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# Effect of unloading on crack growth rate of Zr–2.5Nb tubes

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ABSTRACT

Crack growth rates (CGRs) of a heat-treated Zr–2.5Nb tube were determined using compact tension specimens with 60 ppm *H* at 250 °C under the constant and cyclic loads where the load ratio *R* was changed from 0.13 to 0.68. CGR was the highest under the constant load and decreased under the cyclic load with decreasing *R* despite a decrease of the critical hydride length indicating the enhanced rate of hydride cracking. Hence, the decreased CGR under the cyclic load is due to unloading during the cyclic load inducing the compressive stress at the crack tip. This compressive stress suppresses hydride nucleation rate, leading it to govern the CGR, according to Kim's new model. Evidence is provided by citing Simpson's experiment demonstrating that unloading from 15 MPa $\sqrt{m}$ decreased the CGR of a cold-worked Zr–2.5Nb tube but annealing did the reverse. This study demonstrates for the first time that the retarded CGR due to an overload during the DHC tests is understood in view of crack growth kinetics using Kim's model.



## 1. Introduction

Delayed hydride cracking (DHC) of zirconium alloys is a subcritical crack growth mechanism that requires the following three processes: nucleation, growth and cracking of hydrides [1–3]. In other words, all the three processes should occur consecutively for a crack to grow in any hydride forming metals. Thus, the DHC velocity or crack growth rate (CGR) is governed by the rate of the slowest process among them, which is the core of Kim's new DHC model [1,2]. Nevertheless, all the DHC models [4–7] have proposed that DHC is simply a diffusion-controlled process ignoring the rates of nucleation and cracking of hydrides. They have claimed that CGR is governed either by the rate for the hydride crack tip solubility to increase to the terminal solid solubility (TSSP) [7–9] or by the hydride growth rate [4,5] both of which are determined primarily by hydrogen diffusion, assuming that hydrogen diffusion is dictated not only by temperature but also by the stress gradient or the difference in applied stresses [4–6]. These imperfect claims are the cause of many unresolved issues related to DHC that the old DHC models cannot explain to date: the constant crack growth rate with $K_I$ [10], a rapid drop of the CGR above 300 °C [11] despite higher hydride growth rates at higher temperatures, DHC arrest above 180 °C when approached by heating [11], the activation energy for CGR [12], a linear increase of CGR with hydrogen supersaturation and with load ratio, *R* under the cyclic load [13,14], the anisotropic CGR with orientation [12,15], the neutron fluence dependence of CGR in Zr–2.5Nb tubes [16], the effect of striation spacing on CGR [17], and an increase of the threshold stress intensity factor or $K_{IH}$ and a decrease of CGR after unloading when compared to that before unloading [10]. Recently, Kim [1,2,18–20] put forth a new DHC model termed Kim's model demonstrating for the first time that the CGR of zirconium alloys is kinetically determined by the rate of the slowest process among the three processes involved in DHC: nucleation, growth and cracking of hydrides and the driving force is the gradient of hydrogen concentration. Thus, the difference between the old DHC models [4–9] and Kim's new model [1,2,18–20] is in the driving force for DHC and the rate-determining step of the CGR: the former claims that the CGR is determined simply by the one process or the so-called diffusion-controlled process and the driving force for DHC is the stress gradient but the latter shows that the CGR is governed by the rate of the slowest process among the three processes involved in DHC and the driving force is the concentration gradient. If the CGR were determined only by the diffusion-controlled process like the old DHC models, a rapid decrease of CGR above 300 °C for a Zr–2.5Nb tube could not be understood with the old DHC models. Furthermore, considering that zirconium alloys with hydrogen is a closed system, the stress applied at a crack tip cannot cause hydrogen to diffuse there from the bulk unless cooling is applied: an analogy is that milk cannot come out of a milk carton despite a pumping force being applied through a straw if it is a closed system [18–20]. This fact definitively shows that the old DHC models claiming that the stress gradient is a driving force for hydrogen to move to the stressed region from the unstressed region are incorrect, leading to an incomplete understanding of DHC in zirconium alloys. Using Kim's model, however, Kim has elucidated all of these unresolved issues described above [1,2,13,21–28] except the last

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issue that remains yet to be resolved [9]. More detailed explanations about the differences between two DHC models have been given in [18–20].

