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# **Formulation of Constitutive Laws for Deformation During Irradiation**

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FORMULATION OF CONSTITUTIVE LAWS  
FOR DEFORMATION DURING IRRADIATION

by

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## Abstract

Constitutive laws for radiation-induced deformation are derived based on a phenomenological approach which involves only simple principles of continuum mechanics and avoids detailed mechanistic assumptions. This approach is applied to materials which are initially in an isotropic condition that serves as a reference state. As specific examples of this approach we consider in detail three forms of constitutive laws. In the first case it is assumed that the deformation rate depends on the current stress as the only tensor variable. For the second case we postulate a dependence on the current stress tensor and the strain tensor as produced during the previous radiation-induced deformation. Finally, the third case deals with a dependence on stress and strain as accumulated during cold-working. In both the latter cases, the material is rendered anisotropic with respect to the reference state. It is shown for the last two cases that the phenomenological approach provides a proper formulation and a clear distinction of such phenomena as stress-affected swelling, irradiation creep, radiation-induced anisotropic growth, and creep-swelling interaction. It also supplies the superposition rules for these phenomena when the material is subject to tri-axial stresses which change and redistribute with time.

The constitutive laws as obtained from the continuum approach are not completely suitable for modelling structural materials. Since they are polycrystalline in nature, the constitutive properties may vary from grain to grain, and it becomes necessary to derive a macroscopic constitutive

law for the aggregate. This is accomplished by considering an aggregate of many viscoelastic elements linked in parallel. The resulting constitutive law for the aggregate depends on the strain history. Upon sudden load changes, the aggregate exhibits anelastic transient creep. It is shown that the magnitude of the anelastic strain is related to the range of variability for the constitutive properties of the grains.

Constitutive laws for irradiation creep and stress-induced swelling have been proposed in the past, and they were to a large extent based on mechanistic models [1]. As a result, the constitutive equations are not only linked to rather specific model assumptions, but they also depend on many parameters which characterize point defects, lattice properties, and the microstructure. Needless to say, numerical values for these parameters are very rarely known. In addition to the multitude of parameters needed for just one mechanistic model, there are several atomistic processes which have been invoked as the cause of irradiation creep and stress-induced swelling [2]. It is in fact quite possible that several mechanisms contribute to the radiation-induced deformation, and that additional mechanisms may have to be discovered to understand the experimental data.

In view of this situation, it is desirable to approach the subject matter from a more phenomenological point of view without taking recourse to detailed mechanistic models. This can be done, as shown in this paper, with the formalism and tools provided by modern continuum mechanics. This discipline offers a few guiding principles of great generality which are particularly useful to find the proper tensorial relationships for multi-axial deformations. The disadvantage of this approach is that it is too general in the sense of supplying us with very extensive lists of possible constitutive laws. If the selection of the particular law were to be made on the basis of experimental tests alone, the number of tests would be prohibitively large indeed. Therefore,

in the author's opinion, an optimal strategy is reached by combining the phenomenological approach with insights gained from mechanistic models. This will reduce, as demonstrated in this paper, the extensive list provided by continuum mechanics. There are other shortcomings with the phenomenological approach related directly to the basic assumption of a continuum. If one assumes, as it is usually done, that the rate of deformation at a given location depends only on the values of stresses, temperature, etc., at that same location, and that the same constitutive law applies to all locations, then certain deformation phenomena are excluded. Indeed, the last assumption is in conflict with the heterogeneous nature of real polycrystalline materials where each region or grain must be assigned different constitutive properties. By averaging over the constitutive properties of grains we can derive a macroscopic constitutive law for a polycrystalline aggregate and thereby overcome one of the more severe shortcomings of the phenomenological approach. As will be shown in the last section, the macroscopic constitutive law contains then anelastic effects not present in the corresponding microscopic law.

The major advantage of the phenomenological approach is that one can derive constitutive laws of a general form which include swelling, stress-affected swelling, irradiation creep, anisotropic shape changes, and creep-swelling coupling effects such that all these phenomena are clearly identified without being counted twice or overlooked. Furthermore, they are already formulated for any state of stress. To obtain that tri-axial generality

from mechanistic models involves a great deal of labor and extreme care. Hence, the phenomenological approach offers an additional check for the correctness of tri-axial deformation laws extracted from models. Furthermore, it also provides the framework to extend measured constitutive equations to their most general tri-axial form as needed for structural analysis in reactor design. And finally, it defines the above mentioned phenomena so that appropriate experiments can be designed to measure them.

