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DEFORMATION AND FRACTURE MAP METHODOLOGY
FOR PREDICTING CLADDING BEHAVIOR
DURING DRY STORAGE

B. A. Chin
M. A. Khan
J. C. L. Tarn

Auburn University

E. R. Gilbert, Program Manager

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Pacific Northwest Laboratory
Richland, Washington 99352

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SUMMARY

The licensing of interim dry storage of light-water reactor spent fuel requires assurance that release limits of radioactive materials are not exceeded. The extent to which Zircaloy cladding can be relied upon as a barrier to prevent release of radioactive spent fuel and fission products depends upon its integrity. The internal pressure from helium and fission gases could become a source of hoop stress for creep rupture if pressures and temperatures were sufficiently high. Consequently, it is of interest to predict the condition of spent fuel cladding during interim storage for periods up to 40 years. To develop this prediction, deformation and fracture theories were used to develop maps. Where available, experimental deformation and fracture data were used to test the validity of the maps. Predictive equations were then developed and cumulative damage methodology was used to take credit for the declining temperature of spent fuel during storage. This methodology was then used to predict storage temperatures below which creep rupture would not be expected to occur except in fuel rods with pre-existing flaws. Predictions were also made and compared with results from tests conducted under abnormal conditions.

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INTRODUCTION

Zircaloy has been used extensively as a cladding material in the nuclear industry. The dry storage of Zircaloy clad spent light-water reactor (LWR) fuel requires the ability to predict its long term behavior under low temperature/low stress conditions. Under these conditions creep rupture is the failure mode that will probably limit time in dry storage of spent fuel in an inert gas environment. Because cracks generally progress from the inside of the cladding wall to the exterior, there is little or no impact of the cover gas on the creep rupture mode. However, the cover gas may have an effect once a failure occurs. If the crack is sufficiently large and the temperature high enough, oxidation of the fuel will occur if the cover gas is oxidizing. However, with an inert cover gas, little or no impact is expected. Unfortunately, few experimental data exist for anticipated dry storage conditions because of the long test times required.

Previous predictions of Zircaloy cladding failure under dry storage conditions have been based on direct extrapolation of existing short-time data (Blackburn et al. 1978; Einziger, Bosi and Miller 1980) or have assumed a single phenomenon, such as stress corrosion cracking (Miller 1981), to be the dominant failure mechanism. The direct extrapolation method ignores changes in the deformation and failure mechanisms as the stress and temperature conditions are altered and resulting failure times change. Such analyses are very useful for interpolation between existing data fields but often lead to erroneous results when extrapolated to regimes involving different deformation and failure phenomena.

A method that has been successfully used in predicting deformation and failure mechanisms in unirradiated and irradiated 316 stainless steel (SS) is the use of deformation and fracture mechanism maps. These maps simultaneously display several deformation or failure modes in the form of a schematic diagram of stress/temperature space. Such maps enable prediction of the dominant deformation mechanism and failure mode for various temperatures as well as varying irradiation histories. The deformation map was suggested by Weertman and Weertman in 1965, and the fracture map was first suggested by Wray in 1969.

Ashby and Raj later improved and further developed the concepts of the deformation (Ashby 1972) and fracture maps (Ashby and Raj 1975).

The deformation and fracture maps are usually constructed with normalized stress on the ordinate and homologous temperature on the abscissa. Two distinct approaches to constructing the maps have been adopted. The first approach is to place on the map a large number of experimental points (derived from tests on different specimens) with identification of the deformation or fracture mode associated with each point. Lines are then drawn on the map to indicate the approximate boundaries between the different deformation or fracture processes. This experimental method contrasts with the model based method in which a computer is used to numerically evaluate different theoretical expressions for deformation or fracture. In the generation of the model-based fracture map, values of time to failure are numerically evaluated for possible fracture mechanisms under the assumption that the mechanism with the shortest failure time controls the fracture. Similarly, deformation maps are generated by numerically evaluating different deformation mechanisms and determining which deformation mechanism has the highest deformation rate. In the construction of deformation and fracture maps the various mechanisms are assumed to operate independently. This concept is an over-simplification of the actual process because mixed modes of fracture occur and damage introduced by one mechanism can affect another. However, the deformation and fracture maps do allow one to predict dominant deformation and failure modes over a wide range of stress/temperature space and represent a reasonable approach in the absence of more detailed information.

The ultimate application of these deformation and fracture maps is a prediction of the time to rupture of Zircaloy clad spent LWR fuel subjected to various storage conditions. During dry storage, cask (a container with multiple fuel rods and an inert atmosphere) temperatures are anticipated to decrease with increasing time due to a decay in the heat generation rate of the spent fuel. In this report, a life fraction rule is used to account for damage by this decaying temperature function. This life fraction rule is combined with the results of the deformation and fracture maps to predict maximum allowable initial storage temperatures to achieve a 40 to 100 year life.

This report also discusses the effects of cladding flaw size and analyses of data from assembly B02 of the EMAD test, abnormal dry storage events tests done by Pacific Northwest Laboratory (PNL)^(a) and Battelle Columbus Laboratories (BCL), and dry storage tests conducted by the Federal Republic of Germany (FRG) and by Westinghouse Hanford Company.

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CONCLUSIONS

The following conclusions resulted from this study.

- Fracture and deformation maps provide acceptable methods for extrapolating creep-rupture data over large ranges of temperature and stress.
- Maximum initial dry storage temperatures of 429°C for 1-year-old fuel cladding and 385°C for 5-year-old fuel cladding subjected to an initial stress of 70 MPa are predicted. These predictions are based on achieving a 40-year life in dry storage.
- Predictions indicate that over 90% of the damage accumulated during the first five years of dry storage.
- The use of a constant cladding stress to model the behavior of Zircaloy cladding in dry storage is appropriate. Change in cladding stress resulting from decaying temperatures is a secondary effect.
- The primary effect of irradiation on the fracture properties of Zircaloy is a loss of ductility. This causes an expansion of the predicted triple point cracking regime to lower stresses.
- The effect of cladding flaw size on predicted initial maximum permissible dry storage temperatures is expected to be significant.
- Annealing out of irradiation damage during initial storage of spent fuel rods may be advantageous to long-term storage reliability.
- A postulated pre-existing cladding flaw in a rod contained in assembly B02 is estimated to have extended more than 73% through the wall for failure to have occurred during dry storage.
- Federal Republic of Germany data are in agreement with deformation and fracture map predictions. FRG strain rate data are best predicted by lower boundary strain rate predictions.
- Federal Republic of Germany and Westinghouse irradiated rod tests indicate fracture map predictions may be overly conservative for temperatures above 425°C.

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RECOMMENDATIONS

The results of the studies described in this report indicate that storage of light-water reactor Zircaloy-clad spent fuel in inert gas should result in a low incidence of stress rupture under certain conditions. This would be true for storage times up to 40 years if initial temperatures range up to 420°C for fuel cooled only one year in a storage pool and if, as in most spent fuel, the hoop stress is less than 70 MPa. For fuel cooled five years under similar storage conditions, the limit is 380°C. The temperatures are sensitive to the specific thermal properties of the storage installation and fuel conditions. Flaws in the spent fuel cladding would be expected to increase the frequency of fuel failures by creep rupture, although the failure mode is expected to be slow fission gas leakage through a pin hole crack.

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DEFORMATION MAP CONSTRUCTION

An extensive survey of the literature was performed to identify possible mechanisms of deformation in the Zircaloy alloy system. Based upon this survey, nine mechanisms were identified for investigation. These mechanisms were placed into four generalized categories: high-stress deformation, diffusion-controlled dislocation creep, grain boundary sliding, and diffusional creep. The nine processes and their classifications are shown in Table 1.

The high-stress deformation mechanisms category includes the theoretical strength of the material lattice (Evans and Rawlings 1969) that serves as an upper limit to deformation by dislocation motion. Below this upper limit, high-stress deformation will occur principally by conservative dislocation motion or glide. The constitutive relations describing dislocation glide, formulated by Ashby (Ashby and Frost 1975) and listed in Table 2, were adopted for this study.