In most practical cases, a tensile overload retards not only the CGR in zirconium alloys during the DHC tests but also the fatigue crack growth rate in metals [29,30], the cause of which is unclear to date. A considerable decrease in the fatigue crack growth rate due to overload is suggested to occur due either to the crack closure [31,32] or to compressive stresses or strains [33,34] arising from unloading of the overload. However, the role of the crack closure or the compressive stress in the crack growth rate remains yet to be understood because of incomplete understanding of crack growth kinetics. The aim of this study is to resolve the effect of unloading on the CGR of zirconium alloys, which comes in last among the unresolved issues as listed above. To this end, CGRs of the Zr–2.5Nb tubes were determined at a constant temperature under the cyclic load with the load ratio, $R$ changing from 0.13 to 0.66 where the extent of unloading became higher at the lower $R$. More direct evidence for the effect of unloading after an overload is provided using Simpson's experiment [10] investigating the effect on the CGR of a Zr–2.5Nb tube of the stress states of the pre-fatigue crack tip by unloading or annealing after the formation of a pre-fatigue crack.

## 2. Experimental procedures

The DHC tests were conducted at a constant temperature of 250 °C under the cyclic and constant loads on the 17 mm compact tension specimens shown in Fig. 1 that were taken from a heat-treated Zr–2.5Nb tube (RBMK type) [13,14]. These specimens were pre-charged to 60 ppm of hydrogen using an electrolytic method followed by homogenization treatments and then pre-fatigued to introduce a 1.7 mm fatigue crack at load ratio, $R$ of 0.1. The maximum stress intensity factor, $K_I$ was 12 MPa $\sqrt{m}$ at the initial stage of the pre-fatigue crack and then decreased to 10 MPa $\sqrt{m}$ after the fatigue crack grew to 1.7 mm. A thermal cycle was applied during the DHC tests where the test temperature of 250 °C was approached by cooling from 315 °C at a cooling rate of 1.5 °C/min. To eliminate the fatigue crack tip effect on the CGR, a DHC crack

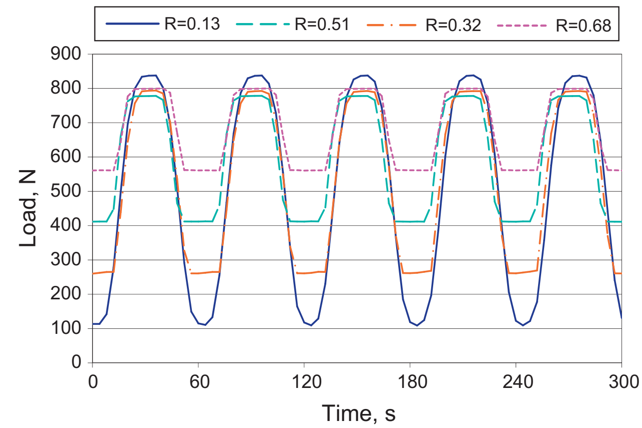


**Fig. 2.** Loading curves of 1 cycle/min with the load ratio $R$ changing from 0.13 to 0.68.

was made ahead of a fatigue crack by applying 15 MPa $\sqrt{m}$ for 15 min upon arrival at 250 °C. Then, the tension–tension cyclic loads were applied with 1 cycle/min to the CT specimens where load ratio, $R$ was varied from 0.13 to 0.68 by changing the minimum load, as shown in Fig. 2. Furthermore, the constant load termed $R = 1$ was applied to investigate the effect of the absence of unloading on the CGR. Note that the maximum load under the cyclic and constant loads was kept the same irrespective of $R$ (Fig. 2). The detailed test conditions for cyclic loading are given in Table 1 [13,14]. Crack growth during the DHC tests was monitored using a potential drop versus crack length calibration curve. After completion of the DHC tests, the actual crack lengths were determined from the fracture surfaces using a stereoscope. Details of the DHC test method are given elsewhere [35].

