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Creep fracture of zirconium alloys

Troy A. Hayes [a,*], Robert S. Rosen [b], Michael E. Kassner [c]

[a] *Exponent Failure Analysis Associates, 149 Commonwealth Drive, Menlo Park, CA 94025, USA*

[b] *Defense Nuclear Facilities Safety Board, 625 Indiana Avenue, NW, Suite 700, Washington, DC 20004, USA*

[c] *University of Southern California, Aerospace and Mechanical Engineering Department, Olin Hall 430, Los Angeles, CA 90089, USA*

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Abstract

Theoretical approaches utilized to predict the lifetime of spent nuclear fuel in interim dry storage assumed that diffusion controlled cavity growth controlled the failure time under these conditions. DCCG, however, fails to account for the fact that the failure time is related to the strain rate in Zircaloys according to the Monkman–Grant relationship. This paper will show that constrained cavity growth, which can account for the Monkman–Grant relationship but was not considered in the spent nuclear fuel lifetime prediction models, is more relevant to failure of spent nuclear fuel in dry storage. Contrary to reports in the past, constant stress creep tests performed in this study on Zircaloy-2 suggest that creep cavity nucleation and/or growth occurs prior to tertiary creep. Constant strain rate creep rupture tests on Zircaloy-2 show strong evidence of extensive cavity nucleation and growth near and at the fracture surface, indicating a creep cavitation failure mechanism under these conditions.

1. Introduction

Upon removal from reactor, spent nuclear fuel (SNF) rods are placed in wet storage for a period of generally 5 years or more. The wet storage capacity at commercial nuclear power plants is inadequate to accommodate all the SNF until a permanent disposal site becomes available. This has led to the development of interim dry storage, where SNF rods are placed inside a canister, usually concrete with a stainless steel liner, in air or an inert atmosphere while awaiting permanent disposal. An increasing number of rods are now being placed in interim dry storage due to delays in the availability of a permanent disposal site (mined geologic repository). Commercial SNF typically consists of uranium dioxide pellets surrounded by a thin cladding, usually a zirconium-based alloy such as Zircaloy-2 or Zircaloy-4. The Nuclear Regulatory Commission (NRC) formerly required potential dry storage licensees to calculate the maximum allowable temperature for the storage of spent nuclear fuel based on creep rupture models [1]. The NRC then accepted calculations based on a strain-limit approach as an alternative for the model-based calculations [2]. The NRC subsequently abandoned all 'creep theoretical' models, as well as the strain-limit approach, in favor

* Corresponding author. Tel.: +1 650 688 7127.
E-mail address: thayes@exponent.com (T.A. Hayes).

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of temperature limits that are based on an examination of spent fuel rods after 15 years of dry storage [3,4].

There is a belief in the nuclear industry that zirconium alloys are resistant to void formation and that creep cavitation plays little or no role in the creep failure of these alloys [5,6], contrary to the results of a study by Keusseyan et al. [7]. The objectives of this study were to evaluate the relevant failure mechanisms for zirconium alloys under SNF interim dry storage conditions and to determine whether creep cavity nucleation and growth occur in Zircaloy-2 under these conditions. A creep theoretical evaluation, constant strain rate creep fracture tests, and constant stress creep tests were employed to meet these objectives.

2. Creep fracture model

Zircaloy cladding in interim dry storage is subjected to stresses up to approximately 100 MPa and temperatures as high as 450 °C. It has been suggested that a failure mechanism known as diffusion-controlled cavity growth (DCCG) as originally proposed by Hull and Rimmer [8], and later modified by Raj and Ashby [9] and Speight and Beere [10], controls the failure of zirconium alloys under these conditions [11,12]. The rupture time according to this model is given by [19]

$$t_{\mathrm{f}} = \frac{3\pi^{1/2}}{32}\frac{F_{\mathrm{v}}(\alpha)}{F_{\mathrm{B}}(\alpha)^{3/2}}\int_{A_{\min}}^{A_{\max}}\frac{\mathrm{d}A}{f(A)}\frac{\lambda^3 kT}{\Omega D_{\mathrm{gb}}\delta\sigma}, \tag{1}$$

where t_{f} is the fracture time, F_{v} and F_{B} are functions of the grain boundary/cavity interfacial angle, α, A is the area fraction of grain boundaries occupied by cavities, k is Boltzman's constant, T is temperature, Ω is the atomic volume, D_{gb} is the grain boundary diffusion coefficient, δ is the grain boundary width, σ is the applied stress and λ is the average cavity spacing. Although this model has been shown to be applicable under very limited conditions (see, for example, Refs. [13–16]), it does not appear to apply to many engineering materials because, as is apparent in Eq. (1), the failure time according to this model is not a function of the strain rate. It is known, however, that the failure time in many engineering metals and alloys (including Zircaloy-2 [17]) is related to the strain rate by the Monkman–Grant relationship, or

$$\dot{\varepsilon}_{\mathrm{ss}}t_{\mathrm{f}} = K, \tag{2}$$

where K is the Monkman–Grant constant, t_{f} is the fracture time, and $\dot{\varepsilon}_{\mathrm{ss}}$ is the steady-state strain rate. Thus, pure, or 'unconstrained' DCCG may not be applicable to zirconium alloys. Strain-based creep cavitation models, however, do not appear to be able to account for the magnitude of cavity growth observed in practice. Instead, a constrained cavity growth model, which can lead to significant cavity growth without large far-field strains and can account for the Monkman–Grant relationship, may be the most relevant for predicting the lifetime of nuclear fuel rods during interim dry storage. Constrained cavity growth is discussed in detail elsewhere [18]. Briefly, unconstrained cavity growth assumes that every grain boundary that lies nearly perpendicular to the applied stress undergoes uniform cavitation. A more realistic assumption is that the cavitating grain boundaries in a metal are interspersed among many cavity-free grains. As cavity growth proceeds on isolated grain boundaries, the adjacent grains begin to elongate in order to accommodate the local increase in volume created by the grain boundary cavities. The stress driving the growth then becomes 'constrained' by surrounding grains that do not contain cavities and do not, therefore, undergo the same local strain. Once the stress is relieved or 'shed' by the constraint of surrounding grains, cavity growth ceases until the surrounding grains deform and the stress on the cavitated grain boundaries rises again. The time for cavity coalescence to occur was calculated by Reidel [19] to be

$$t_{\mathrm{c}} = 0.004\frac{kT\lambda^3}{\Omega\delta D_{\mathrm{gb}}\sigma} + 0.3\frac{(1+3/n)^{1/2}\lambda}{\dot{\varepsilon}g}, \tag{3}$$

where t_{c} is the time to coalescence, g is the grain size, $\dot{\varepsilon}$ is the strain rate, n is the power law (stress) exponent and the other factors are consistent with Eq. (1), above. The range of conditions under which constrained cavity growth is rate controlling was calculated and compared to the conditions relevant to the interim dry storage of SNF. Fig. 1 indicates the 'constrained limit stress' (σ_{c}) below which constrained cavity growth is predicted to control cavity growth and, ultimately, fracture, along with the conditions relevant to the dry storage of SNF.

The time to coalescence was assumed to be 'constrained' when the creep-controlled portion of Eq. (3) (the second term) was approximately equal to the diffusion-controlled portion of Eq. (3) (the first term). As the applied stress is decreased below the