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APPLICATION OF THE GENERALISED THEORY OF YIELDING CREEP TO IRRADIATION CREEP IN ZIRCONIUM ALLOYS

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The theory of yielding creep, in its generalised form of accelerated thermal creep, is extended to include the temperature, neutron flux and time dependence of irradiation creep. The theory is consistent with the author's data at 78 °K and 40 °C and with Canadian data at 300 °C. The growth coefficient, G, of Zircaloy-2 at the respective temperatures is deduced to be 2.3, 1.2 and 1.5. Direct measurement by Buckley gives $G \approx 1.2$ at ~ 100 °C.

If the short term thermal creep behaviour continues to reactor lifetimes of 200 000 h the analysis suggests that linear extrapolation of the irradiation creep rate over-estimates the final strain by a factor of five.

The insensitivity to neutron irradiation of creep processes which are controlled by diffusion is discussed, e.g. Nabarro-Herring creep, the climb of edge dislocations and jogs in screw dislocations.

L'intérêt du fluage par "yielding" dans sa forme généralisée de fluage thermique accéléré est étendu afin d'inclure la relation entre température, flux de neutrons et temps, et le fluage par irradiation. La théorie est d'accord avec les données de l'auteur à 78 °K et 40 °C et avec les données Canadiennes à 300 °C. Le coefficient de croissance G du Zircaloy-2 aux températures respectives a été déduit être égal à 2,3, 1,2 et 1,5 respectivement. La mesure directe par Buckley donne $G \approx 1{,}2$ à 100 °C environ.

Si le comportement au fluage thermique à court terme se poursuit pour des durées de vie en réacteur de 200.000 heures, l'analyse suggère que l'extrapolation linéaire de la vitesse de fluage sous irradiation, surestime la déformation finale d'un facteur égal à 5.

L'insensibilité à l'irradiation neutronique des processus de fluage qui sont contrôlés par la diffusion est discutée, c'est-à-dire le fluage type Nabarro-Herring par montée des dislocations et des crans dans les dislocations vis.

Die Theorie des Kriechens unter Zugspannungen in ihrer verallgemeinerten Form des beschleunigten thermischen Kriechens wird auf das Kriechen unter Bestrahlung ausgedehnt, unter Berücksichtigung der Temperatur des Flusses und der Zeitabhängigkeit. Die Daten der Autoren bei 78 °K und 40 °C stimmen mit kanadischen Daten bei 300 °C und mit der Theorie überein. Der Wachstumskoeffizient G von Zirkaloy-2 bei diesen Temperaturen hat den Wert 2,3, 1,2 und 1,5. Direkte Messungen von Buckley ergeben $G \approx 1.2$ bei ~ 100 °C.

Wenn man von dem kurzzeiten, thermischen Kriechverhalten auf Reaktorstandzeiten von 200 000 Stunden linear extrapoliert, so zeigt sich, dass für die Kriechgeschwindigkeit unter Bestrahlung die Bruchdehnung um den Faktor fünf zu hoch liegt.

Die Unempfindlichkeit gegenüber Neutronenbestrahlung von Kriechprozessen, welche durch Diffusion bestimmt werden (z.B. das Nabarro-Herring-Kriechen, das Klettern von Versetzungen und Unstetigkeiten bei Schraubenversetzungen), wird diskutiert.

1. Introduction

In a single crystal with an anisotropic structure, neutron irradiation can cause a continuing change of shape. This is termed "growth". In a polycrystal the growth of each grain is restricted by its neighbours, which differ in orientation, and large stresses can be generated throughout the material. In the absence of sufficient ductility the polycrystal disintegrates to a powder. Beryllium oxide is an example. When the grains are sufficiently ductile, each deforms plastically and the material remains continuous. If an external stress is imposed on such a polycrystal it creeps at a rate which depends both on the applied stress and on the internal stresses. Uranium and graphite are two well known examples. If, as one measurement indicates, α-zirconium alloys show growth,

they must also show the irradiation creep discussed here. The only necessary physical conditions are that the material is continuous, ductile and polycrystalline.

In the absence of any information it will be assumed that the growth coefficient is independent of stress.

Two criteria may be used to analyse the deformation of a grain in the polycrystal. The first, appropriate to low stresses, is time dependent; the deformation rate is made to depend on the local internal stress through a creep equation. The analysis then gives an enhancement of the thermal creep rate. The second is time independent, and appropriate to stresses sufficiently high for a "plastic flow" approximation to be used. The rate of deformation of a grain then depends upon the constraints imposed by its neighbours, and the overall effect is a creep rate which has no relation to the thermal creep rate but is additional to it. This is termed yielding creep [1]). The second criterion is the asymptote of the first, both mathematically and physically. This is illustrated by applying the analysis to thermal creep data at high stresses and to the irradiation data at 40 °C.

