



A theoretical model for delayed hydride cracking velocity considering the temperature history and temperature gradients

Jingyu Zhang, Jiacheng Zhu, Shurong Ding*

Institute of Mechanics and Computational Engineering, Department of Aeronautics and Astronautics, Fudan University, Shanghai 200433, China



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ABSTRACT

Delayed hydride cracking (DHC) threatens the safety of Zircaloy components. The DHC velocity in the stage of stable crack growth needs to be studied. Based on the recent research, with a new idea the theoretical model is developed for the DHC velocity considering the temperature history and temperature gradient. The stress field ahead of the crack tip is calculated based on the fracture mechanics theory of second-order estimate of plastic zone size, and the redistributed stresses are statically equivalent with the stresses obtained from Linear Elastic Fracture Mechanics. It is confirmed that a simple linear function of the equivalent axisymmetric temperature distribution can be effectively and easily calibrated for predictions of the DHC velocity at different temperature gradients along the cracking direction. The developed model is validated to be effective because the predicted velocities agree well with some experimental results for the cases with and without temperature gradients, whether the test temperature is approached by heating or cooling. The theoretical results indicate that (1) with a higher positive temperature gradient along the cracking direction, the DHC velocity increases and a higher crack arrest temperature will occur; while a negative temperature gradient will remarkably depress DHC process; (2) The total hydrogen concentration in the hydrogen source has an important effect on the DHC velocity, and the crack arrest temperature can be evidently lowered for the cases with a small quantity of hydrogen atoms there; (3) At higher temperatures, negative flux contribution from the hydrogen concentration gradient in the hydrogen source is found to be the main mechanism of crack arrest.

1. Introduction

Zirconium alloys are widely used as the materials of cladding tubes and pressure tubes in nuclear reactors, because of their combination of high strength, high corrosion resistance and low neutron absorption cross-section [1]. But they are susceptible to hydride embrittlement, which is caused by the precipitation of hydride platelets. As one type of hydride embrittlement, delayed hydride cracking (DHC) is one of the most important failure mechanisms, and it is a time-dependent cracking process resulting from the stress assisted diffusion of hydrogen to the crack tip [2].

A part of hydrogen atoms, generated due to the corrosion reaction of zirconium with the coolant water, are absorbed on the outer surface of the tubes and diffuse in Zircaloy components, driven by the gradients of hydrogen concentration, temperature and hydrostatic stress. When the hydrogen concentration exceeds its terminal solid solubility (TSS), the brittle hydrides will precipitate owing to the reaction of zirconium with hydrogen. For the Zircaloy components containing cracks and other defects, the hydrogen atoms tend to diffuse towards the location of

stress concentration with peak tensile stresses, driven by the high hydrostatic stress gradient. Simultaneously, when plenty of hydrogen atoms are absorbed or immersed in the components, high tensile stresses lead to low TSS in the location of stress concentration, which results in low concentration of hydrogen atoms in solid solution. Thus, also induced by the above concentration gradient of movable hydrogen atoms, the hydrogen atoms are inclined to diffuse towards the location of stress concentration and further precipitate as hydrides. Nearby the crack tip, the volume fraction of hydrides is locally increased. When the volume fraction of hydrides ahead of the crack tip increases to a certain level, the hydrided area will be cracked and new crack tip forms.

In CANDU and RBMK reactors, Zr-2.5Nb pressure tubes (PTs) containing cracks are susceptible to failure by DHC [2]. A number of experiments demonstrated that DHC only happened for the cases with a stress intensity factor (SIF) in excess of a threshold value K_I^H , while the DHC velocity varied slightly in a wide SIF range, which was called the steady crack growth stage [1,3]. So below a threshold stress intensity factor K_I^H , the tubes will be safe [4]. At the steady crack growth stage, the cracking will occur when the hydrided area has reached a critical

* Corresponding author.

E-mail address: dingshurong@fudan.edu.cn (S. Ding).

length [5,6], which means that a period of waiting time should be experienced before new cracking. So, this cracking mode is called delayed hydride cracking, a type of subcritical crack growth. The threshold stress intensity factor K_I^H and the DHC velocity are two important parameters to evaluate DHC behavior. Zircaloy tubes are important barriers to prevent nuclear leakage. It is significant to keep their structural integrity during the in-pile operation stage and also the long-term storage period of spent fuels. It is of practical value to develop theoretical models for predicting the DHC behavior.

A number of theoretical models [3,7–11] were proposed for predicting the DHC velocity. Dutton and Puls [7] developed a theoretical model, which, for the first time, mathematically described the diffusion behavior of hydrogen in DHC process, considering the gradients of hydrogen concentration and hydrostatic stress in a steady-state axisymmetrical hydrogen diffusion equation. The hydrogen atoms were assumed to diffuse to the crack tip and then hydrides precipitated when the concentration of hydrogen exceeded TSSP, and this model was called Diffusion First Model [12]. Kim et al. [10,11] developed another model which was called Precipitation First Model by McRae et al. [12]. In this model, the first step in DHC process was the precipitation of hydrides at the crack tip due to the reduced hydrogen terminal solid solubility there, which resulted from the large tensile stresses ahead of the crack tip. Then, the precipitation-induced concentration gradient between the bulk and the crack tip area resulted in the diffusion of hydrogen atoms towards the crack tip. In fact, as we mentioned above, the gradient of hydrostatic stress and the induced gradient of movable hydrogen concentration both contribute to DHC velocity. The fundamental mechanism is due to the existence of stress gradients. It is very important that the terminal solid solubility of hydrogen can be effectively and quantitatively described as a function of stresses. Dutton and Puls [7] made important achievements in this direction. Simultaneously, the contributions of hydrostatic stress gradient and movable hydrogen concentration gradient to the DHC velocity need to be analyzed.

