

An Investigation on γ -U Phase Stability and Thermal Compatibility of Dispersion Fuel Meats Prepared with Atomized U-16at.%Mo, U-14at.%Mo-2at.%Ru and U-14at.%Mo-2at.%Os

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ABSTRACT

The γ -U phase stability and the thermal compatibility of atomized U-16at.%Mo and U-14at.%Mo-2at.%X(: Ru, Os) dispersion fuel meats at an elevated temperature have been investigated. The atomized U-14at.%Mo-2at.%Os powder appeared to have longer decomposition time than the atomized U-16at.%Mo and U-14at.%Mo-2at.%Ru powders. The inter-diffusion of atomized U-14at.%Mo-2at.%Os particles with Al matrix were shown to form thinner and more uniform layers than the other alloy particles. The Al atoms seems to diffuse more easily along the decomposed lamella inter-phases than the cell boundaries of γ phase. The addition of Os instead of Mo is expected to have a beneficial effect on increasing U-density of fuel due to better γ -U phase stability and thermal compatibility.

1. Introduction

Recently, high uranium-density dispersion fuel of low enriched uranium (LEU) has been developed to replace fuels of highly enriched uranium (HEU) for use in high performance research and test reactors [1-4]. The requiring U-density has been known to be up to 8~9 g-U/cm³ [5].

In general, two methods can be considered to obtain high uranium-density dispersion fuels. The first challenge is the discovery or development of uranium compounds or alloys having the highest possible uranium density that can be fabricated in dispersion, and has acceptable irradiation behavior. The second challenge is the achievement of maximum volume fraction of fuel particles in the fuel meats. Since earlier studies have shown the γ -U phase to behave rather stably under irradiation, uranium alloys that retain the γ -U phase are preferred [4]. One group of promising candidates is the uranium-molybdenum alloys, which have a relatively large range of γ -U phase that is retained in a metastable state at the temperature at which the fuel is fabricated and irradiated [6].

Several transition metals, particularly 4d and 5d elements in Group IV to VIII, form solid solutions with γ -U, and this cubic phase can be retained in its metastable state upon cooling. The γ stabilizing power of these elements increases with atomic number as d-electrons participate in bonding through hybridization with s and p atomic orbitals.

However, their solubility decreases as the size difference with uranium atoms becomes larger, and the increased bond strength promotes intermetallic compound formation. It was reported that small additions of a third element such as Pt have been shown to significantly retard phase decompositions in U-Mo [7]. It was further found that a small amount of elements to the right of Mo in the periodic table had a powerful stabilizing effect when added to U-Mo alloys [8,9]. The results of the previous paper mentioned that Ru, Pd, Os and Pt have a positive effect relative to Mo.

Therefore, in this study, the γ -phase stability and the thermal compatibility of dispersion fuel meats prepared with atomized U-16at.%Mo, U-14at.%Mo-2at.%Ru and U-14at.%Mo-2at.%Os were examined by X-ray diffraction analysis (XRD), scanning electron microscopy (SEM) and dispersive spectrometry X-ray analysis (EDX) after isothermal annealing at an elevated temperature.

2. Experimental procedure

Three kinds of fuel powders, which are U-16at.%Mo(U-Mo), U-14at.%Mo-2at.%Ru(U-Mo-Ru) and U-14at.%Mo-2at.%Os(U-Mo-Os), were atomized using depleted uranium lump (99.9 %), molybdenum (99.7 %), ruthenium (99.9 %) and osmium(99.9 %). The alloying elements were induction-melted in high-temperature-resistant ceramic crucibles. The molten metal was heated to approximately 200 °C higher than the melting point and was fed through a small nozzle onto a rotating disk. Alloy melt droplets, which are formed from melt by centrifugal force, were spread from the disk toward the atomizing chamber wall by flowing argon gas during flying.

Fuel meat rods, consisting of 45vol.% fuel powder in an aluminum matrix, were prepared by extruding the compacts, which were prepared by pressing the blended powders at the temperature of 400°C and the pressure of 65 kgf/cm². These extruded specimens of 25mm length and 6.5mm diameter were cut, vacuum-sealed in quartz tubes and annealed for various times such as 0.5, 1, 5, 20, 100 and 500 hours at 500°C for 500 hours.