To corroborate the effect of unloading, we cited Simpson's experiment [10] where the CGR of a cold-worked Zr–2.5Nb tube (CANDU type) were determined under the constant load at 120 °C with and without unloading from 15 MPa $\sqrt{m}$, or after annealing at 475 °C. The DHC tests were conducted on the as-received compact tension (CT) specimens taken from the CANDU

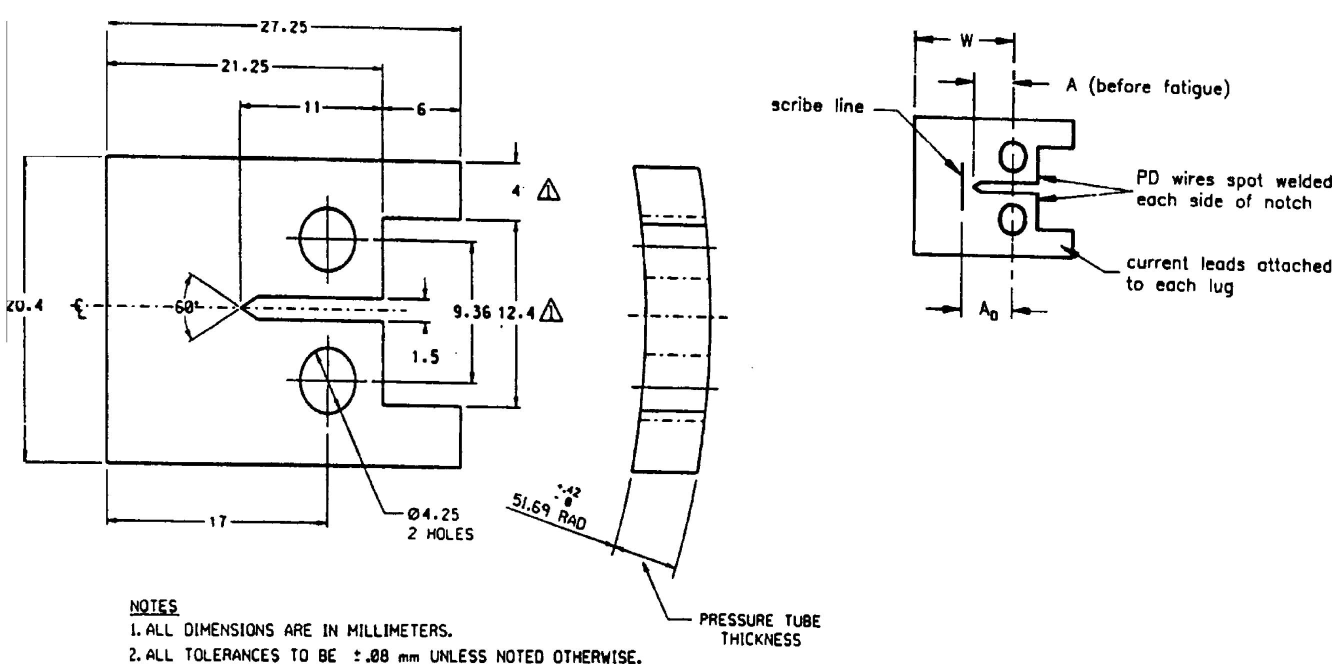


**Fig. 1.** Dimensions of 17 mm CT specimen used in this study.

**Table 1**
Summary of DHC test results under the constant and cyclic loads at 250 °C.

| Specimen ID | DHC crack length (mm) | Incubation time (min)[a] | Cracking time (min)[a] | DHC velocity (m/s) | Initial $K_{Imin}/K_{Imax}$ (MPa m$^{1/2}$) | Final $K_{Imin}/K_{Imax}$ (MPa m$^{1/2}$) | Load ratio $R$ $P_{min}/P_{max}$ |
|---|---|---|---|---|---|---|---|
| R4.0 | 1.003 | 30 | 3473 | 4.81E−09 | 2.1/15.29 | 2.54/18.56 | 0.13 |
| R4.4 | 1.157 | 28 | 2411 | 8.00E−09 | 4.75/14.88 | 5.95/18.62 | 0.32 |
| R4.6 | 1.506 | 42 | 1583 | 1.59E−08 | 7.63/14.85 | 10.3/20.15 | 0.51 |
| R4.7 | 1.528 | 55 | 1195 | 2.13E−08 | 9.9/15.27 | 10.3/20.15 | 0.65 |
| R4.3 | 1.681 | 78 | 1220 | 2.16E−08 | 10.27/15.03 | 14.06/20.57 | 0.68 |
