Zircalloy Fuel Cladding Integrity During Dry Storage

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INTRODUCTION

Pursuant to Ch. 10, Sec. 72.72(h) of the code of Federal Regulations, the licensee of an interim spent fuel storage facility (ISFSI) must protect the spent fuel cladding against degradation and gross rupture. For the case where spent fuel is stored in casks under dry storage conditions, the only parameters that the cask designer has at his disposal to prevent such degradation is an inert internal cask environment and both the initial and decay temperatures of the spent fuel. The latter are governed by the type, quantity, specific power, and age of the spent fuel assemblies and by the passive heat dissipation properties of the storage cask.

The problem faced by reviewers of Topical Safety Analysis Reports (TSARs) was the lack of credible evidence showing that the integrity of the spent fuel cladding could be guaranteed over a design life of the ISFSI of at least twenty years. A considerable amount of study, under U.S. Department of Energy and Nuclear Regulatory Commission sponsorship, had been devoted to assessing spent fuel cladding integrity under dry storage conditions. Some of these studies are summarized in reports by Johnson and Gilbert, 1983, Blackburn et al., 1978 and Einziger and Cook, 1984. A major goal of these studies was to provide potential applicants with dry storage temperature limits that would assure compliance with the regulatory requirement cited above. These studies covered dry storage demonstrations and laboratory tests of Zircalloy material behavior involving the effects of corrosion, hydrating, and creep. Many of these studies were referenced in the TSARs to justify the design limit temperatures for dry storage. However, it was the opinion of the reviewers of the TSARs that the dry storage demonstrations involved too few assemblies observed for too short a time to permit a confident prediction of long-term behavior.

In view of this situation, the reviewers conducted their own investigation to determine whether the storage conditions specified in the TSAR assured compliance with the regulation. The reviewers interpreted the regulatory requirement to mean that not only should gross rupture be avoided during the design storage life of the ISFSI but also that the ability of the cladding to maintain its structural integrity during normal handling procedures at the end of storage life also be assured.

After reviewing the current research relating to spent fuel cladding damage mechanisms, the reviewers concluded that a diffusion controlled cavity growth (DCCG) creep mechanism was the only damage mode applicable to dry storage of spent fuel that could lead to gross rupture of the cladding. Under the influence of the temperature and stress due to fuel rod internal pressure, the DCCG damage mechanism progresses by the nucleation and growth of cavities along the Zircalloy grain boundaries. This damage mechanism is insidious since it can progress without external evidence of damage, may not cause pin holes or cracks to relieve the internal rod pressure, and manifests itself by a sudden nonductile type of fracture.

DIFFUSION CONTROLLED CAVITY GROWTH

A number of investigators of creep deformation phenomena have observed the presence of intercrystalline cracks and grain boundary cavities during post-failure microscopic examination of creep specimens. Greenwood et al., 1954, first suggested that intercrystalline cracking may be caused by prior formation of these grain boundary cavities. Experiments were conducted with alpha-brass, copper, and magnesium wherein each material was subjected to high, intermediate, and low strain rates over a range of temperatures. These experiments showed that a decrease in ductility accompanied a decreasing strain rate. This is illustrated for alpha-brass in Fig. 1. Microscopic examination of the fractured specimens indicated that decreased ductility, due either to increased temperature or to a decreased strain rate, correlated strongly with an increased density of grain boundary cavities. Significantly, cavities were prevalent in grain boundaries normal to the load axis. The straining that occurred subsequent to cavitation apparently caused cavities to link up and form continuous cracks. An illustration of this behavior is shown in Fig. 2 which depicts a micrograph of an alpha-brass creep specimen in the rupture region and Fig. 3 which shows cavity formation one inch away from the fracture surface.

Based upon the assumption that creep cavitation occurs primarily at grain boundaries and favors those boundaries normal to the axis of applied stress, Ballufi and Seigle, 1957, postulated that cavities nucleated at the grain boundaries grow by the migration of vacancies from the grain boundary into the cavity. The driving force for this migration arises from the chemical potential gradient established between the vacancy concentration in the boundary and at the cavity.