The micro-structural examinations for the fuel meats were carried out by scanning electron microscopy (SEM). Phase analysis was done by X-ray diffraction and energy dispersive spectrometry X-ray analysis (EDX).

3. Results and Discussion

3. 1. γ -U phase stability

Fig. 1 shows SEM micro-graphs and X-ray diffraction patterns of as-atomized U-Mo powder, U-Mo-Ru powder and U-Mo-Os powder. All as-atomized powders appeared to have the fine cell structure with the size of 2 ~5 μ m due to the rapid cooling effect of centrifugal atomization. The Mo content of cell boundaries was analyzed to be lower than that of the cell interior. This is a result of segregation when the alloy is cooled through the solidus-liquidus line gap [10]. A slight gradient of Mo concentration in the adjacent region of inter-phase seems to induce a slight difference of Mo content between them.

The results of X-ray diffraction patterns are confirming that all as-atomized powders have body-centered-cubic γ phase, irrespective of powder composition. The atomized powder, rapidly solidified from the melt, appeared to retain a gamma phase as a meta-stable state.

Fig. 2 shows the scanning electron microscopy (SEM) images of the 45vol.% fuel meats of U-Mo, U-Mo-Ru and U-Mo-Os after isothermal annealing at 500°C for 0.5 hour. The microstructures of three kinds of fuel particles were examined to maintain cell structure without any change. In the case of isothermal annealing at 500°C for 1hour, a small change, which is a slight decomposition of γ phase into lamellar structure, took place in U-Mo-Ru fuel meat as shown in Fig 3(b). However, fuel particles of U-Mo and U-Mo-Os were still retained as the cell structure as shown in Fig. 3(a) and 3(c). The decomposition of γ phase was formed to initiate at the cell boundary. This phenomenon is assumed to happen due to the lower Mo content of the cell boundary.

Fig. 4 is SEM micro-graphs of fuel particles when the isothermal annealing time was extended to 5 hours. The decomposition of γ phase in U-Mo particles was observed to initiate at the cell boundaries. In U-Mo-Ru particles a considerable amount of γ phase was revealed to be decomposed to lamellar structure. However, fuel particles of U-Mo-Os appeared to maintain the initial γ phase.

After isothermal annealing the three kinds of fuel meats at 500°C for 20 hours, micro-graphs were obtained as shown in Fig. 5. After this time even in U-Mo-Os particles, a decomposition of γ phase was observed to occur. Two kinds of particles, U-Mo and U-Mo-Ru, were revealed to have decomposed almost completely. The micro-graphs of fuel meats after isothermal annealing at 500°C for 100 hours are shown as in Figure 6. All of U-Mo, U-Mo-Ru and U-Mo-Os were entirely decomposed into lamellar structure. Table 1 shows the decomposition times of the fuel meats at elevated temperatures. From these results, it appears that U-Mo-Os particles have the highest stability of γ phase among the three kinds of fuel powders.

3. 2. Thermal compatibility

Scanning electron microscopy (SEM) images of the 45vol.% fuel meats with U-Mo, U-Mo-Ru and U-Mo-Os after isothermal annealing at 500°C for 0.5h are shown in Fig. 7. U-Mo-Ru particles appeared to react with Al matrix leaving interaction layers and deeply reacted local regions, which protruded into the matrix area. In U-Mo particles very thin interaction layers or traces were observed at the interface. But U-Mo-Os particles seem to have not reacted with Al matrix. After isothermal annealing at 500°C for 1hour, U-Mo-Os particles were observed to have still no interaction layer formation. U-Mo and U-Mo-Ru particles revealed to form thicker interaction layers and larger intensively reacted local regions. The number of the intensively reacted regions seems to increase a little with increasing annealing time. After isothermal annealing at 500 °C for 5hour, the U-Mo-Os particles were shown to form interaction layers with Al matrix. The other particles formed thicker layers and larger intensively reacted local regions at this longer time as shown in Fig. 8.