In the following we carry out this approach with a few examples to demonstrate its potential and value. We restrict ourselves to materials which had at one time an isotropic state that is used as a reference state. Consequently, the derived results are applicable only to cubic polycrystalline materials. However, if they have a texture because of cold-working prior to the irradiation, this particular anisotropy can also be dealt with in the present approach.

#### Notation

It will be expedient for the following to introduce first some conventions and notations which are extensively used in continuum mechanics. Tensors, such as the stress tensor  $\sigma_{ij}$  ( $i, j=1,2,3$ ) are simply denoted by  $\underline{\sigma}$ . A product of two tensors, as for example the stress and the strain tensor, will be written as  $\underline{\sigma\epsilon}$ , and stands for

$$\underline{\sigma\epsilon} = \sum_k \sigma_{ik} \epsilon_{kj} .$$

This product is again a tensor, not necessarily symmetrical, and we can form a scalar quantity, called the trace

$$\text{tr } \underline{\sigma\epsilon} = \sum_{i,k} \sigma_{ik} \epsilon_{ki} .$$



If a product is formed with identical tensors, we simply write  $\underline{\sigma}^2$ , meaning  $\underline{\sigma}\underline{\sigma}$ . Finally, the unit tensor is denoted by  $\underline{1}$  or by  $\delta_{ij}$ , where  $\delta_{ij}$  has the usual meaning of the Kronecker symbol:  $\delta_{ij} = 1$  if  $i = j$  and zero otherwise. Note that  $\sum_k \delta_{ik} \delta_{kj} = \underline{11} = \underline{1} = \delta_{ij}$ , and that  $\text{tr } \underline{1} = 3$ . Any tensor can be decomposed into an isotropic part and a deviatoric part. In the case of the stress and strain tensor, we have

$$\underline{\sigma} = \underline{s} + \underline{1} \frac{1}{3} \text{tr } \underline{\sigma} \quad (1)$$

and

$$\underline{\varepsilon} = \underline{e} + \underline{1} \frac{1}{3} \text{tr } \underline{\varepsilon} . \quad (2)$$

Special significance in the present context is given to

$$\text{tr } \underline{\varepsilon} = \frac{\Delta v}{v} = S , \quad (3)$$

which is equal to the swelling, and to

$$\frac{1}{3} \text{tr } \underline{\sigma} = \sigma_H , \quad (4)$$

which is the hydrostatic stress. According to the definitions of the deviatoric stress  $\underline{s}$  and deviatoric strain  $\underline{e}$ , Eqs. (1) and (2), their traces vanish, i.e.  $\text{tr } \underline{s} = \text{tr } \underline{e} = 0$ .

It is well known that the individual components of a tensor, say  $\sigma_{ij}$ , depend on the coordinate system used. However, there are three basic invariants

$$\text{tr } \underline{\sigma}, \text{tr } \underline{\sigma}^2, \text{tr } \underline{\sigma}^3$$

which are the same in each coordinate system. The invariants of the deviatoric tensor  $\underline{s}$  are of course related to those of  $\underline{\sigma}$ ; for example

$$\text{tr } \underline{s}^2 = \text{tr } \underline{\sigma}^2 - 3 \sigma_H^2. \quad (5)$$

Hence, we shall consider the three invariants

$$\sigma_H, \text{tr } \underline{s}^2, \text{tr } \underline{s}^3 \quad (6)$$

as our basic stress parameters. Among these,  $\sigma_H$  and

$$\text{tr } \underline{s}^2 = \frac{2}{3} \sigma_{eq}^2$$

have a physical meaning, whereas  $\text{tr } \underline{\sigma}^3$  has no clearly defined meaning except that it vanishes if the stress state is only uniaxial or biaxial.  $\sigma_{eq}$  is of course the maximum shear stress connected with the stress tensor.

An important thermodynamic quantity is the mechanical power

$$\text{tr } \underline{\sigma} \dot{\underline{\epsilon}} = \sum_{i,k} \sigma_{ik} \dot{\epsilon}_{ki} \quad (7)$$

where  $\dot{\underline{\epsilon}}$  is the deformation rate tensor. This quantity represents the rate of dissipation of the mechanical work associated with the deformation at a given point. In modern continuum mechanics, the basic thermodynamic laws are postulated to hold at each material point. Hence, the first law of thermodynamics is given by

$$\rho \dot{u} = \text{tr } \underline{\sigma} \dot{\underline{\epsilon}} + \text{div } \vec{q} + \rho r \quad (8)$$

where  $\rho$  is the mass density,  $\dot{u}$  is the rate of internal energy change,  $\vec{q}$  is the heat flux, and  $r$  is the rate of energy per unit mass deposited at each point by the radiation. The second law of thermodynamics is assumed to hold locally in the form of the Clausius-Duhem inequality [3]

$$\rho (\dot{\eta} - \dot{u}) + \text{tr } \underline{\underline{\sigma}} \dot{\underline{\underline{\epsilon}}} + \frac{1}{T} \vec{q} \cdot \text{grad } T \geq 0 \quad (9)$$

where  $\dot{\eta}$  is the rate of local entropy production.

