ANALYSIS OF IRRADIATION-INDUCED
CREEP OF STAINLESS STEEL IN FAST
SPECTRUM REACTORS*

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ABSTRACT

The available stainless steel irradiation creep data from the EBR-II and DFR reactors were evaluated. Transient and steady state creep were observed. The steady state creep rate data are linear with flux and time. The stress dependence of the solution-annealed types 304L, 304, and M316 stainless steels is linear, whereas the 20 percent cold-worked type M316 stainless steel exhibit linear behavior for equivalent stresses below 15,000 psi and a higher stress exponent for higher stresses. For solution-annealed materials, irradiation-induced creep increases in the order M316, 304 and 304L.

The results of this analysis indicate that irradiation creep strain, ϵ_i , can be described by an equation of the form:

$$\epsilon_i = A\sigma\bar{E}\phi + (B\sigma + C\sigma^3) \bar{E}\phi t$$

where $\bar{E}$ is the average neutron energy, ϕ the total neutron flux, σ the applied stress, and A, B, and C are material constants. The first term in the above equation defines the transient irradiation creep component. The second term defines the steady state irradiation creep component. The fast reactor irradiation creep data are consistent with the concept of volume conservative strain.

Significant high fluence irradiation creep information can be obtained from residual stress measurements on irradiated tubing. An analysis of this technique is presented including consideration of the fluence dependence of irradiation creep and swelling.

ANALYSIS OF IRRADIATION-INDUCED CREEP OF STAINLESS STEEL IN FAST SPECTRUM REACTORS

1.0 Introduction

The design of fast reactor core components requires a knowledge of structural material creep properties under irradiation. Initial irradiation creep design positions were strongly influenced by Hasketh's model for the collapse of vacancy clusters into dislocation loops.[1] McElroy, et al.[2] and Gilbert[3] evaluated thermal and fast spectrum stainless steel irradiation creep data using damage function analysis, and concluded that irradiation creep is associated with an isotropic volume increase. Wolfer et al.[4] concluded that there is a relationship between swelling and irradiation creep, and that irradiation creep should be considered to give rise only to volume conservative strains. It was recommended that stress-induced deformation of metals during fast neutron irradiation be separated into isotropic and volume conservative strains. The dependence of the isotropic strains on stress is considered as a stress-effect on swelling.[5,6]

Recently, extensive fast spectrum irradiation creep data have become available from EBR-II[7] and DFR[8]. This paper presents an analysis of the data and provides a basis for design application of the data as well as for theoretical interpretation of the results. The available irradiation creep data cover a limited fluence ($<2 \times 10^{22}$) and temperature range (250-380°C). In addition, a method will be discussed for extracting high fluence irradiation creep data from residual stress measurements on irradiated fuel and capsule tubing.

2.0 Data Analysis

The EBR-II type 304L stainless steel pressurized tube data (chemical composition given in Table I) demonstrated steady state irradiation creep by the linearity of the irradiation creep strain-time data.[7] The demonstration of steady state irradiation creep in the case of the solution-annealed (SA) and 20% cold-worked (CW) DFR helical springs[8] is much more complex because each spring is loaded with a different stress and placed at different reactor positions, which extend from the hard neutron spectrum core out to softer spectrum regions. To demonstrate steady state irradiation creep, it is first necessary to determine the flux and stress dependencies. Walters et al.[7] showed that the EBR-II pressurized tube data are linear with respect to flux and stress. Nosedale and Lewthwaite[8] demonstrated that the DFR springs exhibited a linear stress dependence.

Table 1

Compositions of the Stainless Steels Used in Irradiation Tests

Alloy	C	Cr	Ni	Mo	Mn	Si	Co	Cu	P	S	Remarks
304 (EN58E)	.03	17.6	11	-	-	-	-	-	-	-	DFR Springs
304L	.023	18.43	10.05	.10	1.33	.55	.15	.15	.005	.013	EBR-II Pressurized Tubes
M316 (Batch 1)	.036	16.70	13.7	2.29	1.87	.64	-	-	.006	.01	DFR Springs
M316 (Batch 2)	.038	17.4	13.75	2.41	1.81	.63	-	-	.008	.006	"
M316 (Batch 3)	.036	16.84	13.25	2.25	1.67	.44	.022	-	.013	.01	"
316	.04/.06	17.0/18.0	13.0/ 14.0	2.0/ 3.0	1.5/ 2.0	.75 max.	.05 max.	.10 max.	.02 max.	.01 max.	FFTF Specification*

* Shown for comparative purposes.

