

## Evaluation of irradiation-induced creep rate: application to the vacancy dislocation loop contribution

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**Abstract.** Irradiation (as in a nuclear reactor) drastically affects the defect structure and its time evolution in a material, and induces new creep mechanisms in it. We present a formalism to evaluate the contribution to creep owing to such mechanisms. Beginning with the phenomenological constitutive relation for the strain appropriate to a given mechanism, we put in simple statistical considerations to derive an expression for the corresponding creep rate. This formal expression is in terms of the defect production rate and a non-equilibrium probability distribution function involving the pertinent properties of the defect type concerned. A convenient approximation scheme for practical calculations is employed, that also makes contact with standard rate theory and provides a proper interpretation for the variables occurring there. As an illustration, we evaluate the contribution to irradiation-induced creep from the orientation-dependent shrinkage of vacancy dislocation loops in an applied stress field. The circumstances inducing transient and non-transient creep are clarified and a numerical estimate is given for the latter component.

**Keywords.** Stress; creep; irradiation; vacancy dislocation loops; rate theory; distribution functions.

### 1. Introduction

An important mechanical property of a material is its time-dependent deformation under a constant applied stress, known as creep. This phenomenon must be understood in terms of the stress-assisted kinetics of various species of interacting defects in the material. In particular, the continual infusion and subsequent evolution of defects in the presence of irradiation introduces additional complications in the process of deformation. This causes an enhancement of creep under irradiation, with important implications for fast reactor design. The basic mechanisms responsible for this enhancement have been a subject of considerable debate. A vast literature exists on the topic (Piercy 1968, Hesketh 1972, Heald and Speight 1974, Bullough 1975, Gittus 1975, Wolfer and Ashkin 1976, Harries 1977), and many of the models proposed have been reviewed comprehensively by Gilbert (1971) and more recently in Cundy *et al* (1977). Much of the work on the subject has been phenomenological. This is of course a consequence of the highly complex physical situation, with a host of interacting defect types evolving under a variety of physical conditions such as the irradiation dose, its rate, the temperature of the specimen, its structure and metallurgical history, and so on. It is therefore natural that most of the models developed in order to go beyond empiricism refer to rather idealized cases, in which a very small number of physical mechanisms are assumed to be at work.

Even after a specific creep mechanism has been stipulated, the calculation of the creep rate requires two additional problems to be surmounted. The first is the calculation of the evolution rate of the defect structure, i.e., of a single, given defect. A comprehensive theory derived on the basis of statistical considerations would indeed be desirable, but such an approach is extremely difficult, except perhaps in the simplest idealized cases. A reasonably good compromise is provided by the rate theory model of Bullough *et al* (1975). (We shall show that such a model matches a more formal distribution function approach upto the level of its first moment.) The second problem is one of statistics, amounting to evaluating the creep rate due to a distribution of similar defects when the contribution due to a single defect is known. For stationary distributions, this is little more than a simple enumeration. However, under irradiation the defect distributions evolve in time, and this must be taken into account. In general, of course, the two problems listed above are coupled to each other, putting exact solutions well-nigh out of reach.

In this paper, we attempt to develop the requisite theoretical *methodology* with a view to aiding explicit calculations in any given case. For convenience of illustration, we work in the framework of a particular problem, namely, the evaluation of the contribution to irradiation-induced creep from vacancy dislocation loops evolving under an applied stress. We emphasise, however, that the general approach is intended to be applicable in several other instances as well, with appropriate modification of detail (see, for instance, Venkataraman and Balakrishnan 1977 for further remarks in this regard).

In § 2, we describe a simple statistical approach\* to the problem at hand, and obtain an expression for the strain rate that stands as the basis underlying the corresponding constitutive relation written down phenomenologically. An assumption regarding the basic distribution function then leads to a simplified theory amenable to practical calculations, besides making contact with rate theory (Bullough *et al* 1975) and elucidating how the latter may be interpreted as a special case of our formalism. In § 3, we apply our theory to an explicit calculation of the creep rate arising from the orientation-dependent shrinkage of vacancy dislocation loops under an applied stress. For the purposes of numerical estimation in the present instance, it is sufficient to work in the lowest approximation to the general theory (i.e., the equivalent of rate theory), and this is what we do. Certain defects in earlier work on the vacancy dislocation loop problem (Krishan and Ray 1975), also pointed out subsequently by Weiner (1976) and Lewthwaite (1976), are rectified in the present approach. The concluding remarks summarise the work and offer a perspective of our approach.