Since experiments can be designed with nearly uniform temperature we may omit the last term from the inequality (9). In the absence of stress, the inequality (9) implies that  $\dot{\eta} \geq \dot{u}$ . Upon load application the elastic deformation makes a contribution to  $\dot{u}$  which cancels the elastic part of  $\text{tr } \underline{\underline{\sigma}} \dot{\underline{\underline{\epsilon}}}$ . Hence, in the following  $\underline{\underline{\epsilon}}$  denotes the inelastic strain tensor if not stated otherwise. If we assume that the inelastic deformation does not change the internal energy  $u$  of the material, then

$$\text{tr } \underline{\underline{\sigma}} \dot{\underline{\underline{\epsilon}}} \geq 0 . \quad (10)$$

The condition (10) implies that in a uni-axial tensile experiment  $\dot{\epsilon}$  is always positive even after the tensile stress  $\sigma$  is reduced to a lower positive value. Strain recovery, i.e. a negative  $\dot{\epsilon}$ , was however observed in a load drop experiment [4]. We will show in the last section of this paper that this anelastic strain recovery is a consequence of the inhomogeneity of polycrystalline materials, and that the macroscopic constitutive law for them does not satisfy the inequality (10). Only, the more general inequality (9) is applicable in this case.

### Dependence on Current Stress

Suppose that the rate of deformation  $\dot{\underline{\epsilon}}$  is a function of the current stress  $\underline{\sigma}$ , the displacement rate  $\phi$ , the time  $t$ , and a set of microstructural parameters,  $m$ . We write then

$$\dot{\underline{\epsilon}} = \underline{f}(\underline{\sigma}, \phi, t, m) . \quad (11)$$

If the material can be considered as isotropic, as in the case of a polycrystal with no texture, the constitutive law must be the same in any coordinate system. As a consequence, as Rivlin and Ericksen [4] have shown, the most general constitutive law of the form (11) for an isotropic material is given by

$$\dot{\underline{\epsilon}} = f_0 \underline{1} + f_1 \underline{\sigma} + f_2 \underline{\sigma}^2 , \quad (12)$$

where  $f_0$  to  $f_2$  are scalar functions depending on  $\phi$ ,  $t$ ,  $m$ , and the three stress invariants listed in (6). In order to separate  $\dot{\underline{\epsilon}}$  into swelling and creep we make use of the decompositions of the tensors  $\dot{\underline{\epsilon}}$  and  $\underline{\sigma}$  as given by Eqs. (1) and (2). We obtain then instead of Eq. (12) the equations

$$\begin{aligned} \dot{\underline{S}} &= 3 f_0 + 3 f_1 \sigma_H + f_2 (3 \sigma_H^2 - \text{tr } \underline{s}^2) \\ &= \psi_0(\phi, t, m, \sigma_H, \text{tr } \underline{s}^2, \text{tr } \underline{s}^3) \end{aligned} \quad (13)$$

and

$$\begin{aligned} \dot{\underline{\epsilon}} &= (f_1 + 2 f_2 \sigma_H) \underline{s} + f_2 (\underline{s}^2 - \underline{1} \frac{1}{3} \text{tr } \underline{s}^2) \\ &= \psi_1 \underline{s} + \psi_2 (\underline{s}^2 - \underline{1} \frac{1}{3} \text{tr } \underline{s}^2) , \end{aligned} \quad (14)$$

where we have introduced three new functions  $\psi_0$ ,  $\psi_1$ , and  $\psi_2$  which are linear combinations of the old functions  $f_0$ ,  $f_1$  and  $f_2$ .

Eq. (13) gives the general form for the swelling rate in an isotropic material under stress. With regard to irradiation creep, the directional contributions can be divided into two parts. The first term in Eq. (14) gives rise to creep strains which are colinear with those of the stress. The second term, however, gives contributions to creep in other directions. We can demonstrate this most clearly in a torsion experiment. In a cylindrical coordinate system with the axis assignment  $(r, \theta, z) = (1, 2, 3)$ , the deviatoric stress tensor is given by

$$\underline{s} = \begin{bmatrix} 0 & \tau & 0 \\ \tau & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}$$

so that

$$\underline{s}^2 - \frac{1}{3} \text{tr } \underline{s}^2 = \frac{1}{3} \begin{bmatrix} \tau^2 & 0 & 0 \\ 0 & \tau^2 & 0 \\ 0 & 0 & -2\tau^2 \end{bmatrix}.$$

Therefore, whereas the first term in Eq. (14) gives rise only to shear strains, the second term produces also normal strains. Consequently, if axial and radial deformations were observed in a torsion creep experiment it would indicate that the second term in Eq. (14) is required for a constitutive law.

