Formation of ODS Layer on Zirconium-based Fuel Cladding Tubes for Application as ATF Cladding

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1. Introduction

Accident tolerant fuel (ATF) cladding is being developed globally after the Fukushima accident with the demands for the nuclear fuel having higher safety at normal operation conditions as well as even in a severe accident conditions. Korea Atomic Energy Research Institute (KAERI) is one of the leading organizations for developing ATF claddings [1,2]. Various ATF concepts have been developed via a national R&D project with the grant of Ministry of Science and ICT from 2012. Recently, a new project was launched to develop the irradiation test rod using ATF pellets and cladding in collaboration with research institute, nuclear fuel vendor, and universities under the funding of Ministry of Trade, Industry and Energy. This paper explains one of concepts being developed in the latter project, i.e., zirconium alloy based cladding consisting of oxide dispersion strengthened (ODS) layer and surface coating.

The ODS treatment was proposed to increase the strength of the Zr-based alloy up to high temperatures [2-6]. High-power laser beam was exposed on the zirconium surface previously coated by oxides (typically Y_2O_3). The dispersed oxide layer was formed by the penetration of oxide particles into Zr alloys. According to our previous investigations [3-6], the tensile strength of Zircaloy-4 was increased by up to 20% with the formation of a thin dispersed oxide layer with a thickness less than 10% of that of the Zircaloy-4 substrate. In this paper, the ODS treatment was performed using various zirconium-based alloy tubes, e.g., Zircaloy-4, KNF-M, and HANA-6. It is investigated the formation of ODS layer and mechanical properties depending on the processing conditions and materials.

2. Methods and Results

2.1. Experimental for ODS Treatment

Three kinds of Zr-based tubes (Zircaloy-4, KNF-M, and HANA-6) with an outer diameter of 9.5 mm and a wall thickness of 0.57 mm were used. The tubes were cut in length of 500 mm and cleaned with alcohol and acetone. Y_2O_3 was coated on the cleaned Zircaloy-4 tubes by a dip-coating method. To prepare the dip solution, Y_2O_3 powder was dissolved in ethyl-alcohol at a solute content of 10%. The solution was mixed for 24 h using zirconia balls. The prepared solution was coated on zirconium tubes using a spray gun. The coated tubes were dried in atmospheric conditions for 24 h. Since any kinds of binder were not applied, it was cautious that the coating could be easily removed during handling.

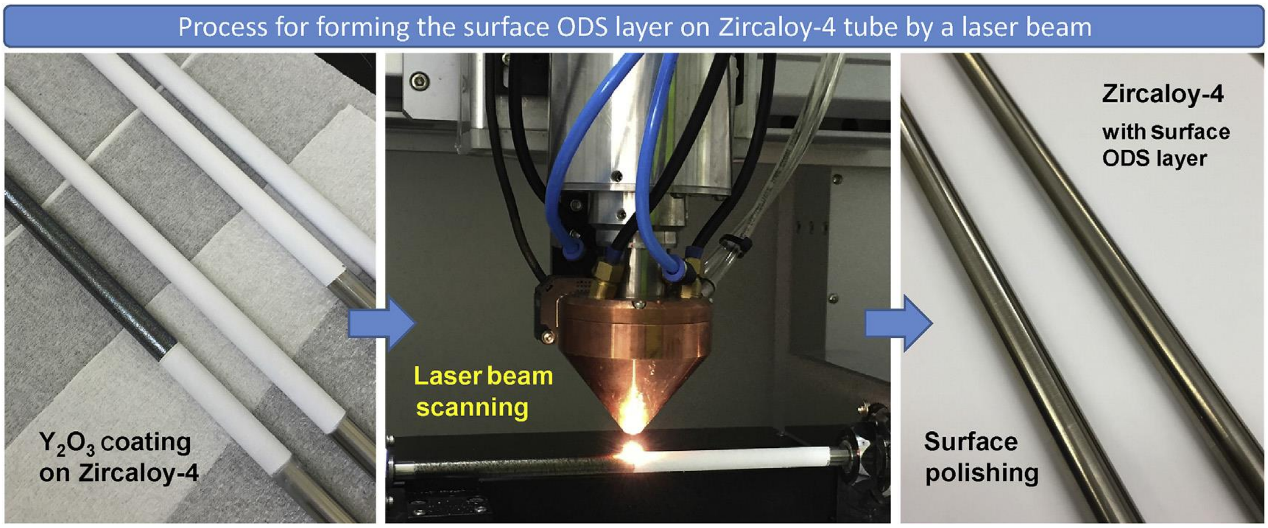


Fig. 1. Fabrication process for forming a surface ODS layer on Zircaloy-4 tube using a laser beam [6].

