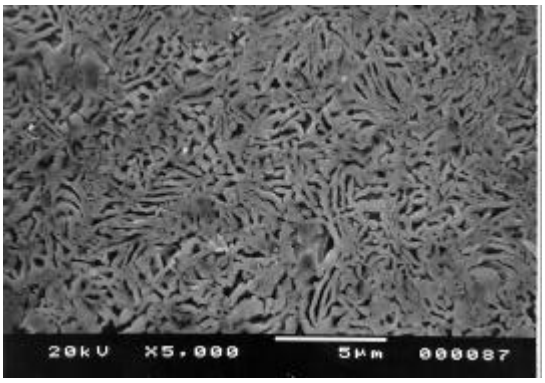
Fig. 4. Cross-sectional SEM images of the atomized U-(5.4~10 wt%) Mo powders after annealing at 500℃ for 1 hour.



(a) U-5.4 wt% Mo



(b) U-7 wt% Mo



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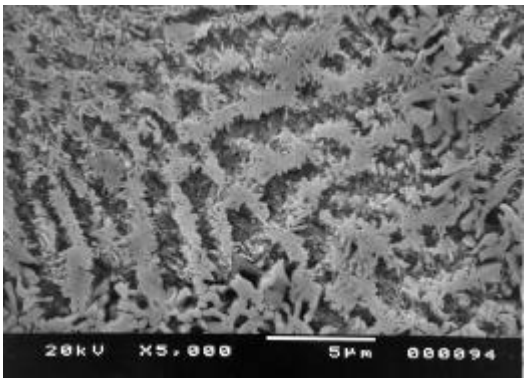

(d) U-10 wt% Mo

Fig. 5. Cross-sectional SEM images of the atomized U-(5.4~10 wt%) Mo powders after annealing at 500℃ for 100 hours.