| R4.1 | 1.531 | 43 | 826 | 3.09E−08 | 15.68 | 21.25 | 1 |
| R4.2 | 1.123 | 253 | 527 | 3.27E−08 | 15.1 | 18.7 | 1 |

[a] Note that the incubation time is the time taken to start DHC after loading and the cracking time is the time from the onset of DHC to the end of DHC.

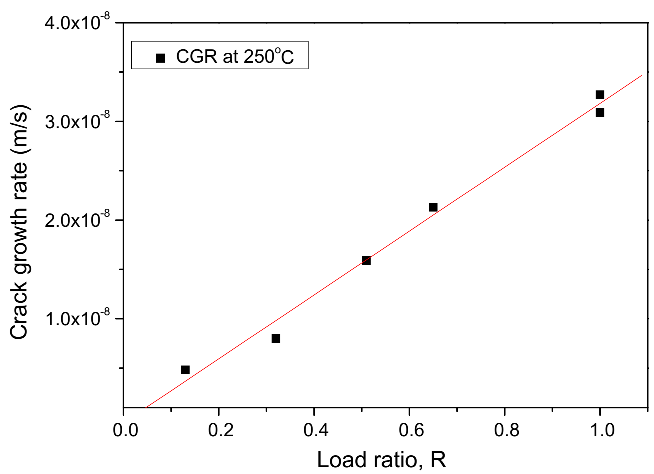


**Fig. 3.** DHC crack growth rate of a heat-treated Zr–2.5Nb tube at 250 °C under the constant and cyclic loads with the load ratio, $R$ changing from 0.13 to 0.68.

Zr–2.5Nb tube with around 10 ppm $H$ with the test temperature being approached by heating. The applied stress intensity factor $K_I$ starting from <6 MPa $\sqrt{m}$ increased in steps of 1–1.5 MPa $\sqrt{m}$ until cracking commenced. To change the stress state at a crack tip, three different procedures were applied: the first one was to produce a pre-fatigue crack under a maximum final $K_I$ of <5 MPa $\sqrt{m}$, the second was to produce a pre-fatigue crack under a maximum final $K_I$ of <5 MPa $\sqrt{m}$ followed by annealing at 475 °C to relieve any residual stresses remaining at the pre-fatigue crack tip, and the third was to apply unloading when $K_I$ had reached about 15 MPa $\sqrt{m}$ and then to reapply the load in steps [10].