2. The stress dependence of irradiation creep

The theory of accelerated thermal creep has been given by Blackburn [2]) who gives a complete expression appropriate to large applied stresses, and by Anderson and Bishop [3]) who give a linear approximation appropriate to small stresses. The complete expression is essential in the present case. Using the assumption that the stress in a randomly orientated aggregate of grains is the sum of the applied stress and a uniform internal stress, Blackburn derives a factor, here called f, by which the thermal creep is accelerated by neutron irradiation.

$$f = \frac{1}{\pi}\left(\frac{3}{2}\right)^{(n+1)/2}\left(\frac{I}{w}\right)^{n} \times \int_0^{\pi}\int_0^{1}\left\{\frac{2w}{3I} + \frac{(\cos^2\psi)(1-x^2)-x^2}{\sqrt{2}}\right\} \times \left\{1 + \frac{(w/2)[(\cos^2\psi)(1-x^2)-x^2]}{I} + \frac{2w^2}{3I^2}\right\}^{(n-1)/2} dx\,d\psi. \qquad (1)$$

This has been computed as a function of applied stress w and internal stress I for various values of n, which is the power of the stress to which the thermal creep rate is proportional. Values of f are given in table 1 and are shown graphically as a function of w/I in fig. 1. The abscissa of fig. 1 is given in this general form because the internal stress is a function of both the thermal creep parameters and the growth rate under neutron irradiation.

In general the index n is a variable, increasing

TABLE 1

The acceleration factor f as a function of n and w/I.

w/I ↓ \ n →	1.5	2.0	3.0	4.0	5.0	7.0	9.0	11.0
0.1	3.84	14.7	211	2960	41 040	7.62×10^{6}	1.38×10^{9}	2.42×10^{11}
0.15	3.14	9.83	94.4	889	8 340	6.86×10^{5}	5.38×10^{7}	4.41×10^{9}
0.25	2.43	5.93	34.6	198	1 122	35 400	1.14×10^{6}	3.41×10^{7}
0.50	1.75	3.06	9.40	28.6	87.4	820	8370	86 400
0.75	1.47	2.18	4.74	10.5	23.4	118	666	3,467
1.00	1.33	1.76	3.10	5.68	10.5	37.4	148	544
1.5	1.18	1.39	1.94	2.85	4.21	9.86	25.0	62.3
2.5	1.07	1.15	1.34	1.62	1.97	3.10	5.18	8.86
5.0	1.02	1.04	1.08	1.15	1.22	1.43	1.72	2.12
7.5	1.01	1.02	1.04	1.07	1.10	1.18	1.30	1.44
10.0	1.005	1.010	1.02	1.04	1.06	1.10	1.16	1.24

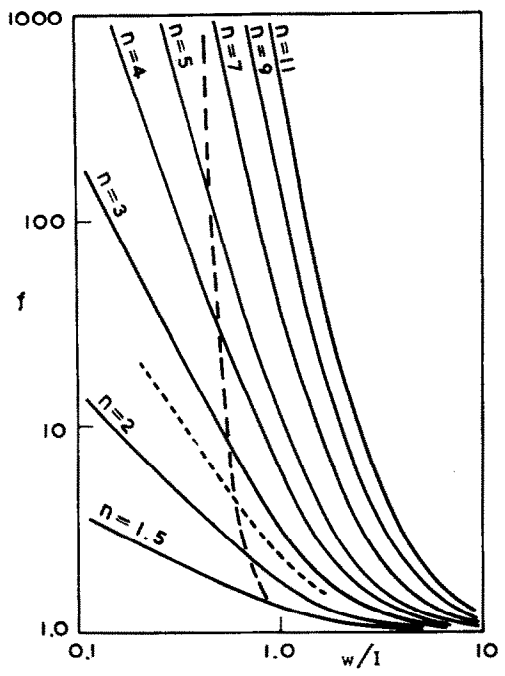

Fig. 1. The acceleration factor f as a function of the applied stress w and the internal stress I, for various values of n. The dotted and broken lines show respectively the variation of f with w when $I = 170$ MN m^{-2}, and the variation of f with I when $w = 100$ MN m^{-2}, assuming thermal creep according to eq. (2).

with increasing stress. Data by Wood [4]) on the thermal creep rate of cold worked Zircaloy-2 at 300 °C. 10 000 h and stresses below 200 MN m^{-2}* appear to be well expressed by

$$\dot{\varepsilon} = 1.1 \times 10^{-12} \sinh (w/w_0) \quad [\mathrm{s}^{-1}], \qquad (2)$$

where $w_0 = 60$ MN m^{-2}. At stresses above 200 MN m^{-2} the creep rate increases more rapidly with stress. When n is a variable it is important to choose the value appropriate to I rather than to w, because the uniform stress I exists in each grain together with a random tensor addition of w, i.e. the individual stresses are distributed about I and not about w. Thus if the internal stress remains constant and the applied stress varies, one simply follows a line of constant n, but if the applied stress remains constant and the internal stress varies one follows the steeper line shown in fig. 1. Experimentally this does not produce the dramatic changes of acceleration that might at first sight be expected, because, as will be seen in eq. (5), the internal stress now changes less rapidly with neutron flux than when n is constant. In fig. 1, along the curve for constant applied stress, the change in acceleration is commensurate with the change in neutron flux; each changes by three orders of magnitude as the internal stress changes by a factor of two, from 120 to 240 MN m^{-2}.