The diffusion-controlled dislocation creep processes included formulations for high-temperature climb (Sherby and Burke 1968), low-temperature climb (Ashby and Frost 1975), and Harper-Dorn diffusional climb (Harper and Dorn

TABLE 1. Deformation Mechanisms

<u>Category</u>	<u>Mechanism</u>	<u>Abbreviation</u>
High-Stress Deformation	Theoretical Tensile Stress	TH
	Dislocation Glide	DG
Diffusion-Controlled Dislocation Creep	High Temperature Dislocation Climb	HT
	Low Temperature Dislocation Climb	LT
	Harper-Dorn Diffusional Climb	HD
Grain Boundary Sliding	GBS - grain boundary diffusion controlled	GBS
	GBS - lattice diffusion controlled	GBL
Diffusional Creep	Nabarro-Herring	NH
	Coble	CO
Athermal		ATH

TABLE 2. Constitutive Equations

Theoretical strength	$\dot{\epsilon}_{TH} = \infty \text{ for } \sigma > \sigma_{TH} \quad \text{where } \sigma_{TH} = \left(\frac{E\gamma_s}{a_0} \right)^{1/2} \approx \frac{E}{10}$ $\dot{\epsilon}_{TH} = 0 \text{ for } \sigma < \sigma_{TH}$
Dislocation glide	$\dot{\epsilon}_{DG} = \exp \left[-(S - \sigma) \frac{ba}{kT} \right] \text{ for } \sigma > \sigma_0$ $\dot{\epsilon}_{DG} = 0 \text{ for } \sigma < \sigma_0$
High-temperature climb	$\dot{\epsilon}_{HT} = A_{\ell} D_{o\ell} \exp \left(\frac{-Q_{\ell}}{RT} \right) \left(\frac{Eb}{kT} \right) \left(\frac{\sigma}{E} \right)^n$
Low-temperature climb	$\dot{\epsilon}_{LT} = 50 A_{\ell} D_{oc} \exp \left(\frac{-Q_c}{RT} \right) \left(\frac{Eb}{kT} \right) \left(\frac{\sigma}{E} \right)^{n+2}$
Harper-Dorn climb	$\dot{\epsilon}_{HD} = A_{HD} D_{o\ell} \exp \left(\frac{-Q_{\ell}}{RT} \right) \left(\frac{Eb}{kT} \right) \left(\frac{\sigma}{E} \right)$
Grain boundary sliding (grain boundary diffusion)	$\dot{\epsilon}_{GBS} = A_{GBS} D_{ogb} \exp \left(\frac{-Q_{gb}}{RT} \right) \left(\frac{Eb}{kT} \right) \left(\frac{b}{d} \right)^3 \left(\frac{\sigma}{E} \right)^2$
Grain boundary sliding (lattice diffusion)	$\dot{\epsilon}_{GBL} = A_{GBL} D_{o\ell} \exp \left(\frac{-Q_{\ell}}{RT} \right) \left(\frac{Eb}{kT} \right) \left(\frac{b}{d} \right)^2 \left(\frac{\sigma}{E} \right)^2$
Nabarro-Herring creep	$\dot{\epsilon}_{NH} = A_{NH} D_{o\ell} \exp \left(\frac{-Q_{\ell}}{RT} \right) \left(\frac{Eb}{kT} \right) \left(\frac{b}{d} \right)^2 \left(\frac{\sigma}{E} \right)$
Coble creep	$\dot{\epsilon}_{CO} = A_{CO} D_{ogb} \exp \left(\frac{-Q_{gb}}{RT} \right) \left(\frac{Eb}{kT} \right) \left(\frac{b}{d} \right)^3 \left(\frac{\sigma}{E} \right)$

1957). All three formulations use a power law expression to describe the relationship between strain rate and stress. These mechanisms are described below.

As temperature rises, the dislocations within the material attain the ability to climb as well as to glide. If a dislocation gliding through the material meets a discrete obstruction to glide, it can overcome the obstacle by climbing to the next plane and continuing the glide there. In this case, climb of the dislocation, rather than glide, is the rate controlling factor. At high temperatures, this climb is accomplished by lattice diffusion. This high-temperature climb has been shown, empirically, to have a σ^n stress dependence. At low temperatures, low-temperature climb, which has a σ^{n+2} type stress dependence, is dominant. This mechanism is controlled by transport of matter via dislocation core diffusion.

In investigating the creep behavior of aluminum near its melting point, Harper and Dorn (1957) observed steady-state creep at very low stresses but at much higher strain rates than can be achieved through diffusional creep (Nabarro-Herring and Coble) alone. They then showed in a quantitative manner that this behavior was due to dislocation climb and that the creep rate was indeed linearly dependent upon stress.

Only recently has the contribution of grain boundary sliding to the deformation of Zircaloy been proposed (Bocek, Hoffman and Peterson 1977; Burton, Donaldson and Reynolds 1979). This mechanism is dominant when the strain rate is low enough to allow diffusion to remove the incompatibilities generated when the grains slide against each other. At high temperatures, the creep rate is lattice diffusion controlled; at lower temperatures its behavior is controlled by grain boundary diffusion (GBD). In this investigation two constitutive relations are used to describe the grain boundary sliding (GBS) processes. The first describes GBS that is controlled by GBD; the second describes GBS controlled by lattice diffusion (GBL) (Luthy, White and Sherby 1979). Theoretical equations similar to those used in this study have been proposed by other investigations (Mukherjee 1971; Stowell, Robertson and Watts 1969). The primary difference is that in the work being reported the strain rate ($\dot{\epsilon}$) is assumed to be proportional to the inverse cube of the grain size (d) when GBD

dominates and proportional to the inverse square when GBL dominates. These grain size dependencies are based upon the data from several studies (Stowell, Robertson and Watts 1969; Herriot, Suery and Baudalet 1972; Holt and Backofen 1966).

The diffusional creep category of mechanisms is expected to be important in the low stress region of Zircaloy deformation. Two mechanisms, NH and CO, were included in the development of the deformation maps. In small grain materials at high temperature, the existence of a stress gradient can cause the transport of matter from one side of the grain to the other as indicated in Figure 1. This transport of atoms results in a net change of shape (creep) of the grain. The creep rate in this case is proportional to the applied stress and is limited by GBL. Nabarro (1948), and later Herring (1950), developed the

FIGURE 1. Nabarro-Herring and Coble Creep. The applied stress provides the driving force for the diffusion of atoms along paths similar to those shown above. Nabarro-Herring creep requires lattice diffusion. Coble creep results from grain boundary diffusion.

analyses that allow the calculation of the creep rate for this mechanism. At temperatures lower than those at which NH creep occurs, the stress gradient can still cause a transport of matter. This transport, due to the lower temperature, occurs along the grain boundaries, rather than through the lattice, because the activation energy for GBD is much lower than that for GBL. This phenomena was first quantified by Coble (1963). It is illustrated in Figure 1.

The equations and the coefficients for these mechanisms are listed in Tables 2, 3, and 4.

With the use of quantified equations for each of the deformation mechanisms, a deformation map was produced by finding the portion of stress/temperature space where a particular mechanism predicted the highest strain rate. Analytical solutions were derived for each of the boundaries between regions. Figure 2 is the resulting deformation map. The limits of $\ln(\sigma/E)$ were chosen so that stresses from 5 to 500 MPa would be included. The temperature limits were chosen to restrict consideration to the α -phase of zirconium ($T < 865^\circ\text{C}$) and to incorporate temperatures down to 50°C . Lines of constant stress from 5 to 500 MPa and temperature marks in $^\circ\text{C}$ have been added to the right and top axes of the graph for interpretive convenience.

The map consists of regions dominated by ATH, LT, HT, GBS, CO, and NH deformation mechanisms. The theoretical strength and dislocation glide deformation regions do not show up on the plot because they occur at values of σ/E higher than shown ($\ln \sigma/E > -3.73$). HD climb does not appear because NH creep dominates for this grain size. GBL is also narrowly eliminated by slightly higher HT and NH deformation rates.

From a quick glance at the deformation map it appears that LT, GBS, and CO mechanisms dominate the stress/temperature space. However, the inverse temperature axis crowds a wide range of temperatures to the right side of the map. For instance, over 80% of the temperatures are on the right half of the map. Also, the decrease in Young's modulus with increasing temperature causes the constant stress contours to curve upward. The position of dominant deformation

TABLE 3. Symbols and Coefficient Values for Deformation Equations

$\dot{\epsilon}$	= strain rate
σ	= stress
T	= temperature
R	= gas constant = 8.3144 J/mole-K
k	= Boltzmann's constant = 1.38×10^{-29} MJ/K
b	= Burger's vector = 3.23×10^{-10} m
d	= grain size = 5.0×10^{-6} m
T_m	= melting temperature of Zircaloy = 2125K
E	= Young's modulus $(11.81 - 14.59 T/T_m) \times 10^4$ MPa, $T_m/T > 4.06$ for $T < 865^\circ\text{C}$ $(11.09 - 11.61 T/T_m) \times 10^4$ MPa, $T_m/T < 4.06$
n	= stress exponent = 5
Q	= activation energy
Q_ℓ	= 250 kJ/mol
Q_{gb}	= 175 kJ/mol
Q_c	= 180 kJ/mol
D_o	= diffusivity coefficient
$D_{o\ell}$	= 2.00×10^{-4} m ² /s
D_{ogb}	= 3.89×10^{-6} m ² /s
D_{oc}	= 2.26×10^{-6} m ² /s
A	= strain rate coefficient
A_ℓ	= 7.4×10^7
A_{CO}	= 5.362×10^2
A_{GBS}	= 8.85×10^6
A_{NH}	= 0.92
A_{GBL}	= 5×10^2
S	= $\frac{Eb}{3\ell}$
ℓ	= obstacle spacing
a	= activation area
σ_o	= cutoff stress = 2.4×10^{-2} E