## 3. Results and discussion

The CGR at 250 °C of the heat-treated Zr–2.5Nb tube was the highest under the constant load and decreased under the cyclic load with decreasing $R$, as shown in Fig. 3. Given the absence of unloading under the constant load, it is evident that the highest

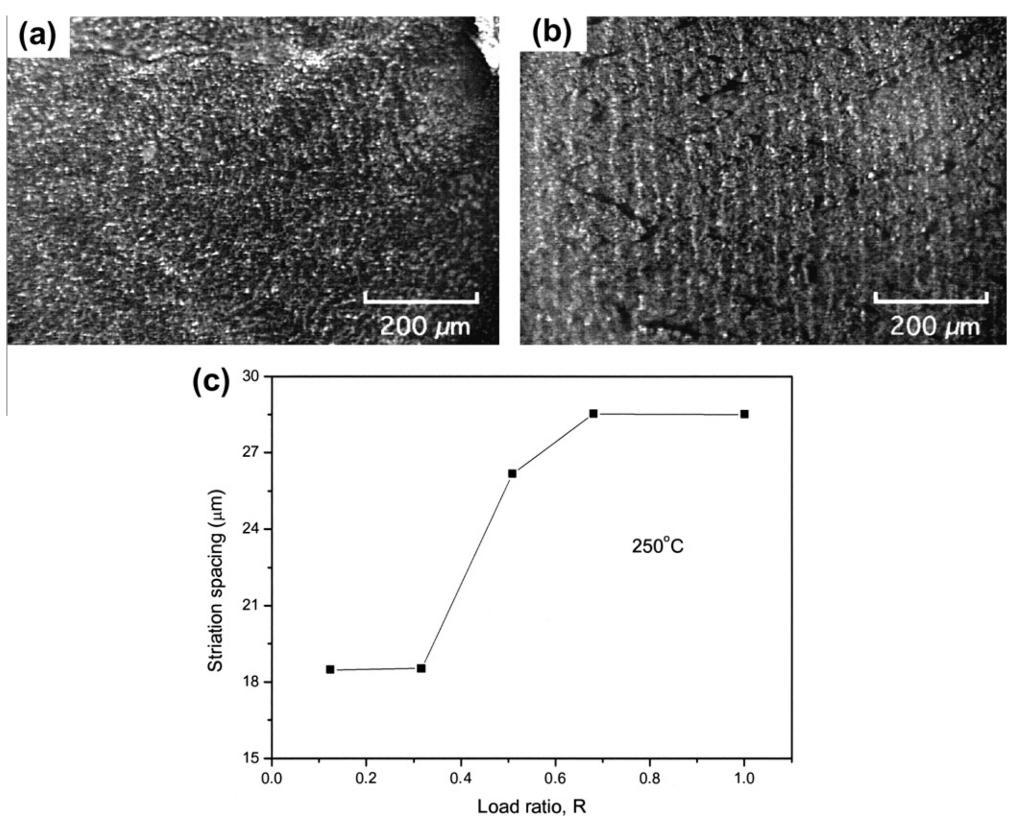


**Fig. 4.** Striations with the load ratio, $R$ on the fracture surfaces of the heat-treated Zr–2.5Nb tubes after DHC tests at 250 °C: (a) $R$ = 0.319, (b) $R$ = 0.68 and (c) the striation spacing that decreased with decreasing $R$.

CGR under the constant load is due to the absence of unloading during the DHC tests. In contrast, the extent of unloading that increased with decreasing $R$ under the cyclic load was the cause of a linear dependence of the CGR on the $R$, as shown in Fig. 3. To assess the effect of the cyclic load on the cracking of hydrides precipitated at a crack tip, we investigated a change of striation spacing with $R$. As shown in Fig. 4, the striations which are a typical fracture pattern of DHC under the constant load were observed even in the cyclic load with $R$ changing from 0.13 to 0.68. From the striations shown in Fig. 4a, the striation spacing was measured as a function of $R$. As shown in Fig. 4b, the striation spacing became narrower with decreasing $R$. Given that the striation spacing represents the critical hydride length above which the cracking of hydrides occurs [25,36]. It is obvious that the critical hydride length became smaller with decreasing $R$. In other words, the cyclic load enhances the rate of hydride cracking when compared to the constant load, causing the hydrides to easily fracture at a smaller length.