The flux dependence of the DFR springs were examined by plotting the ratio of shear strain to shear stress, γ/τ versus the mean neutron energy flux, $E\phi$ for approximately the same equivalent full power times. Figure 1 shows that the data are linear for a typical example of SA M316.

The linear flux and stress dependencies were used to determine the time dependence of creep strain for the helical springs. Although each spring was loaded with a different shear stress, these stresses formed groups centered at 2.0, 4.0, 5.5 and 8.0 ksi for SA M316 (Batch 1), and 3.0, 5.0, 9.0 and 12.5 ksi for CW M316 (Batch 1). Figures 2 and 3 illustrate the linearity of the flux and stress normalized shear strains for CW and SA M316 as a function of the time at power. Only a small stress normalization was necessary to adjust the shear strains to these stress levels. This method minimized both transient strain effects and the high stress non-linear stress correction to the steady state creep strain in the case of CW M316, which will be discussed further below.

The dashed line on Figure 2 represents sequential measurements on a particular spring. The data points in Figure 3 show an apparent increase in irradiation creep after 2000 hours. This tendency could be simply due to data scatter or possibly a small amount of swelling in SA 316 starting after 2000 hours ($7 \times 10^{21} \text{ n/cm}^2$). For example, swelling of 0.02% would be sufficient to increase the creep rate by the amount observed.

In order to compare the SA 304L pressurized tube and the SA M316 and SA 304 spring irradiation creep data, all data were converted to equivalent stress and strain. To convert the hoop irradiation creep strain rates to corresponding equivalent strain rates, a value for Poisson's ratio is necessary. Figure 4 shows the resulting creep rates when Poisson's ratio is 0.5. The results show variations in creep rates with alloy composition. The irradiation creep rates increase in the order M316, 304 and 304L which is also the order of decreasing strength of these alloys. The creep model of McElroy et al.^[2] and Gilbert^[3] is associated with a Poisson's ratio of -0.75. This is a consequence of the influence of the thermal reactor data, and as a result at 280°C the model overpredicts the fast neutron spectrum SA M316 and SA 304 irradiation creep rates by factors of 5 and 3, respectively.

A creep rate temperature dependence given approximately by $1/T$ has been proposed.^[1,2,3,8] For the temperatures under consideration this is a weak temperature dependence, and this is supported by the axial strain profiles of the two EBR-II pressurized tubes at 15 ksi^[7]. The strain profiles were symmetrical above and below the core midplane for temperature differences of 30 to 70°C.

Figure 1. Stress & Flux Dependence of Irradiation Creep Strain

Figure 2. Time Dependence of Irradiation Creep Strain for CW M316

Figure 3. Time Dependence of Irradiation Creep Strain for SA M316

Figure 4. Comparison of DFR & EBR-II Irradiation Creep Data For Solution-Annealed Stainless Steels

Figure 5 shows that the stress-dependence of the steady state irradiation creep rate for the Batch 1 CW M316 helical springs is initially linear at low stresses, and becomes non-linear at higher stresses. The non-linear high stress effect may also be observed with springs at different stress levels irradiated for the same time. Figures 6 and 7 show the departure from linearity by the total shear strain (i.e. transient and steady state components) for the CW M316 springs in rigs 240/4 and 240/10.^[8] Since the transient strain for shear stresses greater than 3.0 ksi is a linear function of the stress (see Figure 8), then the high stress non-linear deviation shown in Figures 6 and 7 is associated with the steady state creep strain.

The SA M316 total shear strain spring data irradiated for 427 hours also show a non-linearity at high stress levels. However, the transient irradiation shear strain is not a linear function of the stress, but remains essentially constant until 8.0 ksi, when it increases rapidly (see Figure 8). This is in good agreement with the results for the SA 304L pressurized tubes, and suggests that the high stress non-linearity of the total shear strain SA material is due to the discontinuous increase in the transient irradiation creep component.

3.0 Residual Stresses In Irradiated Fuel Pin Capsules and Cladding

Pennell^[10] proposed a method to assess the relationship between swelling and irradiation creep. The cylindrical tubing used in fuel pins or capsules is subjected to temperature gradient of the order of 400 to 800°C/cm. A substantial amount of differential swelling may take place in the tubing leading to swelling stresses. Concurrent with differential swelling, irradiation creep will occur to relax these stresses. Depending on the thermal stress, the swelling rate, the temperature dependence of swelling, and the irradiation creep rate, residual stress will be present after the irradiation. By slitting the tube longitudinally, the residual stress can be calculated from the change in the slit width. If $\Delta\sigma_R$ is the residual stress, as defined by the stress on the outside minus the stress on the inside wall of the cylinder, then the change in the slit width, δ is given by:

$$\delta = \frac{(1-\nu^2)\pi D^2 \Delta\sigma_R}{2Ed} \quad (1)$$

where ν is the Poisson's ratio, E the elastic modulus, D the diameter of the tubing and d the wall thickness.

