Additive Manufacturing 22 (2018) 75–85

Contents lists available at ScienceDirect

Additive Manufacturing

journal homepage: www.elsevier.com/locate/addma

Microstructure and mechanical characteristics of surface oxide dispersion-strengthened Zircaloy-4 cladding tube

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ARTICLE INFO

Keywords:
Additive manufacturing
Accident tolerant fuel
Laser processing
ODS
Zircloy-4
LOCA

ABSTRACT

To increase the mechanical strength of Zircaloy-4 cladding at high temperatures, partial oxide dispersion-strengthened (ODS) treatment of the cladding tube surface was achieved by using laser processing technology. The microstructural characteristics and stability of the ODS layer formed on the Zircaloy-4 cladding surface were analyzed at temperatures up to 1000 °C. Ring tensile and loss-of-coolant accident (LOCA) simulation tests were performed to evaluate the mechanical properties of the surface ODS treated Zircaloy-4 cladding tube. The formation and uniform distribution of Y_2O_3 particles formed in the Zr matrix were identified, and the stability of the particles was confirmed up to 1000 °C. When compared to the reference Zircaloy-4 cladding tube, the surface ODS treated Zircaloy-4 cladding tube showed improved mechanical properties at both room temperature and 500 °C, as well as under LOCA simulation conditions.

1. Introduction

For a long time, Zr-based alloys have been used as a fuel cladding and other components of fuel assembly in light water reactors (LWRs) because they have good corrosion resistance, mechanical properties, and stability under the irradiation conditions that occur during reactor operation. Because commercial Zr-based alloys must prevent the release of fission products in nuclear fuel during normal operating conditions, the development of such alloys has focused on alloy performance under normal operating conditions in order to provide a safe, reliable, and economical operation. Of course, it has been mentioned that fuel cladding should maintain its nature during a postulated design-based accident such as a loss-of-coolant accident (LOCA) [1]. The accident behavior of Zr alloy cladding has been evaluated by using the high-temperature oxidation test and the LOCA simulation test [2–5]. From these studies, it is known that the oxidation rate of Zr-based alloys increases considerably at high temperatures, and the cladding failure owing to ballooning and rupture behavior is mentioned in the accident conditions.

Serious reactor damage caused by a hydrogen explosion was experienced in March 2011, at Fukushima nuclear power plant. Hydrogen is generated by the corrosion reaction of Zr-based alloys used as part of the fuel assembly in the accident conditions. Thus, it is expected that the hydrogen generation rate will increase owing to the increased oxidation rate of Zr-based alloys. In addition, radioactivity is released into the environment from the failed cladding caused by the ballooning and rupture of the Zr-based alloy [6]. After the event at Fukushima, accident tolerant fuels (ATFs) for LWRs became a hot topic in the nuclear industry. To suppress hydrogen explosions under accident conditions, various ATF concepts such as FeCrAl cladding [7], ZrMo cladding [8], cladding coatings [9,10], and SiC_f/SiC cladding [11] have been suggested and are being developed. However, the same problems with cladding coatings and FeCrAl cladding in terms of ballooning and rupture during LOCA need to be addressed, although such materials have a good oxidation resistance at high temperatures up to 1200 °C when compared to the Zr alloy cladding. Thus, the strength at high temperatures should be increased in FeCrAl or coated Zr alloys in order to resist ballooning and rupture of the fuel cladding.

Nowadays, additive manufacturing (AM) technology has emerged as a potential for innovative production of industrial components in the manufacturing industry [12], and it is commonly known as 3D printing. In addition, laser-based additive manufacturing (LBAM) processes can be utilized to generate functional parts [13]. This work focused on the fabrication of oxide dispersion-strengthening (ODS) structure on the Zircaloy-4 cladding tube surface based on the laser processing tech-

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https://doi.org/10.1016/j.addma.2018.05.002
Received 19 January 2017; Received in revised form 4 March 2018; Accepted 1 May 2018
Available online 02 May 2018

Table 1
Laser processing conditions to fabricate a reaction layer between Y_2O_3 particles and Zircaloy-4 cladding tube substrate.

Parameters	Value
Wavelength (nm)	1064
Scanning speed (mm/s)	3
Laser beam size (μm)	245
Working power (W)	80, 100, and 120
Hatching overlap (%)	30–40
Ar (inert gas) flow (cc/min)	50

nology. The target of ODS structure is to increase the strength of metal-based alloys at high temperatures because the oxide particles in the matrix are very stable, without phase transformation or dissolution at high temperatures [14–16]. In the previous study, it was confirmed that the strength of Zircaloy-4 sheet at room temperature and 500 °C was considerably increased by partial ODS treatment using laser beam scanning (LBS) method [17]. However, Zircaloy-4 alloys are used as fuel cladding tube in the nuclear industry. As such, it is fabricated into the seamless tubes in 0.57 mm thick, 9.5 mm diameter, and 4 m long, and we used the Zircaloy-4 cladding tube in this feasibility study. In addition, the stability of ODS particles must be confirmed because cladding temperatures exceed 1000 °C under the accident conditions of reactors. This study therefore focused on ODS treatment on the Zircloy-4 cladding tube surface as a function of the various parameters such as laser energy, rotation speed of the tubes, cooling conditions, and inert gas flow. To identify the effect of ODS on the strength and microstructure of Zircaloy-4, ring tensile tests and microstructural observations at both room temperature and high temperatures were performed with the prepared samples.