TABLE 4. Reduced Deformation Equations

High Temperature Climb	$\ln \dot{\epsilon}_{HT} = 5 \cdot \ln(\sigma/E) + 55.75 - 14.15 \left(\frac{T_m}{T} \right) + \ln \left(\frac{T_m}{T} \right) + \ln \left(\frac{E}{10^4} \right)$
Low Temperature Climb	$\ln \dot{\epsilon}_{LT} = 7 \cdot \ln(\sigma/E) + 55.18 - 10.19 \left(\frac{T_m}{T} \right) + \ln \left(\frac{T_m}{T} \right) + \ln \left(\frac{E}{10^4} \right)$
Grain Boundary Sliding (Grain Boundary Diffusion)	$\ln \dot{\epsilon}_{GBS} = 2 \cdot \ln(\sigma/E) + 20.74 - 9.9036 \left(\frac{T_m}{T} \right) + \ln \left(\frac{T_m}{T} \right) + \ln \left(\frac{E}{10^4} \right)$
Nabarro-Herring	$\ln \dot{\epsilon}_{NH} = \ln(\sigma/E) + 18.25 - 14.15 \left(\frac{T_m}{T} \right) + \ln \left(\frac{T_m}{T} \right) + \ln \left(\frac{E}{10^4} \right)$
Coble	$\ln \dot{\epsilon}_{CO} = \ln(\sigma/E) + 11.03 - 9.9036 \left(\frac{T_m}{T} \right) + \ln \left(\frac{T_m}{T} \right) + \ln \left(\frac{E}{10^4} \right)$

regions is generally as expected. The map predicts the following general dominance of stress/temperature space.

Mechanisms	Stress	Temperature
Coble	low	low
Nabarro-Herring	low	high
Grain boundary Sliding	intermediate	low
Athermal	high	low
Low Temperature Climb	high	intermediate
High Temperature Climb	high	high

Figure 3 allows a comparison of the deformation map predictions with the experimental data field. Experimental data (from sources listed in Appendix A) were analyzed and the value of the stress exponent 'n' calculated (from least square linear fits of $\ln \dot{\epsilon}$ versus $\ln \sigma$ plots); the activation energy 'Q' was also used whenever it was provided. These data were then categorized according to n and Q values.

$1 < n < 3, Q_{gb}$	Grain Boundary Sliding (boundary diffusion)
$1 < n < 3, Q_l$	Grain Boundary sliding (lattice diffusion)
$3 < n < 7, Q_l$	High Temperature Climb
$7 < n < 9, Q_c$	Low Temperature Climb.

FIGURE 2. Deformation Map with Constant Stress and Strain Rate Contours

FIGURE 3. Deformation Map with Experimental Data

These categories allowed the evaluation of the probable location of the boundaries between these mechanisms. Different symbols were used to plot points having different n and Q characteristics. For a point to be placed in one of the four categories, both n and Q criteria did not have to be met. For instance, several investigators took data at different stresses but at the same temperature. From these, data stress exponents were calculated, but activation energies were not. Based upon the stress exponent and the temperature of the data a deformation mechanism was assigned. Only unirradiated data are included on Figure 3. Few or no postirradiation creep data were available for comparison with the deformation maps.

That this map (Figure 3) is in good agreement with the characteristic activation energies and stress exponents which define the boundaries between the mechanisms is obvious when one notes that the vast majority of the symbols denoting each mechanism lie inside the boundary of that mechanism. The fact that any overlap between the symbols occurs at or near the boundaries separating the mechanisms those symbols represent also supports the contention that the deformation map is in good agreement with the experimental data. A boundary is defined as that region where the two competing mechanisms predict the same strain rates. Thus, it is expected that more than one mechanism would be observed at or near a boundary.

The predicted steady-state strain rate and the available strain rate data field were also compared. Over the stress and temperature field, the predictions match the data very well.

The deformation and fracture map of Figure 2 contains a region described as athermal creep. The existence of the athermal region was established during a comparative examination of the strain rate data and the predicted strain rates at various temperatures. It was noted that at lower temperatures and above certain stresses the strain rate was essentially independent of temperature. This effect was so pronounced, in fact, that superimposing the experimental data from various low temperatures showed that the data at each stress occupied only a small range of strain rates. The athermal region was added to the map by assuming that the strain rate within this region was independent of temperature and was a function only of homologous stress; in other words, the

temperature and was a function only of homologous stress; in other words, the strain rate within the region at any homologous stress would be constant and equal to the strain rate at that homologous stress on the boundary of the region. The boundary of this region was determined by the locus of stresses above which the strain rate was judged to be independent of temperature for each temperature on the deformation map. Constant strain rate contours of the deformation map, shown in Figure 2, illustrate this point.

Since initial development, the deformation and fracture maps in this report have gone through many stages of development. This development is documented by Khan, Madsen, and Chin (1985) and by Keusseyan (1985).

FRACTURE MAP CONSTRUCTION

Unfortunately, the methods of fracture map development are not as advanced as those used in deformation map generation. There is much debate over the formulation of equations describing different fracture mechanisms. One transgranular and three intergranular processes of fracture were considered in this study. The equations used to describe the fracture processes are shown in Tables 5, 6, and 7.

At the higher end of the spectrum of stresses is ductile transgranular fracture. In this mechanism the presence of a 'hard' inclusion disturbs the elastic and plastic displacement fields and thus leads to the build up of local stresses at the inclusion-matrix interface. Once these local stresses reach a critical value, the inclusion either separates from the matrix or fractures. Either way a hole is nucleated. Once a void has nucleated further, plasticity

TABLE 5. Fracture Equations

Theoretical shear strength	$\sigma_{th} = \left[\frac{E\gamma_f}{a_o} \right]^{1/2} \approx \frac{E}{10}$
Transgranular fracture	$t_f^{TG} = \left[\epsilon_n + \left(\frac{1}{4.937} \right) \left(\frac{n}{n-1} \right) \ln \left(\frac{0.38}{f_v^{1/2}} - 1 \right) \right] \dot{\epsilon}^{-1}$
Triple-point cracking	$t_f^{TP} = \frac{\gamma_f}{E d \xi} \left(\frac{\sigma}{E} \right)^{-1} \dot{\epsilon}^{-1}$
Cavitation--diffusional growth	$t_f^{CD} = \frac{2.525 \times 10^{-3} \ell^3}{\delta D_{ogb} b^2 \exp \frac{-Q_{gb}}{RT}} \left(\frac{kT}{Eb} \right) \left(\frac{\sigma}{E} \right)^{-1}$
Cavitation--power law growth	$t_f^{CP} = \left[\frac{(1 - 0.78 P_o \ell_o)}{4.87} \right] \dot{\epsilon}^{-1}$

TABLE 6. Symbols and Coefficient Values for Fracture Equations

t_f	= time to fracture
ϵ_n	= hole nucleation strain = 0.08
f_v	= volume fraction of intragranular inclusions = 0.025
P_0	= average particle diameter = 100 Å
λ_0	= particle spacing along boundary = 2.0×10^{-6} m
ξ	= $\dot{\epsilon}_{GBS}/\dot{\epsilon}$ = contribution of grain boundary strain rate to total strain rate = 0.2
λ	= average cavity spacing = 2.6×10^{-6} m (6 per grain segment)
δ	= width of grain boundary = 1.6×10^{-8} m (50 Burger's vectors)
γ_f	= free surface energy created by fracture = 35 J/m ²
a_0	= lattice spacing
σ_{th}	= maximum theoretical stress

TABLE 7. Reduced Fracture Equations

Transgranular Fracture	$\ln t_f^{TG} = -1.797 - \ln \dot{\epsilon}$
Triple-Point Cracking	$\ln t_f^{TP} = -5.655 - \ln \dot{\epsilon} - \ln \frac{\sigma}{E} - \ln \frac{E}{10^4}$
Cavitation--Diffusional Growth	$\ln t_f^{CD} = +4.15 - \ln \dot{\epsilon}_{GBS} + \ln \frac{\sigma}{E}$
Cavitation--Power Law Growth	$\ln t_f^{CP} = -1.587 - \ln \dot{\epsilon}$

makes it grow until it joins with other voids and a fracture path is created. This process is illustrated in Figure 4 and explained by Ashby et al. (1979).

