OVERVIEW OF ACCIDENT TOLERANT FUEL DEVELOPMENT FOR LWRS

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ABSTRACT

Regarding the ATF cladding, surface modification technology consists of a coating technique for increasing oxidation resistance and an oxide dispersion strengthened (ODS) for improving high temperature strength. Current coating technologies are combined with arc ion plating and 3D laser coating to increase the bonding strength between the coating material and zirconium matrix. The ODS structure is realized by 3D laser scanning technology using oxide powder. The prototype tube is manufactured with the developed technology and performance evaluation is performed in both out-of-pile and in-pile test conditions. Surface-modified zirconium cladding has excellent oxidation resistance and deformation resistance in an accident environment. The surface-modified zirconium cladding was evaluated to have excellent or comparable performances (corrosion, creep, fatigue, and wear) to existing zirconium cladding in normal conditions. For the ATF pellet, microcell UO_2 pellets are being developed to enhance the thermal conductivity and/or retention capability of the fission products of UO_2 pellets. The fuel temperature can be effectively decreased by the enhanced thermal conductivity. So far, the research reactor test in Halden has continued without any problems up to 290 PFD for the surface modified cladding and microcell UO_2 pellet.

1. Introduction

Since the Fukushima accident, the need for accident tolerant fuel (ATF) has been highlighted [1-3]. Regarding the fuel cladding, the ATF cladding prevents the hydrogen explosion by suppressing hydrogen generation, which was caused by oxidation of Zr-based alloy, at high-temperature condition in case of accident. However, the ATF cladding should have good performance even under normal operating conditions of the nuclear power plant. In order to apply ATF cladding to commercial nuclear power plants, several factors (manufacturing, performance at normal and accident, economy, and compatibility with commercial reactors, etc.) must be considered. Korea's strategy to develop ATF cladding is to apply surface modification to existing zirconium alloys in the short term and to apply SiC_f/SiC cladding in the long term [3]. Surface-modified cladding is a concept that takes into account the lifetime of currently operating nuclear power plants and the period required for commercial application after development. A consideration in surface-modified cladding is how to improve high-temperature oxidation resistance and deformation resistance for commercial zirconium cladding. The easiest option for improving oxidation resistance at high-temperatures is to coat the oxidation resistance material on the surface of the cladding. Such a method is also developed by AREVA and Westinghouse [4, 5]. An important technique in coating methods is the development of coating materials and coating techniques that are superior. However, coated cladding has increased oxidation resistance at high-temperatures, but has little effect on high-temperature strength increase. To overcome this shortcomings, we are introducing oxide dispersion strengthened (ODS) technology. Therefore, Korea's near-term surface-modified cladding technology is combined with coating technology for oxidation resistance enhancement and ODS technology for high-temperature strength enhancement [6].

Microcell UO_2 pellets are expected to enhance the performance and safety of current LWR fuels under normal operational conditions as well as during transients/accidents. A high thermal conductive pellet can reduce not only the fission products (FPs) diffusivity/mobility but also the thermal stress by lowering the pellet temperature and temperature gradient. A lessening of stored energy in low temperature pellets significantly increases the safety margin under accident conditions. An improvement in FPs retention capability leads to a reduction of the inner surface cladding corrosion caused by FPs as well as the internal pressure of the fuel rod. A soft/ductile thin wall facilitates the fast creep deformation of the pellets, thereby reducing the mechanical loading of the cladding under operational transients [7-10].

This work introduces the results of the study on the surface-modified cladding and microcell UO_2 pellets which is being developed by the near-term application technology in Korea.

2. Surface-Modified Technology Development for ATF Cladding

2.1 ODS technology

There are various methods of increasing the strength of a metal material. Among them, a technique of strengthening the dispersion of oxides in metal matrix (oxide dispersion strengthened; ODS) is widely applied as a method of greatly increasing the high-temperature strength. However, it is difficult to make an ODS structure and the cost increases greatly. In order to make the ODS structure on the Zr alloy, a new technique as shown in Fig. 1 was developed at KAERI [11, 12]. The conventional ODS manufacturing process at the top of the figure involves mechanical mixing using powder raw materials and heat treatment. The intermediate product is then processed to form the final product. These processes are costly and time consuming and difficult to control product quality. And there was no research on applicability to Zr-based alloys. On the other hand, the laser ODS manufacturing technology offers the possibility to manufacture products in a short time compared to existing technologies. This can also make an ODS structure for a locally desired region. In case of the cladding tube, it is designed to make the ODS structure external region after considering pellet cladding mechanical interaction (PCMI).