The specific value, $n = 1$, is of interest because it is relevant to diffusion creep, which occurs at significant rates in zirconium and its alloys [5]). It is important that in this case $f = 1$ for all values of w/I, i.e. there is no acceleration of the thermal creep rate for any value of the applied stress.

It is convenient to define as m the stress exponent of the irradiation creep rate $\dot{\varepsilon}_\phi$. Then

$$\dot{\varepsilon}_\phi = f\dot{\varepsilon}, \qquad (3)$$

whence

$$w^m \propto f w^n.$$

Now f is a dimensionless function of w/I, so that

$$f \propto (I/w)^{(n-m)}. \qquad (4)$$

Inspection of fig. 1 shows that as $w/I \to 0$, $m \to 1$ and $f \propto (I/w)^{(n-1)}$, while as $w/I \to \infty$, $m \to n$ and $f \to 1$.

The internal stress has the asymptotic value [2])

$$I = \left(\frac{2}{3}\right)^{1/2} \left[\frac{2\gamma}{A\sqrt{3}}\right]^{1/n} = \left(\frac{2}{3}\right)^{1/2} \left[\frac{2G\sigma\phi}{A\sqrt{3}}\right]^{1/n}, \qquad (5)$$

where γ is the irradiation growth rate, G is the growth coefficient, σ is the collision cross-section of the atoms in a neutron flux ϕ, and A is the constant relating the thermal strain rate $\dot{\varepsilon}$ to the applied stress; $A = \dot{\varepsilon} w^{-n}$. The time taken to approach this asymptotic value is $\sim I/(E\gamma)$, where E is Young's modulus. In both my experiments and those of Fidleris and Williams [6]) this time is calculated to be about

* 1 MN m^{-2} ≡ 10.2 kg/cm^2 ≡ 145 psi.

8000 h. The creep rates in the two sets of experiments are measured continuously up to 2700 h and 4500 h respectively. It is therefore an approximation to use this asymptotic value but I shall do so both for simplicity and because the form of Blackburn's eq. (6) indicates that after a rapid initial rise the internal stress changes only slowly.

Values of f for two stress levels, if measured in the same neutron flux, may be matched to one, and only one, of the family of lines in fig. 1. The ordinates of the two points are fixed, and the slope of the line joining them is fixed by the ratio of the two stresses. The absolute placing of the two points along the abscissa is determined by the internal stress. Thus matching to a line in fig. 1 gives the internal stress, from which eq. (5) gives the growth coefficient G.

Experiments have been performed in the Herald reactor on helical springs of annealed polycrystalline Zircaloy-2 wire. The temperature was 43 °C and the neutron flux (>1.0 MeV) was 0.13 n/m^2·sec (1.3×10^{13} neutrons/cm^2·sec). The spring diameter was 16 mm, the wire diameter was 1 mm and the grain size was 10 μm. Three loads were used, which gave shear stresses at the wire periphery of 24, 51 and 60 MN m^{-2}. Other experimental details will be published elsewhere. Fig. 2 shows irradiation creep curves for two specimens at the highest stress and the reproducibility is to be noted. The corresponding thermal creep curve for an unirradiated specimen is also shown. Comparison of the irradiation creep rate at 100 days with the thermal creep rate following irradiation gives the experimental values $f = 16 \pm 1$ at 51 MN m^{-2} and $f = 10 \pm 1$ at 60 MN m^{-2}. At the lowest stress the post-irradiation creep is too slow to give a reliable value. Matching these two values to fig. 1 gives $n \approx 10$ and $I \approx 250$ MN m^{-2}. This value of n differs greatly from the value $n = 1.6$ which best describes the laboratory data at these applied stresses. Its high value is appropriate to creep at the internal stress, which may be identified with the yield point, measured in shear, of 210 MN m^{-2}. Thus in these experimental conditions the asymptotic case is approached, and it is in fact equally possible to resolve the total irradiation creep into additive thermal and irradiation components.

