Journal of Nuclear Materials 468 (2016) 171–177

Contents lists available at ScienceDirect

Journal of Nuclear Materials

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

Influence of microstructure modification on the circumferential creep of Zr–Nb–Sn–Fe cladding tubes

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HIGHLIGHTS

- Effect of microstructure modification on creep in Zr–Nb–Sn–Fe tubes was studied.
- Creep activation energy in annealed tubes was larger than in stress-relieved tubes.
- Lower dislocation density in lager grains was observed after creep in annealed tubes.
- Larson–Miller parameter of annealed tube was larger than that of stress-relieved one.
- Creep life of tubes was extended through microstructure modification by annealing.

ARTICLE INFO

Article history:
Received 15 July 2015
Received in revised form
14 November 2015
Accepted 17 November 2015
Available online 22 November 2015

Keywords:
Zirconium alloy
Fuel cladding tube
Heat treatment
Creep properties

ABSTRACT

Out-of-reactor, non-irradiated thermal creep performances and lives of annealed and stress-relieved Zr-1.02Nb-0.69Sn-0.12Fe cladding tubes were studied and compared. The creep rates of annealed Zr-1.02Nb-0.69Sn-0.12Fe cladding tubes were appreciably slower than those of stress-relieved annealed counterpart. The stress exponent increased slightly from 5.1 to 6.1 in the stress-relieved cladding to 5.3–6.3 in the annealed cladding. The creep activation energy of the annealed Zr-1.02Nb-0.69Sn-0.12Fe alloy (300–330 kJ/mol) was larger compared to that of the stress-relieved alloy (210–260 kJ/mol). The creep activation energy of annealed alloy is close to that of self-diffusion in α-Zr (336 kJ/mol). The smaller activation energy in the stress-relieved alloy is attributed to the increasing contribution of faster diffusion path such as grain boundaries and dislocations. The presence of dislocation arrays with higher dislocation density and smaller grain size in the stress-relived alloy was confirmed by TEM analysis. The creep rupture time increased dramatically in the annealed Zr–1Nb- 0.7Sn-0.1Fe alloy compared to that of stress-relieved alloy, supporting the decrease of creep rate by annealing. The creep life of Zr-1.02Nb-0.69Sn-0.12Fe claddings can be extended through microstructure modification by annealing at intermediate temperatures in which dislocation creep dominates.

1. Introduction

One of the major current challenges in the nuclear energy industry lies in reducing maintenance and fuel cycle costs, while enhancing safety features [1]. Extending burnup, if the technology to prevent the potential fuel failure is available, may be the most economical method to meet these objectives. Potential fuel failure causes related to increased burnup include corrosion, creep and hydrogen pick-up/redistribution in zirconium alloy claddings. The Zr alloy claddings plays an important role not only as the barrier material that prevents nuclear fission products from being released into the circulating cooling water, but also as the conducting material that transfers fission-generated heat readily to the cooling water. In order to secure the nuclear fuel integrity in a more harsh environment of a higher burn-up nuclear fuel, the enhancement of high temperature creep resistance of Zr cladding tubes as well as the reductions of corrosion and hydrogen pickup are needed [2–7]. Nuclear cladding tubes should also withstand the local stress caused by the pellet-clad mechanical interaction (PCMI) acting on the inner surface of claddings as well as the stress resulting from the pressurized coolant acting on the outer surface in the corrosive environment.

Zr–Nb based alloys have been used as nuclear fuel claddings in nuclear power reactors because of their excellent corrosion

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http://dx.doi.org/10.1016/j.jnucmat.2015.11.029
0022-3115/

resistance and good mechanical properties. Nb has been reported to increase not only corrosion and radiation-induced growth [4–11] resistance but also creep resistance. The addition of Sn is known to increase the creep resistance and high temperature mechanical strength, but to decrease in-reactor corrosion resistance [2,8,9,11]. The decrease of Sn content in Zr–Nb–Sn–Fe alloys from 1.0 wt. % (Zr-1.0Nb-1.0Sn-0.1Fe) to 0.7 wt. % (Zr-1.0Nb-0.7Sn-0.1Fe) is known to enhance the corrosion resistance, but at the expense of the creep resistance [11]. The performance of Zr alloy cladding tubes can be modified by the modification of alloying elements as well as by modification of thermo-mechanical processing schedules. The microstructural details of Zr alloy claddings including the dislocation density, the grain size, the crystallographic texture, the grain shape, and the distribution and size of second phase particles are greatly modified by the accumulation of the cold work energy by cold pilgering and the release of it by heat treatment [6,8].

Post-cold-pilgering heat treatment of Zr alloy claddings is known to decrease the tensile strength, but to enhance the resistances to corrosion and irradiation-induced growth [6,8]. Some Zr alloy tube manufacturers heat-treat cladding tubes under stress relieving condition to enhance the strength and creep properties whereas other manufacturers heat-treat cladding tubes under recrystallization conditions to improve the corrosion resistance properties [6,10] and irradiation growth resistance. Creep resistance and mechanical performance of cladding tubes can be modified not only by the modification of the alloy composition, but also by microstructure modifications in the same Zr alloys [10,11]. In this study, the effects of high temperature annealing and the resultant microstructural evolution on the non-irradiated, out-of-reactor, thermal creep resistance of Zr-1.0Nb-0.7Sn-0.1Fe cladding tubes were studied.