Researchers have made numerous attempts at developing a theoretical basis for predicting the rate of cavity growth. All are based upon the fundamental mechanism of vacancy diffusion but differ with regard to boundary and initial conditions. The theoretical basis adopted by the reviewers was inspired by Chin, 1983, who, under contract to the Batelle Pacific Northwest Laboratory, developed a fracture map for Zircalloy that showed diffusion controlled cavity growth to be the dominant damage mechanism under the temperature and pressure conditions that prevail during dry storage of spent fuel. The fracture map, in turn, was based upon the theoretical work of Raj and Ashby, 1975. Chin further applied the fracture map to predict time to rupture of the Zircalloy clad spent fuel rods under various storage conditions. To account for the decrease in temperature with time due to a decreasing rate of radioactive decay, Chin used an incremental life fraction rule to model the decrease in damage rate with time.

The method adopted by the reviewers to assure compliance with the regulation invoked the Raj/Ashby model for grain boundary creep cavitation and deviates from the Chin approach only to make it more amenable for the reviewers to assess conformity to their interpretation of the regulation. Thus, whereas Chin based his approach on the Raj/Ashby formulation for time to rupture, the reviewers adopted an upper limit to the amount of damage the cladding should sustain at the end of this design storage life. This acceptance criterion was adopted not only for ensuring that the cladding had sufficient residual strength to withstand normal handling loads on the fuel assemblies, but also to avoid the need to establish a creep rupture criterion that is subject to considerable uncertainty. Developing this acceptance criterion required that the final Raj/Ashby equations be modified somewhat. An additional advantage of modifying the equations was that it allowed the time dependent functions to be directly integrated rather than adopting a life fraction rule.

REVIEW PROCEDURE

The measure of cladding damage was defined as the area fraction of decohesion at the grain boundaries and can be ascertained by the following expression

$$\int_{A_i}^{A_f} \frac{dA}{f(A)} = \int_0^{t_f} G(t)dt \tag{1}$$

where A_i is the initial area fraction of decohesion due to the nucleation of stable cavities and A_f as the area fraction of decohesion that occurs over the period of time t_f. The details of the derivation of Eq. (1) appear in the report by Schwartz and Witte, 1987. Furthermore,

$$F(A) = \frac{(1 - (\frac{A_i}{A})^{1/2} \sin\alpha)(1 - A)}{A^{1/2}\left(\frac{1}{2}\ln\frac{1}{A} - \frac{3}{4} - A\left(1 - \frac{A}{4}\right)\right)} \tag{2}$$

$$G(t) = \frac{32}{3\Pi}\frac{F_B^{3/2}(a)}{F_v(a)} \cdot \frac{\Omega\delta\sigma_\infty(t)}{k\lambda^3} \cdot \frac{D_{gb}(t)}{T(t)} \tag{3}$$

The terms of Eqs. (2) and (3) are defined as follows with reference to Fig. 4.

α = grain boundary cavity dihedral angle

= $\cos^{-1}(\gamma_B/2\gamma) \approx 75°$

γ = free surface energy

γ_B = grain boundary surface energy

$\Omega =$ atomic volume $\approx 3.37 \times 10^{-29}$ m^3/atm

$\delta =$ grain boundary thickness $\approx 9.69 \times 10^{-10}$ m

$\sigma_\infty =$ stress on cladding = pr/h

p = fuel rod internal pressure

r = fuel rod radius

h = cladding thickness

k = Boltzman's constant = 1.3802×10^{-16} crg/deg K

$\lambda =$ average cavity spacing – 10 – 20 x 10^{-6} m

$D_{gb} =$ grain boundary diffusion rate $\approx$

$= 5.9 \times 10^{-6}$ (exp(– 131/RT(t)) m^2/sec

R = gas constant

T = absolute temperature

$F_B(\alpha) = \pi \sin^2 \alpha$

$F_v(\alpha) = 2\pi/3\ (2 - 3 \cos \alpha - \cos^2 \alpha)$

It is essential that the applicant provide in the TSAR the maximum initial storage temperature, the assumed fuel rod internal pressure and a temperature decay curve for the design life of the ISFSI.