Fig. 9 shows micro-graphs of fuel meats after annealing at 500°C for 20 hours. The interaction layer of U-Mo-Ru particles appeared to have increased considerably and in very non-uniform. Intensively reacted local regions seem to have grown in both directions toward the center of the particles and matrix area. Some interaction regions were separated into several particles from the unreacted parts. Some small U-Mo-Ru particles reacted with Al matrix entirely. In the U-Mo dispersion fuel meat relatively thin interaction layers were found at the interfaces of most particles with matrix. But some particles were observed to have intensively reacted local regions. As shown in Fig. 9 U-Mo-Os particles formed relatively thin uniform interaction layers. These kinds of interaction layers have

been known to form by inter-diffusion of matrix Al and atoms of fuel particles. Among these atoms, Al is considered to have been the highest diffusivity due to lightest atomic mass. Al diffusion in fuel particles occurs mainly along defects such as grain boundaries. It was reported that Al diffusion along cell boundaries of γ phase in U-10% Mo particles was relatively slow. Three kinds of fuel particles, which were isothermal annealed, would have different fractions of decomposed phases. According to the above mentioned results, U-Mo-Os have the most γ phase stability. Whereas U-Mo-Ru has less γ phase stability than U-Mo and U-Mo-Os. The thermal compatibility of fuel meats seems to have a relation to the γ phase stability. U-Mo-Os fuel meat with the most γ phase stability was shown to have the best thermal compatibility with the thinnest uniform interaction layers but the U-Mo-Ru fuel meat with the least γ phase stability revealed to have the lowest thermal compatibility with the thickest interaction layers and intensively reacted local regions. Generally the fuel particles with lower γ phase stability would contain a larger volume fraction of the decomposed lamellar structure. Accordingly Al atoms are assumed to diffuse more easily along decomposed phase boundaries than γ phase cell boundaries.

The area scanning analysis of annealed U-Mo fuel meat annealed, using energy dispersive X-ray spectroscopy (EDX), indicated that unreacted regions were composed of 87.6at.%U and 12.4at% Mo, whereas reacted layer consisted of 27.6at.%U, 3.8at% Mo and 68.6at.%Al that is a composition equal to (U, Mo)Al₃.

After sufficient annealing at 500C for 500 hours, the variation of interaction layers is shown in Fig. 10. The U-Mo-Os fuel meat shows a distinctly more uniform and thinner interaction layer than the other fuel meats. U-Mo and U-Mo-Ru fuel meats have intensively reacted local regions increasing in frequency with increasing annealing time. Hence, it seems that the U-Mo-Os fuel meat has the highest thermal compatibility among the three kinds of fuel meats. The U-Mo -Os dispersion fuel would be a promising candidate from the view point of increasing U-density by replacing Mo with Os.

4. Conclusion

From the isothermal annealing tests of three kinds of fuel meats, which are U-Mo, U-Mo-Ru and U-Mo-Os fuel meats, the following conclusions could be obtained.

- U-Mo-Os has better γ -U phase stability than U-Mo and U-Mo-Ru. U-Mo-Ru has lower γ -U phase stability than U-Mo.
- U-Mo-Os fuel meat has the best thermal compatibility in the three kinds of fuel meats.
- The thermal compatibility of fuel meats appears to be related to the γ phase stability. Fuel meat with more γ phase stability was shown to have better thermal compatibility forming thinner uniform interaction layers.
- Aluminum atoms are assumed to diffuse more easily along decomposed phase boundaries than γ phase cell boundaries.

The U-Mo -Os dispersion fuel would be a promising candidate from the view point of increasing U-density by replacing Mo with Os.

Acknowledgments

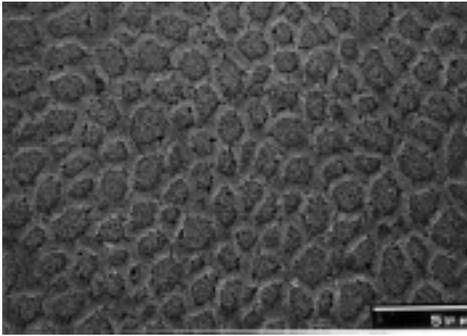
This project has been performed under the Nuclear R&D supported by the Ministry of Science and Technology, Korea.