Fig. 2 shows the results of the photograph to make ODS structure using laser, microstructures of ODS-treated sample and the high-temperature deformation behavior of the manufactured ODS cladding and reference Zircaloy-4 (Zry-4) cladding. The fine oxide particles were evenly distributed about 80 μm thick on the outside of the cladding tube as shown in Fig. 2(a). And the oxide particles were identified as Y_2O_3 phase from the energy dispersive spectrometer (EDS) and selective area diffraction (SAD) analysis.

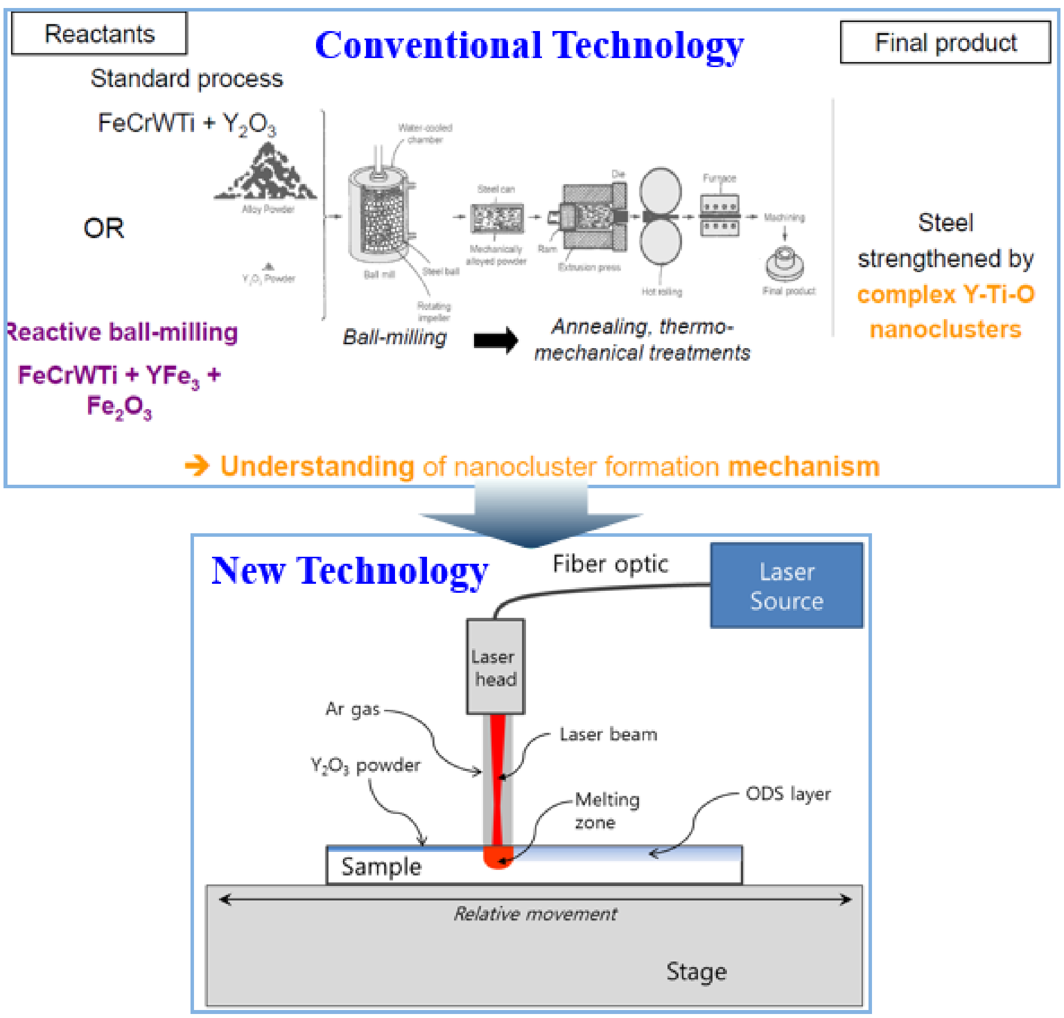


Fig. 1 Schematic drawing to compare ODS manufacturing between conventional technology and new technology

