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Microstructure evolution and recrystallization behavior of cold-rolled Zr−1Sn−0.3Nb−0.3Fe−0.1Cr alloy during annealing

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Abstract: The effects of cold-rolling reduction, annealing temperature, and time on recrystallization behavior and kinetics of cold-rolled Zr−1Sn−0.3Nb−0.3Fe−0.1Cr alloy were investigated using the Vickers hardness test, scanning electronic microscopy (SEM), transmission electron microscopy (TEM) and electron backscatter diffractometry (EBSD). The results show that the rate of the recrystallization increased with increasing annealing temperature and rolling reduction. Recrystallized grains nucleated preferentially at sites with high density dislocation and deformation stored energy and then grew into integral grains. Recrystallization texture changed from $\langle 10\bar{1}0\rangle$ //RD to $\langle 11\bar{2}0\rangle$ //RD. The grain orientation changed from random orientation to the orientation with the maximum misorientation around 30°. Recrystallization kinetics and maps were constructed based on the Johnson–Mehl– Avrami–Kolmogorov (JMAK) equation to derive parameters sensitive to the microstructure. The activation energies for recrystallization of 30%, 50% and 70% cold-rolling reductions were determined to be 240, 249 and 180 kJ/mol, respectively.
Key words: zirconium alloy; recrystallization; microstructure; texture evolution; kinetics

1 Introduction

Zirconium alloys have been used in nuclear reactors as fuel cladding for several decades owing to their low absorption cross-section for thermal neutrons, acceptable mechanical properties, and excellent corrosion resistance. However, in order to reduce the cost of nuclear power, it is necessary to extend the refueling cycle. To meet the requirements of increased burn-up and extended fuel cycle, new zirconium alloys have been developed and tested under out-of-pile and in-pile conditions. These alloys are synthesized by adding alloying elements such as Fe, Cr, Ni and Cu in Zr−Sn−Nb alloys, resulting in ZIRLO (Zr−1.0Sn−1.0Nb−0.1Fe) [1], M5(Zr−1.0Nb) [2], E635 (Zr−1.3Sn−1.0Nb−0.35Fe) [3] or Zr−1Sn−0.3Nb−0.3Fe−0.1Cr alloy [4]. In particular, the Zr−1Sn−0.3Nb−0.3Fe−0.1Cr alloy is a kind of Zr−Sn−Nb alloy with low Nb content. Compared to the zirconium alloys with high Nb content, the Zr−1Sn−0.3Nb−0.3Fe−0.1Cr alloy can undergo relatively high thermal mechanical processes. However, when preparing the alloy it is beneficial to obtaining a fine and uniform distribution of recrystallized microstructure and second phase particles that have a great influence on the corrosion resistance [4−6]. Therefore, the alloy processing must be further looked into to guarantee a uniform microstructure.

Service conditions of nuclear reactors require that the zirconium alloy has a uniform microstructure after processing, which involves smelting, hot working, intermediate annealing, cold working, and recrystallization annealing. The final annealing step is necessary for releasing the residual stresses and/or improving the mechanical properties and corrosion resistance [4,7−9]. This is an important step, since it has been clearly established that the recrystallized Zr−1Sn−0.3Nb−0.3Fe−0.1Cr alloy is more suitable for nuclear applications than the as-deformed ones due to the higher irradiation creep resistance and lower irradiation growth [4,10]. In addition, the recrystallized

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microstructure can restore the ductility of Zr−Sn−0.3Nb−0.3Fe−0.1Cr alloy, which facilitates the following manufacturing operations of cladding materials. Therefore, it is important and necessary to investigate the influence of annealing on recrystallization mechanisms and microstructural characteristics of Zr−1Sn−0.3Nb−0.3Fe−0.1Cr alloys.