Reference

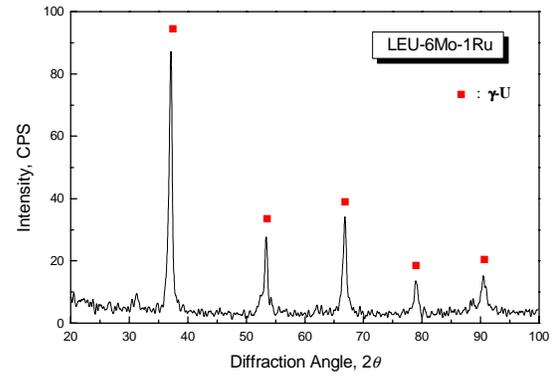
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| | 0.5h | 1h | 5h | 20h | 100h |
|----------------|-------------|-----------------------|-----------------------|-----------------------|-----------------|
| U-Mo | Cell | Cell | Cell+ lamellar | lamellar | Lamellar |
| U-Mo-Ru | Cell | Cell+ lamellar | lamellar | lamellar | Lamellar |
| U-Mo-Os | Cell | Cell | Cell | Cell+ lamellar | Lamellar |

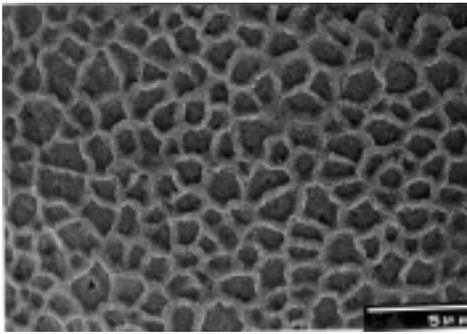
Table. 1. The decomposition time of the fuel meat at an elevated temperature.



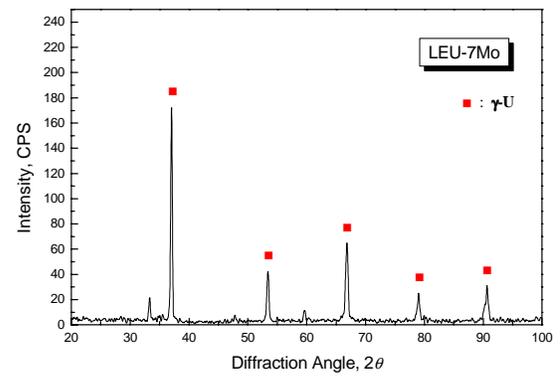
(a)



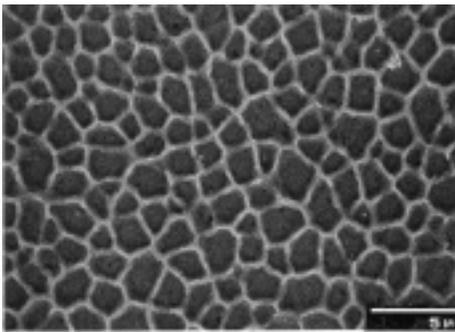
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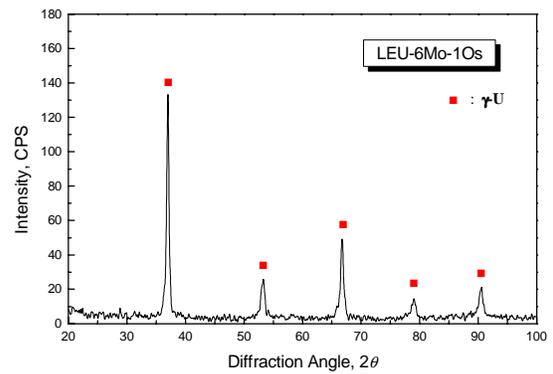
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(e)