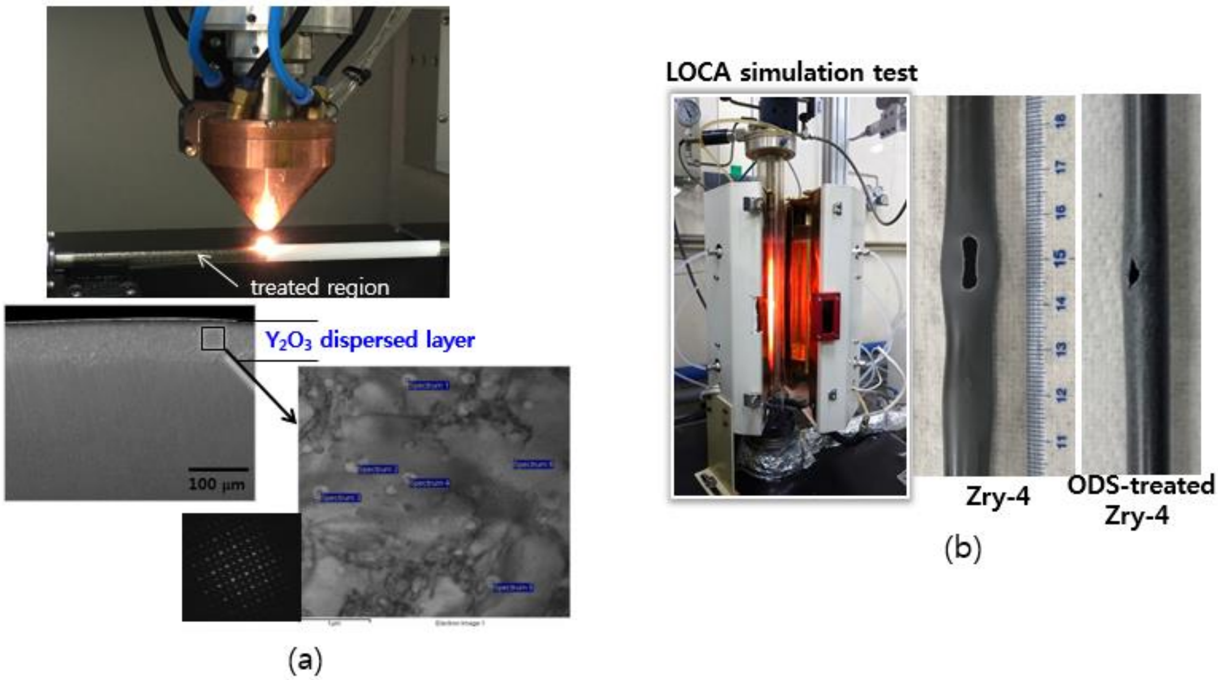
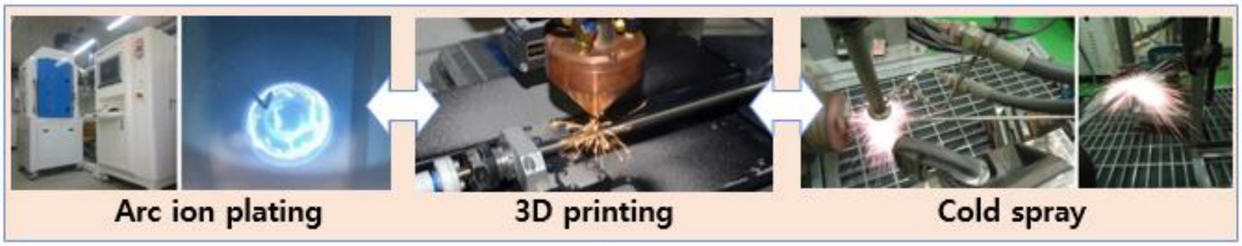


