In-situ TEM observation of the dissolution and re-precipitation behavior of hydrides in Zircaloy-4

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Abstract
This work presents delta hydride dissolution and re-precipitation behaviors in Zircaloy-4 by in-situ heating and cooling observation in the transmission electron microscope (TEM). The hydride dissolves in the range of 473-673 K, depending on the hydrogen content. A dislocation loop is left nearby where the hydride finally disappears. The re-precipitated hydrides, normally exhibit a different growth direction from the original ones, preferentially nucleate nearby the dislocation loop during fast cooling.

Keywords: Zircaloy-4, hydride, dissolution, precipitation, in situ observation

1. Introduction

Zirconium alloys exhibit a strong affinity of hydrogen, which leads to hydride precipitated out from the cladding tubes after a long-time service. As a brittle phase, the formation of hydrides is considered to be detrimental to materials' performance, especially under stress. Although the critical solid solution and critical precipitation temperature of hydrides were determined by the previous studies, there is a lack of detailed descriptions about the dissolution and re-precipitation behaviors of individual hydrides. In situ electron microscopy is an intuitive and effective method to study the dissolution and precipitation of hydrides. However, the work on this aspect is relatively limited. Therefore, the purpose of this study is to investigate the decomposition and re-precipitation behaviors of hydrides in Zircaloy-4 with different hydrogen contents by in-situ TEM observation during heating and cooling.

2. Experiment

Zircaloy-4 cladding tube was used as experimental material. It was firstly cold-rolled to a sheet and annealed at 600°C for 4 h to acquire a fully recrystallized microstructure. The φ-3mm disk specimens were prepared and subsequently subjected to hydrogen-charging processing performed in a vacuum furnace under a gas atmosphere (3.5 vol.% H in Ar) at 673 K with various pressures. In-situ TEM observation was performed using a JEM-2000F microscope and a Gatan heating holder. The heating and cooling rates were estimated as 4 K/min (up to 623 K) and 24 K/min.

3. Conclusion

Results showed that the dissolution behaviors of hydride were different, depending on the absorbed hydrogen content. In the low hydrogen content case, the hydrides completely decomposed at ~523 K, while in the high hydrogen case, the hydride partially decomposed even at 623 K. The hydrogen moves into the matrix during heating through bulk diffusion or channel (dislocation and grain boundary) diffusion. It is also noted that after the dissolution of hydride, a dislocation loop was always left nearby, which might play a role in promoting the re-precipitation of hydride upon cooling because of the Korotkoff effect. The structure and crystallographic features of the re-precipitated hydrides were analyzed and compared with the original one. Moreover, the effect of cooling rate on the precipitation of hydrides will be also discussed.