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"The Use of State Variables in the
Description of Irradiation Creep and
Deformation of Metals"

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ABSTRACT

Our understanding and our mode of description of the effects of irradiation on metal creep and deformation are not yet really satisfactory. This is due in part to the limitations on experimentation in radiation environment. Because of such limitations, theoretical considerations must play a strong role in the interpretation and in the representation of the phenomena. Reliable representations are necessary if we are to extrapolate the measured behavior beyond the experimental ranges of stress, temperature, flux and time duration. Virtually all of the theoretical considerations currently employed are based on micro-mechanical models for the deformation behavior. While the fundamental mechanisms proposed may in fact be correctly identified, it is not at all clear that the quantitative conclusions from them are accurate. After all, the detailed micro-mechanical description of deformation even in the absence of irradiation is still moot.

The recent theoretical and experimental development of a plastic equation of state for metal deformation has led to the identification of some of the principal micro-mechanisms in phenomenological terms. The role of the individual mechanisms can be related to the state variables of the description, and those variables are directly accessible measurable quantities.

In this paper we shall explore how irradiation might affect this description. It will be shown that the radiation flux and the radiation fluence are expected to affect different components of the equation of state. The resultant description makes considerable use of the information developed in radiation-free environment.

Our ability to predict the deformation response of metals under neutron irradiation conditions is still far from satisfactory. This is largely due to the limitations on experimentation in radiation environment. Because of the limitations on the range of test variables it has been necessary to find ways to extrapolate beyond the experimental ranges of stress, temperature, flux, and exposure time. This is clearly a problem for the application of theoretical understanding. Most of the theoretical considerations currently employed are based on micro-mechanical models for the deformation behavior. Nichols[1] has recently reviewed a number of the theoretical models in relationship to the experimental work. None of the models offers a sufficiently complete description for deformation behavior. While many of the fundamental mechanisms may in fact be correctly identified, it is not clear that the resultant quantitative conclusions are fully reliable. This is not much different from the situation for purely thermal creep behavior.

The present paper proposes that considerably greater use can be made of thermal environment test data, for both irradiated and unirradiated material, in order to characterize the behavior under irradiation. Since considerably more data can be collected over broader ranges of test variables out-of-reactor, it is clear that any success of such utilization would be highly advantageous.

The basis for the proposal is a phenomenological theory that has so far had considerable success in describing deformation in thermal environment. The theory has been subjected to experimental test over broad ranges of stress and temperature. Materials that have been tested so far include high-purity Al, 1100 Al, α -Fe, Nb, Ni, 316 Stainless Steel, 304 Stainless Steel, and Zircalloy-4.

The central idea of the phenomenological theory is the deduction from experiment of the existence of state variables that can specify the current deformation state of the material. The theory was first described by Hart[2]

in an analysis of the deformation process that stated the experimental tests that would answer the questions posed in the theory. A sequence of experimental investigations resulted in a firm verification of the state variable formulation. A discussion and review of that work was published by Hart et al[3]. The formulation to that point was concerned mainly with the non-transient deformation behavior. The rôle of recoverable (anelastic) strain was recognized and discussed from the beginning of the work, but the detailed relationship between transient behavior and recoverable strain was only recently confirmed.

Now a rather complete model for metal deformation is available that adequately describes transient response to rapid load change as well as the more steady non-transient flow. This model has been described as full three-dimensional constitutive relations by Hart[4].

In the following we shall first present a brief description of these constitutive relations and the state variables they depend upon. We shall then state the proposed extension to radiation environment, and will describe the application to some of the available experimental data. Finally we shall suggest experimental procedures that would most efficiently implement the application.

The Deformation Model

We shall limit our description here to uniaxial deformation for simplicity. The model represents a synthesis of three deformation mechanisms whose mutual relationship is most conveniently shown in the rheological diagram of Fig. 1. The three mechanisms are represented there by the three elements labeled 1, 2, and 3. In our discussion they will be referred to as the α -element (or stored strain element), $\bar{\alpha}$ -element (or barrier penetration element), and $\bar{c}$ -element (or glide friction element), respectively. The elements and their relationships can be taken to represent some familiar micro-mechanical processes according to the following considerations.

The dislocation flux that is responsible for the non-elastic deformation of the metal grain matrix must traverse both large regions of relatively

"good" crystal and also strong barriers to dislocation motion due to dislocation tangles and other concentrated defects. In the good regions dislocation motion is limited by glide friction. Passage through or around strong barriers can occur either mechanically at sufficiently high stress or by thermal activation at lower stress. The barrier can be second phase inclusions, disordered regions, or dislocation tangles generated by plastic straining. The dislocation tangle barriers represent the cumulative effect of strain hardening.