Fig. 2 Photo for making the ODS structure using laser and microstuructural observation of ODS structure (a) and the high-temperature deformation behavior in both commercial Zry-4 and surface ODS-treated Zry-4 tube

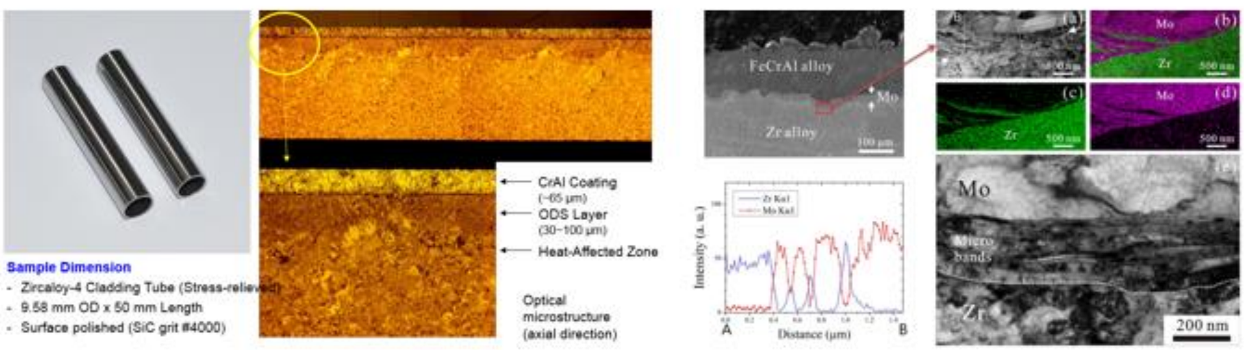
For this sample, the ballooning and rupture behaviors were evaluated using LOCA simulation test equipment as shown in Fig. 2(b). The hoop stress of the cladding tube was 60MPa which was maintained during the test before the cladding rupture, and the heating rate was 5℃/s during the ramp. The internal pressure is controlled by measuring with a pressure gage in real time, and te pressure gas is shut off immediately when rupture occurs. This is a method of applying a constant internal pressure regardless of whether or not there is a dummy pellets in the cladding tube. The burst appeared in both test samples during the ramp; however, the burst temperature of the surface ODS-treated Zry-4 cladding tube (829 °C) was higher than that of the reference Zry-4 cladding tube (743 °C). The burst opening size of the reference Zry-4 cladding was higher than that of the surface ODS-treated Zry-4 cladding, and the burst strain of the reference Zry-4 cladding (48%) was higher than that of the surface ODS-treated Zry-4 cladding (7%). Although the surface ODS layer thickness was approximately 80 μm, which was contained in the total cladding thickness of 570 μm, the cladding properties such as burst temperature and burst strain were considerably improved by the ODS treatment using laser. The ballooning and rupture behavior results for ODS or coated cladding were published in literature [13].

2.2 Coating technology

Coating methods that are being studied on the concept of surface-modified cladding include arc ion plating, 3D laser printing, and cold spraying [14-16]. Fig. 3 shows the each coating process, sample appearance after coating, and cross-sectional microstructure of the coated sample.



(a)



(b)

Fig. 3 Photos of three kinds of coating processes (a), sample appearance after coating process and cross-sectional microstructure of the coated sample (b)

Each of the coating methods has advantages and disadvantages in terms of performance, economy, or manufacturability, so that it can be selected or combined according to purposes. In particular, the chemical and physical properties of the coating material and the substrate material must be considered. An important factor to be considered with the coating technology in the coating concept is the coating material. Cr, CrAl alloy, and FeCrAl alloy are studied as coating materials for Zr alloy cladding among various materials having good oxidation resistance [3-5]. Among them, FeCrAl alloy has applied diffusion barrier layer (Nb, Cr, and Mo etc.) due to the low eutectic temperature with Zr (~945^{o}C). Our coatings are focused on the Cr, CrAl alloy and FeCrAl/Cr (or Mo) layers using arc ion plating, 3D laser or cold spray method. These coating techniques were applied to each of the Zr cladding tubes to evaluate their corrosion performance under normal and high temperature oxidation conditions.

