ENHANCED RADIAL THERMAL CONDUCTIVITY OF UO2 FUEL PELLETS WITH MOLYBDENUM MICROPLATES

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ABSTRACT

It is important to enhance the thermal conductivity of UO_2 fuel pellets, and which can lead to reduce fission product release, and to increase safety and operation margin of the fuels. In this study, metallic micro-sized molybdenum plates(microplate) were dispersed in UO_2 pellet and horizontally aligned for enhancing the thermal conductivity efficiently in radial direction. The aligned Mo microplates work as heat conducting paths in the fuel pellet, enhancing the thermal conductivity in radial direction, doubled as that of an UO_2 pellet, in the case of a pellet with 5 vol.% Mo microplates at 1000^oC. The thermal properties and microstructures of the pellets with the shape of Mo microplates included were investigated.

1. Introduction

Through the decades of developments of nuclear fuel pellets, many of efforts have been focused on increasing the economic efficiency of the LWR power generation such as, increasing the fuel discharged burn-up, extending the fuel cycle, and up-rating the maximum power. However, in the wake of Fukushima accident, it becomes more important recently, and well-known that the current LWR fuel should be tolerable to severe accidents to mitigate their consequence with maintaining the performances. Thus, various concepts of new fuels are being suggested and developed under the name of accident tolerant fuels (ATF).

One of the current issues for nuclear UO_2 fuel pellet is about its low thermal conductivity. The low thermal conductivity leads to increase thermal gradient in the fuel pellet and centerline temperature when in operation. Enhancing the thermal conductivity of UO_2 fuel pellet is greatly attractive in the aspect of fuel performance [1–3] and also for its safety margin. The fuel pellets having high thermal conductivity can lower fuel temperature and reduce the mobility of the fission gases [4–6]. In addition, a reduced temperature gradient within the pellet probably enhances the dimensional stability, with lower thermal stress of the fuel pellet, thus the pellet cladding mechanical interaction (PCMI) and even in fuel fragmentation, relocation and dispersal (FFRD) can be mitigated. A thermal margin gained from the high thermal conductivity of pellet would be utilized in a safe operation of LWR. There have been efforts on enhancing the thermal conductivity of the fuel pellet. One of the methods is introducing high thermal conductive materials into fuel pellets. Yang et al. [7] have shown experimentally that the thermal conductivity of a UO_2 pellet can be increased substantially by providing a UO_2 pellet with connected tungsten channel. KAERI has also developed micro-cell UO_2 fuel pellets consist of granules enveloped by thin metallic cell walls [8-10]. The metallic cell walls in pellets are continuously connected to each other, enhancing thermal conductivity.

In this study, to enhance the thermal conductivity in radial direction, molybdenum metal microplate was dispersed in a UO_2 fuel pellet. Micrometer-sized thin Mo plates were prepared and aligned horizontally in a UO_2 pellet to have enhanced thermal conductivity by an aid of heat transfer paths in radial direction. Moreover, the compatibility in the fuel fabrication process can be enhanced, due to the simple pellet fabrication method. The thermal properties of the pellets were characterized with the microstructures of the fuel composite.

2. Experimental

2.1 Mo Microplate Preparation

Mo microplate UO_2 pellets were fabricated by composing UO_2 powder and Mo microplate powders. Mo microplate powders were prepared by milling spherical Mo powder particles. Mo metal powder (SIGMA-ALDRICH, 99.9%) was milled in a planetary milling machine with optimizing conditions to have thin and wide plate shapes. Three kinds of Mo particle size powders were used in this study, 1-5, 3-7 and 10 micrometers; the sample were labeled as MP1, MP3 and MP10, respectively. The microplate powder samples were characterized with SEM analysis, the thickness and the diameter were measured and also the aspect ratio of samples were calculated.

2.2 Fuel Pellet Fabrication

The three kind of UO_2-Mo composite fuel pellets were fabricated with three different microplate samples MP1, MP3 and MP10. 5 vol.% of Mo microplate powder was simply mixed in a tubular mixer with UO_2 powder. The powder mixture was compacted using a uniaxial press at about 300 MPa, and the pelletized green body was sintered at 1730 ^{o}C for 4hrs in a flowing H_2 atmosphere. Two kinds of pellets were also prepared for comparison, one was bare UO_2 pellet sample and the other was UO_2 pellet with 5 vol.% Mo spherical particles, of which same fabrication method was used as others.
The sintered densities of Mo microplate pellets were determined using an immersion method. The microstructure of the sintered pellet was observed using optical microscopy and SEM to specify the dispersion and alignment of the microplates in the UO_2 matrix and structural integrity.

2.3 Thermal Conductivity Analysis

Thermal conductivities of the pellets were characterized by LFA method. The thermal diffusivity of a sample pellet was measured by a laser flash apparatus(LFA427, Netzsch), and the thermal conductivity of the sample was calculated with measured values of density, specific heat capacity and the thickness of the sample. In order to measure the effective radial thermal conductivity, the sample was sliced in axial direction with having thin and parallel surfaces. The atmosphere and temperature range of measuring conditions were Ar gas and 200-1200^{o}C, respectively. The thermal conductivities were measured then compared.

