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Study of decomposition and reactions with aluminum matrix of dispersed atomized U-10 wt% Mo alloy

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Abstract

A neutron diffraction study of atomized U-10 wt% Mo alloy showed that homogenizing annealing above 560 °C retards the decomposition of the metastable γ-phase at 400 and 500 °C. In fuel with U-10 wt% Mo alloy particles dispersed in the Al matrix, Al diffuses along the grain boundaries and reacts with α-U phase formed from the decomposed γ-phase at 400 and 500 °C. The formation of Al_3U and Al_2U phases was observed. Neutron diffraction did not detect formation of a ternary Al–U–Mo compound.

1. Introduction

The U–Mo alloy is one of the prospective materials for low-enriched uranium fuel with high loading because a solid solution of Mo in the γ-U phase possesses acceptable irradiation and mechanical properties and is formed over a wide range of Mo concentration [1]. Below 560 °C the γ-phase can exist only in a metastable state because, according to the equilibrium phase diagram of U–Mo system, below 560 °C the γ-phase decomposes into α-U and γ′-phases (U_2Mo) [2]. Therefore, in the fuel fabrication and its service, it is very important to retain the metastable γ-phase below 560 °C.

In fuel with U–Mo alloy particles dispersed in the Al matrix, along with decomposition of the γ-phase, the reaction of U–Mo alloy phases with the Al matrix may proceed. Formation of new intermetallic compounds and the accompanying processes in the Al matrix can change the properties of fuel composition. In particular, it was shown that the inter-diffusion process between dispersed fuel and the Al matrix may result in the formation of pores in the Al matrix [3].

It was reported that U-10 wt% Mo alloy prepared by the centrifugal atomization method retains the high-temperature γ-U phase with a cubic (bcc) structure at room temperature and has physical properties acceptable for reactor fuels [4]. A neutron diffraction study of U-10 wt% Mo and U-5 wt% Mo alloys showed that as-atomized samples consisted of a mixture of γ-phases with different Mo contents [5,6]. Uniformity in composition of the γ-phase was achieved after homogenizing annealing at a temperature above 560 °C followed by quenching [6].

The main aim of this work was to study the phase composition of the product of the reaction of U-10 wt% Mo alloy prepared by the centrifugal atomization method within an Al matrix at 400 and 500 °C using neutron diffraction. Since at these temperatures the γ-phase decomposes and the Al matrix reacts with the product of decomposition, we studied the decomposition of the γ-phase and the influence of homogenizing annealing on γ-phase stability.

Neutron diffraction is the most appropriate method for this study because the high-penetration ability of neutrons allows us to study bulk (several mm) samples. Because of the high-X-ray absorption of U atoms,

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X-rays have a penetration depth of about the 2 μm which is not appropriate for the study of U–Mo fuel.

2. Experiment

Depleted uranium of 99.9% purity and 99.7% pure molybdenum were used to prepare U–Mo alloys by induction melting. Powders of these alloys were prepared by the centrifugal atomization method using a rotating graphite disk. To obtain a uniform single γ-phase state, the as-atomized powder was homogenized by annealing at 800 °C for 10 h and then quenched. As-atomized and homogenized powder samples were annealed at 500 °C for 10, 50 h to study the decomposition of the γ-phase.

Dispersion fuel rods were prepared by blending U-10 wt% Mo (~37 vol.%) and Al powders, and following extrusion at 400 °C. The rods were annealed at 400 and 500 °C for different times from 100 h to 2000 h to study the dispersed fuel-matrix reaction.

Neutron diffraction patterns from 5° to 155° were measured at room temperature using the high-resolution powder diffractometer (HRPD) at the HANARO reactor of KAERI. The detector bank consists of 32 tube He_3 detectors and 10′ Soller collimators before detectors. A 20′ in-pile collimator and 30′ collimator after Ge (3 3 1) monochromator at a 90° take-off angle were used to obtain monochromatic neutrons with a wavelength of 0.18339 nm. Powder samples were measured in a vanadium can, 8mm in diameter and 40 mm in height.

