Effect of Molybdenum Melting on the UO_2-Mo Composite Pellet Integrity

Jae Ho Yang*, Dong Seok Kim, Heung Soo Lee, Dong-Joo Kim, Ji-Hae Yoon
KAERI, 111 Daedeok-daero 989beon-gil, Yuseong-gu, Daejeon, 34057, Korea
* Corresponding author: yangjh@kaeri.re.kr

1. Introduction

UO_2–Mo micro-plate or microcell pellets, in which small amounts of molybdenum metal particles are arranged in the form of directionally aligned plates or continuous networks, are actively studied worldwide [1-5] as a promising accident tolerant fuel candidate for light water reactors, because they have improved thermal conductivity. This fuel design might be easily implemented in nuclear power plants in the near term, as it inherits the superior safety performance of UO_2 nuclear fuel and can leverage existing knowledge and infrastructure.

Since the melting temperature of molybdenum is about 200 K lower than that of UO_2, there is a concern that preferential melting of molybdenum in the event of power excess may reduce the thermal safety margin of the fuel system.

The aim of this study is to investigate experimentally the change in the dimension and microstructure of UO_2–Mo pellets when exposed to high-temperature where the Mo melts. The impact of Mo melting on the integrity of UO_2–Mo pellets in the event of an abnormal power transient or accident is discussed. Our out-of-pile assessments are expected to contribute to the evaluation of this material for use as an accident-tolerant fuel.

2. Experimentals

The UO_2-3vol% Mo micro-plate composite pellets for high temperature annealing test were prepared by conventional sintering process. The pellet dimension is similar to that of nuclear fuel pellet.

Annealing tests were performed in high-frequency induction heating apparatus at various temperatures. The two pellet samples were placed in a tungsten crucible and then heated to the target temperature in an Ar atmosphere at a heating rate of ~80 - ~120 K/min. All the samples were annealed for ~10 min. After annealing, the samples were cooled to room temperature in an Ar atmosphere. Temperature change of the samples during the annealing test was measured at the pellet surface using a pyrometer.

The morphology changes of the samples after annealing test were characterized by SEM (Tescan Vega3) with an attached EDS system (Oxford Instruments, Inca X-act). During the SEM investigation, EDS was used for elemental analysis using the analysis procedure provided in the AZtec software (Oxford Instruments).

3. Results

Fig. 1 (a) shows a SEM image of polished surface of the as-fabricated pellet. Bright matrix is UO_2 and darker inclusions are Mo plate. The density of as-fabricated pellet was measured to be 97.4%TD. A number of pores are observed around the Mo plates and UO_2 matrix. Figs. 1(b), 1(c) and 1(d) show the microstructures change after the annealing tests under the temperature profiles of Fig. 2(a), 2(b) and 2(c), respectively. About 1% of TD increase was observed for the both pellets annealed at 2530 and 2670 °C. In the case of the pellet heat-treated at 2730 °C, the density was increased about 1.5 %. Figs. 1(b)-1(d) obviously indicate the coarsening of pores and decrease of pore density. Therefore, the observed density increase is mainly due to the densification of UO_2 matrix during the annealing. The morphology change of Mo plates in the pellet shown in Fig. 1(d), which was annealed at 100 K higher than melting temperature of Mo, reveals that Mo was melted.

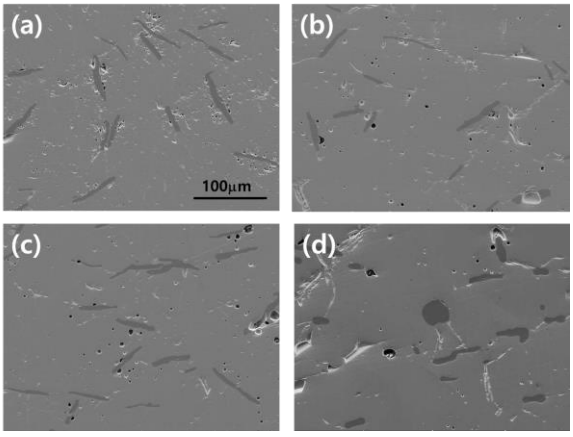

Fig. 1. SEM microstructures of UO_2-Mo pellets; (a) as-fabricate, and annealed at (b) 2530°C, (c) 2670°C, and (d) 2730°C

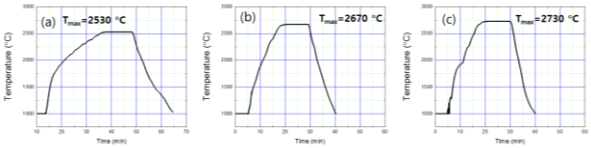

Fig. 2. Annealing temperature profiles. (a) T_{max}=2530°C, (b) T_{max}=2670°C, (c) T_{max}=2730°C

Fig. 3 shows the elemental mapping images of pellet annealed at 2730°C. SEM images and EDS analysis reveal the sharp UO_2/Mo interface and oxygen-free Mo

particle, indicating the absence of noticeable chemical interactions between UO_2 and molted Mo. Observed chemical stability is consistent with thermodynamic calculations [5].