Experiments with thin-walled pressurized tubes are also suitable to test for the existence of the second term in Eq. (14). If  $\sigma_\theta$  denotes the hoop stress, then the hoop strain rate is given by

$$\dot{\epsilon}_\theta = \frac{1}{2} \psi_1 \sigma_\theta + \frac{1}{6} \psi_2 \sigma_\theta^2$$

and the axial strain rate by

$$\dot{\epsilon}_z = -\frac{1}{12} \psi_2 \sigma_\theta^2 .$$

It should not be overlooked that  $\psi_1$  and  $\psi_2$  are in general dependent on the stress invariants. Thus, the two terms in Eq. (14) are not necessarily linear and quadratic in the stress.

The requirement of positive mechanical power imposes an important restriction on the selection of the functions  $\psi_0$  to  $\psi_2$ . The condition (10), applicable to the stress-induced deformation rate only, can be written as

$$\text{tr} (\dot{\underline{\epsilon}} - \dot{\underline{\epsilon}}_0) \underline{\sigma} \geq 0 , \quad (15)$$

where  $\dot{\underline{\epsilon}}_0$  is the stress-free deformation rate. Decomposition of the tensors gives

$$\text{tr} (\dot{\underline{\epsilon}} - \dot{\underline{\epsilon}}_0) \underline{s} + (\dot{s} - \dot{s}_0) \sigma_H \geq 0 ,$$

and since this inequality must be valid for any stress state, it follows that

$$\text{tr} (\dot{\underline{\epsilon}} - \dot{\underline{\epsilon}}_0) \underline{s} \geq 0 \quad (16)$$

and

$$(\dot{S} - \dot{S}_0) \sigma_H \geq 0 , \quad (17)$$

where  $\dot{S}_0$  is the stress-free swelling rate. With the constitutive law of Eq. (14), the condition (16) gives

$$\psi_1 \text{tr } \underline{s}^2 + \psi_2 \text{tr } \underline{s}^3 \geq 0$$

or in the case of uniaxial loading

$$\psi_1 + \psi_2 \frac{1}{3} \sigma \geq 0 .$$

In order to satisfy the last condition for a compressive stress of arbitrary value,  $\psi_2$  must either depend on the stress invariants in such a way as to change sign for compressive stresses, or else it must be zero.

The condition (17) asserts that a compressive hydrostatic stress reduces the swelling rate below its value for the stress-free case. Of course, the function  $\psi_0$  must be chosen so that  $S \geq 0$ .

#### Dependence on Current Stress and Strain

There are several experimental observations, discussed recently [5], which indicate that previous swelling and irradiation creep affects subsequent deformation. To include this in a constitutive law we may postulate the following dependence:

$$\dot{\underline{\epsilon}} = \underline{f} (\underline{\sigma}, \underline{\epsilon}, \phi, t, m) . \quad (18)$$

Again, it can be shown [4] that for a material which was originally (i.e. for  $\epsilon=0$ ) in an isotropic state, the most general law of the form of Eq. (18) is

$$\begin{aligned} \dot{\underline{\epsilon}} = & f_0 \underline{1} + f_1 \underline{\sigma} + f_2 \underline{\epsilon} + f_3 \underline{\sigma}^2 + f_4 \underline{\epsilon}^2 \\ & + f_5 (\underline{\sigma}\underline{\epsilon} + \underline{\epsilon}\underline{\sigma}) + f_6 (\underline{\sigma}\underline{\epsilon}^2 + \underline{\epsilon}^2\underline{\sigma}) \\ & + f_7 (\underline{\sigma}^2\underline{\epsilon} + \underline{\epsilon}\underline{\sigma}^2) + f_8 (\underline{\sigma}^2\underline{\epsilon}^2 + \underline{\epsilon}^2\underline{\sigma}^2) \end{aligned} \quad (19)$$

where the nine functions  $f_0$  to  $f_8$  may depend on  $\phi$ ,  $t$ ,  $m$  and the following list of ten invariants:

$$\begin{aligned} & \sigma_H, \text{tr } \underline{\sigma}^2, \text{tr } \underline{\sigma}^3, S, \text{tr } \underline{\epsilon}^2, \text{tr } \underline{\epsilon}^3, \\ & \text{tr } \underline{\sigma}\underline{\epsilon}, \text{tr } \underline{\sigma}\underline{\epsilon}^2, \text{tr } \underline{\sigma}^2\underline{\epsilon}, \text{tr } \underline{\sigma}^2\underline{\epsilon}^2. \end{aligned}$$

The constitutive law of Eq. (19) is already exceedingly complicated so that preliminary experimental information or other theoretical insight must be used to simplify it and reduce it to a tractable form.