Transgranular creep controlled fracture is found at somewhat lower stresses and at higher temperatures. This mechanism is very similar to ductile transgranular fracture. Here also, voids nucleate at inclusions within the matrix and grow as the material creeps until coalescence occurs and the material fractures. Void nucleation in transgranular creep follows the steps described earlier for ductile fracture, i.e., stress is concentrated at hard inclusions due to plastic flow until the local stress reaches a critical level.

FIGURE 4. Ductile Transgranular and Transgranular Creep Fracture Mechanisms. Ductile transgranular and transgranular creep fracture are initiated when holes nucleate at inclusions within the matrix. These holes grow until they coalesce and the material fractures.

In this case, however, if the temperature is high enough to allow diffusion, there is transport of matter from those areas on the inclusion surface that are in compression to those areas that are in tension. This transport tends to stabilize the plastic flow and thus postpone the coalescence of the voids. These two transgranular mechanisms, along with the three intergranular mechanisms discussed below, are described in detail by Ashby et al. (1979).

Experimental observations of intergranular fracture have shown that there are two distinct characteristics of intergranular failure: wedge cracking and cavitation. One constitutive equation for wedge or triple-point cracking is presented; cavitation failure is described by two processes: power-law and diffusional growth.

Based upon experimental observations, triple-point cracking occurs at low strain and relatively high stress (Gandhi and Ashby 1979; Williams 1967; Morris and Harries 1977). In this mechanism, the application of a tensile stress causes sliding of grains along the grain boundaries, giving rise to a stress concentration at triple points. This stress concentration can be relieved by

the nucleation of a crack or change of the grain boundary orientation (Figure 5). Once the crack has been nucleated at the triple point, it continues to grow by sliding of the grain boundaries.

Cavitation failure, on the other hand, has been experimentally observed under conditions of low stress and high temperatures. Here, cavities are nucleated and grow on the grain boundaries; the cavities continue linking up until final fracture occurs. The nucleation process of intergranular cavities is not well understood; therefore, cavitation models usually assume a fixed number of cavity nuclei with growth of these cavities beginning upon the application of stress. Experimental evidence for creep-induced cavities at grain boundary inhomogeneities in Zircaloy-4 has been found (Keusseyan 1985).

Two mechanisms of cavity growth were considered in this study: diffusional growth (Hull and Rimmer 1959; Speight and Harris 1967; Speight and Beere 1975; Raj and Ashby 1975) and power-law growth (Monkman and Grant 1956; Nix, Matlock and DiMelfi 1977). Diffusional growth models are based on the fact that vacancies diffuse along the grain boundaries, preferentially being absorbed into cavities (Figure 6). This movement of vacancies is equivalent to a reverse flow of atoms from the cavity between two adjacent grains.

FIGURE 5. Triple-Point Cracking Mechanism. As the material deforms, voids nucleate at triple points. Further deformation causes growth until voids coalesce and a fracture path is created.

FIGURE 6. Diffusion-Controlled Cavitation Fracture Mechanism. In diffusion-controlled cavitation, cavities nucleate at irregularities and inclusions on the grain boundaries. These cavities then grow by diffusion along the grain boundaries.

Unfortunately, the predicted proportionality between $\dot{\epsilon}$ and t_f of the diffusional growth model does not match a large body of data where the following experimental relation is found

$$\dot{\epsilon} t_f = K \quad (1)$$

where K is the Monkman-Grant constant. To explain this inverse dependence between creep rate and time to fracture, mechanisms in which cavity growth is controlled by power-law creep have been proposed. At lower temperatures and higher stresses, the diffusion field of each cavity is hypothesized to extend less than halfway to the next cavity, as depicted in Figure 7. Thus, the cavities are separated by a bridge of material that must deform if the cavities are to grow. The growth of each cavity is thus controlled by the creep of the surrounding matrix of material, which is in turn controlled by a power-law-type behavior as is illustrated in Figure 7. To describe this creep-controlled fracture behavior, the formulation of Raj (1978) was adopted in this study. Mechanisms involving dynamic recrystallization and dynamic fracture were not

FIGURE 7. Power-Law-Controlled Cavitation Fracture Mechanism. If the diffusion field of a cavity on the grain boundary extends less than half way to the next cavity, the cavitation behavior is power-law-controlled. The cavities are encased in a cage of material that deforms according to a power law function.

included in this analysis. It is anticipated that conditions under which these mechanisms occur will not be seen during dry storage, except in accident situations.

Using the fracture equations with the appropriate coefficients, analytical solutions for the boundaries between dominant fracture regions were developed. A fracture map was then generated using the strain rate predictions of the deformation map. This fracture map is depicted in Figure 8. Direct substitution of strain rates from the deformation map into the fracture equations results in a map that is totally dominated by triple-point cracking for stresses greater than 150 MPa. This phenomenon results from the assumption that the contribution of GBS to the creep rate is a constant 20% ($\xi = 0.2$). To alleviate this problem, an upper limit stress of 200 MPa was chosen for triple

FIGURE 8. Fracture Map

point cracking. At this stress, the GBS contribution was placed at zero. Hence, transgranular fracture becomes the dominant mechanism for higher stresses.

In the fracture map, as in the deformation maps, the theoretical strength of the material, which is higher than the σ/E values shown, does not appear. The fracture modes are positioned on the map as expected with the following general characteristics relative to stress and temperature space:

<u>Mechanisms</u>	<u>Stress</u>	<u>Temperature</u>
Transgranular	high	---
Triple-point cracking	high intermediate	low
Cavitation Power Law	low intermediate	---
Cavitation Diffusion	low	---

With the use of the constant strain rate contours from the deformation maps, time to failure curves can be generated. These time to failure contours generated from the fracture equation are shown in Figure 9. As can be seen from the shape of these curves, the transition from cavitation power law (CPL) growth to cavitation diffusional (CDL) growth has the greatest effect on the predicted time to failure of all fracture mode transitions.

Because the fracture mechanisms are dependent on the rate at which deformation is accumulated, a change in the deformation map is usually reflected in the fracture map. Often, both the regions of dominance and the times to fracture are altered. For example, the introduction of the athermal deformation region caused the CPL fracture mechanism to remain dominant at lower temperatures and stresses by increasing the strain rates at the stress and temperatures that now fall within the athermal deformation region. The athermal region also has a small effect on the triple-point-cracking region, extending its dominance to lower temperatures and stresses.

Expressions to describe the ductile transgranular fracture region were developed based on experimental yield stress and ultimate tensile strength data (Lee and Park 1978). This region was added to the map by plotting the locus of the yield stresses, one from each temperature, onto the fracture map.

The effect of the athermal deformation region and ductile transgranular fracture region on the predicted times to failure can be seen in Figure 9. This figure is a plot of log stress versus log fracture time for various temperatures. Predicted fracture times are slightly decreased at the highest temperatures and increased for the lowest temperatures. The effect of the athermal deformation region is clearly seen by a collapse of different temperature fracture lines to a single line at higher stresses. This line extends from approximately 70 MPa at a fracture time of 10^7 seconds to about 300 MPa at a fracture time of approximately 10^{20} seconds. At this point the effects of the addition of the ductile fracture region begin to become apparent. The presence of this region reduces the slope of the time to fracture lines by a factor that approaches zero as the temperature approaches 675°C. This variation is caused by the temperature dependence of the yield and ultimate tensile strengths of the material.

FIGURE 9. Time to Fracture Contours Derived from the Fracture Map. The numbers across the contours are the temperatures, of each contour.

COMPARISON WITH EXPERIMENTAL RESULTS

DEFORMATION MAP

The experimental strain rates were placed on a series of stress versus strain rate plots with each plot containing strain rate predictions for a specific temperature. Two of these, at 350°C and at 400°C, are included as Figures 10 and 11.

To analyze the accuracy of these predictions a statistical correlation was developed.

$$\text{Correlation} = 1/N \sum_{i=1}^N (\ln \dot{\epsilon}_{\text{exp}} - \ln \dot{\epsilon}_{\text{pred}}) \quad (2)$$

where N = Total number of data points at the temperature

$$\begin{aligned} &= 1/N \sum_{i=1}^N \ln \dot{\epsilon}_{\text{exp}} / \dot{\epsilon}_{\text{pred}} \\ &= \ln (\dot{\epsilon}_{\text{exp}} / \dot{\epsilon}_{\text{pred}})_{\text{avg}} \end{aligned} \quad (3)$$

The value of this correlation provides a measure of the average error associated with the predictions at each temperature. A value of negative one implies that on the average, the strain rate at that temperature is over predicted by a factor of 2.71; a value of positive one implies an under prediction by a factor of 2.71; a value of zero implies that the prediction is correct on the average.

This specific correlation was used because, of all the various formulations available in the literature or developed by the author, this one was felt to best represent the various characteristics of the data and the predictions. However, this correlation is not considered nor implied to be foolproof.