Fig. 4 shows the corrosion behavior of two types of surface-modified Zr cladding in a PWR simulation loop condition at 360^{o}C. Corrosion tests were performed in a 360^{o}C water state under a saturated pressure of 18.9 MPa using a pressurized water reactor (PWR) simulated water loop (Li: 2.2 ppm, B: 650 ppm, O2: <5ppb, and H2: ~3 ppm). The corrosion behavior of prepared samples was evaluated by weight measurements taken at periodic intervals.

Regarding the corrosion behavior of a FeCrAl/Cr-coated sample with 3D laser coating for FeCrAl and arc ion plating for Cr and a CrAl-coated sample with arc ion plating, the flaking or galvanic corrosion phenomenon was not observed in all tested samples at the Zr matrix and coated material interface. The corrosion weight gain of FeCrAl/Cr-coated samples and CrAl-coated samples is the lower than that of an uncoated samples after 120 days. Because the corrosion test specimen is coated only on the outer surface of the cladding, the weight increase in the test can be seem to be the weight increase of Zry-4 in the uncoated area.

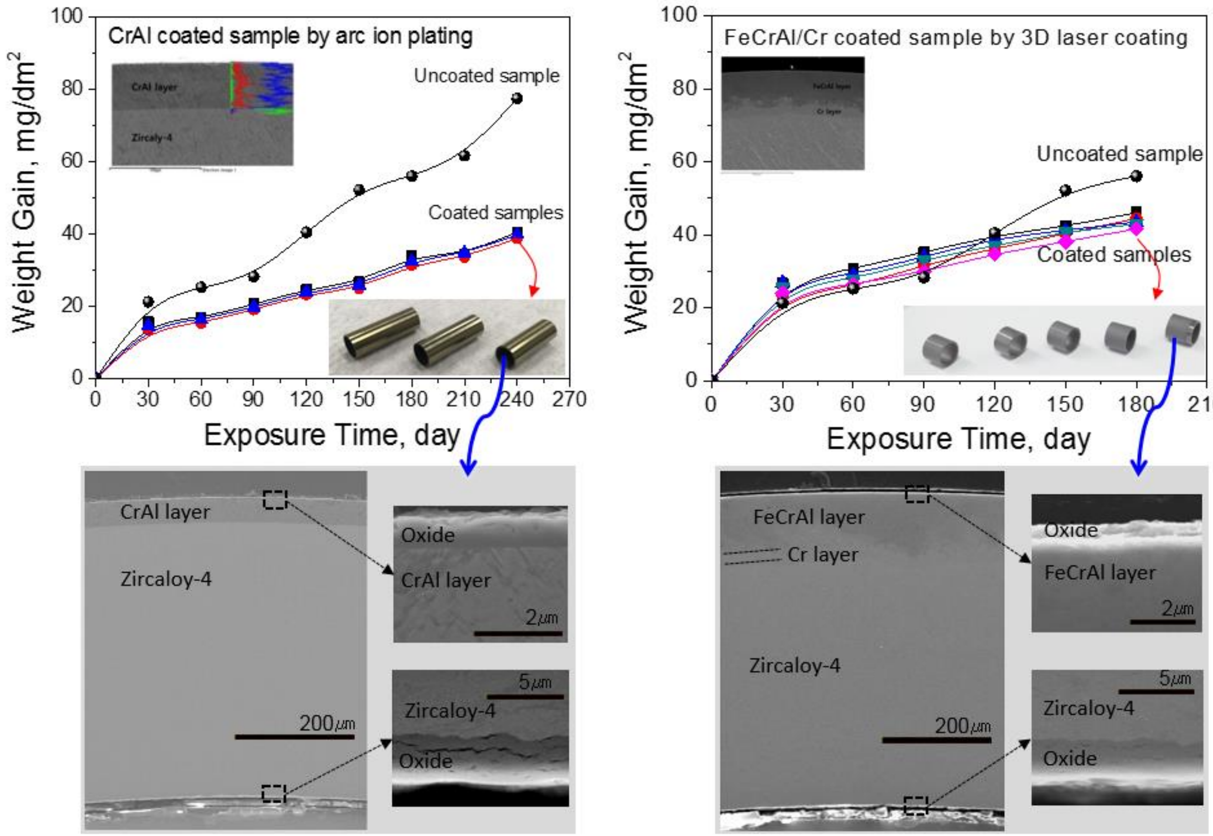


Fig. 4 Corrosion behavior of surface-modified Zry-4 cladding and reference Zry-4 cladding in PWR simulation loop condition at 360^{o}C [13]