Fidleris and Williams [6]) have measured irradiation creep in Zircaloy-2 at 300 °C. The last column of table 3 of their paper gives $n \approx 5$ for the two higher stresses. This differs from the value given by eq. (2), but I shall prefer it because it is specific to their material. Their

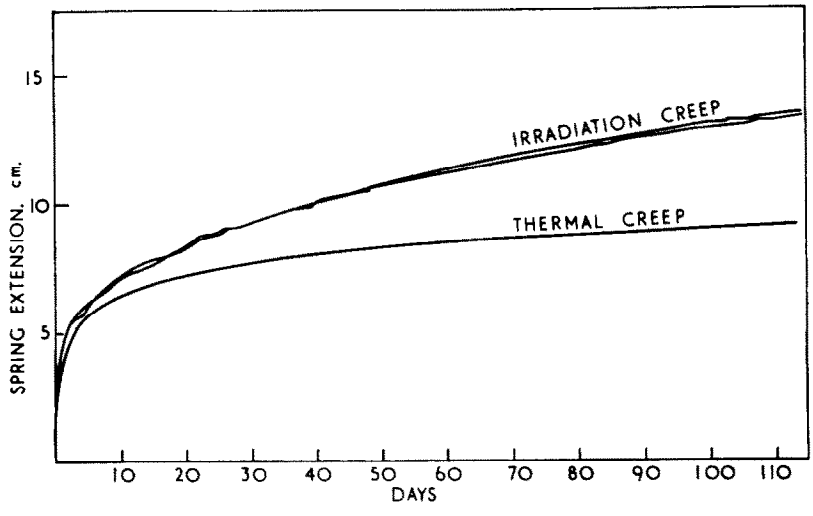

Fig. 2. Two irradiation creep curves for Zircaloy-2 in nominally identical conditions, together with the corresponding thermal creep curve.

irradiation tests R2, R4, R6 and R9, when taken together, give an average acceleration $f = 10.8$ for an average stress of 170 MN m^{-2}. This is shown as the solid point in fig. 3. Placing it on the theoretical curve for $n = 5$ gives $I = 173$ MN m^{-2}. The individual tests may now be related to the theoretical curve. Fig. 3 shows that there is good agreement.

In comparing irradiation and thermal creep data it seems essential to restrict comparison to rates measured in one specimen or in specimens of identical material. The variation of thermal creep parameters from one group of specimens to another is too great to permit freer comparison.

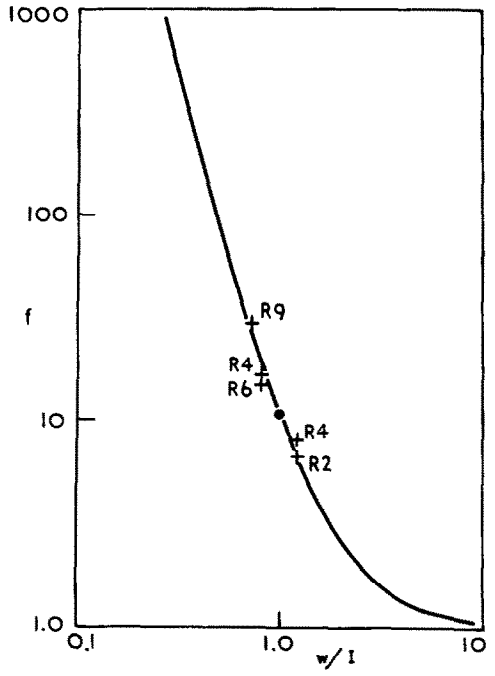

Fig. 3. Comparison of theory and experiment [6]) at 300 °C (section 2).

3. The single crystal growth coefficient

Rearranging eq. (5) and putting $\dot{\varepsilon} = Aw^n$ gives the irradiation growth rate,

$$\gamma = \tfrac{1}{2}\sqrt{3}\left(\frac{3}{2}\right)^{n/2}\left(\frac{I}{w}\right)^{n}\dot{\varepsilon}. \qquad (6)$$

The parameters n and $\dot{\varepsilon}$ are obtained from the thermal data and (I/w) is obtained by matching f to the thermal data. Putting $\sigma = 4$ barn and using the known fast fluxes gives, for my experiments

$$G_{70^\circ\mathrm{K}} = 2.3$$

and

$$G_{40^\circ\mathrm{C}} = 1.2,$$

and for those of Fidleris and Williams [6])

$$G_{300^\circ\mathrm{C}} = 1.5.$$

These values are directly comparable with the value $G \approx 1.2$ measured at ~ 100 °C from dimensional changes [7]).