To form the ODS layer, the Y_2O_3-coated cladding tubes were scanned by a continuous wave diode laser. There are two kinds of lasers: one is a laser with the beam diameter of 230 μm, and the other is that of 1 mm. The ODS layer was formed at a laser beam power of 120–200 W and scan speed of 10 mm/s. The laser beam was scanned continuously along the circumferential direction with an overlap distance of 0.2–0.8 mm. To prevent oxidation, Ar gas was continuously blown on the samples' surfaces during laser processing. Cooling water was supplied to the inside of the tubes to release the induced heat. The representative process was presented in Fig. 1 [6].

2.2. Microstructures (230 μm Laser Beam)

Fig. 2 shows the cross-sectional microstructures of the ODS treated Zircaloy-4 samples. The ODS layer was observed at the surface with dark contrasts. The microstructure along the axial direction revealed the formation of a dispersed oxide layer 50–140 μm in thickness in the surface region. The helical laser beam scans with a fixed offset distance of 0.4 mm produced the wavy interface. The thermal energy induced by a laser beam is known to form a heat-affected zone (HAZ); however, the formed HAZ is not distinguishable from the Zr matrix in these micrographs.

Fig. 3 shows the cross-sectional microstructures of the ODS treated KNF-M and HANA-6 samples. In the case of Zircaloy-4, the as-received tubes showed a cold-worked and stress-relieved microstructure (Fig. 2). In the case of KNF-M and HANA-6, recrystallized microstructures were observed in the as-received tubes. ODS layer was developed about 100 μm in thickness in the surface region. Below the ODS layer, HAZ was form up to 260 μm in depth from the surface. The dispersed oxide particles were confirmed using an electroscope analysis, as shown in Fig. 4 observed in the case of HANA-6.

2.3. Microstructures (1 mm Laser Beam)

Fig. 5 shows the cross-sectional microstructures of the ODS treated Zircaloy-4 samples. The ODS layer was very thin (<10 μm in thickness) when the laser beam power was 180 W, and it was increased to 45–75 μm in thickness as the beam power increased to 200 W. In the case of the laser with 1 mm beam diameter, HAZ was developed largely. Because the induced thermal energy was hardly dissipated, the HAZ was enlarged as compared to the microstructures shown in Figs 2 and 3 obtained using the laser with small beam diameter.

2.4. Mechanical Properties

Fig. 6 shows stress–strain curves for fresh and ODS Zircaloy-4 samples during ring tension tests. Tests were performed at room temperature (RT) and elevated temperatures of 380 and 500℃. In the test at RT, the ultimate tensile strength of fresh and ODS Zircaloy-4 was about 790 MPa and about 870 MPa. ODS samples exhibited higher tensile strength than fresh Zircaloy-4 samples. Tensile elongation, on the contrary, decreased dramatically, showing an abrupt drop in the applied tensile load.

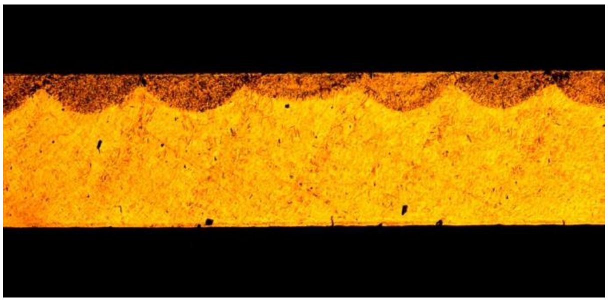
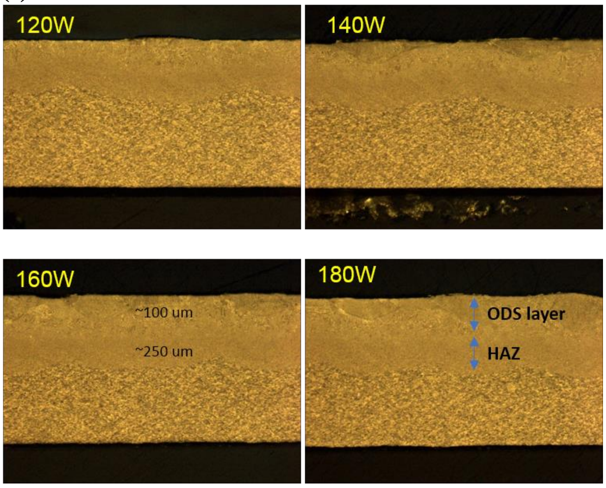