Firstly, all mechanistic models for stress-affected swelling [6] show that for an isotropic material, i.e., for  $\underline{\epsilon} = 0$ , the effect depends on  $\sigma_H$  only. Hence, if we assume this to be true, then  $f_3 \equiv 0$ . Secondly, most data on irradiation creep for stainless steels [7] confirm, after proper subtraction of thermal creep, a linear stress dependence. Therefore,  $f_7 = f_8 = f_3 \equiv 0$ . Finally, the anisotropy created by previous deformation is to a first approximation linear in the strain tensor  $\underline{\epsilon}$ . In fact, most of the current mechanistic models for irradiation creep lead to this conclusion. Accordingly, we assume that  $f_4 = f_6 = f_8 \equiv 0$ . There remains then the constitutive law



$$\dot{\underline{\epsilon}} = f_0 \underline{1} + f_1 \underline{\sigma} + f_2 \underline{\epsilon} + f_5 (\underline{\epsilon}\underline{\sigma} + \underline{\sigma}\underline{\epsilon}) , \quad (20)$$

which is a drastic simplification of the one in Eq. (19), but it still incorporates important features discussed in [5] . To illuminate these, we employ again the decomposition of tensors into isotropic and deviatoric parts, and we obtain

$$\dot{S} = \psi_0 (\sigma_H, S, \text{tr } \underline{es}, \phi, t, m) \quad (21)$$

and

$$\dot{\underline{\epsilon}} = \psi_1 \underline{s} + \psi_2 \underline{e} + \psi_3 (\underline{es} + \underline{se} - \underline{1} \frac{2}{3} \text{tr } \underline{es}) . \quad (22)$$

Based on our assumption of a linear stress dependence of the irradiation creep rate,  $\dot{\underline{\epsilon}}$ , the functions  $\psi_1$  and  $\psi_3$  are independent of stress, whereas  $\psi_2$  may be a linear function of stress, i.e. of  $\sigma_H$  and  $\text{tr } \underline{es}$ . Let us first consider the swelling law of Eq. (21). We see that  $\dot{S}$  depends on  $S$ . This is not unreasonable, since the void structure present will certainly influence the subsequent swelling. A more interesting effect on  $\dot{S}$  is brought about by the dependence on  $\text{tr } \underline{es}$ . Its physical meaning can be illuminated by the following example.

Suppose that one irradiates two square metal sheets loaded, say, in tension along the 1-direction. At time  $t_0$  the load direction is changed on the second

sample into the 2-direction, and the irradiation is continued. The deviatoric strain tensor before time  $t_0$  is

$$\underline{e}(t_0) = \begin{bmatrix} \epsilon & 0 & 0 \\ 0 & -\frac{1}{2}\epsilon & 0 \\ 0 & 0 & -\frac{1}{2}\epsilon \end{bmatrix}$$

and the deviatoric stress tensors after time  $t_0$  are

$$\underline{s}^{(1)} = \begin{bmatrix} \frac{2}{3}\sigma & 0 & 0 \\ 0 & -\frac{1}{3}\sigma & 0 \\ 0 & 0 & -\frac{1}{3}\sigma \end{bmatrix}, \quad \underline{s}^{(2)} = \begin{bmatrix} -\frac{1}{3}\sigma & 0 & 0 \\ 0 & \frac{2}{3}\sigma & 0 \\ 0 & 0 & -\frac{1}{3}\sigma \end{bmatrix}.$$

For the first sample,  $\text{tr } \underline{e}\underline{s}^{(1)} = \epsilon\sigma$ , but for the second sample,  $\text{tr } \underline{e}\underline{s}^{(2)} = -\frac{1}{2}\epsilon\sigma$ . If  $\psi_0$  is linear in  $\text{tr } \underline{e}\underline{s}$ , then the swelling rate should decrease in the second sample after the rotation of the load direction. Such an effect is indeed expected if irradiation creep results in a dislocation loop structure which is preferentially aligned perpendicular to the tensile stress direction as was observed recently [8]. Upon rotation of the stress direction most loops are aligned parallel to the tension axis, and their capture efficiency is reduced according to theoretical calculations [1]. This results in a lower bias for swelling. An isotropic or random loop orientation would give no change in bias.