4. Temperature dependence

The irradiation creep is

$$\dot{\varepsilon}_\phi = f\dot{\varepsilon} = fAw^n, \qquad (7)$$

whence, if n is independent of temperature,

$$\frac{1}{\dot{\varepsilon}_\phi}\frac{\partial\dot{\varepsilon}_\phi}{\partial T} = \frac{1}{f}\frac{\partial f}{\partial T} + \frac{1}{A}\frac{\partial A}{\partial T}.$$

The temperature dependence of f is obtained by substituting expression (4) into eq. (5) and differentiating,

$$\frac{1}{f}\frac{\partial f}{\partial T} = \frac{n-m}{n}\left(\frac{1}{\gamma}\frac{\partial\gamma}{\partial T} - \frac{1}{A}\frac{\partial A}{\partial T}\right).$$

Thus

$$\frac{1}{\dot{\varepsilon}_\phi}\frac{\partial\dot{\varepsilon}_\phi}{\partial T} = \frac{n-m}{n}\frac{1}{\gamma}\frac{\partial\gamma}{\partial T} + \frac{m}{n}\frac{1}{A}\frac{\partial A}{\partial T}. \qquad (8)$$

Let us ignore the first term on the right, on the ground that the rather uncertain values of the previous section show it to be small. Then, at small applied stresses or in very high fluxes, when w/I is small and $m \to 1$,

$$\frac{1}{\dot{\varepsilon}_\phi}\frac{\partial\dot{\varepsilon}_\phi}{\partial T} \approx \frac{1}{n}\frac{1}{\dot{\varepsilon}}\frac{\partial\dot{\varepsilon}}{\partial T}, \qquad (9)$$

while at the other extreme, of high applied stresses or small neutron fluxes,

$$\frac{1}{\dot{\varepsilon}_\phi}\frac{\partial\dot{\varepsilon}_\phi}{\partial T} \approx \frac{1}{\dot{\varepsilon}}\frac{\partial\dot{\varepsilon}}{\partial T}. \qquad (10)$$

The temperature dependence is thus a variable quantity. It involves the parameters

A, γ, m and n and their derivatives. It is thus clear that a measured value does not have a simple physical interpretation as the activation energy of a single process.

The temperature dependences measured [8, 9] at 207 MN m^{-2} and 97 MN m^{-2} differ in the expected sense and by the magnitude expected from eq. (8). The gradient of the line in fig. 3 shows that at the respective stresses $m \approx 2.5$ and $m \approx 1.7$. The ratio of these two values is that of the temperature sensitivities, 4 : 3. Also the temperature dependence of irradiation creep is about half that of the thermal creep rate, which is consistent with the m/n ratio for this data.

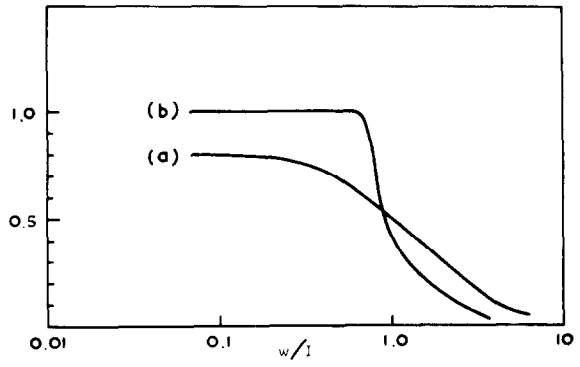

Fig. 4. The index of the flux dependence $(n-m)/n$, as a function of w/I. For curve (a) $n=5$. For curve (b) $w=100$ MN m^{-2} and n varies according to eq. (2). For other values of w this second curve is shifted horizontally so that each point on it relates to the same value of I as shown here.

5. Flux dependence

Differentiation of eqs. (4), (5) and (7) gives

$$\frac{1}{\dot{\varepsilon}_\phi}\frac{\partial \dot{\varepsilon}_\phi}{\partial \phi} = \frac{1}{f}\frac{\partial f}{\partial \phi} = \frac{n-m}{n}\frac{1}{\gamma}\frac{\partial \gamma}{\partial \phi} = \frac{n-m}{n}\frac{1}{\phi},$$

and integrating,

$$\dot{\varepsilon}_\phi \propto \phi^{(n-m)/n}. \qquad (11)$$

The index $(n\text{-}m)/n$ is shown as a function of w/I in fig. 4 for two cases, both of which refer to data at 300 °C. Curve (a) is that calculated by putting $n=5$ and taking values of m given by eq. (1). At around $w/I \sim 1$ the curve is appropriate to the data of fig. 3 and predicts a flux variation of $\phi^{\frac{1}{2}}$ for the conditions of Fidleris and Williams' experiments. Curve (b) is for a constant applied stress of 100 MN m^{-2} and for a changing internal stress which produces creep within each grain according to eq. (2). For other values of applied stress the curve has to be displaced horizontally so that w/I is changed but I remains unchanged. The difference between the two curves demonstrates the sensitivity to the thermal creep parameters. Both curves show the insensitivity to neutron flux at high applied stresses and curve (b) shows, in its plateau, where the asymptotic condition of simple yielding creep is reached.

