Effect of Crack Tip Stresses on Delayed Hydride Cracking in Zr-2.5Nb Tubes

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1. Introduction

Delayed hydride cracking (DHC) tests have shown that the DHC velocity becomes faster in zirconium alloys with a higher yield stress [1-3]. To account for this yield stress effect on the DHC velocity, they suggested a simple hypothesis that increased crack tip stresses due to a higher yield stress would raise the difference in hydrogen concentration between the crack tip and the bulk region and accordingly the DHC velocity [2,3]. This hypothesis is also applied to account for a big leap in the DHC velocity of zirconium alloys after neutron irradiation [4]. It should be noted that this is based on the old DHC models [5.6] that the driving force for DHC is the stress gradient. Puls predicted that an increase in the yield stress of a cold-worked Zr-2.5Nb tube due to neutron irradiation by about 300 MPa causes an increase of its DHC velocity by an order of magnitude or 2 to 3 times depending on the accommodation energy values [6].

Recently, we proposed a new DHC model that a driving force for DHC is not the stress gradient but the concentration gradient arising from the stress-induced precipitation of hydrides at the crack tip [7-9]. Our new DHC model and the supporting experimental results have demonstrated that the DHC velocity is governed primarily by hydrogen diffusion at below 300 ^oC [7,9]. Since hydrogen diffusion in Zr-2.5Nb tubes is dictated primarily by the distribution of the β-phase, the DHC velocity of the irradiated Zr-2.5Nb tube must be determined mainly by the distribution of the β-phase, not by the increased yield stress [9], which is in contrast with the hypothesis of the previous DHC models[1-3].

In short, a controversy exists as to the effect on the DHC velocity of zirconium alloys of a change in the crack tip stresses by irradiation hardening or cold working or annealing. The aim of this study is to resolve this controversy and furthermore to prove the validity of our DHC model. To this end, we cited Pan et al.'s experiment [10] where the delayed hydride cracking velocity, the tensile strengths at 240 ^oC and the Nb concentration in the β-Zr phase for the same Zr-2.5Nb tube specimens were investigated as a function of neutron fluence (E > 1MeV) after irradiation tests at lower temperatures ranging from 250 to below 285 ^oC in a high flux reactor.

2. Results and discussion

Figs. 1 and 2 show the DHC velocity and the tensile strengths at 240 ^oC for the same Zr-2.5Nb tube with increasing neutron fluences up to 26.1x10^{25} n/m^2, which were reproduced using Pan's data [10]. The DHC velocity at 240 ^oC increased gradually at fluences below 5x10^{25} n/m^2 and leveled off to a constant value above it while the transverse tensile stress at 240 ^oC for the same tube had a sharp rise at below 0.5x10^{25} n/m^2 and then a very slow increase with increasing fluences. In other words, the neutron fluence dependency of the DHC velocity had nothing to do with that of the tensile stress, which is in contrast with the old DHC models' hypothesis. If the crack tip stresses governed the DHC velocity as with the old DHC models, then, the former should have been similar to the latter since the crack tip stress increases proportionally with the increased tensile stresses due to radiation hardening.

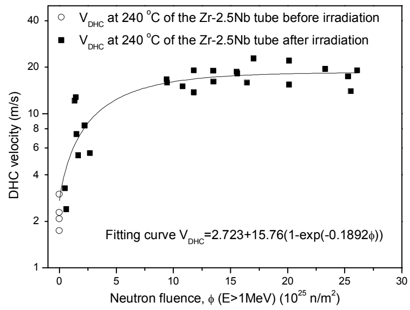

Fig. 1. DHC velocity at 240 ^oC of the Zr-2.5Nb with neutron fluences (E>1MeV) after irradiation tests in a high flux reactor at 250 to below 285 ^oC (reproduced using Pan's data [10]).

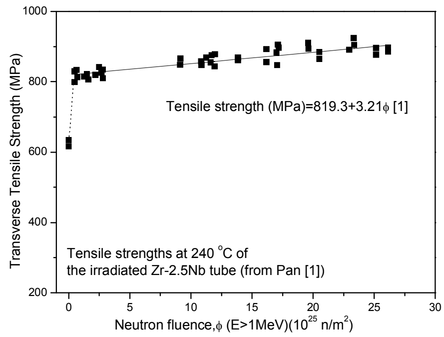

Fig. 2. Transverse tensile strengths at 240 ^oC with neutron fluences (E> 1MeV) of the same Zr-2.5Nb tube as described in Fig. 2 (reproduced using Pan's data [10]).