A scanning electron microscope (SEM) was used to characterize the microstructure of the atomized particles and fuel meat. Samples for SEM were prepared by polishing with 1 μm diamond paste.

3. Results and discussion

3.1. Influence of homogenizing annealing on decomposition of γ-phase

The results of phase analyses of as-atomized and homogenized U-10 wt% Mo powder after annealing at 400 and 500 °C are presented in Table 1. The Rietveld refinement of each pattern using FULLPROF program(Version 3.2) converged to good agreement ($\chi^2 =$ 1.87–2.21). The analysis of the as-atomized sample showed that the sample crystallized mainly (~90%) in the cubic γ-phase but it contained a mixture of γ-phases with different Mo content. A small amount (<1%) of UO was observed in the neutron diffraction pattern. After homogenizing annealing at 800 °C for 10 h, the sample became uniform. These results confirm our previous studies of U–Mo alloy powders prepared by the centrifugal atomization method [5,6].

Table 1
Phase composition (wt%) of as-atomized and homogenized U-10 wt% Mo alloy after annealing at 500 °C

Sample	γ	U_2Mo	α	UO	UO_2
Atomized, 500 °C for 10 h	81.39	9.16	9.46		
Atomized, 500 °C for 50 h	38.93	41.50	19.56		
Homogenized sample (atomized, 800 °C for 10 h)	97.50			2.50	
Homogenized, 500 °C for 10 h	95.33	0.90	2.92	0.67	0.18
Homogenized, 500 °C for 50 h	79.52	11.99	8.06	0.19	0.23

Homogenizing annealing resulted in coarsening of grains from ~2 μm in as-atomized powder to ~25 μm in homogenized powder.

After annealing at 500 °C for 10 h, both the as-atomized and homogenized samples partially decomposed into α-U and U_2Mo phases. The degree of decomposition for the homogenized sample was lower than the as-atomized sample. This difference was distinctly observed after annealing at 500 °C for 50 h (see Table 1 and Fig. 1).

Only 38.93% of the metastable γ-phase remained in the as-atomized sample while 79.52% remained in the homogenized sample. It is natural to suppose that the U_2Mo phase is formed in the central part while the α-U phase is formed at the periphery of the grain. After 500 h annealing of the as-synthesized sample at 500 °C, the decomposition was completed and no γ-phase remained in the sample.

The decomposition of the γ phase into α-U and U_2Mo is connected with the redistribution of Mo in the original grain of the γ-phase. The regions depleted with molybdenum (α-U) and enriched with molybdenum (U_2Mo) are formed. This process is reversed with respect to the homogenization and therefore it is natural that homogenization retards the decomposition process. Coarsening of grains during homogenization also retards the process of decomposition.

3.2. U-10 wt% Mo dispersed fuel reaction with Al matrix

In Figs. 2 and 3, the back-scattering electron images of the as-atomized and homogenized U-10 wt% Mo dispersion fuel samples after annealing at 500 °C are shown.

One can see that the thickness of the fuel particle-Al interaction layer (dark-gray) increases with time. In the homogenized U-10 wt% Mo dispersion fuel sample, the interaction layer is thinner than in the as-atomized dis-