There is very little data on flux dependence. That by Ross-Ross and Hunt [9], appears to me to show an index of 0.75. The value calculated from the thermal data is 0.7. Both values are uncertain, the first because of the limited flux range and the second because of the uncertainty of the thermal data. I have in hand measurements at 40 °C. Ideally such measurements require running a reactor at several power levels to avoid the changes in flux spectrum which occur on moving from one reactor facility to another. Subsequently, transfer from, say, a materials testing reactor to a fast reactor could allow identification of the part of the spectrum responsible for irradiation growth.

The most abrupt variation of neutron flux is that occurring when the reactor is switched on or off. In making experimental comparisons the second is to be preferred because transient creep is avoided. As indicated in section 2 the rapidity of transient creep lessens the effect of irradiation. Suppose that the neutron flux ceases instantaneously and that the stress within each grain relaxes. During relaxation there is, overall, conservation of the sum of the

elastic and creep strains within each grain, so that

$$\frac{1}{E}\frac{\partial I}{\partial T} + \dot{\varepsilon}_I = 0. \qquad (12)$$

While the internal stress persists, the polycrystal accumulates a creep strain in excess of the thermal creep strain, and in a time interval Δt this is

$$\Delta\varepsilon_x = \dot{\varepsilon}(f-1)\Delta t.$$

Substituting from eq. (12), rearranging and integrating,

$$\varepsilon_x = \varepsilon_e \int_I^0 (f-1)(w/I)^n(-\mathrm{d}I/w), \qquad (13)$$

where ε_e is the elastic strain created by the applied stress w. In this integral the values of f and n are those appropriate to the internal stress.

In the asymptotic condition of yielding creep n is large for all appreciable values of $(f-1)$ and the integral remains small. This is no longer true when there is thermal creep. Numerical integration for the constant values $n = 5$, 4, 3 and 2, and allowing the internal stress to relax from the initial value $I = w$, gives $\varepsilon_x \approx 1000\,\varepsilon_e$, $100\,\varepsilon_e$, $10\,\varepsilon_e$ and ε_e respectively. If n varies according to eq. (2) then $\varepsilon_x \approx \varepsilon_e$. This strong variation of ε_x makes it necessary to know the thermal creep parameters with some accuracy, and they must be those of the irradiated material rather than the unirradiated material. Most existing data applies to the latter.

In my experiments at 78 °K and 40 °C the irradiation creep ceases with the abruptness expected from the asymptotic condition. In those of Fidleris and Williams [6]) there is some variability; their fig. 5 shows two reactor shutdowns during which the creep rate did not decrease and one during which it did. Of the section (e)–(f) of their fig. 4 they remark that it "suggests a gradual decrease of creep rate once the neutron flux is removed".

6. Time dependence

The thermal creep rate is in general not constant with time, and at the comparatively low stresses which exist in structural components it is often an inverse function of time. Say

$$A \propto t^{-k}. \qquad (14)$$

Eq. (5) shows that as A decreases the internal stress I, and therefore the acceleration f, increases. Thus a reactor experiment will show a less marked decrease of creep rate with time than the corresponding laboratory experiment. However, only in the asymptotic condition of simple yielding creep is the irradiation creep rate constant. Following section 4,

$$\frac{1}{\dot{\varepsilon}_\phi}\frac{\partial\theta_\phi}{\partial A} = \frac{1}{A} + \frac{1}{f}\frac{\partial f}{\partial A},$$

which from eqs. (4) and (5) results in $m/(nA)$.

Substituting from eq. (14) and integrating twice,

$$\varepsilon_\phi \propto t^{1-km/n}, \text{ if } n \neq km. \qquad (15)$$

In designing reactor pressure tubes it is necessary to extrapolate from the flux of the test reactor to that of the power reactor, and from experiments of 2000 h duration to reactor lifetimes of 200 000 h. The second of these extrapolations is the more hazardous. There is no assurance that k does not change with time; for example the material used in Fidleris and Williams' experiments received 20% prior cold work and its structure cannot be assumed to be stable. Again, thermal creep may well consist of a dislocation glide component and a diffusion creep component, and k will approach zero as the second becomes predominant. From his high temperature measurements Bernstein [10]) estimates that at 138 MN m^{-2} and 300 °C, material of grain size 12–15 μm will creep at 3×10^{-12}/s by grain boundary diffusion. This rate is not far short of the value measured by Fidleris and Williams, though it differs in having $k = 0$ rather than $k = 1$.