The results of Figs. 3 and 4 showed that the cyclic load enhanced the rate of hydride cracking but decreased the CGR. In other words, in the cyclic load, the CGR decreased with decreasing $R$ despite the enhanced rate of hydride cracking. This fact demonstrates that it is not the rate of hydride cracking but the rate of hydride nucleation that governs the CGR according to Kim's model given that the rate of hydride growth that is constant at a constant temperature does not change independent of $R$. As the extent of unloading arising from the cyclic load increases with decreasing $R$, a linear dependence of the CGR on the $R$ as shown in Fig. 3 corroborates that the CGR is retarded in proportion with the extent of unloading. Hence, considering that nucleation of hydrides is restrained by the compressive stress, unloading in the cyclic load induces the compressive stress at a crack tip [33,34] the magnitude of which increases with decreasing $R$, suppressing the rate of hydride nucleation so as to cause it to become the slowest among the three processes involved in DHC. This rationale explains a linear dependence of the CGR on the $R$ (Fig. 3) despite the faster rate of hydride cracking with decreasing $R$, as shown in Fig. 4, which arises due to an increase in the compressive stress with $R$.

Definitive evidence for the effect of unloading is provided by Simpson's experiment [10] where CGRs of a cold-worked Zr–2.5Nb (termed CANDU) tube were determined under the constant load at 120 °C without and with unloading from 15 MPa $\sqrt{m}$ and with annealing at 475 °C after the formation of a pre-fatigue crack. Without unloading, as shown in Fig. 5, the CGR of the CANDU Zr–2.5Nb tube was $2.1 \times 10^{-9}$ m/s independent of $K_I$ above $K_I$ of 10 MPa $\sqrt{m}$. In contrast, after unloading from 15 MPa $\sqrt{m}$, for example, it decreased to $7.1 \times 10^{-10}$ m/s at 14.9 MPa $\sqrt{m}$ (corresponding to point A in Fig. 5), then increased rapidly with increasing $K_I$ and became constant at $K_I$ of over 18 MPa $\sqrt{m}$ (Fig. 5). It is interesting to note that unloading not only decreased the CGR but also increased $K_{IH}$ to 14 MPa $\sqrt{m}$ so that a crack did not grow at 10 MPa $\sqrt{m}$ where a crack was seen to grow in case of the absence of unloading. However, annealing decreased $K_{IH}$ to around 6 MPa $\sqrt{m}$ and increased the CGR to, for example, $3.1 \times 10^{-9}$ m/s at 14.9 MPa $\sqrt{m}$, as indicated by the vertical dotted line in Fig. 5.

The results of Fig. 5 clearly showed that unloading decreased the CGR and increased $K_{IH}$ when compared to those without unloading. Therefore, it is definitively evident that a decrease in the CGR under the cyclic load as shown in Fig. 3 is due to unloading. Given that $K_{IH}$ is related to the resistance of hydride cracking [37], the increased $K_{IH}$ due to unloading shows that unloading under the constant load suppresses the rate of hydride cracking, which is in contrast with the enhanced hydride cracking rate under the cyclic load despite the same effect of unloading. In other words, the CGR is decreased due to unloading regardless of whether unloading enhances the hydride cracking rate under the cyclic load

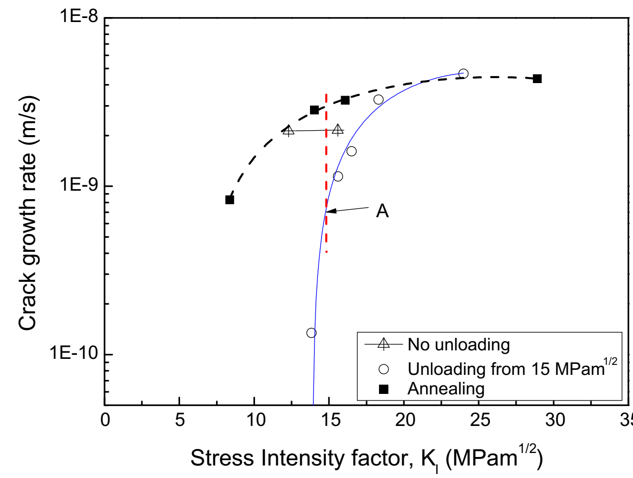


**Fig. 5.** Crack growth rate of a cold-worked Zr–2.5Nb tube without and with unloading from 15 MPa $\sqrt{m}$, and with annealing at 475 °C after pre-fatigue cracking (reproduced from [10]).