## 2. General Theory

### 2.1. Introduction

The first step is the association of the strain at time  $t$  with the corresponding value of a suitable physical variable, depending on the problem. For instance, the relevant

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\*Statistical considerations *have* been used earlier in isolated problems in the field (see, e.g. the references in Gittus 1975). By such considerations we mean, of course, much more than the introduction of ad hoc, static distributions for quantities otherwise treated deterministically, such as dislocation length segments, grain sizes, etc.

variable may be the total area of vacancy dislocation loops of a given orientation (as in the rest of this paper), or the volume of neutron or fission-fragment spikes, etc. (see Gilbert 1971 for various examples). In the model chosen for illustration (Krishan and Ray 1975, Lewthwaite 1976), creep occurs because of the change in the area of vacancy dislocation loops (referred to simply as 'loops' hereafter). The loops are formed in the core regions of displacement cascades produced by irradiation. The nucleation of the loops occurs with uniform probability in all orientations independent of the applied stress. Once formed, the loops shrink by absorbing interstitials and by thermal emission of vacancies. The emission probability is influenced by the applied stress, leading to an orientation dependence in the loop population. To evaluate the strain rate and the creep (by this we shall mean the deviatoric part of the strain tensor) due to such a distribution of loops, we must write first an appropriate 'constitutive' relation between the strain  $e(t)$  and  $A(t)$ , the area at time  $t$  of a single loop, or rather, the change in area as compared to the initial area  $A(0)$  at some fiducial instant of time, taken to be  $t=0$ . Thus

$$e(t) = k[A(t) - A(0)], \quad (1)$$

where  $k$  is a quantity that is, in general, the product of a number of time-independent parameters involving some geometrical factors, orientation dependence, etc. Such a constitutive relation is also applicable to many of the models proposed for irradiation creep, e.g., creep due to the stress-induced preferential absorption of interstitials (Heald and Speight 1974), exhaustion creep (Welch and Smoluchowski 1972), etc. This fact underlies the generality we claim for the formalism.\* In what follows, we evaluate the total strain  $\epsilon(t)$  due to *all* the loops. This will depend on both the constitutive relation (1) as well as the manner in which the loop distribution evolves in time.

## 2.2. Formula for the strain rate

We assume that irradiation, and the attendant loop formation, commences at  $t=0$ . Let  $\rho(A_0, t_0)dA_0dt_0$  be the number of loops with areas in the range  $(A_0, A_0+dA_0)$  that are nucleated\*\* in unit volume of the material in the time interval  $(t_0, t_0+dt_0)$ . The subsequent evolution of the loops is specified by a probability distribution function  $W$ : given that a loop nucleates with area  $A_0$  at time  $t_0$ , the probability that it has an area in the range  $(A, A+dA)$  at time  $t$  is  $W(A, t | A_0, t_0) dA$ . (For shrinking loops, the range of physical interest is  $0 \leq A \leq A_0$ ). The distribution function may have a further implicit dependence on other variables such as the orientation  $(\theta, \phi)$  of the loops (e.g., when a uniaxial stress is applied). For simplicity we shall not indicate this dependence explicitly until it is needed in § 3.

We are interested in deriving a formal expression for the strain rate in terms of the general distribution  $W$ . The initial condition pertinent to our model is

$$W(A, t_0 | A_0, t_0) = \delta(A - A_0); \quad (2)$$

\*However, in certain cases, the basic constitutive relation (obtained on physical grounds) is necessarily a differential one, involving  $e(t)$ . In such cases,  $e(t)$  will involve an integral over the previous history of the appropriate variable.

\*\*Note that  $\rho$  is the rate of production of the actual defect species of interest. Its relation to the irradiation dose rate must be worked out separately and fed into the calculations. See, e.g., Bullough *et al* (1975).

for  $t > t_0$ ,  $W$  describes the distribution in area of loops that shrink continuously to zero area starting from the given initial area  $A_0$ . We shall ignore the effect of the small fraction of loops that may undergo discontinuous jumps in area, e.g., by coalescing with other loops. In the presence of continual irradiation, we should not expect  $W$  to be a stationary distribution, i.e., a function of the difference  $(t - t_0)$  alone, as far as its time-dependence is concerned. This non-stationarity introduces certain complications into the problem.