Bearing in mind this uncertainty, consider the example [6]) $k = 1$ and $n = 5$. For a design stress of 110 MN m^{-2} and the flux of the CANDU reactor one expects $m = 1.3$. Extrapolation from time t_1 to a time t_2 by eq. (15) shows that the

final creep strain is less than that given by a linear extrapolation. The two differ by the factor

$$\frac{n}{n - km}\left(\frac{t_1}{t_2}\right)^{km/n} . \qquad (16)$$

If also, as suggested in section 5, the creep rate varies as $\phi^{0.7}$ rather than $\phi^{1.0}$ the two extrapolations together give a creep strain at 200 000 h which is less by a factor of five than that of linear extrapolations from 2000 h.

7. Discussion

It is worth emphasising that this form of creep is a direct consequence of growth; no physical assumption is required other than that the specimen is a continuous, ductile polycrystal. The one direct measurement of growth which has been made [7]) gives a value similar to those deduced here.

Attention has been drawn to the strong dependence of the calculated acceleration on the thermal creep parameters, yet correspondence with authors of other papers in this issue, indicates considerable uncertainty in the pre-irradiation values and greater uncertainty in the post-irradiation values. Equally, one may choose from a wide range of irradiation creep rates. These uncertainties are a handicap in testing the hypothesis made here, and so one returns to the fundamental question, does growth exist in zirconium alloys?

8. Diffusion controlled creep

Neutron irradiation produces defects which may diffuse through the crystal lattice and it is of interest to consider its effect on those creep processes which are controlled by diffusion. These include Nabarro-Herring creep [11]), the climb controlled creep of edge dislocations [12]) and creep by the motion of jogged screw dislocations [13]). These have the common feature that the creep rate is proportional to the self-diffusion coefficient. This originates in the equations as the product of the mobility of a defect, such as a vacancy, and the equilibrium concentration of the defect, at whatever sources and sinks are under consideration, and in the absence of stress. These sources may be surfaces, grain boundaries, edge dislocations or jogs in screws.

In the absence of neutron irradiation it does not matter if one discards the restriction that the defect concentration is that at the sources, and not at some remote point in the lattice. In the presence of neutron irradiation it is essential to retain it. The homogeneous and adiabatic formation of vacancies and interstitials by the collision of neutrons with lattice atoms produces concentration gradients of these defects into all the sinks. Thus, away from the sinks, up the concentration gradients, the concentration and hence the self-diffusion is increased, e.g. Dienes and Damask [14]), but the diffusion coefficient which appears in the creep equation remains unaltered. Neutron irradiation has no effect on the equilibrium defect concentrations at the sources, since these are determined by the energy required to form a defect by thermal fluctuations, and its only effect on the mobility of a defect is to decrease it in very high fluxes by providing a defect of opposite sign which annihilates the defect before it has jumped.

Alternatively one may say that so long as there is no mutual recombination of defects, each pursues a random walk, and the defects migrate without preference to all available sinks. In a uniform stress field, in the absence of stress gradients, this walk is unaffected by stress. Nor is the homogeneity of the creation of defects affected by stress. Thus the defects created by neutron irradiation do not affect the creep rate.

This conclusion has already been stated by Nabarro [11]) and by Mosedale [15]). The opposite conclusion has been stated by Schoeck [16]), Brinkman and Wiedersich [17]), who, together with several textbooks, have followed Schoeck, by Fidleris and Williams [6]), and Holmes [18]). It is of interest to note that Conrad and Schoeck looked for an increase in creep rate due to an increase in vacancy concentration but failed to find it [19]). Since a proposal similar to Schoeck's

is being made by Piercy [20]), I shall expand the point of view expressed above, and consider three cases, diffusion with no recombination, recombination with fixed sinks distributed throughout the material, and mutual recombination of vacancies and interstitials. It will be seen that neutron irradiation affects only the last of these.

Consider diffusion in one dimension. (The physics does not change in other geometries). The diffusion equation is

$$\partial c/\partial t = (a^2\nu/\alpha)(\partial^2 c/\partial x^2),$$

where a is the interatomic spacing, ν is the jump frequency and α is a coordination number. In this equation $\partial c/\partial t$ is the flux of defects into a region, so that in steady conditions under neutron irradiation

$$\partial c/\partial t = -\phi\sigma M,$$

in the absence of recombination (M is the average number of atoms displaced in a neutron collision).

$$\partial c/\partial t = -(\phi\sigma M - B_1 a^2 \nu c/\alpha),$$

for recombination with fixed sinks, and

$$\partial c/\partial t = -(\phi\sigma M - B_2 a^2 \nu c^2/\alpha),$$

for mutual recombination. In the last case the two diffusion equations for vacancies V and interstitials I are respectively

$$\phi\sigma M - B_2 a^2 \nu c_V c_I/\alpha = (-a\nu_V/\alpha)(\partial^2 c_V/\partial x^2),$$

and

$$\phi\sigma M - B_2 a^2 \nu c_V c_I/\alpha = (-a\nu_I/\alpha)(\partial^2 c_I/\partial x^2).$$

These differ only in the frequency constants ν_I and ν_V so that c_I and c_V differ only in scale. Put $D = \alpha\phi\sigma M/(a^2\nu)$ and $\partial c/\partial x = p$.