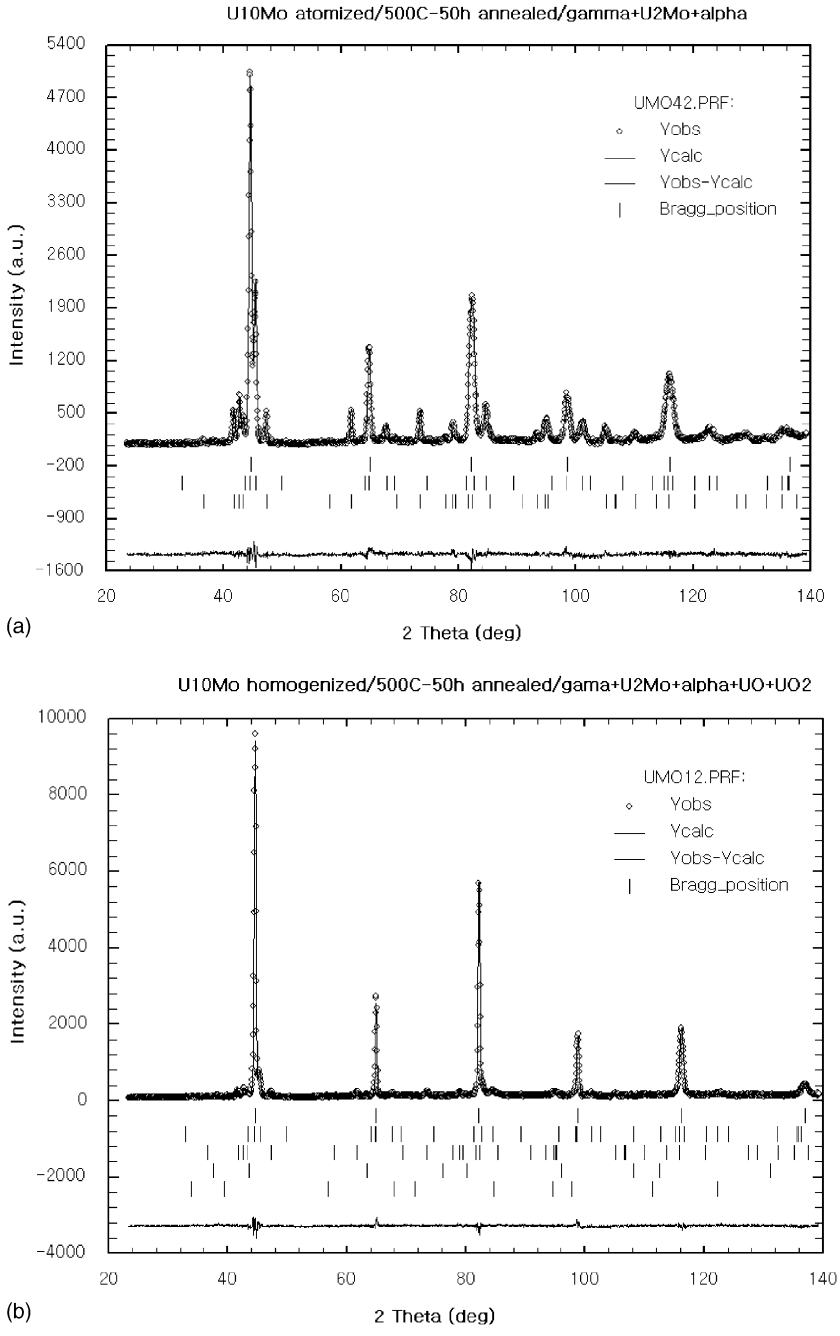

Fig. 1. Neutron diffraction pattern of as-atomized (a) and homogenized (b) U-10 wt% Mo alloy after annealing at 500 °C for 50 h.

persion fuel sample. After annealing at 500 °C for 500 h, the interaction layer covers all or the main part of a fuel grain (Fig. 3). In the as-atomized U-10 wt% Mo dispersion fuel sample, the grains entirely consisted of the product of fuel-Al interaction and were uniform dark-gray on the electron images. The phase composition of fuel rods with the as-atomized U-10 wt% Mo alloy dispersed in the Al matrix after annealing at 400 and 500 °C is shown in Table 2. It is determined by the analysis of neutron diffraction patterns.