FIGURE 10. Strain Rate Contours at 350°C with Experimental Data. The middle curve is the actual prediction; the other two give strain rates that are larger and smaller by an order of magnitude.

FIGURE 11. Strain Rate Contours at 400°C with Experimental Data. The middle curve is the actual prediction; the other two curves give strain rates that are larger and smaller by an order of magnitude.

This correlation was then compared with a standard deviation of the data for a given temperature. The standard deviation is defined as follows:

$$\text{Standard Deviation} = \frac{\left[\sum_{i=1}^N (\text{Mean} - \ln \dot{\epsilon}_{\text{exp}})^2 \right]^{1/2}}{N} \quad (4)$$

where the Mean is defined as

$$\text{Mean} = \frac{\sum_{i=1}^N \ln \dot{\epsilon}_{\text{exp}}}{N} \quad (5)$$

The correlation and the standard deviation are plotted in Figure 12. As seen from this figure, the absolute value of the error correlation is smaller than the standard deviation of the data at most of the temperatures considered. The apparent error, as defined by this correlation, can be misleading though, because this correlation can have a lopsided value simply due to lack of data at a specific temperature. Figures 12, 13, and 14 illustrate this point. From Figure 12 it would seem that as the temperature gets higher the correlation deviates more and more from zero. At 600°C the prediction seems to be the worst. But an examination of Figure 13 shows that there is very little data at 600°C. At 800°C (Figure 14) though, there is significantly more data and also a much better correlation. If, however, the data from Luthy, White and Sherby (1979) were removed from the 800°C prediction, this prediction would become quite similar to the 600°C prediction with a correspondingly large absolute value of the correlation. Conversely, the addition of data to the 600°C correlation would improve the correlation at this temperature significantly. The logical conclusion of these arguments is that Figure 12 should be used to identify systematic trends in deviation and that an accurate analysis of the strain rate predictions requires an examination of the predicted strain rate plots.

FIGURE 12. Statistical Evaluation of the Deformation Map. A comparison of the data shows the predicted strain rates to be accurate.

FIGURE 13. Strain Rate Contours at 600°C with Experimental Data

FRACTURE MAP

The experimental data were plotted on a series of fracture submaps in stress versus time to fracture space. Each one of these maps depicts a specific temperature. In addition to the experimental data at that temperature, the locus of predicted fracture times for the complete range of stress was also plotted on the map. Figures 15 and 16 show these maps for 350°C and 400°C.

To analyze the accuracy of the predictions a statistical correlation similar to the one developed for the deformation map was developed.

FIGURE 14. Strain Rate Contours at 800°C with Experimental Data

$$\text{Correlation} = 1/N \sum_{i=1}^N (\ln t_{\text{pred}} - \ln t_{\text{exp}}) \quad (6)$$

where N = Total number of data points at the temperature

$$\begin{aligned} \text{Correlation} &= 1/N \sum_{i=1}^N \ln t_{\text{pred}}/t_{\text{exp}} \\ &= \ln (t_{\text{pred}}/t_{\text{exp}})_{\text{avg}} \end{aligned} \quad (7)$$

FIGURE 15. Time to Fracture Contour at 350°C with Experimental Data. The center curve is the actual prediction; the other two curves give times to fracture that are larger and smaller by an order of magnitude.

FIGURE 16. Time to Fracture Contour at 400°C with Experimental Data. The center curve is the actual prediction; the other two curves give times to fracture that are larger and smaller by an order of magnitude.

Again, the value of this correlation gives the average error in the predictions for a given temperature. A value of positive one implies that, on the average, at this temperature the stress predicted for a given time to fracture is larger than the experimental stress by a factor of 2.71. This specific correlation was chosen because it best represented the relationship between the data field and the predictions. It should again be recognized that this correlation should be used as a guide to determine trends in deviation. Figure 17 is a plot of this correlation versus temperature. As can be seen from this figure, the average predictions at the lower temperatures are exceptionally good, but deviations tend to increase as the temperature rises. The relatively large numbers at the higher end of the temperature scale are somewhat misleading because of the way the correlation is defined. For example, at 700°C the correlation coefficient is -0.5 but Figure 18 shows the prediction to be in

FIGURE 17. Statistical Evaluation of the Fracture Map. The correlation constant provides an estimate of the accuracy of the time-to-fracture predictions.

FIGURE 18. Time to Fracture Contour at 700°C with Experimental Data. The center curve gives the actual fracture times at this temperature; the other two curves give fracture times that are larger and smaller by an order of magnitude.

excellent agreement with the data. This discrepancy is the result of the large slope of the predicted time to fracture curve at the lower stresses. This high slope causes a large error in the predicted stresses for even a small deviation in the fracture times. Generally, the predictions throughout the stress-temperature envelope are exceptionally good. The best predictions, however, lie in the temperature regime of most interest for dry storage (250 to 400°C).

DISCUSSION OF ENVIRONMENTAL AND IRRADIATION EFFECTS

The modeling of the effects of irradiation and stress corrosion cracking is based upon data from three sets: data generated by Argonne National Laboratory (Yaggee, Mattas, and Neimark 1979), the stress corrosion data from the Kraftwerk Union tests on unirradiated Zircaloy (Peehs and Fleisch 1986; Peehs, Stehle and Steinberg 1979; Peehs et al. 1979), and data gathered at the Chalk River test site on fuel rods tested in air at 250°C (Novak and Hastings 1983). These data sets show that loss of ductility is one of the principal effects of irradiation. Typical ductilities during creep of unirradiated Zircaloy range from 25 to 50%. Post irradiation data (Yaggee, Mattas, and Neimark 1979) show fracture strains between 5 and 12% that correspond to reductions in ductility of 200% to 1000%.

Our analysis predicts that stress corrosion cracking should not be an important factor in dry storage. Post-irradiation stress corrosion cracking data (Yaggee, Mattas, and Neimark 1979; Peehs, Stehle and Steinberg 1979; Peehs, Garzarolli, Hahn, and Steinberg 1979; Peehs and Fleisch 1986) show that at 325°C only stresses of 120 MPa or greater were found to cause stress corrosion cracking. Below 100 MPa all tests were terminated before failure with no indication of the onset of stress corrosion cracking. Similarly tests conducted by Kraftwerk Union in the Federal Republic of Germany show no indication of failure after 36,000 h of testing (Peehs and Fleisch 1986). These tests were conducted at 100 and 300°C and at stresses of 62 and 95 MPa. These results suggest that at the temperatures of interest for dry storage and at stresses below 100 MPa, stress corrosion cracking does not significantly affect fuel cladding performance.

For simplicity and because of the lack of additional post-irradiation data, irradiation was assumed to reduce the ductility of the cladding by a factor of 10 in the stress temperature region of interest for dry storage. Such an assumption is consistent with post-irradiation behavior seen in 316 SS. In a comparison of post-irradiation, in-reactor, and laboratory pressurized tube specimens, only the onset of tertiary creep was found to be affected by

irradiation (Lovell, Chin, and Gilbert 1980; Chin 1983). Because the steady-state creep rate is not affected by irradiation and since tertiary creep is theoretically associated with the onset of fracture of the material, it is consistent to assume that the primary effect of irradiation is a reduction in ductility that leads to a reduction by some constant factor in the fracture time. This assumption of a loss in ductility at fracture also leads to the enhancement of the triple-point-cracking region to lower stresses because triple-point cracking is a relatively low strain (to fracture) mechanism. Irradiation also moves the boundary of the ductile transgranular fracture region upward by increasing the yield strength of the material.

In addition to stress and temperature changes that occur during the dry storage of spent fuel, there will be Zircaloy microstructural adjustments that are anticipated to change the material properties. These adjustments are the result of recovery of irradiation damage, including irradiation enhanced dislocation density, point defect concentration, and other effects that are "annealed" out as a function of time in dry storage. This recovery is time and temperature dependent and is an important factor when storage temperatures exceed 350°C. As the irradiation damage is annealed out, the material properties are anticipated to approach those of unirradiated material. The principal effect of annealing in the dry storage temperature-stress regime is a recovery of ductility.

To model this annealing effect, a recovery factor was developed based upon experimental data generated by the Federal Republic of Germany (Steinberg, Weidinger, and Schaa 1984) on annealing of unirradiated cold worked Zircaloy cladding. Activation energy and time constants for this recovery factor were based upon the FRG data. The recovery factor is:

$$r = 1 - 0.9 \left[\frac{1}{1 + \sum_{i=1}^n K \Delta t_i \exp - Q/RT_i} \right] \quad (8)$$

where $k = 7.36 \times 10^{24} \text{ yr}^{-1}$

Δt_i = time at temperature, T , in years

$Q = 40,000 \text{ R}$

R = gas constant

This factor is 0.1 at time $t = 0$ and approaches 1 for high temperatures as time approaches infinity. In other words, for the irradiated material that is discharged from wet pool storage, the r factor = 0.1, which, in the model, signifies that the irradiated material has one tenth the strain to fracture of unirradiated material. As annealing out of irradiation damage occurs, this r factor approaches 1, which is the ductility of the unirradiated material. Figure 19 shows the time and temperature dependence of this recovery factor.

