Microstructure and Hydrogen Solubility Effects on the Creep Behavior of Zircaloy-4

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1. Introduction

Spent nuclear fuel (SNF) undergoes a vacuum drying process before being placed in dry storage. During this process, the cladding is exposed to elevated temperatures and internal pressure due to the presence of helium gas and various fission products. As a result, the cladding experiences creep, a time-dependent deformation that occurs under sustained stress. Understanding the creep behavior of hydrogen-containing cladding is essential for ensuring the structural integrity and safety of spent fuel storage systems.

However, previous studies have not sufficiently addressed the effects of varying hydrogen concentrations on creep behavior under consistent temperature and pressure conditions, especially when considering terminal solid solubility for dissolution (TSSD) as a key parameter. Additionally, creep behavior is expected to vary depending on both cladding type and recrystallization extent [1]. This study aims to evaluate the creep response of low-burnup Zircaloy-4 spent fuel cladding, which typically exhibits hydrogen concentrations, by conducting systematic creep tests that consider the influence of recrystallization.

2. Methods and Results

2.1 Material preparation

Cold-worked stress-relieved (CWSR) Zircaloy-4 cladding tubes, with an outer diameter of 9.5 mm, a thickness of 0.57 mm, and a gauge length of 4 mm, were used in this study. To obtain specimens with different microstructures and grain sizes, ring-shaped samples underwent annealing in a Sieverts apparatus.

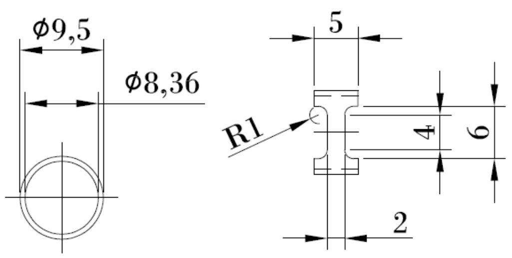

Fig. 1. Schematic of ring specimens.

2.2 creep test

Creep strain was measured using a linear variable differential transformer (LVDT) positioned at the center of the equipment, connected to the main controller. Instead of a standard dog-bone specimen, a custom-designed semicylindrical jig was used for ring specimens.

Before mounting the ring specimen onto the jig, a thin layer of lubricant was applied to the contact surface to minimize friction and ensure uniform stress distribution [2]. Temperature monitoring was performed using a comprehensive system with K-type thermocouples. The test procedure started with heating the specimen to 450°C at a rate of 5°C/min, followed by a 1-hour stabilization period. After stabilization, a load of 27.9 kg, corresponding to 120 MPa of stress, was applied.

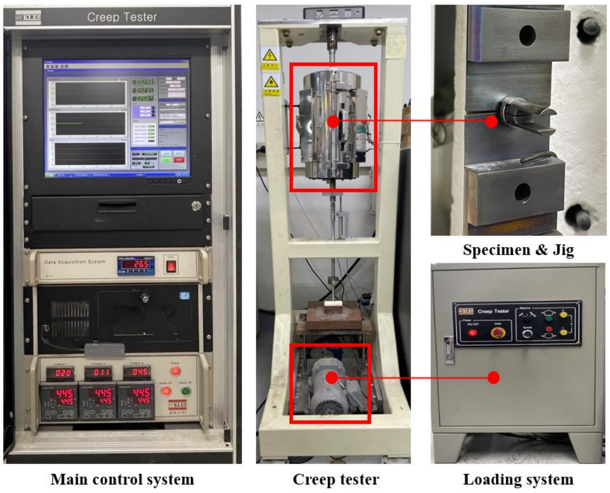

Fig. 2. Creep testing equipment and jig assembly.