Fig. 5 shows the evaluation of the high temperature oxidation properties of the CrAl-coated Zry-4 cladding by arc ion plating, using uncoated Zry-4 cladding as a reference. Because the coating is applied only on the outer surface, the oxidation test of two specimens was focused on the outer surface of the tube. When the prepared specimens were maintained at 1200^{o}C for 3000 s in a steam environment and cooled to 800^{o}C and then water quenched, the CrAl-coated Zry-4 cladding remained without damage, while uncoated Zry-4 cladding was damaged severely by thermal shock.

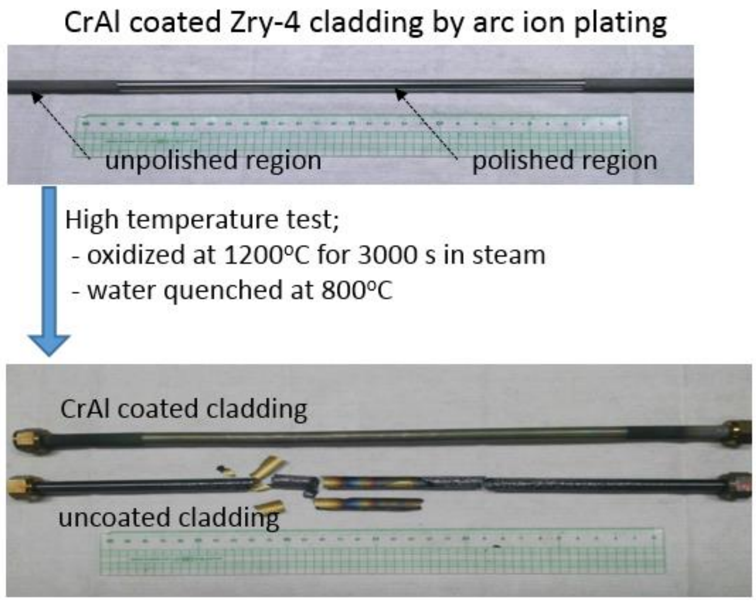


Fig. 5 High temperature oxidation and water quenching behavior of CrAl-coated Zry-4 cladding and uncoated Zry-4 cladding

2.3 Performance evaluations

To summarize the various performances, the surface-modified Zr cladding shows the improved performance (corrosion/oxidation, creep, wear, LOCA) and shows the reasonable performance (welding) when compared to commercial Zr cladding [6,11-17]. Regarding the corrosion behavior, the flaking or galvanic corrosion was not observed in the samples between Zr matrix and coated material interface, which can be identified form the corroded sample appearances. An excellent corrosion/oxidation resistance of the CrAl-coated cladding is the result of the stable oxide formations by an optimized compositional ratio between Cr and Al. It is confirmed that $CrAl_2O_4$ phase is stabilized under the normal operation condition and accident conditions [13]. A good corrosion/oxidation resistance of the FeCrAl/Cr-coated cladding is due to the inherent properties of the FeCrAl alloy. Improved mechanical properties such as creep and wear resistance were shown in the surface-modified Zr cladding, because the CrAl alloy had a higher strength than Zr alloy as well as the ODS structure layer had a good strength up to high temperatures. At the high-temperature, cladding strength is considerably improved by a uniform distribution of the Y_2O_3 particles although the thickness of ODS layer was 100 microns. Table 1 shows the performance test result for our study.