FIGURE 19. The Time and Temperature Dependence of Recovery Factor

LIFE FRACTION RULE

Of particular interest to cask designers is the maximum allowable initial storage temperature of the cask. This of course will be dependent on the spent fuel condition (burnup, wet pool storage time, etc.), cask loading, and specifics of cask design. All passive storage concepts to date project a decrease in storage temperature with time because of the decay in the heat generation rate of the fuel. To account for this anticipated decrease in storage temperature, a life fraction rule was used to calculate accumulated damage for decreasing temperature conditions. The life fraction rule can be expressed as

$$1 = \frac{\Delta t_1}{\tau_1} + \frac{\Delta t_2}{\tau_2} + \frac{\Delta t_3}{\tau_3} + \dots , \quad (9)$$

or

$$\sum_{i=1}^N \frac{\Delta t_i}{\tau_i} = 1$$

where Δt_i is the time spent at the i^{th} temperature, and τ_i is the time required to fracture a specimen under isothermal (at temperature), isostress testing conditions. When the cumulative damage fraction--the summation of the life fractions ($\Delta t_i/\tau_i$)--reaches one, the material is assumed to have failed.

To model the anticipated cask storage temperature as a function of time, an analysis of projected and measured fuel storage temperatures was made (Johnson and Gilbert 1983). The data were found to belong to a single family of curves having a constant slope when plotted as a log temperature versus log time. By defining time as the elapsed time after discharge from the reactor, the data can be expressed by the equation

$$T = T_0 t^Y \quad \text{for } t > 1 \text{ yr} , \quad (10)$$

where T is the temperature at time t , and T_0 and γ are constants. Again, time t is defined as the elapsed time after the end of irradiation. The value of γ was chosen as -0.1935 based upon available measured and projected temperature decay data. It is the value used in all predictions unless otherwise noted. The constant T_0 controls the initial temperature at the start of dry storage. It is determined by solving for T_0 in Equation (10) for a given temperature at the beginning of dry storage and a given elapsed time after the end of irradiation. A comparison of the predictions of Equation (10) and the data are shown in Figure 20.

FIGURE 20. Comparison of Predicted and Actual Dry Storage Temperature Histories

RESULTS AND DISCUSSION

MAXIMUM INITIAL DRY STORAGE TEMPERATURE^(a) FOR ISOTHERMAL HISTORY

Deformation and fracture maps provide important insight into predicting deformation and fracture modes for different dry storage conditions. Although isothermal spent fuel storage is not anticipated, the maximum allowable isothermal storage temperature provides an important benchmark. Placing constant time to failure contours on a fracture map allows selection of a desired life of an element and determination of the loci of constant stress and temperature points below which failure will not occur. Figure 21 is such a map based on the modified deformation and fracture equations for a dry spent fuel storage time of 40 years (unirradiated cladding).

For end-of-life fuel rod stresses of 100 MPa (14 ksi), a storage time of 40 years can be obtained at isothermal temperatures of 320°C or lower. This represents a slight increase from the 290°C maximum obtained from previous deformation and fracture map predictions. Based on the deformation and fracture map methodology presented, the following limits would be predicted for dry storage times of 1, 40, and 1000 years:

<u>Life, yr</u>	<u>Maximum Stress, MPa</u>	<u>Maximum Isothermal Temperature, °C</u>
1	90	400
40	100	320
1000	100	270

MAXIMUM INITIAL DRY STORAGE TEMPERATURE FOR DECAYING TEMPERATURE AND CONSTANT STRESS

It is anticipated that actual spent fuel rods in dry storage will experience a temperature history that can be described by Equation (10) (where T is a function of time spent in wet pool storage) and fuel rod characteristics. To

-
- (a) The maximum initial dry storage temperature is defined as the maximum allowable temperature at the start of dry storage to guarantee a certain lifetime in dry storage. In this report all calculations are based on assuming a lifetime of 40 years in dry storage.

FIGURE 21. Fracture Map Showing Region in Which a Fuel Rod with Isothermal/Decaying Temperature Conditions Would Last 40 Years or More

determine the axial initial storage temperature for spent fuel rods that have experienced various times in wet storage, a computer routine was developed. This computer routine uses the deformation and fracture map methodology in conjunction with the life fraction rule to predict the behavior in dry storage for a given fuel assembly with a known temperature history (a decaying one in this

case). A constant stress of 69 MPa (10 kpsi) was assumed for all the predictions presented in this section. In addition, no damage was assumed to accumulate during wet pool storage. The predictions are based on achieving a 40-year life in dry storage.

Figure 22 shows the projected maximum allowable temperature histories of maximally loaded casks containing fuel cladding that has spent 1 and 5 years in wet pool storage. As expected, the 1-year-old fuel can initially be stored at higher temperatures than the 5-year-old fuel. However, after a short time (1 to 2 years in dry storage), the 5-year curve crosses above the 1-year curve and stays above it for the rest of the storage time. It should be kept in mind that these figures represent maximum allowable temperature histories of maximally loaded casks, not individual assembly temperatures. These curves demonstrate the fact that although an individual rod of 5-year-old fuel will have a lower temperature than an individual rod of 1-year-old fuel, a cask containing 5-year-old rods may be stored at higher temperatures than a cask containing 1-year-old rods and still achieve the same storage life.

FIGURE 22. Predicted Maximum Allowable Temperature as a Function of Time Spent in Dry Storage to Ensure a 40-Year Life

This fact is also responsible for the shape of the cumulative damage fraction curves of Figure 23. Damage is accumulated much more rapidly (due to short times at high temperature) in the 1-year wet-pool-stored fuel than in 5-year-old spent fuel. These results indicate that consolidation of older fuel rods is possible as long as the maximum temperature history for the given age of spent fuel rods is not exceeded. Fuel rod consolidation would reduce the total number of required casks.

Figure 24 shows the maximum initial allowable storage temperature (required to achieve a 40-year life in dry storage) as a function of time spent in wet pool storage. Curves for two constant stresses, 40 and 70 MPa, are given. The large increase in initial dry storage temperatures for 1 and 2 years of spent fuel is due to annealing out of irradiation damage, which occurs rapidly at temperatures above 420°C.

FIGURE 23. Predicted Damage Accumulation as a Function of Time Spent in Dry Storage

FIGURE 24. Predicted Maximum Allowable Initial Dry Storage Temperatures for a Constant Stress and Decaying Temperature History

MAXIMUM INITIAL DRY STORAGE TEMPERATURE FOR DECAYING TEMPERATURE AND DECAYING STRESS

Calculations presented up to this point have assumed that the fuel cladding will experience a constant stress throughout its life in dry storage. This assumption is unrealistic because: 1) as the temperature drops, the fission gas pressure in the fuel rods will drop because of contraction of the gas, 2) as the fuel rods creep, their internal volume will increase and the pressure will drop (in both cases in a manner that can be approximated by the ideal gas law), 3) as the fuel rods accumulate strain, their radii increase, and thus the hoop stress seen by these tubes will increase, and 4) as the radii of the fuel rods increase, wall thicknesses decrease, and the hoop stress increases. These last two effects result from assuming that 1) the fuel rods can be approximated as thin wall cylinders for the purpose of calculating hoop stresses and 2) the volume of metal remains constant during creep.

The initial pressure for the cladding was calculated from the following relationship

$$P(\text{MPa}) = 0.0162 \times \text{Temperature}(^{\circ}\text{K}) - 4.099 \quad (11)$$

which gives the plenum gas pressure as a function of initial rod temperature. This equation was computed from measured end-of-life rod volumes (volume available for gas inside rod) and indicated fission gas release of spent fuel rods. This particular relationship was developed to produce hoop stresses of 70 MPa at the initiation of dry storage. The computer routine developed for the last section was then modified to reflect the above mentioned characteristics. Figure 25 displays the stress decay curves generated using this computer routine for fuel that has spent one and five years in wet pool storage. An interesting point about these curves is their similarity to the temperature decay curves of Figure 22. This is not surprising because stress is a linear function of pressure, which in turn is a linear function of temperature.

The above mentioned subroutine was also used to generate a new set of maximum initial dry storage temperatures. These new temperatures are depicted

FIGURE 25. Predicted Hoop Stress as a Function of Time in Dry Storage

in Figure 26. As can be seen from a comparison of Figures 24 and 26, this new set of assumptions has not affected the predicted maximum initial dry storage temperatures drastically.