2. Experimental procedures

A commercial Zircaloy-4 (Zr1.5Sn0.2Fe0.1Cr in wt%) cladding tube was used as a substrate for ODS treatment using LBS. The initial state of the Zircaloy-4 cladding tube was stress-relief annealing (SRA). Before the ODS treatment, the cladding tube surface (300 mm length) was cleaned using alcohol to remove contamination or other stains on the tube surface and then dried. The Y_2O_3 powders as oxide particles were fabricated at Richest Group Ltd., and mean diameter of powders was less than 1 μm. Because of the very small size of the Y_2O_3 powders, commercial powder feeding equipment could not be used. Thus, these powders were spread onto the Zircaloy-4 cladding tube surface in a suspended state with alcohol, and then dried. From the eddy current test (ECT) measurement, the thickness of the initial Y_2O_3-deposited layer on the Zircaloy-4 cladding tube surface was approximately 50 ± 5 μm.

LBS was applied to create a partial ODS layer on the Zircaloy-4 cladding surface. The LBS conditions used in this work are described in our previous study [17]. A continuous wave (CW) diode laser with a maximum power of 250 W was used, and inert gas (pure Ar) was continuously supplied to the melted zone of the Zircaloy-4 cladding surface to prevent oxidation during LBS. In addition, cooling water was supplied to the inside of the cladding tube to prevent cladding deformation by laser heat. The maximum laser scanning length in the cladding tube was 27 cm. Table 1 shows the detailed parameters of the laser treatment conditions. Here, the helical pattern in the cladding tube surface can be obtained by the combined movement in both the laser beam to the axial direction of tube and the tube rotation with uniform velocity during the processing. The means of the hatching overlap is the overlapped region of laser beam diameter of each scan. Surface roughness of fuel cladding was measured by a stylus profilometer (SV-624, Mitutoyo) with a tracing length of 7 mm and 2 mm cut-off. Tracing was performed in quintuplicate for each sample and the mean value was calculated. The length of surface ODS treated region of Zircaloy-4 cladding tube was ranged from 10 mm to 250 mm to the axial direction. Short length samples were used for microstructural observation and ring tensile tests, and long length samples were used for LOCA simulation test.

The microstructure and phase stability of the partial ODS layer were respectively analyzed using a scanning electron microscope (SEM) equipped with an energy dispersive spectrometer (EDS) and a high-voltage electron microscope (HVEM) of 1250 keV at the Korea Basic Science Institute (KBSI). The thin foil sample used to analyze Y_2O_3 particle stability from room temperature (RT) to high temperature was prepared using focused ion beam (FIB) milling for the ODS-treated tube samples. A single heating holder was used for heating the thin foil sample from RT to 1000 °C. During the in situ heating, the heating rate was carefully controlled to prevent sample bending. The heating rate was 1 °C/min up to target temperatures of 350, 500, 750, 850, and 1000 °C, and a holding time of 30 min was used to obtain good-quality images.

A ring tensile test was performed at RT and 500 °C for the surface ODS-treated tube through laser scanning using an Instron-type tensile testing machine. The ring tensile test was performed following the procedure of ASTM E8-82 [18]. For the ring tensile test, a specific jig for the ring-shaped sample was used [19], and the crosshead speed during the tensile test was 1.7×10^{-3} mm/s for all samples. Two samples were tested for each test condition. The ballooning and rupture behaviors of surface ODS-treated tubes were evaluated using LOCA simulation test equipment [10]. The hoop stress of the cladding tube was 60 MPa which was maintained during the test before the cladding rupture, and the heating rate was 5 ℃/s during the ramp. The mixed phase of steam (50 vol %) and Ar (50 vol %) was applied on the outer region of tested tube. The steam supply rate was 1.0 g/cm^2-min. Due to this test parameter, the steam oxidation, balloon and rupture behavior were shown in the cladding tube in this work. The sequence of this test consisted of preheating at 300 °C, ballooning and rupture by internal tube pressure during the ramp, high-temperature steam oxidation at 1200 °C for 300 s, and water quenching at 800 °C. The internal tube pressure and the temperature variation of the tube samples were continuously measured during the test. After the LOCA test, a 4-pont bend test using a crosshead speed of 1 mm/s was applied to evaluate the bending performance of the surface ODS-treated Zircaloy-4 cladding tube.

3. Results and discussion

3.1. Microstructural characterization

In our previous study, a Y_2O_3 dispersive layer was produced on a Zircaloy-4 sheet surface using LBS [17]. However, in the current study, the Y_2O_3 dispersive layer (ODS layer) had to be applied to Zircaloy-4 cladding tubes because nuclear fuel cladding is tube-shaped, with a length of 4 m, an outer diameter of 9.5 mm, and a wall thickness of 0.57 mm. After researching ODS layer formation for fuel cladding tubes, ODS layer formation on the outer surface of the cladding tube was considered possible, whereas ODS layer formation on the inner surface was considered very difficult due to limitations of the laser equipment at the present time. Also, it was expected that the pellet cladding interaction (PCI) problem would increase if the ODS layer was formed on the inner surface of the cladding tube. Thus, the outer surface of cladding tube was chosen for ODS layer formation.