The anisotropy of the dislocation structure, being a result of the previous deformation, enters into our phenomenological description in the form of a dependence of  $\dot{\underline{\epsilon}}$  on  $\underline{\epsilon}$  itself. The isotropy assumption is made for the reference state only where  $\underline{\epsilon}$  was equal to zero.

Turning now to the irradiation creep law, Eq. (22), we see that the first term,  $\psi_1(S) \underline{\dot{e}}$ , represents the constitutive law as presently used [7]. Although it is independent of  $\underline{e}$ , it may be linked to swelling. However, this does not imply that the irradiation creep law is linear in the swelling  $S$ . The reason is that all the functions  $\psi_0$  to  $\psi_3$  may also depend on the microstructural parameters,  $m$ . It is in fact well known from theoretical models and experiments that void swelling is strongly dependent on the microstructure. Hence, the list of independent parameters in the functions  $\psi_0$  to  $\psi_1$  is redundant. We may solve Eq. (21) for  $S$  and insert it into the function  $\psi_1(S)$ , thereby obtaining a function of the swelling rate and the microstructure, i.e.  $\psi_1(\dot{S}, m)$ . In this way we can recover the previously proposed and widely used relationship for swelling dependent irradiation creep [1].

The second term in Eq. (22) remains even after load removal. In such a situation

$$\underline{\dot{e}} = \psi_2(0, 0, \phi, t, m) \underline{e} ,$$

and if we assume that  $\psi_2$  is time-independent, the integration of this equation gives

$$\underline{e}(t) = \underline{e}(0) \exp(\psi_2 t) . \quad (23)$$

This equation describes a shape change after load removal which is superimposed on the swelling rate  $\dot{S}_0$ . This effect, called "memory-creep", is however

fading with time, i.e.  $\psi_2$  must be a negative quantity to prevent an exponential increase of the anisotropy.

The last term in Eq. (22) can be interpreted as an anisotropic creep behavior caused by the anisotropic loop and dislocation structure that was generated during previous deformation.

#### Dependence on Stress and Cold-Working

Previously accumulated strains may not only include irradiation creep but plastic flow imposed on the material by the cold-working process. We may have to add then an additional tensor variable,  $\underline{c}$ , to the list of parameters in the constitutive law of Eq. (18). However, to avoid further complications, we assume that the cold-working strain tensor  $\underline{c}$  replaces  $\underline{\epsilon}$ . The ensuing law could be suitable for the initial radiation-induced deformation of cold-worked materials. Since plastic flow causes little volume changes,  $\text{tr } \underline{c} \cong 0$ , and the constitutive laws of Eqs. (21) and (22) become the following.

$$\dot{\underline{S}} = \psi_0 (\sigma_H, S, \text{tr } \underline{cs}) \quad (24)$$

$$\begin{aligned} \underline{\dot{\epsilon}} = \psi_1 (S) \underline{s} + \psi_2 (\sigma_H) \underline{c} \\ + \psi_3 (\underline{cs} + \underline{sc} - \frac{1}{3} \text{tr } \underline{cs}) . \end{aligned} \quad (25)$$

The presence of the second term in Eq. (25) implies that dimensional growth is anisotropic in stress-free cold-worked materials, and this was indeed observed [9]. There is no mathematical reason in the present case that this anisotropy be fading. To model it requires that  $\psi_2$  is a decreasing function with time.

### History Dependence

There is no compelling reason other than the desire for simplicity that the deformation rate  $\dot{\underline{\epsilon}}$  should depend only on the current stress  $\underline{\sigma}$  and strain  $\underline{\epsilon}$ . One can, as done recently [10], assume that it depends instead on the entire stress and strain history by introducing superposition integrals. These integrals replace then the independent parameters and tensors. As has been shown by Wineman and Pipkin [11] this added complexity will not change the tensorial relationship between  $\dot{\underline{\epsilon}}$  and  $\underline{\sigma}$ . For example, the constitutive law of Eq. (14) (with  $\psi_2 = 0$ ) is simply replaced by

$$\dot{\underline{\epsilon}}(t) = \int_0^t dt' \psi_1(t') \underline{s}(t')$$

where  $\psi_1(t')$  depends on the stress invariants at the time  $t'$ . We show in the following section that a history dependence with regard to the strain emerges when we consider heterogeneous material.

