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Strengthening of Zircaloy-4 using Y_2O_3 particles by a laser-beam-induced surface treatment process

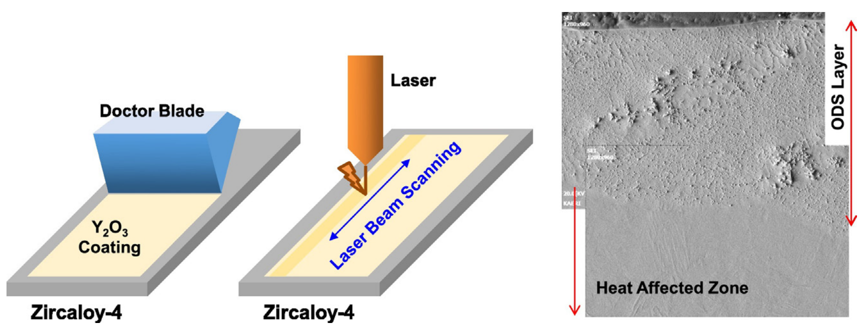
Yang-Il Jung *, Hyun-Gil Kim, Il-Hyun Kim, Sun-Han Kim, Jung-Hwan Park, Dong-Jun Park, Jae-Ho Yang, Yang-Hyun Koo

LWR Fuel Technology Division, Korea Atomic Energy Research Institute, 989-111 Daedeokdaero, Yuseong, Daejeon 34057, Republic of Korea

HIGHLIGHTS

- Laser beam treatment technology was developed for producing the oxide-dispersion-strengthened (ODS) Zircaloy-4.
- ODS Zircaloy-4 with increased strength was obtained by surface treatment to form a dispersed oxide layer.
- The strengthening was more effective at elevated temperature revealing the high toughness without brittle failure.

GRAPHICAL ABSTRACT



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ABSTRACT

The mechanical strength of Zircaloy-4 was enhanced by surface treatment consisting of laser beam scanning of an oxide-coated Zircaloy-4 sheet. An yttrium oxide (Y_2O_3) coating 7–55 μm thick was applied to Zircaloy-4 sheets. Then, a laser beam was used to form a dispersed oxide layer on the Zircaloy-4 metal surface. The thickness of the dispersed oxide layer varied from 80 to 200 μm depending on the process parameters. Tensile tests of samples having both surfaces treated were conducted at room temperature and 380 °C. The tensile strength of Zircaloy-4 at room temperature was increased by up to 20% with the formation of a thin dispersed oxide layer with a thickness < 10% of that of the Zircaloy-4 substrate. However, the tensile elongation of the samples decreased drastically. The decreased ductility became insignificant as the testing temperature increased to 380 °C. The strengthening of the fabricated samples could be explained by two mechanisms: (i) oxide particles dispersed in the metallic matrix and (ii) the phase transformation in Zircaloy-4.

1. Introduction

Zircaloy-4 is a zirconium-based alloy containing a few percent of Sn, Fe, and Cr. It was developed for application in nuclear fuel assembly components and has been used for more than 50 years in commercial nuclear power plants. Zirconium alloys have exhibited good corrosion resistance and irradiation stability in a reactor environment. However, to improve nuclear safety under accident conditions, an increased need for higher strength and oxidation resistance than conventional zirconium alloys motivated the development of enhanced zirconium alloys [1–3]. The tensile strength of Zircaloy-4 exceeds 400 MPa at room temperature (RT); however, the strength decreases to several tens of megapascals as the temperature increases to above 600 °C [4,5]. The

* Corresponding author.
E-mail address: yijung@kaeri.re.kr (Y.-I. Jung).

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softening of Zircaloy-4 at elevated temperatures is an inherent physical characteristic of metallic material. Nevertheless, to preserve the fuel rod geometry in order to maintain the core coolability in accidents, increased high-temperature strength of zirconium alloys is essential.

The concept of oxide-dispersion-strengthened (ODS) alloys was recently applied to Zircaloy-4 [2,3,6] to increase its strength at RT as well as at elevated temperatures. Oxide particles in ODS alloys are thermally stable and resistant to neutron irradiation; thus, oxide dispersion strengthening treatment of Zircaloy-4 is promising for developing fuel cladding materials with enhanced accident tolerance. Yttrium oxide (Y_2O_3) is a typical material used in ODS alloys [6–8]. ODS alloys are generally manufactured through mechanical alloying of the source metal with oxide powders [9] and applying consolidation processes such as hot isostatic pressing [10], hot extrusion [11], and spark plasma sintering [12]. Homogeneous dispersion of oxide particles during the manufacturing process is a key issue. In this study, an ODS layer was formed in the Zircaloy-4 surface region by a laser beam treatment. Surface treatment is advantageous for uniform distribution of oxide particles and control of their volume fractions [6]. Moreover, the surface treatment process is directly applicable to final products such as tubes, strips, and sheets.[1] Y_2O_3-coated Zircaloy-4 plate samples were scanned by a laser beam. A dispersed oxide layer was formed by penetration of Y_2O_3 particles into Zircaloy-4. The dependence of the thickness of the ODS layer on the laser beam power and the thickness of the Y_2O_3 coating was investigated. The effect of oxide dispersion strengthening treatment on the mechanical strength was evaluated by tensile tests at RT and an elevated temperature of 380 °C.