Since the dislocation flux in the good regions will generally exceed the rate of passage through barriers, glide dislocations will tend to pile-up at the barriers. The pile-ups raise the driving force for barrier penetration and generate back stresses (internal stresses) that slow down the dislocation flux through the good regions.

In our model the $\dot{\alpha}$ -element represents the barrier processes, the a -element characterizes the pile-ups as a stored strain, and the $\dot{\epsilon}$ -element represents the glide friction. The resultant mechanical and kinematical relations of the diagram account for the mechanical interactions and the dislocation flux balances of the fundamental processes. These relations will now be stated quantitatively for the thermal deformation régime.

The applied uniaxial stress σ is the sum of the stresses σ_a and σ_f that are operative in the two branches. The observed total uniaxial strain rate $\dot{\epsilon}$ is the same as the strain rate exhibited in the lower branch and equals the sum of the strain rate component $\dot{\alpha}$ and the time rate of change of the stored strain a . Thus there are two constraint equations among the auxiliary variables σ_a , σ_f , $\dot{\alpha}$, and a . These are

$$\sigma = \sigma_a + \sigma_f, \quad (1)$$

$$\dot{\epsilon} = \dot{\alpha} + da/dt. \quad (2)$$

The constitutive equations for the component elements, as deduced from experimental measurement, are as follows:

The α -element is a linear elastic element with a modulus M such that

$$\sigma_a = M \alpha \quad (3)$$

The $\dot{\alpha}$ -element is the plastic-creep element satisfying the relations

$$\ln(\sigma^*/\sigma_a) = (\dot{\epsilon}^*/\dot{\alpha})^\lambda, \quad (4)$$

$$\dot{\epsilon}^* = (\sigma^*/G)^m \cdot f \cdot e^{-Q/RT}, \quad (5)$$

$$d \ln \sigma^*/dt = \Gamma(\sigma_a, \sigma^*) \dot{\alpha} - \mathcal{R}(\sigma^*, T). \quad (6)$$

In these equations T is the absolute temperature, R is the gas constant, G is the modulus of rigidity at temperature T , and λ , f , m , and Q are constants for each material. The functions Γ and $\mathcal{R}$ are to be determined experimentally.

Now the $\dot{\alpha}$ -element depends prominently on the scalar state variable σ^* which we have called the hardness. The strain hardening then appears as an incremental increase of the value of σ^* with strain increments $d\alpha$ and purely thermal recovery is described by $\mathcal{R}$. In our current discussion we shall not be concerned much with $\mathcal{R}$. We shall describe the dependence of $\dot{\alpha}$ on σ_a below graphically.

Finally, the $\dot{\epsilon}$ -element can be represented over fairly large ranges of strain rate $\dot{\epsilon}$ as a power law function of σ_f . We write this for the magnitude of $\dot{\epsilon}$ and σ_f and note that $\dot{\epsilon}$ must have the same sign as σ_f . Thus

$$\dot{\epsilon} = \dot{\alpha}^*(T) (\sigma_f/M)^M. \quad (7)$$

The exponent M is a constant and $\dot{\alpha}^*(T)$ must be measured experimentally.

Some typical values for the constants are: $M=8$, $m=4.5$, $\lambda=0.15$, M is of the order of G , and Q is commonly the same as the activation for self-diffusion of the atomic species.

Now we may make some simplification in this representation in the

consideration of creep at moderately elevated temperatures in the absence of stress transients. Under those circumstances da/dt is small compared to $\dot{\epsilon}$ and to a good approximation $\dot{\alpha} = \dot{\epsilon}$. Then σ_a and σ_f can both be plotted against $\dot{\epsilon}$. This is shown schematically for moderately high homologous temperature in Fig. 2.

Our current interest is in relatively strong materials. For such materials σ^* can be expected to be rather high because of second phase hardening and possibly because of deliberate high strain hardening levels. Under those circumstances σ_f is small compared to σ_a and thus the operative flow stress σ is determined almost entirely by σ_a . This leads us to consider the $\dot{\alpha}$ -element as the flow controlling mechanism, and we can simplify our constitutive equations to the form

$$\ln(\sigma^*/\sigma) = (\dot{\epsilon}^*/\dot{\epsilon})^\lambda, \quad (8)$$

$$d \ln \sigma^*/d\epsilon = F(\sigma, \sigma^*), \quad (9)$$

where we are ignoring the recovery term P_r , and Eq. 5 stands as before.