### Anelastic Effects

It was mentioned in the introduction that the requirement for positive mechanical power was not consistent with strain recovery upon load reduction, an effect which has in fact been observed experimentally for irradiation creep [12]. Nevertheless, one can still retain the requirement that the constitutive law at every material point satisfies the condition of positive mechanical power, if one combines it with the possibility of nonuniformity.

In a polycrystalline material this must in fact be done. Wolfer and Garner [13] have shown recently that the creep mechanism of stress-induced preferential absorption (SIPA) does imply that the creep rate depends strongly on the crystallographic orientation of the grain with respect to the stress. Consequently, a polycrystalline material consists of volume elements with varying irradiation creep properties. And since they form a composite which must deform together, the stresses vary from element to element so as to produce in each the same total deformation rate. Upon unloading, residual stresses reside in the elements, and their subsequent relaxation gives rise to anelastic transient effects.

Although the isotropy assumption is no longer true for the constitutive law of an individual grain, if we average over many grains we arrive at a macroscopic constitutive that is isotropic, provided the grains have random orientation.

In order to treat this in a more quantitative fashion, we model the polycrystalline aggregate as a series of viscoelastic elements connected in parallel, as shown in Fig. 1. The stress in the  $\alpha$ -th element is denoted by  $\sigma_\alpha$  and its irradiation creep rate is given by the simple law  $\dot{\psi}_\alpha \sigma_\alpha$ , where  $\psi_\alpha$  is a constant differing from element to element. We assume that the elastic modulus,  $E$ , of each element is the same. The total strain rate, composed of elastic and creep parts, is then

$$\dot{\epsilon} = \frac{1}{E} \dot{\sigma}_\alpha + \psi_\alpha \sigma_\alpha, \quad (26)$$

and it must be the same in each element to preserve the coherency of the aggregate. Solution of Eq. (26) with the initial condition that there are no initial microstresses, i.e.  $\sigma_\alpha(0) = 0$ , gives

$$\sigma_\alpha(t) = E \int_0^t \dot{\epsilon}(t') \exp \{E \psi_\alpha (t'-t)\} dt'. \quad (27)$$

For a constant creep rate

$$\sigma_\alpha(t) = \frac{\dot{\epsilon}}{\psi_\alpha} [1 - \exp (-E \psi_\alpha t)] . \quad (28)$$

This last expression clearly shows that the stresses in different elements are different. If  $n_\alpha$  is the fraction of elements with the same creep compliance  $\psi_\alpha$ , then the average, or macroscopic stress, can be defined as

$$\sigma(t) = \sum_{\alpha} n_{\alpha} \sigma_{\alpha}(t) . \quad (29)$$

From Eq. (27) we obtain then the relation

$$\sigma(t) = E \int_0^t \dot{\epsilon}(t') \sum_{\alpha} n_{\alpha} \exp \{E \psi_{\alpha} (t'-t)\} dt' ,$$

and, upon integration by parts, we find

$$\epsilon(t) = \frac{\sigma(t)}{E} + \int_0^t \epsilon(t') k(t-t') dt' \quad (30)$$

where the creep response function is defined as

$$k(\tau) = \sum_{\alpha} n_{\alpha} E \psi_{\alpha} \exp (-E \psi_{\alpha} \tau) . \quad (31)$$

For  $\tau = 0$ ,

$$k(0) = E\bar{\psi} \quad (32)$$

where

$$\bar{\psi} = \sum_{\alpha} n_{\alpha} \psi_{\alpha} \quad (33)$$

is the average creep compliance for the aggregate.

We can derive from Eq. (30) a constitutive law for the aggregate of the form discussed in previous sections. To this end, we differentiate Eq. (30) with respect to the time  $t$ . Then

$$\dot{\epsilon}(t) = \frac{\dot{\sigma}}{E} + \epsilon(t) k(0) + \int_0^t \epsilon(t') \frac{d}{dt} k(t-t') dt' ,$$

and upon substituting  $\epsilon(t)$  from Eq. (30), we obtain finally

$$\dot{\epsilon}(t) = \frac{\dot{\sigma}}{E} + \bar{\psi}\sigma + \int_0^t \epsilon(t-\tau) [k(0) k(\tau) + \frac{d}{d\tau} k(\tau)] d\tau . \quad (34)$$



The second term represents the average irradiation creep rate for the aggregate, whereas the last term depends on the strain history.