ACCEPTANCE CRITERION FOR CAVITY GROWTH

The need to handle fuel assemblies for consolidation and storage or disposal at a monitored retrievable storage facility of a geologic repository precludes allowing damage to progress to a condition of zero residual strength. Analysis of cladding resistance to inertia loads performed by Chun et al., 1987, indicate that, for undamaged material, local yielding will not occur for lateral accelerations of less than 73 g at the end of storage life for the most vulnerable fuel assemblies. Damage limited to 15 percent of the original cross section area of the cladding will reduce this acceleration to approximately 62 g, which is the approximate capacity of undamaged Zircalloy at 380°C. A considerable margin still exists, however, between the onset of local yielding and the point at which cladding will actually fail. Nevertheless, in view of the large uncertainty associated with anticipating the point at which failure occurs, prudence dictates limiting damage due to creep cavitation to 15 percent of the original cladding cross section. Thus, the equality stated in Eq. (1) must be achieved for a value of A_f less than or equal to 0.15 when t_f is greater than or equal to design life of the ISFSI.

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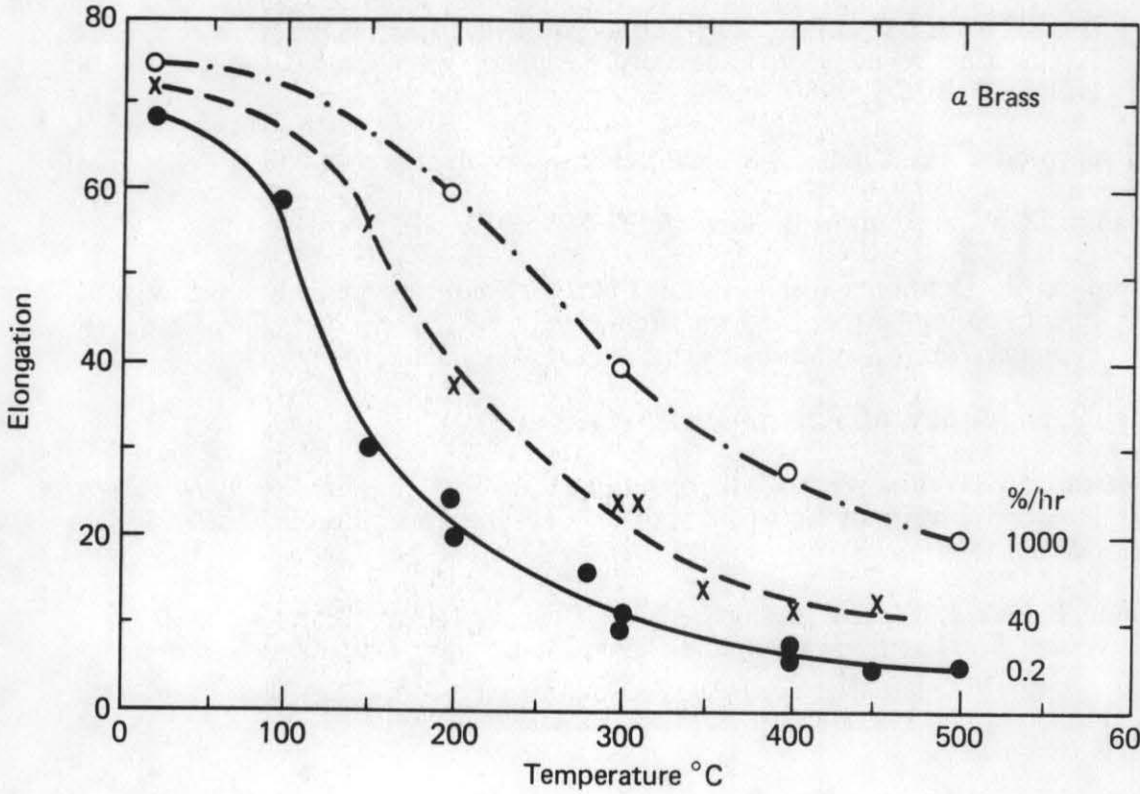

Figure 1 Influence of strain rate and temperature on the ductility of α brass.