Table 1. Summary of performance test results for surface-modified ATF cladding

Item Technology	Corrosion resistance	Creep deformation	Oxidation resistance	Ballooning deformation	Burst temperature
ODS	Slightly decreased	Highly decreased	Slightly decreased	Highly suppressed	Highly increased
CrAl coating	Highly	Slightly	Very-high	Slightly	Slightly

	improved	decreased	improved	suppressed	increased
FeCrAl/Cr coating	Highly improved	Slightly decreased	Highly improved	Slightly suppressed	Slightly increased
ODS+CrAl coating	Highly improved	Highly decreased	Very-highly improved	Highly suppressed	Highly increased
ODS+FeCrAl/Cr coating	Highly improved	Highly decreased	Highly improved	Highly suppressed	Highly increased

An irradiation test of the surface-modified Zr cladding was reached up to 360 FPD (burnup: 16.2MWd/kgU) in Halden research reactor. During the operating period so far, the KAERI rods have operated normally and without any indication of failure.

3. Microcell UO_2 Technology Development for ATF Pellet

Fig. 2(a) shows the shape of a 5 vol% of Mo containing metallic microcell UO_2 pellet. This pellet was fabricated by co-sintering of metal powder over-coated UO_2 granules through a conventional sintering process. The density of the sintered pellets is about 97.5 %T.D. Fig. 2(b) shows a typical microstructure of metallic microcell UO_2 pellets in which the microcell concept was successfully implemented. It is a panoramic image of metallic microcell pellet from end to end (pellet radial direction). The averaged UO_2 grain size is about 8 μm. The metallic cell wall thickness ranges from 4 to 6 μm and the averaged diameter of the UO_2 granules is about 300-400 μm.

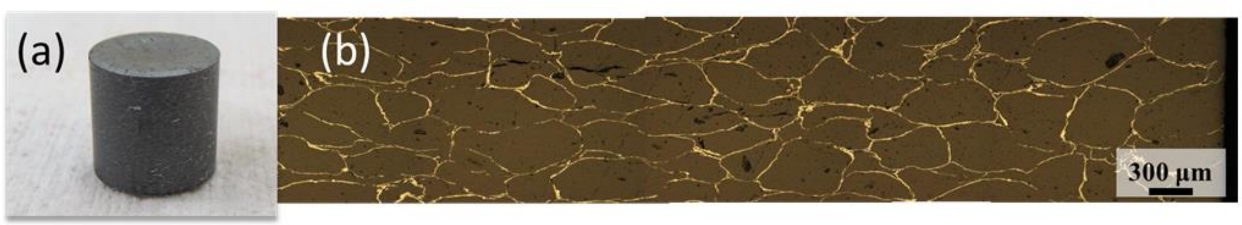


Fig. 6 Pellet shape (a) and microstructure (b) of 5 vol% Mo metallic microcell UO2 pellet (brighter lines are Mo metallic phase).

The main benefit of the metallic microcell UO_2 pellets is an enhanced thermal conductivity. A continuously connected metallic wall can effectively increase the thermal conductivity of UO_2 pellets. Fig. 3 shows an enhancement of the thermal conductivity. The thermal conductivity of 5 vol% of Mo containing microcell UO_2 pellets was increased by about 100 % at 1000 °C, compared to that of a standard UO_2 pellet. The fuel temperature can be effectively decreased by the enhanced thermal conductivity of the pellet. The linear thermal expansion coefficient of the Mo metallic microcell UO_2 pellet decreased with increasing Mo content because of the lower thermal expansion coefficient of Mo ($\sim 4.8\times10^{-6}$ /K at 25 °C) than that of UO_2 ($\sim 10\times10^{-6}$ /K). Under the transient conditions, it is expected that the mechanical stress loading on the cladding due to pellet expansion can be reduced by this feature of the metallic microcell UO_2 pellet.