The residual stress, $\Delta\sigma_R$ is the difference between the residual stress on-power, $\Delta\sigma$ minus the thermal stress:

$$\Delta\sigma_R = \Delta\sigma - \left(\frac{E}{1-\nu}\right) \alpha \Delta T \quad (2)$$

Figure 5. Irradiation Creep Rates of CW M316

Figure 6. Total CW M316 Irradiation Creep Strain Vs. Stress for Rig 240/10

Figure 7. Total CW M316 Irradiation Creep Strain Vs. Stress for Riq 240/4

Figure 8. Transient Irradiation Creep Strain for M316

where α is the linear thermal expansion coefficient and ΔT the temperature difference across the wall. As an example of the evolution of the residual stress with time, let us assume that the irradiation creep rate, $\dot{\epsilon}_1$ is linearly dependent on the stress and flux. Then, the rate of change of the on-power residual stress is given by:

$$\dot{\Delta\sigma} = - \frac{E}{1-\nu} \left[\dot{\epsilon}_1 \Delta\sigma + \frac{2}{3} \Delta T \frac{\partial}{\partial T} \left(\frac{\Delta V}{V} \right) \right] \quad (3)$$

where $\Delta\sigma(t=0) = \frac{E}{1-\nu} \alpha \Delta T$.

If the differential swelling rate $\frac{\partial}{\partial T} \left(\frac{\Delta V}{V} \right)$ changes with time faster than the creep rate, the on-power residual stress builds up continuously until other relaxation mechanisms such as yielding or thermal creep are activated. As an example, Figure 9 shows the residual surface stress, $\Delta\sigma_R/2$ as a function of time for an EBR-II capsule of SA 304L. The creep rate was assumed to be time independent, and the swelling rate given by the theta swelling equation.^[3,11] The effect of irradiation creep rate is shown for: the nominal rate, $\dot{\epsilon}_1$; $2\dot{\epsilon}_1$; and $0.5\dot{\epsilon}_1$. The rapid development of the residual stress during the early part of the irradiation is due to the relaxation of the thermal stresses by irradiation creep. A reversal of the residual stress occurs, however, after sufficient amount of differential swelling has taken place.

An alternate situation is encountered if the creep rate, $\dot{\epsilon}_2$ is proportional to the swelling rate at higher fluences. This form of relationship between swelling and irradiation creep has been considered on theoretical grounds^[4,9] and can be expressed as:

$$\dot{\epsilon}_2 = \dot{\epsilon}_1 + D\sigma \frac{\Delta V}{V} \quad (4)$$

where D is a constant depending on material microstructure, and $\Delta V/V$ is given by % $\Delta V/V$ per unit time. For example, for a swelling rate of 1% $\Delta V/V$ per 10^{22} fast fluence ($E > 0.1$ Mev), a fast flux of 10^{15} , a stress of 10^4 psi, and a D value of 2×10^{-7} , the second term in equation (4) yields a creep rate of 7.2×10^{-5} %/hr. This value may be compared to the creep rates for SA 304L and M316, shown in Figure 4, of 5.4×10^{-5} and 1.7×10^{-5} %/hr, respectively. Thus, at the indicated swelling rate the irradiation creep rates of SA 304L and M316 would be enhanced by 130 and 420%, respectively.

Figure 9B shows the residual stress as a function of fluence for the creep law given by equation (4) and for various values of D . In contrast to the results shown in Figure 9A, a constant residual stress may be reached using equation (4). For this case, the steady state on-power load is given by:

$$\Delta\sigma = - \frac{2}{3} \frac{1}{\dot{\epsilon}_2} \Delta T \frac{\partial}{\partial T} \left(\frac{\Delta V}{V} \right). \quad (5)$$

Figure 9. Residual Hoop Stress on the Outside of a Fuel Pin Capsule

This analysis of residual stress in irradiated tubing demonstrates a method for obtaining high fluence irradiation creep data as well as information on the relationship between swelling and irradiation creep. Residual stress measurements on irradiated tubing are in progress. The preliminary analysis of these results indicates minimum D values (see equation 4) greater than 2×10^{-7} indicating a large increase in irradiation creep rate for high swelling rate values.