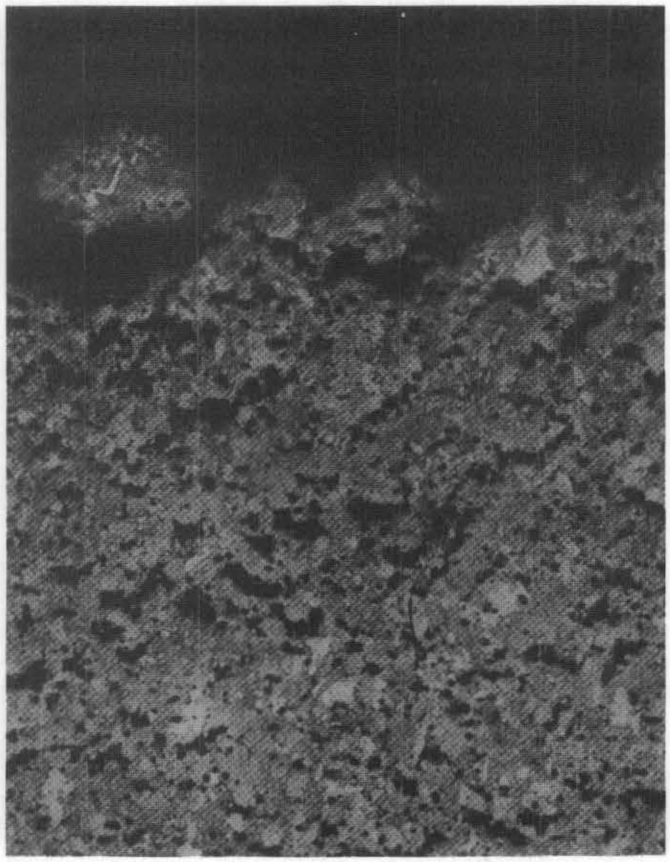

Figure 2 Photomicrograph of creep specimen at the fracture zone.

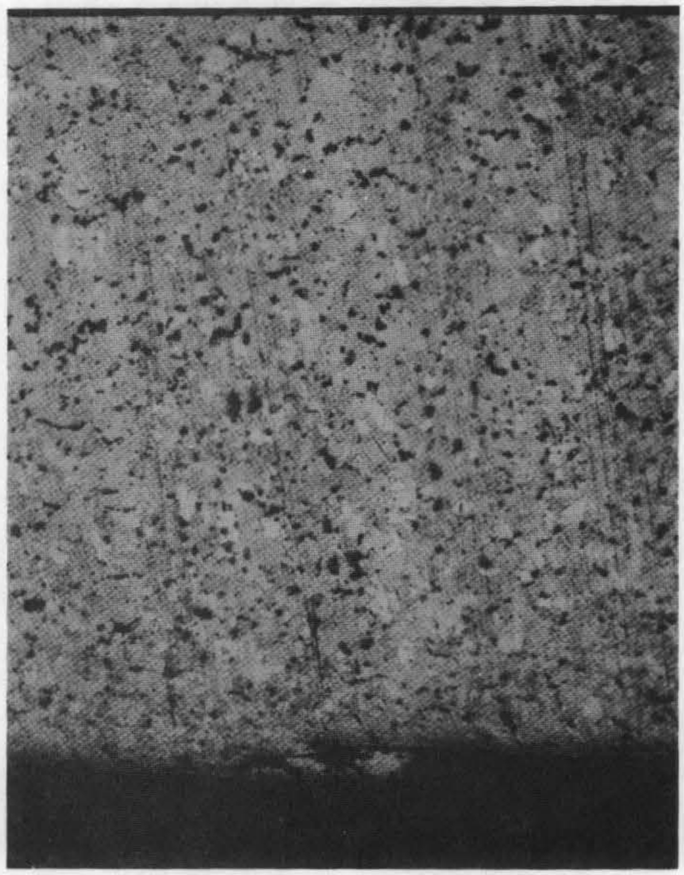

Figure 3 Photomicrograph of creep specimen at the fracture zone.