or suppresses it under the constant load. Given that the CGR of zirconium alloys is governed by the slowest rate of nucleation, growth and cracking of hydrides, according to Kim's model [18–28], a decrease in the CGR by unloading at a constant temperature must be caused primarily by the lowest rate of hydride nucleation. Hence, the rate of hydride nucleation that becomes the slowest is the rate-controlling factor for the CGR in the presence of unloading that induces the compressive stress at a crack tip. Evidence for the compressive stress at a crack tip due to unloading is provided by in situ measurements of the lattice strains at the crack tip [33,34] demonstrating the presence of compressive strains ahead of a crack tip under cyclic loading–unloading that has increased with decreasing $R$. However, if the compressive stress is relieved by annealing, the rates of nucleation and cracking of hydrides are enhanced, thus resulting in a decrease in $K_{IH}$ and an increase in the CGR, as shown in Fig. 5. Conclusively, it is demonstrated that it is the rate of hydride nucleation that governs the CGR of zirconium alloys when unloading arises from the cyclic load or the constant load. Hence, a decrease in the yield strength arising from annealing at 475 °C, if any, cannot affect the crack growth rate considering that the yield strength affects primarily the rate of hydride cracking [1,2,23].

According to the old DHC models claiming that the CGR is governed only by the hydride growth rate and the stress gradient is the driving force for hydrogen diffusion, the compressive stress could prevent hydrogen diffusion to the crack tip completely [10], leading to no DHC or DHC arrest during unloading. Thus, the crack will grow only in the loading cycle producing the tensile region ahead of the crack tip, not in the unloading cycle irrespective of the magnitude of compressive stress. The CGR is expected to be lower under the cyclic load when compared to the constant load. However, according to the old DHC models, the CGR under the cyclic load will be constant irrespective of the load ratio, $R$ because of no hydride growth irrespective of the magnitude of compressive stress that increases with $R$. However, the old DHC models' prediction is in contrast with the results shown in Fig. 4 showing a linear increase in the CGR with $R$. Therefore, it is clear that the old DHC models are too defective to explain a linear decrease of the CGR with $R$.

## 4. Conclusions

The CGR at 250 °C of a heat-treated Zr–2.5Nb tube was the highest under the constant load and decreased under the cyclic load with decreasing load ratio, $R$ due to unloading. The striation spacing decreased with $R$, demonstrating that the cyclic load enhanced the rate of hydride cracking. Given the presence of unloading during the cyclic load, the decreased CGR in the cyclic load is due to unloading in the cyclic load inducing the compressive stress at the crack tip. This compressive stress suppresses the hydride nucleation rate, leading it to govern the CGR, according to Kim's new model. Likewise, a linear decrease in the CGR with decreasing $R$ is also due to a linear dependence of the extent of unloading on $R$ that determines the magnitude of the compressive stress. Consequently it is suggested that the rate of hydride nucleation that becomes the slowest due to the compressive stress induced by unloading governs the CGR under the cyclic load. Definitive evidence for this suggestion is provided using Simpson's experiment demonstrating that unloading from 15 MPa $\sqrt{m}$ decreased the CGR of a cold-worked Zr–2.5Nb tube but increased $K_{IH}$ and annealing relieving the compressive stress at the crack tip increased the CGR and decreased $K_{IH}$. Consequently, it is demonstrated that the compressive stress induced by unloading suppresses the hydride nucleation rate and hence, the CGR under the cyclic load and annealing does the reverse. This study shows for the first time that the retarded CGR due to an overload during DHC tests or during fatigue crack growth tests is understood in view of crack growth kinetics using Kim's model.

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