Fig. 2. Cross-sectional microstructures of ODS Zircaloy-4 samples showing the ODS layer formed at the surface.

(a) KNF-M



(b) HANA-6

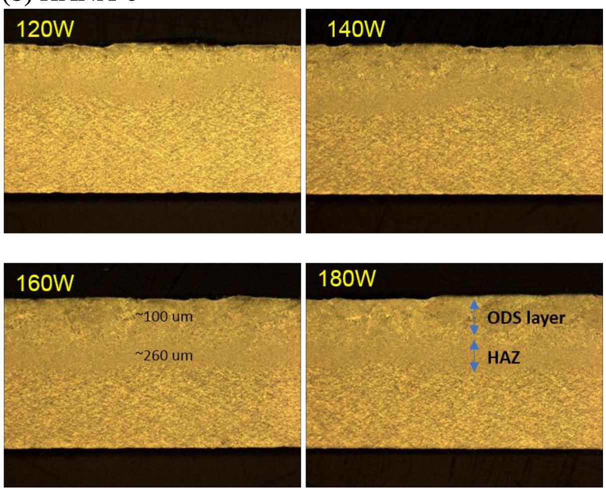


Fig. 3. Cross-sectional microstructures of ODS treated (a) KNF-M and (b) HANA-6 samples.

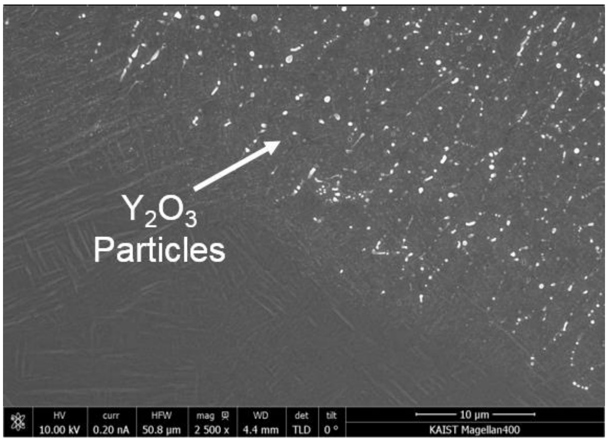


Fig. 4. SEM micrographs of ODS layer in HANA-6 samples.

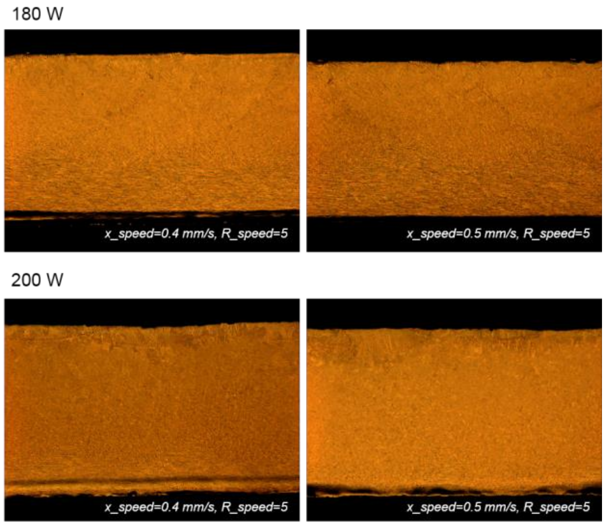


Fig. 5. Cross-sectional microstructures of ODS treated Zircaloy-4 samples using a laser with a 1 mm beam diameter.