It is this form of the deformation model that we shall use to discuss the proposed extension to radiation environment behavior. The principal features of this model are the explicit analytical form of the stress-strain rate relationship (Eq. 8) and the dependence on the state variable σ^* . The auxiliary variable $\dot{\epsilon}^*$ is dependent on σ^* and T in the thermal environment case and is given by Eq. 5. We shall now examine how these relationships might be altered by the effects of radiation.

The Proposed Application to Radiation Environment

It is now proposed that the stress dependence of the strain rate under radiation conditions can also be represented by Eq. 8 and that the appropriate values of σ^* and $\dot{\epsilon}^*$ can be determined as a function of fluence and current flux. Let us consider these two functional dependences separately.

The Fluence Effect

The fluence experienced by a specimen results in a modification of the micro-structure of the material. If an irradiated specimen is tested out-of-reactor, it should behave like a material with altered micro-structure in much the same way as if such alteration had occurred by some other metallurgical processing or aging. This results merely in a change of σ^* and $\dot{\epsilon}^*$. Such effects have already been noted. Woodford[5] found that a low alloy steel when tested near one-third of its melting temperature could be described by Eq. 8 with the same value $\lambda = 0.15$ that was appropriate for high-purity Aluminum. The only difference was the values of σ^* and $\dot{\epsilon}^*$. Aging effects for that alloy resulted in smaller additional change of the values of σ^* and $\dot{\epsilon}^*$. Further strain hardening can be expected to proceed substantially as in the thermal case.

The important point here is that the form of the σ - $\dot{\epsilon}$ curve is insensitive to the details of micro-structure. Even the value of λ is unchanged. The effect of micro-structural change is expressed entirely in terms of displacement of the point $(\sigma^*, \dot{\epsilon}^*)$ in the σ - $\dot{\epsilon}$ space. The same behavior has been found for the effects of radiation as we shall note below.

It is anticipated that there will also be a fluence effect on the friction constant $\dot{\alpha}^*$ in the complete model, but we shall not discuss this further here.

The Flux Effect

The most direct effect of radiation flux is to establish a supersaturation of point defects over the thermal equilibrium concentration. Such supersaturation can increase the rate of those processes that depend on dislocation climb. In his review article Nichols[1] discusses several of the proposals concerning the effect on radiation creep. He considers both those mechanisms that depend on a bias in the defect absorption at dislocations and those that are independent of such bias. In the present treatment we are not attempting to make a detailed micro-mechanical model. We believe, nevertheless, that the strain results from glide, and that the strong barriers or obstacles are passed by diffusive processes. We propose then, that the effect of radiation flux is to cause a positive displacement of $\ln \dot{\epsilon}^*$ in the same way as might be brought about by effective temperature increase. This effective temperature increase can be estimated, but we shall propose that it be measured as a function of

flux and energy. We also expect a flux effect on $\dot{\epsilon}^*$. The effect is probably to increase $\dot{\epsilon}^*$, i.e., to reduce the dislocation friction.

The Proposed Representation

The combined effects of flux and fluence in the non-transient creep range are expected to be represented entirely in terms of the pair of state variables σ^* and $\dot{\epsilon}^*$. These two variables can be considered conveniently as the point $(\ln \sigma^*, \ln \dot{\epsilon}^*)$ in a plot of $\ln \sigma$ vs. $\ln \dot{\epsilon}$. Such a plot is shown at a single temperature in Fig. 3. The three labeled points show schematically the effects we have discussed above. The point labeled 1 represents the state point for unirradiated material at some level of hardness according to prior strain history. The solid line through that point is the locus of state points at other levels of hardness. Point 2 represents the state point for irradiated material, but for deformation behavior in thermal environment. The shift from 2 to 3 represents the displacement to higher $\dot{\epsilon}^*$ value due to radiation flux level. For each of these cases the expected experimental stress-strain rate relationship is given by Eq. 8 with the values of σ^* and $\dot{\epsilon}^*$ determined by the appropriate state point.

Now what remains to be determined is the dependence of the state point displacements on the radiation fluence and flux. That relationship must be determined experimentally.

In the absence of systematic experimental data we cannot state too closely the form of these relations. It is probably, however, that the displacement of the state point due to irradiation hardening can be expressed incrementally in much the same way as strain hardening has been, and that the rate of change of σ^* at high fluence levels becomes progressively much smaller. Of course, this displacement is two-dimensional. The displacement of $\ln \dot{\epsilon}^*$ due to flux should be much like the thermal dependence on temperature, except for possible transients.