3. Result and Discussion

Fig. 1 shows Mo microplates prepared in this study. After the milling process, spherical Mo particles were successfully transformed to microplates; the shape of the microplates were having 0.1~1 micrometer of thin thickness and 5-50 micrometer of wide circular surfaces.

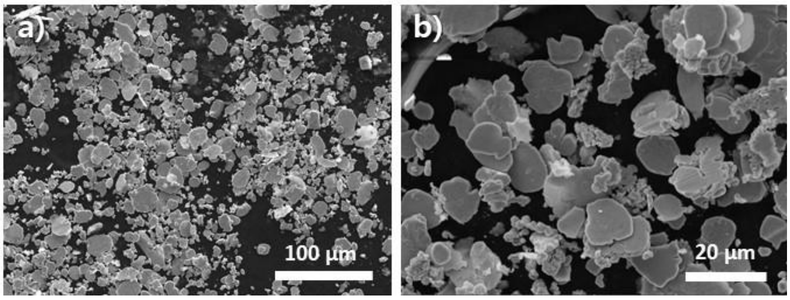

Fig. 1. SEM images of Mo microplates(MP3) with different magnifications. a)500x and b)2000x

Fig. 2 shows three different microplates with varying size distribution. As increasing the particle size of raw powder, the diameter of microplate was increased. Meanwhile, the thickness of the microplates was not changed much, maintained in the range of 0.1-1 micrometer, the aspect ratio of diameter/thickness of the microplates were increased with the raw particle sizes.

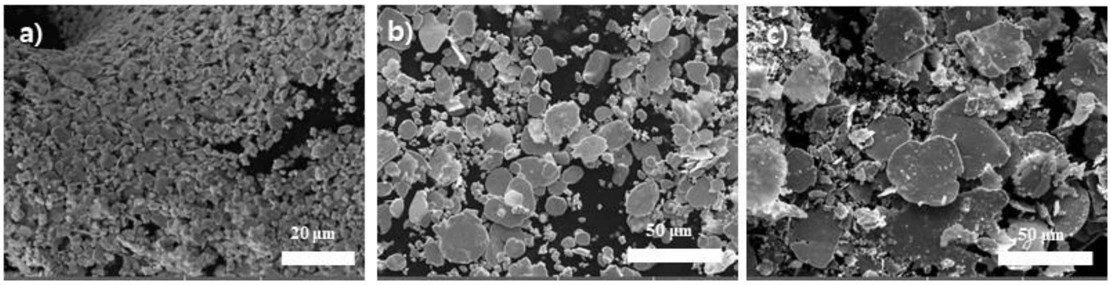

Fig. 2. SEM images of Mo microplates with different size distributions. a)MP1, b)MP3 and b)MP10

Fig. 3 shows the microstructure of a Mo microplate UO_2 pellet. The Mo microplates of bright phase were dispersed homogeneously and aligned in horizontal direction in UO_2 pellet. The structural degradation such as micro-cracks and interfacial gaps could not be found in of UO_2 matrix with addition of Mo microplates.

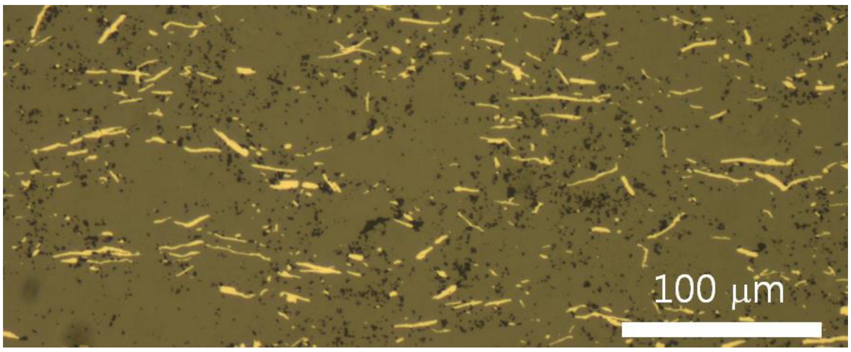

Fig. 3. Microstructure of a Mo microplate UO_2 pellet. (MP10)

Fig. 4 shows the microstructures of UO_2 pellets with varying Mo particles. The sintered densities of pellets were varied in 96.5-97.5% of relative density. Fig. 4a is the microstructure of UO_2 pellet with Mo spherical particle powder and Figs. 4b-d are the microstructures of UO_2 pellet with Mo microplates, MP1, MP3 and MP10 respectively. It was observed that the use of larger particles, i. e. microplates with larger aspect ratios, results in better lateral continuity in the UO_2 matrix in a pellet, which is forming effective thermal conductive paths for radial heat transfer.