Several models [3,8,12–14] have been further developed based on the original Diffusion First Model. The obtained formula for the velocity can be expressed as

$$v = f(D_H, \dots)(E_L - E_l) \quad (1)$$

where D_H is the diffusion coefficient of hydrogen in zirconium alloy; $f(D_H, \dots)$ is a function of D_H and other variables; l and L are the distances away from the crack tip, referred to the sink and source positions of hydrogen; E_L, E_l are the variables determined by the stress-free TSS, stresses and temperatures at the corresponding locations. Puls [3] modified the initial model [7] by considering different stress-free TSS values, that is, TSSP and TSSD, at l and L , respectively. And the stresses at l were calculated according to the HRR theory [15,16]. Shmakov et al. [9,14] gave a DHC model, assuming a bi-linear distribution of normal stress in the plastic zone, and the stress perpendicular to the cracking plane was set to be zero at the crack tip in view of material damage. According to this assumption, the obtained maximum stress was different from that in Ref. [3]. In fact, under the applied stress intensity factor for steady crack growth, the stress perpendicular to the cracking plane is impossible to reach zero in the crack tip, and should be a little lower than the maximum stress in the plastic zone. Sunil et al. [17] found in the experiments that the DHC velocity depended heavily on the stress state of the crack tip area. So, the distribution of stress around the crack tip should be well described in theoretical models for DHC velocity, because they directly affect E_L, E_l .

Sagat et al. [8] improved Puls's model [3] to take account of temperature gradients and temperature history. The temperature near the crack tip was assumed to follow a distribution function of $A + Br^{0.5}$. In the tubes, the real distribution of temperature is possible to be different from this function, and is impossible to be axisymmetrical. As is known that the temperature gradient plays a vital role in DHC velocity, it is very critical for an axisymmetrical diffusion model to use an average (or

equivalent) temperature distribution from the hydrogen sink to the hydrogen source, and establish a relation between the equivalent distribution function and the real distribution along the cracking direction. As for the temperature history, meaning that the test temperature is approached by cooling or heating, different values of accommodation energy owing to the hydride precipitation were used for those at l and L . However, the same stress-free TSS was used in this model and it should be different following cooling or heating [10]. Shmakov et al. [9] considered a higher virtual temperature in the calculation of stress-free TSS at l ; but the effect of anisotropic misfit strain due to hydride precipitation on TSS was not considered.

The steady-state diffusion equation used in these models [3,8,9] was solved by giving the concentration of hydrogen at l and L . l refers to the position of maximum stress in the plastic zone ahead of the crack tip in many papers [3,8]. L denotes the location of hydrogen source, but is not consistent in these models. Puls [3] mentioned it was about 300 μm for hydrogen concentrations of 45 to 90 at.%; Sagat et al. [8] suggested that it was 500 μm ; Shmakov et al. [9] set $L = 20r_p$, where r_p was the length of plastic zone. As L is related to hydride spacing and affected by total concentration of hydrogen, its value should not be fixed and a more reasonable value should be defined.

In most of the current models, the lengths of plastic zone were calculated using a first-order estimate of plastic zone size. In fact, the stresses along the cracking plane are redistributed because of plastic deformation and damage accumulation surrounding the crack tip, so the length of plastic zone should be recalculated and must increase in size according to the fact that the normal stresses should satisfy equilibrium according to the fracture mechanics theory [18].

Besides, in previous studies, the concentration of hydrogen atoms in solid solution at the hydrogen source location of $r = L$ was assumed as TSS. If the original average concentration of all hydrogen atoms is not high enough, the concentration of real moveable hydrogen atoms is possible to be lower than TSS at L . The effects of the hydrogen concentration at the source location on the DHC velocity should be further investigated.

In this study, an improved theoretical model for DHC velocity is developed to involve the effects of temperature history and temperature gradient with a new idea, in which a more reasonable distribution of stress is given according to the theory of elastic-plastic fracture mechanics. The crack arrest temperature can be predicted, and the three stages of delayed hydride cracking can be reflected by the improved model. Some experimental data of DHC velocity for Zr-2.5Nb pressure tubes and Zircaloy-2 cladding tubes are compared with the theoretically predicted results, through which the effectiveness of the improved model is validated. Besides, the effects of temperature gradient, temperature history, the hydrogen concentration at the source location and other parameters are investigated. The contributions of the temperature, hydrostatic stress and hydrogen concentration gradients to the DHC velocity are analyzed. The mechanism of crack arrest at a high temperature value is discussed.

2. A theoretical model for delayed hydride cracking velocity

As illustrated in Fig. 1, the delayed hydride cracking is generally recognized to comprise the following processes: (1) hydrogen atoms, driven by the gradients of hydrogen concentration, hydrostatic stress and temperature, diffuse to the crack tip; (2) hydrogen atoms accumulate in the vicinity of the location undergoing the maximum stresses with a distance r_M away from the crack tip, followed by the nucleation and growth of hydrides when the hydrogen concentration exceeds its terminal solid solubility; (3) the hydride phase fractures after it grows to a critical length l_c with a thickness t ; (4) the above processes repeat themselves. So the temperature field, stress field and hydrogen concentration field all contribute to the DHC velocity. In this section, the tensile stresses at the crack tip zone obtained from classical Linear Elastic Fracture Mechanics are redistributed according to the elasto-

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