The Experimental Routines

In the case of thermal creep behavior the most useful test for measuring the state relations has been the load relaxation test. The same test can certainly be used to investigate fluence effects by out-of-reactor testing.

It would be desirable to utilize load relaxation testing more generally for in-reactor measurement as well.

Application to Available Data

304 Stainless Steel

Post irradiation load relaxation tests have been made by Yamada[6] on 304 Stainless Steel following an exposure of 1.82×10^{22} neutrons/cm² of energy greater than 0.1 MeV. Those measurements at 25°C and 300°C confirmed that the $\dot{\epsilon}$ -element curve was of the same shape as for the unirradiated material and determination of post-irradiation values of σ^* and $\dot{\epsilon}^*$ were made. On the basis of that testing it is possible to predict a thermal σ - $\dot{\epsilon}$ curve at 400°C using a value of Q of 65 Kcal/mol in Eq. 5. That curve is shown in Fig. 4 together with a curve displaced in strain rate to what would correspond to thermal behavior at 700°C. The calculations of Harkness and Li[7] concerning void formation can be used to show that this is a reasonable effective temperature shift at this dose level for the flux conditions characteristic of the creep rate measurements of Slimm *et al*[8]. Those creep rate results are plotted on the displaced curve in Fig. 4.

The consistency of the representation is encouraging. However, our plot implies some conflict with the claim of Newtonian behavior of ref. 8. Our plot would imply a stress exponent closer to three. Clearly, the experimental range of the data is not sufficient to settle this question.

Zirconium Alloys

Load relaxation tests of unirradiated Zirconium alloys [9,10,11] have shown a basic equation of state behavior similar to the other materials tested. There are no such results available for the post-irradiated condition. Post-irradiation values of σ^* can be estimated from post-irradiation tensile test data, but no measurement of $\dot{\epsilon}^*$ is possible.

We show in Fig. 5 plots of two of the radiation creep tests available in the literature, those by Ibrahim[12] and by Gilbert and Mastel[13]. The solid curve is from Eq. 8 with $\lambda = 0.15$ and $\log \sigma^* = 5.2$.

Discussion

We have proposed that a state variable theory that has been successful in describing thermal environment deformation behavior can be extended to the description of radiation environment creep behavior. We have discussed the detailed application only for that part of the model that is concerned with non-transient, elevated temperature creep. That application is consistent with the data so far available. The main strength of the theory is that it is based entirely on the relationships among measurable quantities. The theory is quite explicit in its predictions outside the range of the measurements needed to specify its parameters. It can therefore be tested crucially by those predictions.

As we have noted above, load relaxation testing, both in-and-out-of reactor, is the most efficient way of making the crucial tests. Such tests can directly measure the constant hardness stress-strain rate characteristics.

The load transient behavior that is fully described by the complete model depends principally on the parameters $\dot{a}^*$ and M . Nir et al [14,15] have shown how these parameters are easily measured by load change tests in the anelastic stress range. Such tests can measure the dependence of $\dot{a}^*$ on radiation history and can also determine what effect flux has on that parameter.

The details of carrying out these tests at the required level of precision are to be found in the references already noted.

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Figure Captions

- Fig. 1 A diagram representing the mechanical and kinematical relationships among the deformation mechanisms for inelastic behavior of metals. Element 1 is Hookean; element 2 is the $\dot{\alpha}$ -element; element 3 is a non-linear viscous element.
- Fig. 2 A graph of stress vs. strain rate for transient free behavior at high temperature. The applied stress σ is the sum of σ_a and σ_f . When σ_f is small, σ is nearly the same as σ_a . The hardness σ^* is an asymptote for the σ_a curve at large $\dot{\epsilon}$.
- Fig. 3 Points representing several values of the state variable pair $(\sigma^*, \dot{\epsilon}^*)$. Point 1 is for unirradiated material; point 2 is a result of fluence for out-of-reactor behavior; point 3 includes the shift of strain rate due to flux. Point 1 simply displaces along the solid line with increased straining.
- Fig. 4 Radiation creep rates, according to Slimm et al[8], plotted on a constant hardness curve for 304 Stainless Steel.
- Fig. 5 Radiation creep rates of Zr-2.5 Nb plotted on a constant hardness curve with $\lambda = 0.15$, $\log \sigma^* = 5.2$. The data points are from Ibrahim[12] (2% cold worked) and Gilbert and Mastel[13].

FIG. 2

FIG. 3