Strengthening of Zircaloy-4 by the ODS layer was effective at elevated temperatures. In the test at 380 ℃, the ultimate tensile strength of fresh and ODS Zircaloy-4 were about 500 and 575 MPa. It is about 15% increase. In the test at 500 ℃, the ultimate tensile strength of fresh and ODS Zircaloy-4 was about 385 and 470 MPa. It is more than 20% increase with the formation of an ODS layer. Elongation did not exhibit stress drop or dramatic decrease, which was observed in the test at RT.

3. Conclusions

Surface treatment was performed by a laser beam to form a dispersed oxide layer in Zr-based alloys. Laser beam scanning of a tube coated with Y_2O_3 resulted in the formation of a dispersed oxide layer in the tube's surface region. This surface ODS treatment can improve the mechanical properties of Zr tubes, and thus is useful for manufacturing Zr tubes with enhanced safety and accident tolerance.

Acknowledgment

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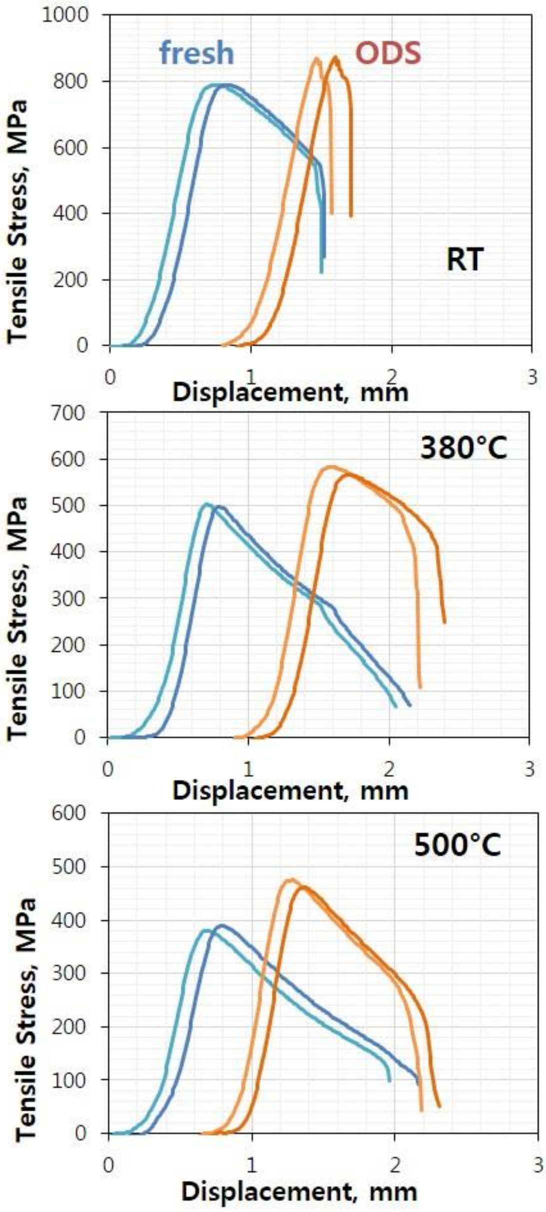

Fig. 6. Stress–strain curves of fresh and ODS Zircaloy-4 samples in ring tensile tests at room temperature (RT), 380℃, and 500℃, respectively [6].

REFERENCES

[1] Y.-H. Koo et al., "KAERI's development of LWR accident-tolerant fuel," Nucl. Technol., 186, 295-304 (2014).

[2] H.-G. Kim et al., "Development status of accident-tolerant fuel for light water reactors in Korea," Nucl. Eng. Technol., 48, 1-15 (2016).

[3] H.-G. Kim et al., "Microstructure and mechanical strength of surface ODS treated Zircaloy-4 sheet using LBS," Nucl. Eng. Technol., 46, 521-528 (2014).

[4] Y.-I. Jung et al., "Strengthening of Zircaloy-4 using Y_2O_3 particles by a laser-beam-induced surface treatment process," Mater. Des., 116, 325-330, (2017).

[5] Y.-I. Jung et al., "Surface treatment to form a dispersed Y_2O_3 layer on Zircaloy-4 tubes," Appl. Surf. Sci., 429, 272-277, (2018).

[6] Y.-I. Jung et al., "Effect of a surface oxide-dispersion-strengthened layer on mechanical strength of zircaloy-4 tubes," Nucl. Eng. Technol., 50, 218-222, (2018).