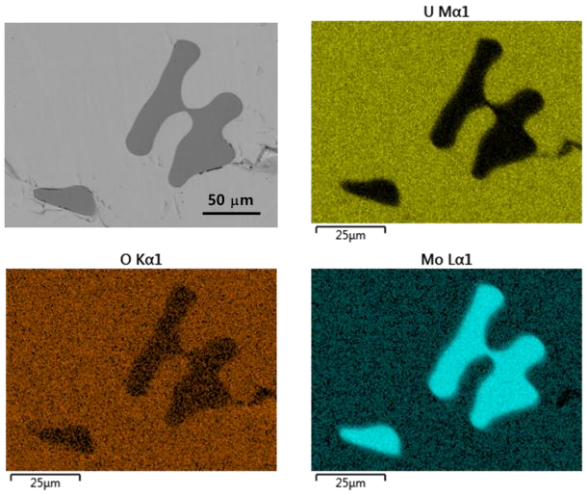

Fig. 3. SEM and EDS elemental mapping images at the selected area of sample annealed at 2730°C

When the UO_2-Mo pellet reaches the temperature where Mo melts, volume expansion owing to the phase transition of Mo from solid to liquid is expected to cause local stress build up, which can initiate crack formation. Solidification of molten Mo may leave pores or gap between Mo and UO_2 that may increase fuel volume and change thermo-physical properties. Contrary to our concerns, the experimental results showed that the density of pellets increased and the formation of cracks, pores and gaps around Mo inclusions were insignificant, even in the sample where Mo has melted. At the high temperature above 2,500 °C, UO_2 can easily deform. The stress generated by the melting or solidification of Mo can be sufficiently relieved by the plastic deformation of UO_2. In addition, mass transportation rate of both UO_2 and Mo will be fast enough to heal the any unstable void. The pore or gap formed at UO_2/Mo interface owing to the phase transformation of Mo seems to be disappeared by the rapid densification.

If UO_2 and Mo chemically react to form low density and low melting phases, the volume of the pellet may increase and the thermal safety may degrade. However, the test results showed that UO_2 is chemically stable with liquid Mo as well as solid Mo. Another important issue is the transport behavior of molten Mo. If molten Mo can easily relocate along the crack or gaps by gravity or temperature gradient, the impact of Mo melting on fuel performance and safety becomes more significant. Fig. 1(d) and Fig.3 (a) reveal that molten molybdenum tends to become spherical or combine with neighbor Mo to reduce interface energy. Nevertheless, the Mo transport through UO_2 matrix was not observed and the relocation of Mo is confined to limited space.

3. Conclusions

Regarding the fuel safety performance of the UO_2-Mo micro-plate pellet system, there are concerns that preferential melting of molybdenum in the event of power excess may reduce the thermal safety margin of the fuel system. Contrary to concerns, annealing experiments on pellets at high temperatures where Mo melts showed no microstructural degradation of pellet, such as formation of internal cracks, chemical reactions, or relocation of molten Mo. The shape and dimensions of the pellet were also maintained without significant changes. Therefore, the impact of Mo melting on the structural stability in UO_2-Mo system seems to be insignificant. However, since the plate shape of Mo was changed, the thermal conductivity will be varied.

Acknowledgments

Funding: This work was supported by the National Research Foundation funded by Ministry of Science and ICT of Korea (grant number 2017M2A8A5015056).

REFERENCES

[1] Dong Seok Kim, Keon Sik Kim, Jong Hun Kim, Dong-Joo Kim, Jae Ho Yang, Jang-Soo Oh, Yang-Hyun Koo, (2017) Aligning Mo Metal Strips in UO_2 Fuel Pellets for Enhancing Radial Thermal Conductivity, Water Reactor Fuel Performance Meeting at: Jeju, Korea
[2] S.C. Finkeldei, J.O. Kiggans, R.D. Hunt, A.T. Nelson, K.A. Terrani, (2019) Fabrication of UO_2-Mo composite fuel with enhanced thermal conductivity from sol–gel feedstock, J. Nucl. Mater. 520, 56-64
[3] Liang Cheng, Rui Gao, Qingdong Xu, Zhenliang Yang, Fangfang Li, Bingqing Li, Yun Wang, Xiangdong Liu, Pengcheng Zhang, Biaojie Yan, (2020) UO_2−Mo−Be composites for Accident Tolerant Fuel: SPS fabrication, microcracks-free in as-fabricated state and superior thermal conductivity, Ceram. Int. 46, 28939-28948, https://doi.org/10.1016/j.ceramint.2020.08.064
[4] J. Buckley, J.D. Turner, T.J. Abram, (2019) Uranium dioxide - Molybdenum composite fuel pellets with enhanced thermal conductivity manufactured via spark plasma sintering, J. Nucl. Mater. 523, 360-368, https://doi.org/10.1016/j.jnucmat.2019.05.059
[5] Jae Ho Yang, Kun Woo Song, Dong Seok Kim, Dong-Joo Kim, Heung Soo Lee, Ji-Hae Yoon, Yang-Hyun Koo, (2021) Thermodynamic evaluation of equilibrium oxygen composition of UO_2-Mo nuclear fuel pellet under high temperature steam, Frontiers in Energy Research 9:66791, DOI: 10.3389/fenrg.2021.667911,