4. Summary

Surface modification technology can be defined as combination of the coating technology for enhanced oxidation resistance and the ODS technology for high temperature strength enhancement. Surface-modified specimens were fabricated, and performance tests were conducted under normal and accident conditions. The improvement of corrosion/oxidation resistance in normal and accident conditions of surface-modified Zr cladding lies in the application of excellent oxidation materials and coating techniques. The improvement of high-temperature deformation resistance was largely achieved by strengthening by ODS treatment. As an ATF pellet, microcell UO2 pellets are being successfully developed to enhance the thermal conductivity and/or retention capability of the fission products of UO_2 pellets. The assessment of the out-of-pile properties of the developed pellet is being performed. Especially, the observation of the in-reactor performance and behavior of the microcell UO_2 pellets is in progress through a Halden irradiation test. The excellent performances of the microcell UO_2 pellets are being shown by the online measurement data of the Halden irradiation test. From the out-of-pile and in-pile tests, cladding and pellet technology was shown to have sufficient potential as an ATF cladding. Based on this results, KHNP, KepcoNF, and KAERI are preparing cooperation system for commercialization of an AFT in Korea.

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4. References

[1] S.M. Bragg-Sitton et al, Advanced Fuels Campaign: Enhanced LWR Accident Tolerant Fuel Performance Matrix, INL/EXT-13-000264, Feb. (2014).

[2] J. Carmack, F. Goldner, S.M. Bragg-Sitton, and L.L. Snead, TopFuel 2013, Charlotte, North Carolina, Sep. 15-19, 2013.

[3] H.G. Kim, J.H. Yang, W.J. Kim, Y.H. Koo, Nucl. Eng. & Technol. 48 (2016) 1.

[4] J. Bischoff, C. Delafoy, C. Vauglin, P. Barberis, C. Roubeyrie, D. Perche, D. Duthoo, F. Schuster, J.-C. Brachet, E.W. Schweitzer, K. Nimishakavi, Nucl. Eng. & Technol. 50 (2018) 223.

[5] M. Sevecek, A. Gurgen, A. Seshadri, Y. Che, M. Wagih, B. Phillips, V. Champagne, K. Shirvan, Nucl. Eng. & Technol. 50 (2018) 229.

[6] H.G. Kim, I.H. Kim, Y.I. Jung, D.J. Park, J.H. Yang, Y.H. Koo, TopFuel 2016, Boise, ID, September 11-15, 2016.

[7] Y.H. Koo, J.H. Yang, J.Y. Park, K.S. Kim, H.G. Kim, D.J. Kim, Y.-I. Jung, K.W. Song, Nucl. Technol, 126 (2014) 295-304.

[8] J.H. Yang, K.S. Kim, D.J. Kim, Y.H. Koo, F2.2 EHPG meeting, 2014.

[9] D.J. Kim, Y.W. Rhee, J.H. Kim, K.S. Kim, J.S. Oh, J.H. Yang, Y.H. Koo, K.W. Song, J. Nucl. Mater, 462 (2015) 289-295.

[10] D.J. Kim, K.S. Kim, D.S. Kim, J.S. Oh, J.H. Kim, J.H. Yang, Y.H. Koo, Nucl. Eng. & Technol, 50 (2018) 253-258.
[11] H.G. Kim, I.H. Kim, Y.I. Jung, D.J. Park, J.Y. Park, Y.H. Koo, Nucl. Eng. & Technol, 46(4) (2014) 521.
[12] H.G. Kim, I.H. Kim, Y.I. Jung, D.J. Park, J.H. Park, J.H. Hoo, Y.H. Koo, Additive Manufacturing 22 (2018) 75.
[13] H.G. Kim, I.H. Kim, Y.I. Jung, D.J. Park, J.H. Park, B.K. Choi, Y.H. Lee, J. Nucl. Mater, 510 (2018) 93.
[14] J.H. Park, H.G. Kim, J.Y. Park, Y.I. Jung, D.J. Park, Y.H. Koo, Surf. Coat. Technol, 280 (2015) 256.
[15] H.G. Kim, I.H. Kim, Y.I. Jung, D.J. Park, J.Y. Park, Y.H. Koo, J. Nucl. Mater, 465 (2015) 531.
[16] D.J. Park, H.G. Kim, Y.I. Jung, J.H. Park, J.H. Yang, Y.H. Koo, J. Nucl. Mater, 482 (2016) 75.
[17] H.G. Kim, I.H. Kim, Y.I. Jung, D.J. Park, J.H. Park, J.H. Yang, Y.H. Koo, WRFPM 2017, Jeju Island, Korea, September 10-14, 2017.