2. Experimental procedures

Zircaloy-4 (Zr-1.5Sn-0.2Fe-0.1Cr, wt.%) alloy sheets with a thickness of 2 mm were used as a substrate. The Zircaloy-4 sheets initially exhibited recrystallized microstructure with an average grain size of ~7 μm. The sheets were cleaned by alcohol and acetone to remove stains and organic contamination on the surface, and then dried in an oven at 80 °C. Y_2O_3 was coated on the prepared Zircaloy-4 sheets by a doctor blade coating method. To form a paste, Y_2O_3 powder (99.9%, 1 μm, Alfa Aesar, USA) was placed in an agate bowl, and mixed with distilled water containing 10 wt.% poly vinyl alcohol (PVA) as a binder. The amount of PVA was 3 wt.% of the Y_2O_3 content. The Y_2O_3 paste was wet-coated on the Zircaloy-4 sheet using a doctor blade with a 0.1 mm gap, and then dried in a vacuum oven at 80 °C for 20 min. The final thickness of the Y_2O_3 coating was measured by an eddy-current isoscope (MP30, Fischer, Germany). The developed Y_2O_3 coating was 7–55 μm in thickness depending on the coating speed and solution dilution.

The Y_2O_3-coated Zircaloy-4 sheet was scanned by a continuous-wave diode laser (PF-1500F, HBL Co., Korea). The wavelength of the emitted laser beam was 1064 nm, and the beam diameter was 230 μm. The maximum operating power was 250 W. The thickness of the ODS alloy layer was controlled by varying the laser beam power. The laser beam power was varied from 150 to 200 W. The laser treatment parameters for the obtained samples were summarized in Table 1. The scan speed (which can also be used to control the thickness) was fixed at 10 mm/s in this investigation. To prevent oxidation during laser beam scanning (LBS), Ar gas was continuously blown on the samples' surfaces. The samples were placed on an aluminum panel with circulating coolant water to release the induced heat during LBS. Laser beam lines were scanned repeatedly with an overlap distance of 0.2 mm to form a rectangular region. The ODS Zircaloy-4 samples were cut cross-sectionally for microstructural observation using an optical microscope (OM) and a scanning electron microscope (SEM). For a mechanical test, a small-size tensile specimen was electro-discharge-machined from the ODS Zircaloy-4 samples. The over-all length of the tensile specimen was 22 mm, and the length of reduced section was 6 mm. The width of grip section was 10 mm, and that of reduced section was 4 mm by a fillet radius of 3 mm. The thickness of the specimen was about 2 mm. Both sides of the Zircaloy-4 sheet were surface treated by oxide dispersion strengthening for the tensile test. The test was performed at RT and 380 °C with a cross-head speed of 1 mm/min (i.e., 2.78×10^{-3} s^{-1}) using a universal testing machine (Instron 3367, USA). The fracture surfaces after the tensile test were also observed by an SEM (EM-20, Coxem, Korea).

[1] It would be difficult to manufacture final products using ODS Zr alloys because they are harder than pristine Zr alloys.

Table 1
Laser treatment parameters for the formation of dispersed oxide layer in Zircaloy-4.

(a) For a fixed laser beam power of 180 W		(b) For a fixed Y_2O_3 coating thickness of 20 μm	
Y_2O_3 coating [μm]	Ref.	Laser beam power [W]	Ref.
7		150	
10	Fig. 5	160	
15	Figs. 2(a) & 3(a)	170	
20	Figs. 2(b) & 3(b)	180	Figs. 2(b) & 3(b)
30	Figs. 5, 6, 7	190	
55	Figs. 5 & 7	2000	

3. Results and discussion

Y_2O_3-coated Zircaloy-4 was scanned by a laser to form a dispersed oxide layer. Fig. 1 shows the appearance of the laser-beam-treated region. Repeated scanning of laser beam lines in the vertical direction formed rectangular regions. The laser beam power was 180 W, and the offset distance between adjacent scans was 0.2 mm. As a shield gas, Ar was blown through a laser nozzle during laser beam scanning. When Ar was blown during laser beam treatment, the resulting surface shone; however, a sooty surface appeared when Ar was not blown. The PVA binder in the Y_2O_3 coating was volatilized by the laser-induced thermal energy and blown away by the Ar gas. Without Ar blowing, the residual carbonaceous binder burned to form the sooty surface. Ar is generally used as a shielding gas to prevent oxidation of a metal substrate. In this case, it was effective for removing the residual binder as well.

Fig. 2 shows the cross-sectional microstructures of Zircaloy-4 after surface treatment for oxide dispersion strengthening. LBS on Y_2O_3-

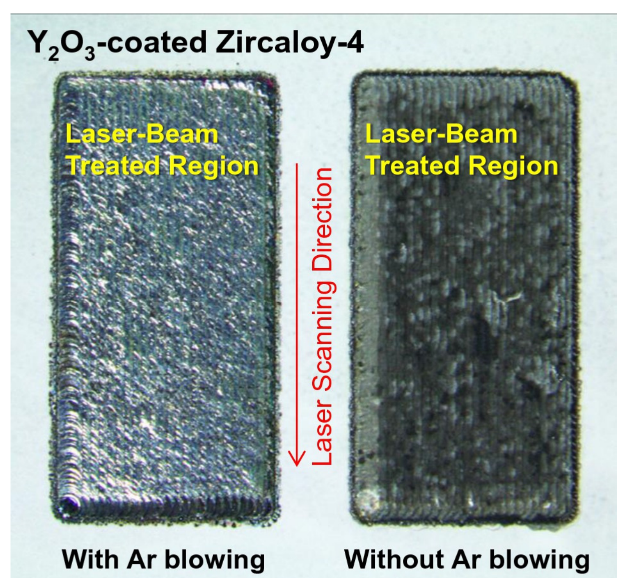

Fig. 1. Appearance of the laser-beam-scanned Zircaloy-4 sheet coated with Y_2O_3 (20 μm) and treated by 180 W laser beam. Left and right regions correspond to laser-beam-treated samples with and without Ar blowing.