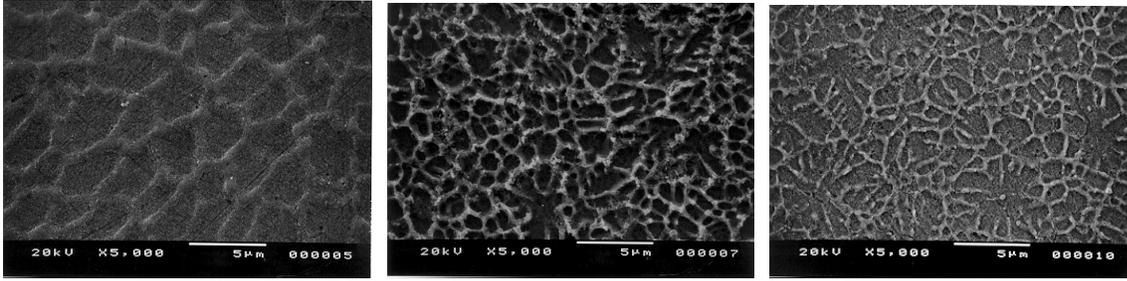


(c)



(f)

Fig. 1. The SEM micrographs and the X-ray diffraction patterns of as-atomized U-Mo, U-Mo-Ru and U-Mo-Os powders; (a),(d) U-Mo, (b),(e) U-Mo-Ru and (c), (f) U-Mo-Os powder.

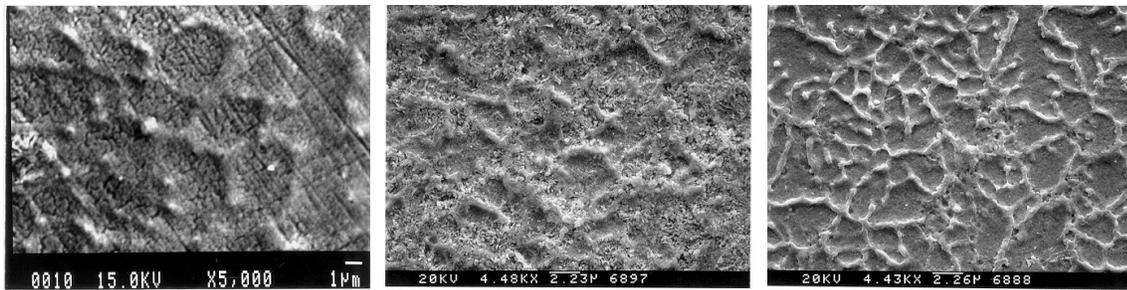


(a) U-16at.%Mo

(b) U-14at.%Mo-2at.%Ru

(c) U-14at.%Mo-2at.%Os

Fig. 2. The scanning electron micrograph of the fuel meats after annealing at 500°C for 0.5 hour.(x5000)

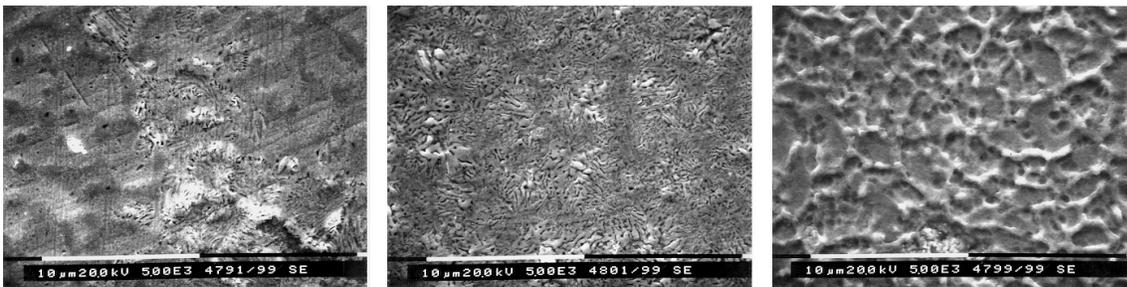


(a) U-16at.%Mo

(b) U-14at.%Mo-2at.%Ru

(c) U-14at.%Mo-2at.%Os

Fig. 3. The scanning electron micrograph of the fuel meats after annealing at 500°C for 1 hour.(x5000)



(a) U-16at.% Mo

(b) U-14at.%Mo-2at.%Ru

(c) U-14at.%Mo-2at.%Os

Fig. 4. The scanning electron micrograph of the fuel meats after annealing at 500°C for 5 hours.(x5000)