2. Experimental details

Zr–Nb–Sn–Fe alloy cladding tubes used in this study with lower Sn content than in regular Zirlo™ (trademark of Westinghouse Electric Co.) have the composition of 1.02 wt.% Nb, 0.69 wt.% Sn, 0.12 wt.% Fe, 0.13 wt.% O and balance Zr. Zr-1.02Nb-0.69Sn-0.12Fe alloy cladding tubes were cold-pilgered with two intermediate heat treatments and finally stress-relieved at 460 °C for 7 h after final cold pilgering. Some cladding tubes were further annealed at 700 °C for 3 h to induce the grain growth. These cladding tubes with different heat treatments will be called stress-relieved and annealed cladding tubes hereafter. It should also be noted that the monotectoid temperature is 610 °C below which βZr phase transforms to αZr + βNb phases [12]. The effect of the heat treatment above (annealed) and below (stress-relieved) the monotectoid temperature on the formation of second phase particles and the grain size will also be studied.

In order to investigate the circumferential thermal creep behaviors of the Zr-1.02Nb-0.69Sn-0.12Fe alloy cladding tubes, the Zr alloy tubes were sectioned perpendicularly to the tube axis in lengths the width of 4 mm. The experimental details of the circumferential creep and mechanical testing are described elsewhere [2,3,9]. In this study, a set of specially designed grips with two half-cylinders was used to strain the cladding tube specimen circumferentially while keeping a round shape of the ring specimens in contact with the specially designed grips. It was experimentally confirmed that the deformation occurred in the left and right-hand sides of the ring within 5 mm width centered on the split line of two half-cylinders and the optimum gage length was assumed to be 5 mm [2,3,9].

Constant stress creep testing machines with the Andrade-Chalmers lever type loading cam profile designed to maintain the constant stress [2] in the creep specimens were used to examine the influence of microstructure modification on creep deformation properties. The creep tests were performed at 450, 475 and 500 °C with the applied stress between 80 MPa and 150 MPa. The temperatures and stresses in the present study were chosen for testing convenience and greater than those expected under normal reactor operating conditions. However, these experimental conditions can be used to study the effect of microstructure modification on the creep life for dislocation creep within a reasonable timeframe (up to 67 days for a single test) and applicable to creep behaviors at lower temperatures. For microstructural analyses, transmission electron microscopy (TEM) specimens were prepared from the stress-relieved and annealed specimens. Using a twin jet polisher with the perchloric acid (10%)–ethanol solution in the temperature of −35 to −45 °C, 3 mm disks were electro-polished until perforation with a voltage of 12–20 V. TEM specimens were examined by a TEM (JEOL-2010) equipped with an energy dispersive X-ray spectroscopy (EDS) detector with an accelerated voltage of 200 kV. For fracture surface analyses, scanning electron microscopy (SEM) were performed using a JEOL JSM-7000F.

3. Results and discussion

Typical TEM microstructures of the stress-relieved (a) and annealed (b) Zr-1.02Nb-0.69Sn-0.12Fe cladding tubes showing the grain morphology and second phase particles are exhibited in Fig. 1. The stress-relieved Zr alloy displayed small equi-axed grains with partially unrecrystallized regions with dislocations (marked by thick white arrows). The regions with dislocation tangles in the stress-relieved tubes are caused by dislocation generation and motion during the pilgering process. In the annealed Zr-1.02Nb-

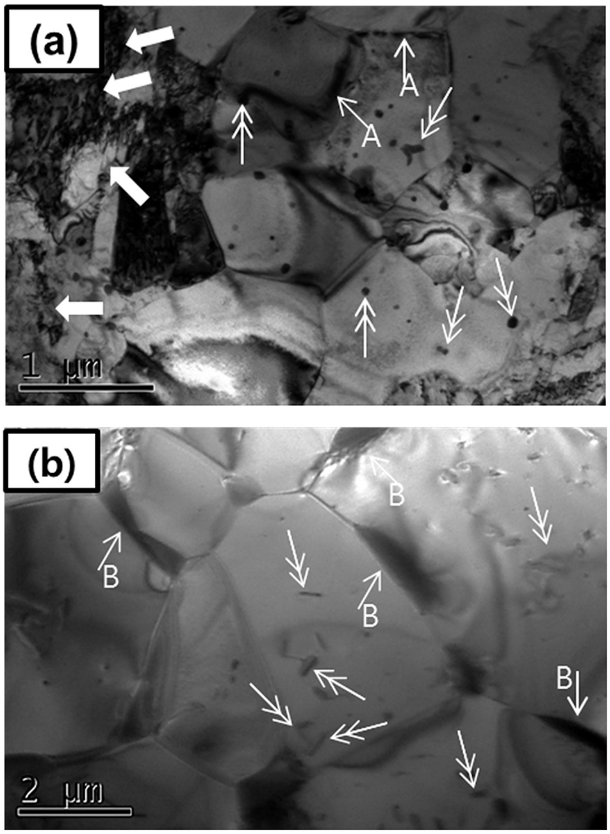

Fig. 1. TEM microstructure of the stress relieved Zr-1.02Nb-0.69Sn-0.12Fe (a) and annealed Zr-1.02Nb-0.69Sn-0.12Fe (b).