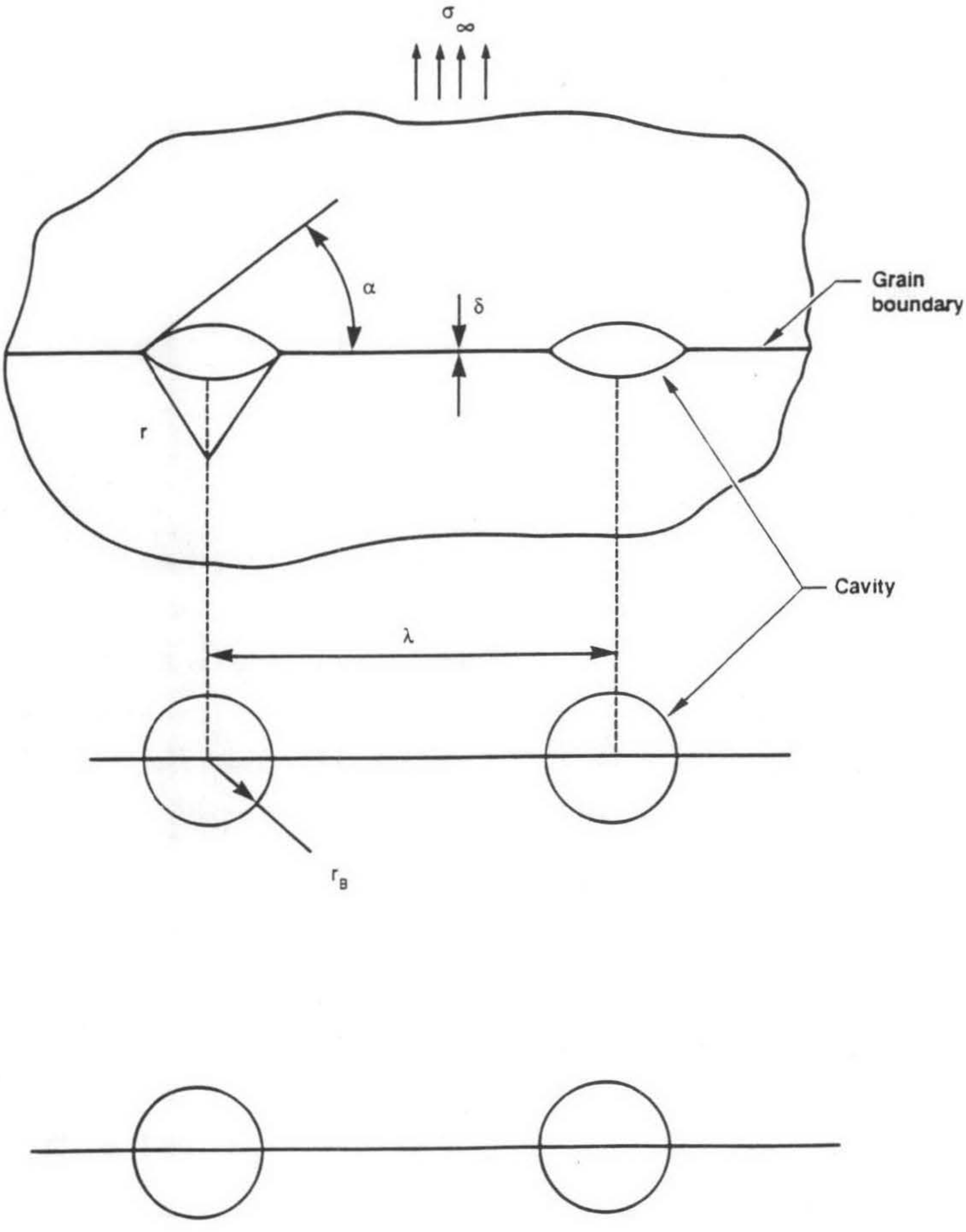

Figure 4 Definitions of symbols r, r_β, α, δ, and σ_m.