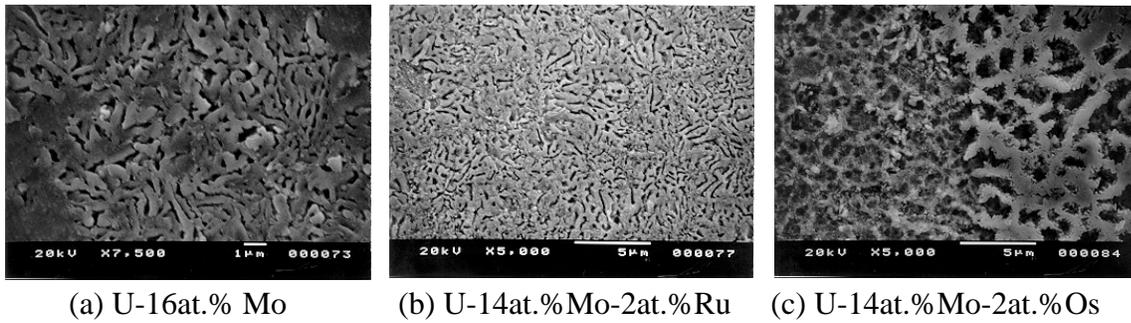


Fig. 5. The scanning electron micrograph of the fuel meats after annealing at 500°C for 20 hours.(x5000)

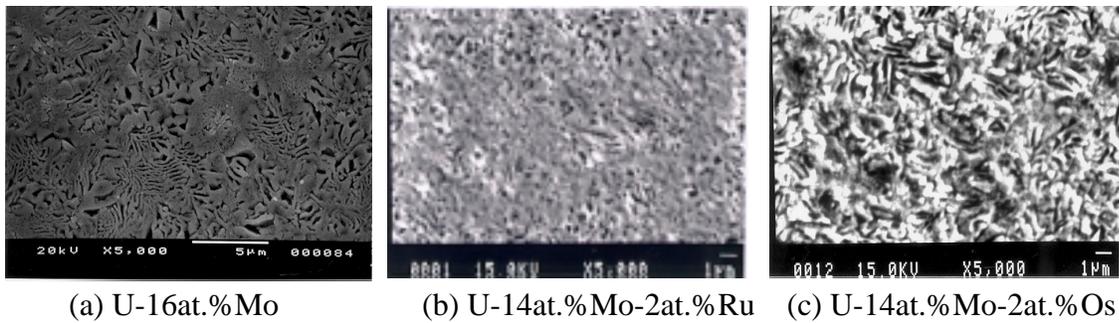


Fig. 6. The scanning electron micrograph of the fuel meats after annealing at 500°C for 100 hours.(x5000)

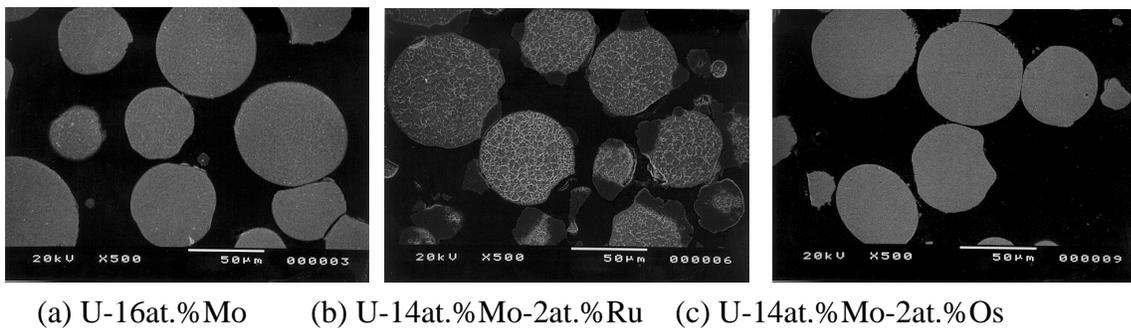


Fig. 7. The scanning electron micrograph of the fuel meats after annealing at 500°C for 0.5 hour.(x500)