The response function  $k(\tau)$  of the aggregate is determined by the distribution  $n_\alpha$  of elements with a given creep compliance  $\psi_\alpha$ . For a polycrystalline material we can assume that an equal number of grains exist with a creep compliance between  $\bar{\psi} - \Delta\psi$  and  $\bar{\psi} + \Delta\psi$ . The sum in Eq. (31) can then be written as an integral, i.e.

$$k(\tau) = \int d\psi n(\psi) E\psi \exp(-E\psi\tau),$$

where  $n(\psi) = 1/(2\Delta\psi)$  for  $\bar{\psi} - \Delta\psi \leq \psi \leq \bar{\psi} + \Delta\psi$  and  $n(\psi) = 0$  otherwise. The evaluation of the integral in the above equation gives the following result:

$$k(\tau) = E\Delta\psi e^{-z} \left\{ \left( \frac{\bar{\psi}}{\Delta\psi} + \frac{1}{z} \right) \frac{\sinh z}{z} - \frac{\cosh z}{z} \right\} \quad (35)$$

where

$$z = E\Delta\psi\tau.$$

It is instructive to solve Eq. (34) for the case of a step-load change. Let us suppose that at time  $t = 0$  the stress is changed from  $\sigma$  to  $\sigma + \Delta\sigma$ , where  $\Delta\sigma$  can be either positive or negative. To compute  $\dot{\epsilon}(t)$  from Eq. (34) Laplace transforms are used. The result of the somewhat lengthy calculations [14] is

$$\dot{\epsilon}(t) \cong \bar{\psi}\Delta\sigma \left\{ 1 - \frac{1}{3} \left( \frac{\Delta\psi}{\bar{\psi}} \right)^2 [1 - \exp(-E\bar{\psi}t)] \right\} + \dot{\epsilon}_0$$

if  $\Delta\psi/\bar{\psi} \ll 1$ . Here,  $\dot{\epsilon}_0$  is the creep rate before the load change.

We see from this expression that the creep rate immediately after the load change is given by  $\dot{\epsilon}(0) = \bar{\psi}\Delta\sigma + \dot{\epsilon}_0$  whereas the asymptotic creep rate for  $t \rightarrow \infty$  is given by

$$\dot{\epsilon}(\infty) = \bar{\psi}\Delta\sigma \left[ 1 - \frac{1}{3} \left( \frac{\Delta\psi}{\bar{\psi}} \right)^2 \right] + \dot{\epsilon}_0 .$$

By measuring these two values of the irradiation creep rate in a load change experiment, it is possible to find the variation of the creep compliances for the different grains according to the formula

$$\frac{\dot{\epsilon}(0) - \dot{\epsilon}(\infty)}{\dot{\epsilon}(0) - \dot{\epsilon}_0} = \frac{1}{3} \left( \frac{\Delta\psi}{\bar{\psi}} \right)^2 . \quad (38)$$

The information about the parameter  $(\Delta\psi/\bar{\psi})$  is not only important for a better understanding of irradiation creep, but is also of practical significance. If certain alloys exhibit a large variation in the irradiation creep compliances of different grains, then large residual microstresses will develop in these materials when subject to loads. Large differences in these microstresses from grain to grain are likely to be detrimental to creep rupture and ductility properties because of enhanced grain boundary fracture.

#### Summary and Conclusions

Using basic principles of continuum mechanics one can derive constitutive laws on a phenomenological basis. No detailed mechanistic description is needed to properly formulate for any state of stress such phenomena as

irradiation creep, stress-induced swelling, anisotropic growth, and the coupling between growth and irradiation creep. This is particularly important when one needs to apply the measured deformation behavior from a pressurized tube or uniaxial specimen to the deformation of actual reactor components.

Information obtained from mechanistic insights, as supplied by microscopy or theoretical modelling, is however very important for making the proper choice among many possible phenomenological laws. As examples of this phenomenological approach the following constitutive laws were considered: the radiation-induced deformation rate depends only on the stress, it depends on the stress and the previous strain, or it depends on the stress and the deformation produced by cold-working.

The various phenomena that can arise for these cases were discussed and in some cases related to experimental observations. It is hoped that these examples not only demonstrate the approach but that they will provide the framework for the analysis of irradiation creep experiments as well as suggestions for future testing. In particular, they suggest the extraction of additional measurements not normally made on irradiation creep specimens. The approach also allows the extrapolation of data into stress states not normally tested.

Not all deformation phenomena can be understood properly by considering a specimen as a homogeneous continuum. We demonstrated this by deriving the anelastic transient irradiation creep from a simple model that consists of viscoelastic elements connected in parallel. This model can serve as a realistic description of the irradiation creep behavior of a polycrystalline material.

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Fig. 1 Viscoelastic model for irradiation creep in a polycrystal.