4.0 Discussion

The analysis of the EBR-II and DFR results presented earlier indicate the following general equation for irradiation induced creep strain, ϵ_i (in/in):

$$\epsilon_i = A\bar{E}\phi + (B\sigma + C\sigma^3) \bar{E}\phi t \quad (6)$$

where $\bar{E}$ is the average neutron energy (MeV), ϕ the flux ($n/cm^2/sec$), σ the stress (psi), t the time (seconds), and A, B and C are materials constants. The first term on the right side of equation (6) is the transient irradiation creep component, and the second term is the steady state irradiation creep component. Equation (6), is considered to be essentially athermal over the temperature range 250-380°C because the temperature dependence appears weak.

Table II lists the values of the constants which are applicable to several different types of stainless steel. The materials constants for irradiation creep of SA and CW stainless steels, shown in Table II, vary considerably depending upon alloy composition and heat treatment. The results reported for SA and CW 304 are based on a limited number of data points^[8] and should be considered as preliminary. A comparison of the steady creep constants of types M316 and 304 in the SA and CW conditions reveals a marked difference between the two steels. The SA and CW M316 stainless steel show similar steady state irradiation creep rates (below equivalent stresses of 15 ksi) whereas the CW 304 stainless steel shows a considerably higher steady state irradiation rate than the SA material. As indicated in Table II the CW 304 show a non-linear stress effect at equivalent stresses of 10 ksi. The greater sensitivity of the CW 304 stainless steel to stress may account for the observed difference between types 304 and M316 stainless steel. However, additional data on type 304 stainless steel are necessary to confirm the observed differences.

Values for the A constant are not reported for SA materials because the strain intercepts tend to be negative at low stresses (below equivalent stresses of 15,000 psi). It is proposed that the negative strain intercepts at low stress for SA stainless steel are related to the densification associated with the precipitation of $M_{23}C_6$ carbide. Out-of-pile densification has been measured at 700°C^[12,13] but not at the temperatures of the reactor experiments (250-380°C).^[7] However, carbide precipitation in austenitic stainless steels is enhanced by neutron irradiation, and the precipitation of $M_{23}C_6$ in SA 304 irradiated in EBR-II has been reported in the temperature interval between 370 and 425°C.^[13] It is proposed that the observed transient is the sum of two opposing components: a diameter decrease due to densification and an increase due to mechanical strain. At low stresses the densification component is larger, and negative strain intercepts result.

Table II

Irradiation Creep Materials Constants

(temperature range 250-380°C)

Material	$A \times 10^{22}$	$B \times 10^{29}$	$C \times 10^{38}$	Maximum Equivalent Stress Level (ksi)
CW M316	*0.12	0.37	0.51	21.7
CW 304	1.2	3-7	**	10.2
SA M316	-	0.47	-	14.7
SA 304	-	0.86	-	13.9
SA 304L	-	1.5	-	23.1

* The transient is more correctly given by

$$A = \frac{0.41 \times 10^3}{\sigma} + 0.12.$$

** Limited data indicate a non-linear high stress effect.

The values for the C constants for SA materials are zero for the maximum stress levels used in the experiments. The results indicate that an enhancement of irradiation creep at high stress levels occurs only in cold-worked materials.

5.0 Conclusions

The experimental results reveal the following main characteristics for irradiation creep in types 304 and M316 stainless steels:

1. A transient irradiation creep component linear with stress and $E\phi$ (only observed in CW 304 and CW M316 stainless steel).
2. At low stress levels, a steady state irradiation creep component linear with stress and $E\phi$ (observed in all materials examined in this study).
3. At high stress levels, a steady state irradiation creep component non-linear with stress (only observed in CW 304 and CW M316 stainless steel).
4. The fast reactor irradiation creep data are consistent with the concept of volume conservative strain.
5. Over the temperature range 250-380°C, the irradiation creep data exhibit only a weak temperature dependence.

The above characteristics of irradiation creep are considered in another paper by two of the authors.^[8] Theoretical models involving preferential loop nucleation and climb controlled glide are proposed to explain the experimental observations.

Residual stress measurements on irradiated capsule or fuel cladding can provide information on irradiation creep at high fluence and temperature. Measurements of this type would also provide information on the relationship between irradiation creep and swelling.

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