It follows from Table 2 that the main process in U-10 wt% Mo dispersed fuel is decomposition of the γ-phase

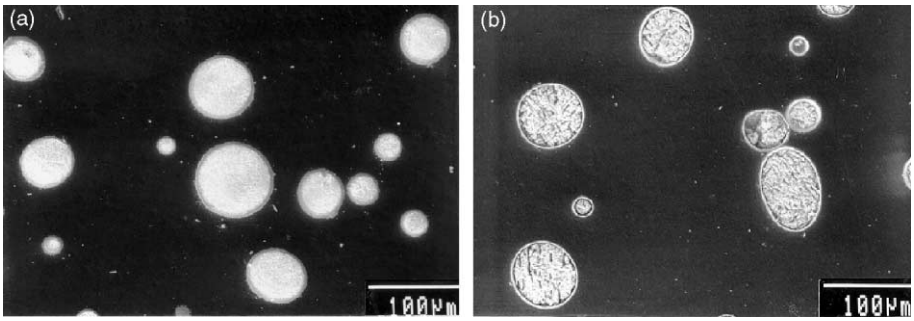

Fig. 2. Back-scattering electron images of the as-atomized (a) and homogenized (b) U-10 wt% Mo dispersion fuel samples after annealing at 500 °C for 10 h.

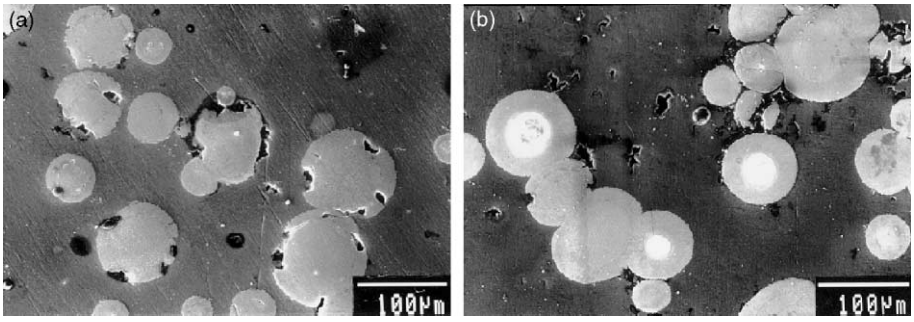

Fig. 3. Back-scattering electron images of the as-atomized (a) and homogenized (b) U-10 wt% Mo dispersion fuel samples after annealing at 500 °C for 500 h.

Table 2
Phase composition (wt%) of as-atomized U-10 wt% Mo alloy dispersed in Al matrix after annealing at 400 and 500 °C

Sample	γ	Al	U_2Mo	α	UAl_2	UAl_3	UO	UO_2
Not annealed	81.15	16.68					2.17	
400 °C for 500 h	10.19	15.96	31.80	40.84	0.72	0.48		
400 °C for 1000 h	10.05	14.38	31.85	41.65	0.65	1.42		
500 °C for 100 h	5.96	12.04	29.38	47.39	2.06	3.17		
500 °C for 500 h	1.67	13.80	27.89	49.88	1.11	5.42		0.23

into the α-U and U_2Mo phases. Decomposition increases with temperature and after annealing at 500 °C for 500 h is practically completed (see Table 2 and Fig. 4).

The Al matrix reacts with α-U and it results in the formation of Al_2U and Al_3U phases. Therefore, the interaction layer is a mixture of α-U, U_2Mo, Al_2U and Al_3U phases. Thus the reaction of U-10 wt% Mo fuel particle with Al matrix at 500 °C can be described as follows. At 500 °C a grain of metastable γ-phase decomposes into the grains of α-U and U_2Mo phases. Grains of the α-U phase are formed on the periphery while grains of the U_2Mo phase in the central part of the original grain of the γ phase. The Al diffuses along grain boundaries and forms grains of Al_2U and Al_3U phases by reaction with α-U grains. Thus the reaction of the Al matrix with a fuel particle is the reaction of diffused Al with α-U. We did not observe the formation of any ternary compound of the Al–U–Mo system.

4. Conclusions

Homogenizing annealing at temperatures above 560 °C of as-atomized U-10 wt% Mo alloy retards the process of the decomposition of the metastable γ-phase at 400 and 500 °C.