Accordingly, the results shown in Figs 1 and 2 demonstrate that the DHC velocity of the zirconium alloys is not dictated by the tensile strengths or the crack tip stresses. This finding definitively corroborates that the old DHC models are illogical.

If hydrogen diffusion governs the DHC velocity as Kim has suggested [9], then, the neutron fluence dependency of the DHC velocity should follow that of the distribution of the β-Zr in the Zr-2.5Nb tube which greatly affects the hydrogen diffusivity [9,11]. Fig. 3 shows the neutron fluence dependency of the Nb concentration in the β-Zr of the same Zr-2.5Nb tube. As expected, the neutron dependency of the Nb concentration in the β-Zr was indeed similar to that of the DHC velocity. Consequently, it is demonstrated that the DHC velocity of the Zr-2.5Nb tube with α-Zr and β-Zr phases is mainly governed by the distribution of the β-Zr or hydrogen diffusion, not by the crack tip stresses that are determined by the yield or tensile stresses of the zirconium matrix. Accordingly, it is concluded that our DHC model is convincing.

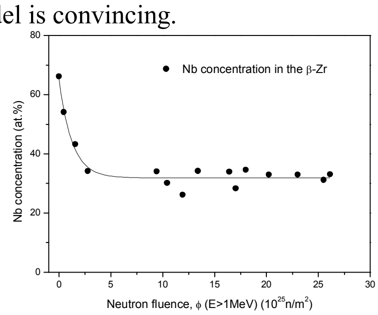

Fig. 3. Nb concentrations in the β-Zr in the same Zr-2.5Nb tube with neutron fluences (E>1MeV) as that described in Fig. 2 (reproduced using Pan's data [10]).

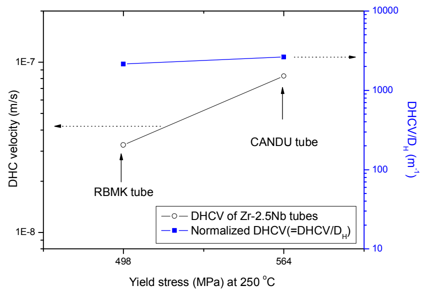

Fig. 4. DHC velocity at 250 ^{o}C of the CANDU and RBMK Zr-2.5Nb tubes with their yield strengths before and after normalization by D_H.

Though many experiments have shown that the DHC velocity of zirconium alloys appear to increase with increasing the yield stress [1-3], these observations may be fortuitous due to the ignorance of a change in hydrogen diffusivity or D_H accompanying annealing treatment or neutron irradiation introduced to change the yield stress of the zirconium alloys [11-12]. To discriminate the yield stress effect on the DHC velocity, it would be better to normalize the DHC velocity by all the factors, as described above, other than the yield strength. Using the DHC tests' results [9], we plotted the yield stress dependencies of the DHC velocity at 250 ^{o}C before and after normalization by D_H for the CANDU and RBMK Zr-2.5Nb tubes as shown in Fig. 4. Before normalization by D_H, the DHC velocity at 250 ^{o}C of the Zr-2.5Nb tube apparently increased with increasing the yield stress. However, normalization by D_H of the DHCV velocity for the CANDU and RBMK Zr-2.5Nb tubes lessened the yield stress dependency so that the normalized DHCV of the Zr-2.5Nb tube became rather flat independent of the magnitude of the yield stress. Consequently, it is clear that the crack tip stress effect on the DHC velocity is of little importance than D_H at temperatures below 300 ^{o}C and that our DHC model is valid.

3. Conclusion

In contrast to the old DHC models, Pan et al.'s in-reactor test results showed that the DHC velocity of a cold-worked Zr-2.5Nb tube is dictated by the Nb concentration in the β-Zr or the decomposition of the β-Zr phase, not by the tensile stress or the crack tip stress. This fact demonstrates that the hydrogen diffusion through the β-Zr phase is more influential in governing the DHC velocity than the crack tip stress, which is consistent with our new DHC model. Furthermore, the DHC velocities at 250 ^{o}C of the CANDU and RBMK tubes that seemed to increase with increasing the yield stress or the crack tip stress was found to be relatively flat against the yield stress after their normalization by D_H. Conclusively, it is clear that the crack tip stress on the DHC velocity of zirconium alloys is of little importance, proving the validity of our new DHC model.

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