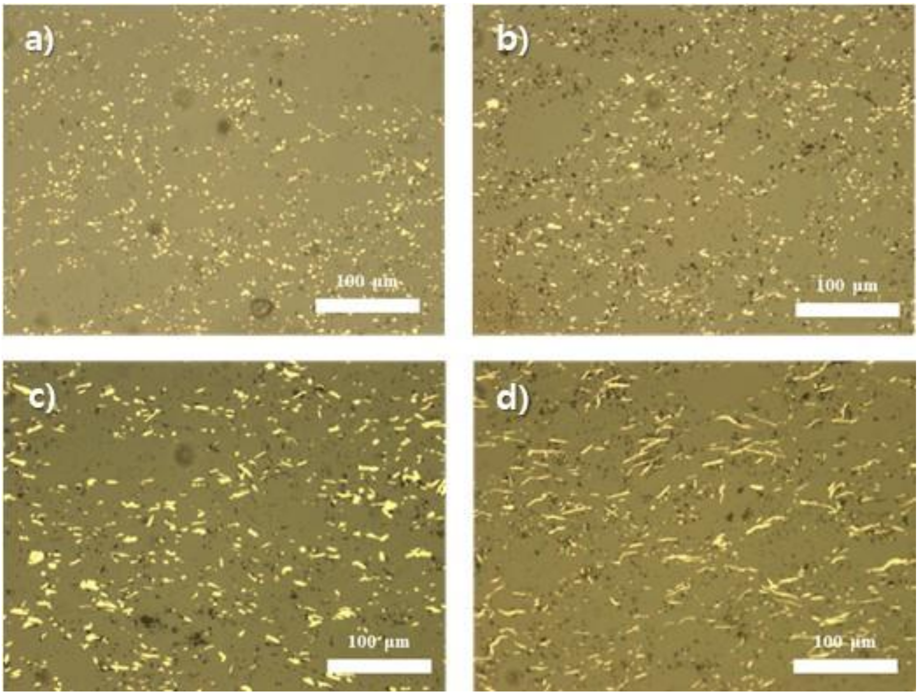

Fig. 4. Microstructure of UO_2 pellets with 5 vol.% Mo metal particles. a) spherical particle, b)MP1 microplate, c)MP3 microplate and d)MP10 microplate

Fig. 5 shows the measured radial thermal conductivities of UO_2 pellets with 5 vol.% Mo metal particles at 1000°C. The radial thermal conductivity was increased as increasing the aspect ratio of microplates, and it is much enhanced compared with bare UO_2, and even also higher than the conductivity of the UO_2 pellet with same amount of spherical Mo particles included. This enhancement of the thermal conductivity of the Mo microplate UO_2 pellet was mainly affected by the shape and arrangement of the metallic particles in the pellet, especially related with the aspect ratio of the microplates. The high thermal conductivity of microplate fuel pellet is comparable to that of the microcell fuel pellet developed in KAERI, the conductivity is about 10 % less than the microcell pellet due to the connectivity of metal microplates. It is considered that the thermal conductivity of microplate pellet can be increased if the size and shape of Mo microplate particle is optimized.

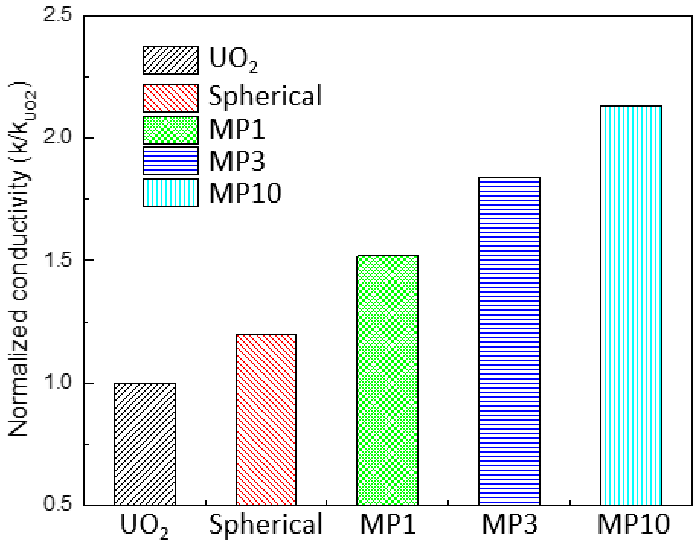

Fig. 5. Comparison of thermal conductivities of UO_2 fuel pellets at 1000°C. The thermal conductivity of UO_2 at 1000°C is 2.79 $Wm^{-1}K^{-1}$

4. Summary

In this study, Mo microplate UO_2 nuclear fuel pellet was fabricated for enhancing the thermal conductivity of the pellet. Mo metal microplates were aligned working as heat conducting paths in the pellet and the microplates with higher aspect ratio enables better horizontal continuity in the UO_2 pellet. Therefore, the thermal conductivity of the UO_2 pellet in radial direction could be enhanced, which can lead to reduce thermal gradient of the pellet when in operation in a reactor. Moreover, the pellet could be achieved by simple UO_2 pellet fabrication method to concern the compatibility with a conventional UO_2 pellet fabrication process. Considering the outstanding fuel pellet characteristics, this Mo microplate UO_2 pellet will be one of the promising fuel concepts of ATF pellets in near future.

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