During the past few decades, several works have been devoted to study the recrystallization mechanisms of zircaloy [8,10−14] and the newly developed Zr−Sn−Nb alloys [4,15] after cold rolling or dynamic recrystallization. LUAN et al [11] studied the growth behavior of second phase particles in Zr−Nb−Sn−Fe−Cr−Cu alloys during aging. They found that linear distribution features of the second phase particles weaken with longer aging time or higher temperatures. Despite the importance of second phase particles, the recrystallization after cold working is responsible for the formation of final microstructure of Zr−1Sn−0.3Nb−0.3Fe−0.1Cr alloy. JUNG et al [10] investigated the recrystallization behavior of HANA-4 and compared with the HANA-6 and constructed the recrystallization maps. LIU et al [13] investigated the microstructural evolution of Zr−Nb−Sn−Fe−Cr alloy at 590 °C for 1 and 2 h, which is insufficient to characterize the details of the recrystallization process. However, as an important cladding material, the recrystallization behavior of Zr−1Sn−0.3Nb−0.3Fe−0.1Cr alloy is still not fully understood.

In this work, the aim is to investigate the recrystallization behavior, microstructure, texture evolution, recrystallization mechanism and kinetics of the Zr−1Sn−0.3Nb−0.3Fe−0.1Cr alloy. In order to do so, the alloy was subjected to different deformation degrees before annealing, which also provides a reference to the hot and cold processing of this new developed Zr-alloy.

2 Experimental

The materials used were received from the Nuclear Power Institute of China, which were hot rolled plates with 5 mm in thickness. Three different reductions of 30%, 50% and 70% were applied in the cold rolling to achieve the thickness values of 3.5, 2.5 and 1.5 mm, respectively. The material was able to accommodate large strain without any evidence of cracking or delamination. The annealing processes subsequent to every cold-rolling were conducted at temperatures of 530, 560 and 580 °C for various time.

The samples were annealed under vacuum atmosphere for the isochronal and isothermal annealing and the annealing parameters are shown in Table 1. The oxidation during annealing was prevented by using quartz tubing for the vacuum. The samples for annealing were cut into 10 mm × 8 mm pieces along the rolling direction.

The hardness of the specimens was measured by a Vickers hardness tester (HV−10B, China) with a load of 3 kg for 15 s. Prior to the hardness tests, the examined surfaces were metallographically prepared using SiC papers (4000 grit in the final step), and then etched with a 10%HF−45%HNO_3−45%H_2O (volume fraction) solution. The hardness was measured on the rolling direction (RD)–transverse direction (TD) plane to evaluate the extent of recrystallization. For the determination of the hardness, 15 measurements were carried out to obtain an average value for each sample. A scanning electron microscope (SEM, FEI-Sirion 200) was employed to characterize the microstructure in RD–ND plane of the specimens. Prior to SEM observations, electro-polishing was conducted at −20 °C under 17 V in a 95%C_2H_5OH−5%$HClO_4$ (volume fraction) solution.

The microstructure and texture were characterized at various stages of annealing and several locations through the thickness of the sample. A combination of two techniques was used in order to have a complete picture of the microstructures present. EBSD was used to measure the grain size, the distribution of grain boundary misorientations and the texture on the surface. A large EBSD data set was acquired on this recrystallized material using an HKL-Technology system (Channel 5 software) (Oxford Instruments HKL, Hobro, Denmark) coupled to a JELL−6300 SEM (scanning electron microscopy). Sample preparation for EBSD measurement included grinding with increasingly finer SiC papers of 240, 600, 800, 1000, 2000, 3000, and 5000 grit size, followed by polishing with 3.5 μm diamond paste. Special care was taken to avoid the formation of twins during grinding and polishing. The samples were further etched with a solution of nitric acid (45%), distilled water (45%) and hydrofluoric acid (10%) for

Table 1 Annealing parameters of cold-rolled Zr−1Sn−0.3Nb− 0.3Fe−0.1Cr alloy sheets

Temperature/°C	Time										
	1 min	3 min	5 min	10 min	20 min	30 min	1 h	2 h	3 h	5 h	10 h
530	×	×	√	√	√	√	√	√	√	√	√
560	√	√	√	√	×	√	√	√	√	√	√
580	√	√	√	√	×	√	√	√	√	√	√

Experiments that have been done are marked with "√", and those have not been done are marked with "×"