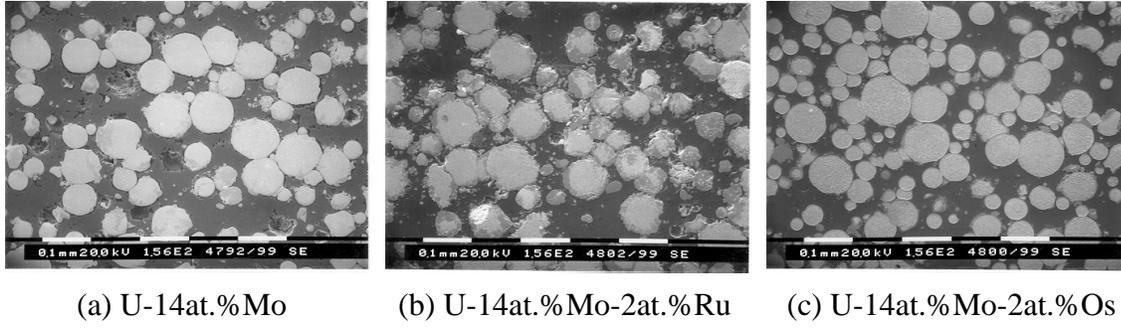


Fig. 8. The scanning electron micrograph of the fuel meats after annealing at 500°C for 5 hour.(x150)

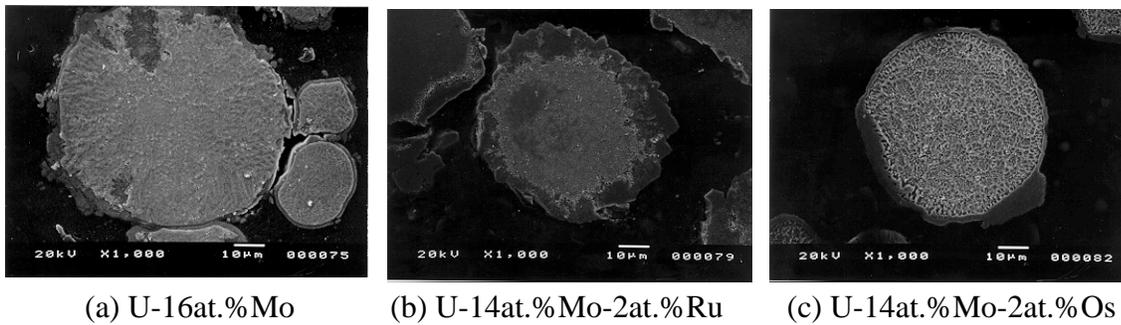


Fig. 9. The scanning electron micrograph of the fuel meats after annealing at 500°C for 20 hours.(x1000)

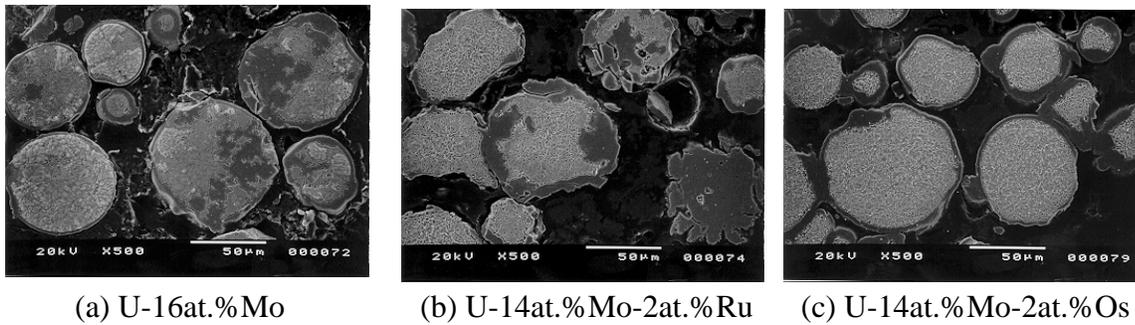


Fig. 10. The scanning electron micrograph of the fuel meats after annealing at 500°C for 500 hours.(x500)