2.3 Results

This study examines the effect of hydrogen concentration on the creep behavior of Zircaloy-4 specimens with varying grain sizes. The samples underwent different annealing treatments, including CWSR and heat treatments at 500°C for 2, 6, 12, and 24 hours, as well as at 700°C for 24 hours, followed by hydrogen charging. Grain sizes were analyzed using Electron Backscatter Diffraction (EBSD), with the results summarized in Table 1. In general, grain growth begins after recrystallization, as newly formed recrystallized grains merge, consuming adjacent grains and increasing in size. As grain size increases, the number of grains decreases, leading to a reduction in grain boundary area and a corresponding decrease in total surface energy. A clear correlation was observed between higher annealing temperatures and durations with an increase in average grain size.

Table 1. Average grain size of Zircaloy-4 at different annealing durations

CWSR	
As-received	1.22 ㎛
Annealed at 500°C	
For 2 hours	1.78 ㎛
For 6 hours	1.97 ㎛
For 12 hours	2.6 ㎛
For 24 hours	3.12 ㎛
Annealed at 700°C	
For 24 hours	5.59 ㎛

Fig. 3 and Fig. 4 illustrate the time-strain curves for all creep-tested specimens under each annealing condition. Rupture strain showed no significant dependence on hydrogen concentration.

Analyzing the effect of hydrogen content, the creep tests of CWSR and 500°C-6h annealed specimens revealed that increasing hydrogen content led to a decrease in the secondary creep rate and an increase in rupture time. However, for the 500°C-12h and 700°C-24h annealed specimens, the secondary creep rate increased while rupture time decreased as hydrogen content approached the TSSD. Beyond TSSD, further hydrogen uptake resulted in a reduction in the secondary creep rate and an extension of rupture time.

By controlling the degree of recrystallization, the relationship between hydrogen content and grain size is evaluated, showing consistency with previous trends in zirconium-based cladding materials. However, a deviation in creep behavior is observed in hydrogenated cladding when the grain size increases from 1.97 μm after recrystallization at 500°C for 6 hours to 2.6 μm after 12 hours.