THE NONIDEAL FUEL ROD

The model developed so far simulates fairly closely the environment that the fuel cladding will be exposed to during dry storage. This model fails to account for the fact that the cladding is not going to be made from a perfect material. In other words, the cladding will contain flaws that penetrate a certain distance into the wall. Few data are available on the flaw size distribution of irradiated Zircaloy fuel cladding. However, Miller, Uckan, and Tasooji (1981) have presented a formulation believed to describe the flaw size distribution in unirradiated Zircaloy cladding. Although the validity of this relationship has yet to be established, it was used to simulate existing flaws:

FIGURE 26. Predicted Maximum Allowable Initial Dry Storage Temperatures for a Decaying Stress and Decaying Temperature History. The initial stress is 70 MPa.

$$\phi(c) = 1 - \exp [-KA_S (1/\sqrt{c})^P] \quad (12)$$

where A_S is the surface area (1.61 m^2 for a PWR bundle), c is the flaw size, $\phi(c)$ is the probability that the cladding contains a flaw size greater than c , and K and P are constants ($1.7 \times 10^{-11} \text{ m}^{0.5875}$ and 5.175 , respectively). A plot of this flaw size distribution versus the ratio of flaw size to wall thickness is shown in Figure 27. Because the validity of the existence of any given flaw size as given by Equation (12) is in question, the following discussion considers three different flaw size to thickness ratios (c/t).

In calculating the maximum initial dry storage temperature for a given flaw size, a worst case analysis approach was taken. The thickness of the entire length of the cladding was assumed to have been reduced by an amount equal to the flaw size. In other words, it was assumed in the calculations that a flaw size of 25% of the wall thickness meant that the wall thickness was now only 75% of its original value. This methodology was then used to generate

FIGURE 27. Probability Distribution of Flaw Sizes at the End of Irradiation Service. Wall thickness is 0.00066 m .

a set of maximum initial dry storage temperatures for three casks, each of which had been designed assuming a certain maximum c/t (in this case 0.25, 0.5, and 0.75).

The resulting predictions are shown in Figure 28. The probabilities of flaws larger than the design value in the casks are 0.1544, 0.0275, and 0.01, respectively. The probabilities project that storage of casks designed on the basis of the 0.75 c/t at the predicted maximum initial dry storage temperature would result in 10 rods out to 1000 failing in 40 years. Similarly, for casks designed on the basis of 0.5 c/t and 0.25 c/t , storage at the associated maximum initial temperatures for 40 years would result in the failure of 28 and 155 rods, respectively.

ANALYSIS OF ASSEMBLY B02: EMAD TEST

The deformation and fracture map methodology was utilized to analyze the behavior of assembly B02 of the EMAD test. This test (Johnson et al. 1985),

FIGURE 28. Predicted Maximum Allowable Initial Dry Storage Temperature for Various Flaw Sizes. The maximum initial dry storage temperatures for three maximum flaw size to wall thickness ratios are shown.

which simulated the dry storage of a pressurized water reactor (PWR) nuclear spent fuel assembly, was conducted at the Nevada Test Site in the Engine Maintenance and Disassembly (EMAD) shielded facility. The objective of the test was to investigate the behavior of Zircaloy-clad spent fuel in air between 200°C and 275°C. An axial temperature gradient, consistent with that predicted for LWR fuel in dry storage, was maintained by electrical heaters. Thermocouples in the assembly were used to measure temperatures that were programmed to simulate the declining decay heat with time in a spent fuel assembly. During the second month of testing, a fuel rod breach was indicated by a large increase in krypton-85 level. This breach was the first to be detected in over 6000 Zircaloy-clad rods that have been monitored in dry storage tests and demonstrations. Assembly 802 is one of 17 Turkey Point 15 x 15 PWR spent fuel assemblies received by EMAD for dry storage testing. The assembly was discharged November 25, 1975, after 827 full-power days of operation. The average core burnup was 25,665 MWd/MTU.

The objectives of the deformation and fracture map analysis were to: 1) predict the damage and strain accumulated during dry storage and 2) determine the size of pre-existing flaw required to cause failure (if the accumulated damage is insufficient to cause failure of a defect-free fuel pin). To analyze the test, a temperature history function was constructed by combining two temperature decay curves each similar to Equation (10). The modeled temperature history is shown in Figure 29. The first temperature versus time section (150 to 100°C) is the temperature history the assembly experienced during silo and vault storage; the second section (300 to 275°C) represents the temperature history of the assembly during the FTT tests.

Assuming a constant stress of 70 MPa generated by internal fission gas pressure, cumulative damage and strain were calculated using the modeled temperature history of Figure 29. The results of the calculation are tabulated in Appendix D and shown in Figures 30 and 31. The predicted cumulative damage is less than 1%, and the accumulated strain is less than 0.01%. The results indicate that breach would not be anticipated for a nondefected fuel rod.

The fact that a single rod did fail indicates that a pre-existing flaw may have been responsible for the breach. Analyses presented in the following

FIGURE 29. B02 Modeled Temperature History

FIGURE 30. Predicted Cumulative Damage for Assembly B02

FIGURE 31. Predicted Cumulative Strains for Assembly B02

section demonstrated the effect that different size pre-storage flaws may have on the maximum initial dry storage temperature. Calculations were performed that took into account the temperature history of assembly B02, assumed a constant internal fission gas pressure equivalent to 70 MPa stress in a nondefected rod, and assumed the large release of krypton-85 to indicate creation of a through-wall crack. Based upon the calculations, the size of the pre-existing through-wall crack is calculated to have a crack depth to wall thickness (c/t) value of 73%. The c/t value is the percentage of radial wall thickness penetrated by a crack.

ANALYSIS OF ABNORMAL DRY STORAGE EVENTS TESTS

This section presents a deformation and fracture map analysis of a series of tests conducted at Pacific Northwest Laboratory (PNL)^(a) and Battelle

(a) Operated for DOE by Battelle Memorial Institute.

Columbus Laboratories (BCL) to investigate the effect of abnormal storage conditions on the behavior of Zircaloy spent fuel cladding (Stahl et al. 1984; Guenther 1983). The tests were conducted on irradiated and unirradiated rod segments, some containing UO_2 and others with stainless steel mandrels to simulate fuel column effects. The fuel rod segments were heated at rates between 9 and 55°C/h to simulate impeded cask convective heat transfer. Such an event may be brought about by a fire in an adjacent area or restriction in natural convection due to a debris bed resulting from tornadoes or earthquakes.

Five out of nine tests were analyzed using the deformation and fracture map methodology to calculate cumulative life fraction, failure temperature, failure stress, and failure strain. A lack of detailed temperatures and pressures as a function of test time for four of the nine rods prevented analysis of those rods.

Detailed internal pressure and temperature versus time data for the ramp heating rate tests were used to obtain the predictions. Table 8 gives a summary of the test data of the five rod segments analyzed. Functions describing the temperature and internal pressure of the test rod segments as a function of test time were developed from the data and served as the input to the deformation and fracture map analyses. The temperature history profiles are given in Figures 32, 36, 40, 44, and 48. Assuming a constant stress and temperature profile across the specimen, the program calculated cumulative rod damage, cladding stress (accounting for wall thinning due to creep), and strain for the temperature and pressure history of the test. Failure time was predicted using the life fraction summation criteria of Equation (10), which was then used to

TABLE 8. Abnormal Dry Storage Events Test Data

Heating Rate, °C/h	Rod (Segment)	Failure Temp., °C	Failure Pressure (MPa)	Maximum Pressure (MPa)	Average Strain, %	Temp. at Max. Pressure, °C
9.9	4F1	800	1.03	7.35	36	490
39.5	T3	801	1.61	7.67	21	500
52.9	1D1	763	1.94	7.73	17	490
55.5	Dummy	710	3.00	8.48	10	500
55.0	(TP-RH)	740	4.10	4.86	17	650

determine failure temperature, failure stress, and failure strain. The resulting cumulative damage (Figures 33, 37, 41, 45, and 49), stress (Figures 34, 38, 42, 46, and 50) and cumulative strain (Figures 35, 39, 43, 47, and 51) predictions are plotted as a function of test time. Table 9 summarizes and compares deformation and fracture map predictions with test results.

In three of five tests the deformation and fracture map methodology predicted failure earlier than it occurred. Good agreement is found between predicted and actual failure stress. Predicted failure temperatures and actual failure temperatures are within 50°C for all but one rod. There is a noted tendency in the data for the predictions to become less conservative at increased heating rates. There is, however, substantial disagreement between actual and predicted failure strain. This is hypothesized to be a result of two factors. First, the deformation and fracture map equations had been developed to describe long-term irradiated material properties. During the ramp heating tests, substantial recovery of the cladding occurred, leading to an annealed material with larger strains at failure. There appears to be approximately a factor-of-10 difference in the predicted and actual strains to failure, which is consistent with the anticipated differences between annealed and irradiated material. The second factor is that the strain listed as the actual value is the numerical average of some very large strains (ballooning in the center at the higher temperature segments of the rod) and some very small strain (towards the ends of the test segments). In the calculations used to make the predictions, a uniform temperature gradient was assumed across the test specimen. This produced a uniform strain. These factors combine to result in the difference between predicted and actual strains.