The reaction of the Al matrix with a dispersed atomized U-10 wt% Mo fuel particle is, in fact, the

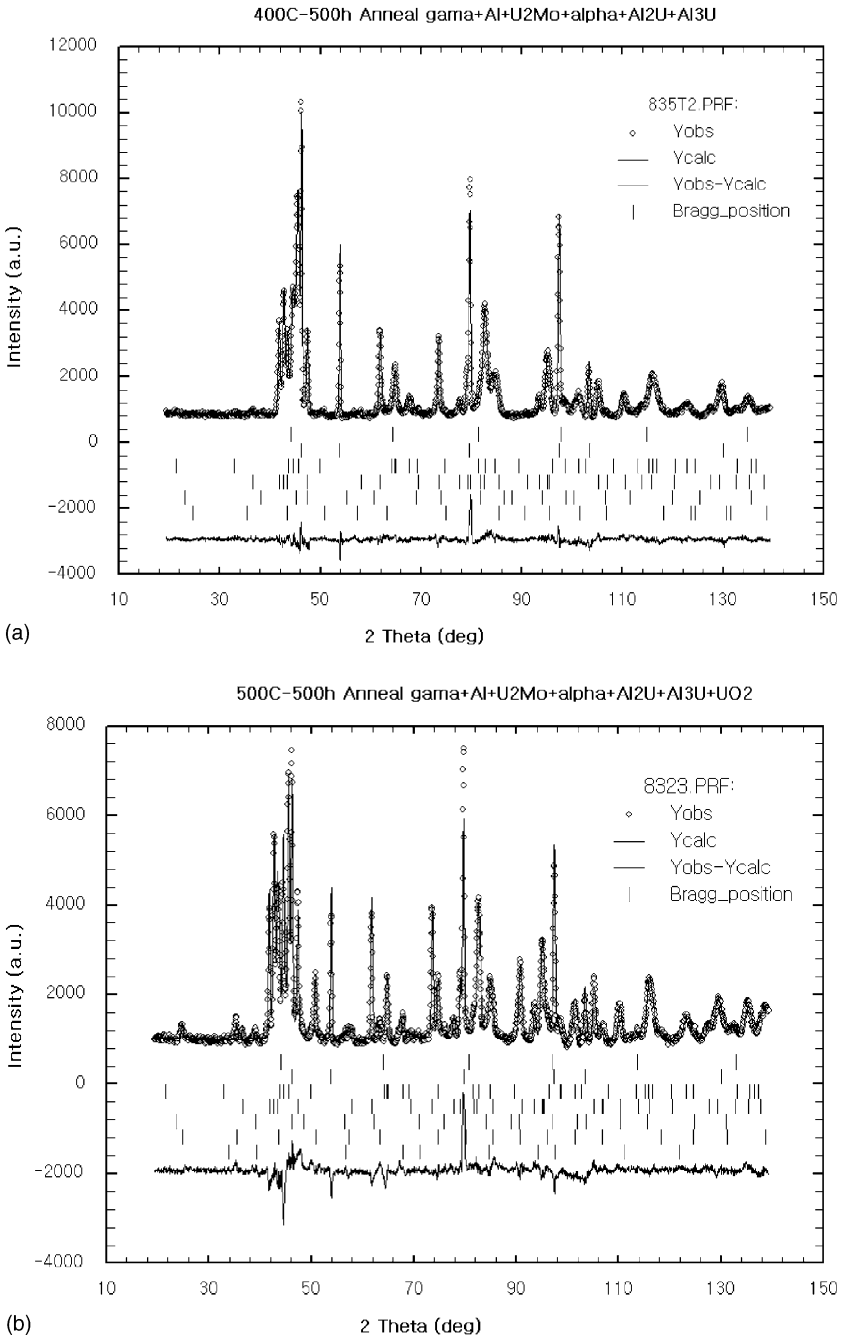

Fig. 4. Neutron diffraction pattern of as-atomized U-10 wt% Mo fuel dispersed in Al matrix after annealing at 400 °C for 500 h (a) and 500 °C for 500 h (b).

reaction of diffused Al with α-U formed from a decomposed γ-phase. It results in the formation of Al_3U and Al_2U phases. A single ternary Al–U–Mo compound was not observed as result of fuel-Al interaction.

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