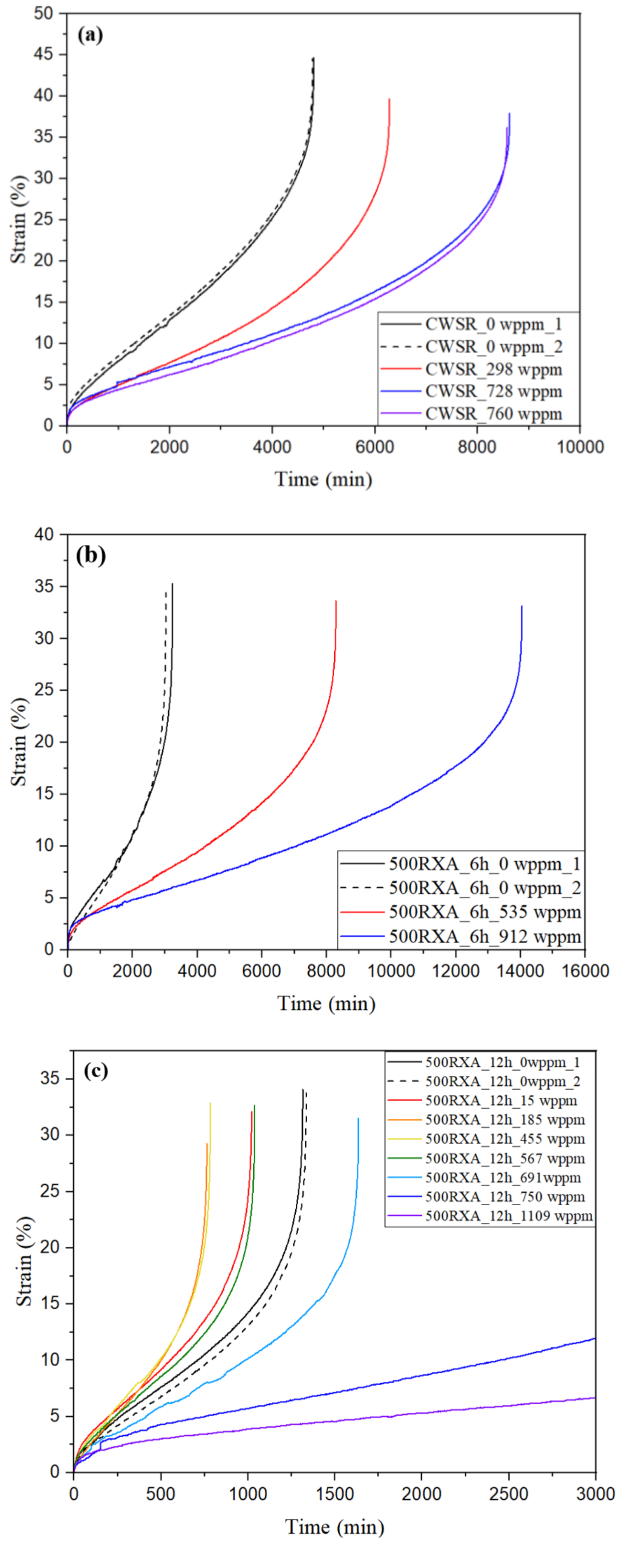

Fig. 3. Creep behavior of Zircaloy-4 at different hydrogen concentrations under 450°C and 120 MPa: (a) cold-worked stress-relieved specimen, (b) recrystallized specimen annealed at 500°C for 6 hours, (c) recrystallized specimen annealed at 500°C for 12 hours.

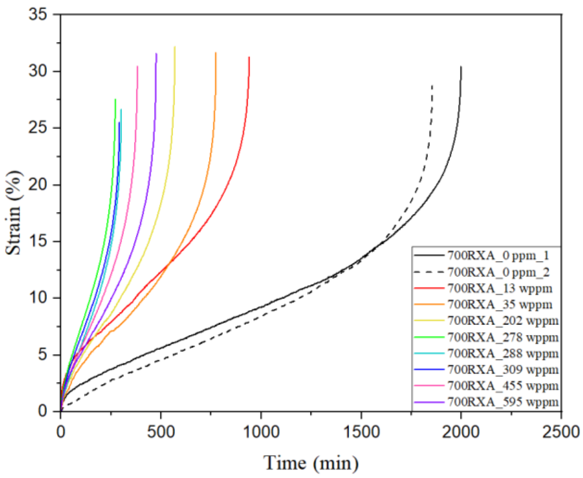

Fig. 4. Creep behavior of recrystallized Zircaloy-4 annealed at 700°C for 24 hours at different hydrogen concentrations, tested at 450°C and 116 MPa.

3. Conclusions

This study examines the NRC's regulatory guidelines for low-burnup fuel cladding, focusing on how hydrogen concentration influences creep behavior in CWSR and RXA Zircaloy-4 with varying grain sizes.

In CWSR cladding, high defect density leads to localized lattice distortion when hydrogen dissolves and migrates near defects, raising activation energy for dislocation movement and enhancing creep resistance. Additionally, its anisotropic structure, formed during pilgering, promotes hydride interconnectivity upon precipitation, further increasing resistance. As a result, creep resistance improves with increasing hydrogen concentration.

In contrast, RXA cladding has lower defect density, allowing hydrogen to distribute more uniformly with minimal lattice distortion. This weakens the inhibition of dislocation motion, lowering activation energy and facilitating deformation. However, once hydrogen concentration exceeds the TSSD, hydride precipitation restricts dislocation movement, initially reducing resistance but recovering as hydrogen content increases.

These findings have regulatory implications, as NRC guidelines permit temperature excursions up to 570°C, where recrystallization could accelerate creep. Future studies will extend similar creep tests (450°C, 120 MPa) to HANA cladding, which has a larger grain size than Zircaloy-4 cladding, to evaluate whether its behavior follows the observed trends. By improving the understanding of hydrogen's impact on creep, this research contributes to the development of standards ensuring the safety and reliability of nuclear fuel cladding.

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