ANALYSIS OF FEDERAL REPUBLIC OF GERMANY AND WESTINGHOUSE TEST DATA

Analysis of Federal Republic of Germany Data

Table 10 is a summary of both unirradiated and irradiated strain rate data from tests conducted by the Federal Republic of Germany (FRG) in support of their dry spent fuel storage effort (Peehs et al. 1979). The data are superimposed on predictions of the deformation and fracture map methodology in Figure 52. These plots are at temperatures of 300, 325, 350 and 375°C.

FIGURE 32. Modeled Temperature Versus Time History: Rod Segment ZR-4F1. Broken line indicates predicted failure temperature and time.

FIGURE 33. Predicted Damage Accumulation Versus Time Test: Rod Segment ZR-4F1. Broken line indicates failure prediction.

FIGURE 34. Predicted Hoop Stress Versus Test Time: Rod Segment ZR-4F1. Broken line indicates failure prediction.

FIGURE 35. Predicted Cumulative Strain Versus Test Time: Rod Segment ZR-4F1. Broken line indicates failure prediction.

FIGURE 36. Modeled Temperature Versus Time History: Rod Segment ZR-T3. Broken line indicates failure prediction.

FIGURE 37. Predicted Damage Accumulation Versus Test Time: Rod Segment ZR-T3. Broken line indicates failure prediction.

FIGURE 38. Predicted Hoop Stress Versus Test Time: Rod Segment ZR-T3. Broken line indicates failure prediction.

FIGURE 39. Predicted Cumulative Strain Versus Test Time: Rod Segment ZR-T3. Broken line indicates failure prediction.

FIGURE 40. Modeled Temperature Versus Time History: Rod Segment ZR-1D1. Broken line indicates failure prediction.

FIGURE 41. Predicted Damage Accumulation Versus Test Time: Rod Segment ZR-1D1. Broken line indicates failure prediction.

FIGURE 42. Predicted Hoop Stress Versus Test Time: Rod Segment ZR-1D1. Broken line indicates failure prediction.

FIGURE 43. Predicted Cumulative Strain Versus Test Time: Rod Segment ZR-1D1. Broken line indicates failure prediction.

FIGURE 44. Modeled Temperature Versus Time History:
Rod Segment ZR-Dummy.

FIGURE 45. Predicted Damage Accumulation Versus Test Time:
Rod Segment ZR-Dummy. Broken line indicates
failure prediction.

FIGURE 46. Predicted Hoop Stress Versus Test Time: Rod Segment ZR-Dummy

FIGURE 47. Predicted Cumulative Strain Versus Test Time: Rod Segment ZR-Dummy

FIGURE 48. Modeled Temperature Versus Time History: Rod Segment ZR-(TP-RH). Broken line indicates failure prediction.

FIGURE 49. Predicted Damage Accumulation Versus Test Time: Rod Segment ZR-(TP-RH). Broken line indicates failure prediction.

FIGURE 50. Predicted Hoop Stress Versus Test Time: Rod Segment ZR-(TP-RH). Broken line indicates failure prediction.

FIGURE 51. Predicted Cumulative Strain Versus Test Time: Rod Segment ZR-(TP-RH). Broken line indicates failure prediction.

TABLE 9. Abnormal Dry Storage Events Test Data

Rod, Segment #	Heating Rate °C/h	Failure Temperature, °C		Failure Stress, MPa		Failure Strain, %	
		Actual	Predicted	Actual	Predicted	Actual	Predicted
4F1	9.9	800	696	14.9	14.1	27	3.8
T3	29.5	801	750	19.5	10.5	23	3.2
101	52.9	763	718	20.9	16.5	17	3.6
Dummy	55.5	710	753	--	6.8	10	1.2
TP-RH	55.0	740	754	--	15.4	17	4.4

TABLE 10. Strain Rate Data from the Federal Republic of Germany

Temperature (°C)	Stress (MPa)	(Sec ⁻¹)
275	200	4.215E-11
300	150	4.81E-11
300	200	8.25E-11
325	150	1.01E-10
325	200	1.953E-10
350	150	2.65E-10
350	200	1.18E-9
375	150	1.1185E-9
375	70	6.09E-11 ^(a)
375	70	4.74E-11 ^(a)
400	150	9.4E-10
400	112	3.61E-10
400	75	2.81E-10
400	70	1.74E-10
400	70	1.625E-10
400	70	1.44E-10
400	70	1.323E-10
400	70	1.87E-10
400	70	1.92E-10
400	70	1.48E-10
400	70	1.63E-10
400	70	1.0025E-9 ^(a)
400	70	9.485E-10 ^(a)

(a) Postpile creep data.

FIGURE 52. Comparison of the Predicted Strain Rates and FRG Experimental Data

Generally, good agreement is found between predictions and data. The data at these temperatures fall towards the lower limits of the data field (lower creep rates), but they are consistent with data of other investigators. Because the FRG data fall towards the lower creep rate side of the data field scatter, strain rates are best predicted by the lower-limit creep rate prediction of the deformation and fracture map models.

The FRG data have also been displayed on plots of stress versus fracture time predictions in Figure 53. These maps are at temperatures of 300, 325, 350 and 375°C. None of the FRG specimens have actually fractured; therefore, the test data represent maximum time of exposure at temperature without fracture. This has been indicated by an arrow pointing to longer times to differentiate between failed and unfailed specimens. Again the fracture maps appear to be conservative; no fractures are reported before predicted failure. At 350 and 400°C some specimens have exceeded the predicted fracture time without indications of failure. However, since the accumulated strain on each of these specimens exceeds 1%, they are all approaching failure. It is therefore concluded that the deformation and fracture maps provide good agreement with FRG data.

Analysis of Westinghouse Postpile Creep Data

The postpile strain rate data generated in support of the dry spent fuel storage program by Westinghouse Hanford Company and Battelle Columbus Laboratories are tabulated in Table 11. Plots of constant strain rate contours and superimposed data are contained in Figure 54. These data are at temperatures of 325, 450, 500, and 575°C. At all temperatures except 325 and 575°C the data agree with predictions of the deformation and fracture map models. However, at 325 and 575°C there are specimens that show a higher than nominally predicted creep rate. The data are not consistent at these temperatures. For instance, at 325°C there are five specimens subjected to nearly equal stress (146 to 157 MPa), but they show creep rates that vary by four orders of magnitude. Similarly, at 575°C there are four specimens in which the stress changes by over a factor of two; yet there is no detectable change in the creep rate. Part of this inconsistency is due to errors in measurement. The strains measured were in many cases on the order of the error of the measurement apparatus. To obtain reliable creep rate data, the tests would have to be conducted for

FIGURE 53. Comparison of the Predicted Fracture Times and the FRG Experimental Data. Errors indicate that specimens have not yet fractured.

TABLE 11. Strain Rate Data from Westinghouse Hanford Company

Temperature (°C)	Stress (MPa)	(Sec ⁻¹)
323	150	4.63E-7(a)
323	152	6.37E-8(a)
323	152	2.076E-8(a)
323	146	3.14E-9(a)
323	157	2.65E-10(a)
425	39.3	1.4E-8(a)
450	40.6	7.0E-9(a)
500	39.45	1.23E-7(a)
500	42.55	1.02E-7(a)
575	39.2	1.88E-8(a)
575	50.1	2.63E-8(a)
575	32.5	1.88E-8(a)
575	22.6	1.94E-8(a)

(a) Postpile creep data.

Source: 323° data are from Einziger and Kohli 1983; data for all other temperatures are from Einziger et al. 1982.

much greater periods of time. The second probable explanation is the inability to control all the unknowns in a hot cell test. Temperature variations and leaks in the pressurization system are likely to have caused errors in the conditions of the test.

The Westinghouse data have also been plotted in Figure 55. These data are at temperatures of 325, 450, 500 and 575°C. Again the extended arrows indicate unfailed specimens. For temperatures above 425°C, the deformation and fracture map methodology predicts failure for points where no fracture has been observed in experimental specimens. Based upon these data, the predictions appear to be too conservative. The data, however, are from the same set that showed inconsistency in the accumulated strain behavior, as indicated above. Therefore, careful sorting is required before any conclusion to alter the fracture map predictions is made.

FIGURE 54. Comparison of the Predicted Strain Rates and the Westinghouse Experimental Data

FIGURE 55. Comparison of the Predicted Fracture Times and the Westinghouse Experimental Data. Arrows indicate that specimens have not fractured.

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