In the absence of recombination

$$D = -p\partial p/\partial c,$$

and

$$Dc = -\tfrac{1}{2}p^2 + \text{constant}.$$

At the point in the lattice, between two sinks, at which $p = 0$, put $x = x_1$ and $c = c_1$. Then

$$p = \{2D(c_1 - c)\}^{\frac{1}{2}}$$

and integrating again,

$$c_1 - c = \tfrac{1}{2}D(x_1 - x)^2.$$

At a sink put $x = x_0$, and $c = c_0$ in the absence of stress. Now let a stress be applied to the material, changing the equilibrium concentration c_0 by Δc_0. Then

$$\begin{aligned}\Delta p_0 &= -D\Delta c_0/p_0,\\ &= -D\Delta c_0\{2D(c_1 - c_0)\}^{-\frac{1}{2}},\\ &= -\Delta c_0/(x_1 - x_0). \qquad (17)\end{aligned}$$

Since this equation does not contain D the change of concentration gradient into the sink is independent of the neutron flux.

When there is recombination at fixed sinks dispersed throughout the material,

$$D - B_1 c = -p\partial p/\partial c.$$

Assume that the distance between sinks is large enough for the saturation concentration to be reached between them (otherwise one approaches the previous case) and put $p \to 0$, $c \to c_1$ as $x \to \infty$. Then $c_1 = D/B_1$.

Integrating,

$$Dc - \tfrac{1}{2}B_1 c^2 = -\tfrac{1}{2}p^2 + \text{constant}.$$

Since $p = 0$ when $c = c_1$,

$$p = (c_1 - c)(D/c_1)^{\frac{1}{2}}.$$

At the sink

$$p_0 = (c_1 - c_0)(D/c_1)^{\frac{1}{2}},$$

and, as before,

$$\begin{aligned}\Delta p_0 &= -(D - B_1 c_0)\Delta c_0/p_0,\\ &= -\sqrt{B_1}\Delta c_0. \qquad (18)\end{aligned}$$

Again the change in concentration gradient, and hence of a diffusion controlled creep rate is independent of neutron flux.

For mutual recombination between vacancies and interstitials

$$D - B_2 c^2 = -p\partial p/\partial c.$$

Again it is assumed that the distance between sinks is large enough for saturation to be reached

between them; $p \to 0$ and $c \to c_1$ as $x \to \infty$. Then $c_1 = (D/B_2)^{\frac{1}{2}}$ and

$$p = \{\tfrac{1}{3}D(4c_1 - 6c + 2c^3/c_1^2)\}^{\frac{1}{2}}.$$

If now $c_1 \gg c_0$,

$$p_0 = (\tfrac{16}{9}D^3/B_2)^{\frac{1}{4}},$$

and

$$\Delta p_0 = -D(1 - c_0^2/c_1^2)\Delta c_0/p_0$$

$$= -\tfrac{3}{4}(B_2 D)^{\frac{1}{4}}\Delta c_0. \qquad (19)$$

In this case the concentration gradient does depend on the neutron flux. Since the creep rate of the three diffusional processes named at the outset is linearly proportional to the flow of defects down this gradient, one has finally, for fluxes high enough for saturation to be reached,

$$\dot{\varepsilon}_\phi \propto \phi^{\frac{1}{4}}. \qquad (20)$$

It should be comparatively easy to distinguish between this flux dependence and the linear flux dependence of simple yielding creep.

Piercy's result [20]) differs in several ways from that given here. It is based on Rosenthal's eq. (43), which is a solution of the three dimensional diffusion equation for a moving point source (of defects in our case). As such it contains no term for the homogeneous creation of defects throughout the lattice. It therefore refers to the case of zero neutron flux. The concentration of defects at points remote from the dislocations which control the creep rate is assumed to be determined not by mutual recombination but by absorption at fixed sinks. The result should therefore be compared with eq. (18) rather than eqs. (19) and (20). It should also be applicable to metals other than zirconium, and in the non-fissile cubic metals its prediction that no strain recovery occurs during post-irradiation thermal annealing is grossly at variance with experiment.

Fidleris and Williams [6]) have already noted that, for Zircaloy, Barrett and Nix's equation [13]) gives a thermal creep rate which is between two and three orders of magnitude too large. Thus, in addition to its independence of neutron irradiation, there is some doubt of the relevance of this mechanism to the thermal creep behaviour.

9. Conclusion

When thermal creep is accelerated by an irradiation growth stress the applied stress, temperature, time and neutron flux become interdependent functions of the creep rate. The irradiation creep data on Zircaloy-2 are consistent with an irradiation growth stress, but it is desirable to extend their range and accuracy.

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