The number of loops per unit volume with areas in the range  $(A, A + dA)$  at time  $t$  is given by\*

$$n(A, t)dA = dA \int_0^{A_{\max}} dA_0 \int_0^t dt_0 \rho(A_0, t_0) W(A, t | A_0, t_0) / I(t; A_0, t_0), \quad (3)$$

where the distribution is normalised on dividing by

$$I(t; A_0, t_0) = \int_0^{A_0} dA W(A, t | A_0, t_0). \quad (4)$$

$A_{\max}$  stands for the upper limit on the area of a freshly nucleated loop. The total area of all the loops at time  $t$  is

$$\begin{aligned} \mathcal{A}(t) &= \int_0^{A_{\max}} dA A n(A, t) \\ &= \int_0^{A_{\max}} dA \int_0^{A_{\max}} dA_0 \int_0^t dt_0 \rho(A_0, t_0) A W(A, t | A_0, t_0) / I(t; A_0, t_0). \end{aligned} \quad (5)$$

(In the case of orientation-dependent evolution of the loops, an integration over all orientations is also necessary, of course.) The strain rate, given by

$$\dot{\epsilon}(t) = k \frac{d\mathcal{A}}{dt}, \quad (6)$$

from (1), is therefore

$$\dot{\epsilon}(t) = k \int_0^{A_{\max}} dA A \rho(A, t) + k \int_0^{A_{\max}} dA \int_0^{A_{\max}} dA_0 \int_0^t dt_0 A \rho(A_0, t_0) \frac{\partial}{\partial t} (W/I). \quad (7)$$

The first term on the right merely represents the contribution of freshly nucleated loops at time  $t$ , while the second term arises from the cumulative effect of loops born at earlier instants of time. Equation (7) is the general formula sought for the strain rate. The derivation is emulated easily in any other case of interest.

\*We should actually indicate the orientation dependence of  $W$ ,  $n$ , etc. As stated earlier, we shall do so subsequently when required. It is assumed that the orientation of a loop remains unaltered throughout its lifetime.

### 2.3. Peaked distributions and connection with rate theory

A first-principles theory would require the derivation and solution of appropriate coupled master equations for the functions  $\rho$  and  $W$  corresponding to each of the interacting defect species involved in the problem. Except in very idealised situations, this is a virtually impossible task, although it may be possible in many situations to 'derive' a generalised Fokker-Planck-like equation for it, and to proceed from that point. However, since the formulas derived above entail only the zeroth and first moments of  $W$  with respect to  $A$ , it is possible, and sufficient, to use a satisfactory approximation to  $W$  that involves the same input parameters as the exact answer would. This approximation amounts to saying that the distribution is peaked extremely sharply around a certain most probable value  $\alpha$  at all times, where  $\alpha$  is itself a function of  $t$  as well as the initial parameters  $A_0$  and  $t_0$ . Thus we simply approximate  $W$  according to

$$W(A, t | A_0, t_0) \approx \delta(A - \alpha(t; A_0, t_0)), \quad (8)$$

with  $\alpha(t_0; A_0, t_0) \equiv A_0$ .

It is at once evident that this is equivalent to the simplest possible 'decoupling' of higher moments. The problem is now reduced to the specification of  $\rho(A_0, t_0)$  and  $\alpha(t; A_0, t_0)$ . Indeed, the rate equation approach may be interpreted as the derivation (from phenomenological considerations) and solution of an 'equation of motion' for the quantity  $\alpha$ . On substituting (10) in (7), we find that the integration over  $A$ ,  $A_0$  and  $t_0$  is restricted to the surface  $A = \alpha(t; A_0, t_0)$ , so that some care must be exercised in simplifying the integrals. It is now necessary to introduce the mean lifetime  $\tau$  of the loops. While the precise expression for  $\tau$  would follow from the actual equation of motion for  $\alpha$ , we may record here a few general properties of  $\tau$ . Since  $W$  is a non-stationary distribution,  $\tau$  will depend on the time of nucleation  $t_0$ , although the dependence may be a weak one. There may be a weak dependence on  $A_0$  as well. In practice, the shrinkage is rapid enough to cause  $\alpha$  to reach the value zero for a finite value of  $t$ . The lifetime is defined then by

$$\alpha(t_0 + \tau; A_0, t_0) \equiv 0. \quad (9)$$

Inserting (8) in (5) for  $A(t)$  and integrating over  $A$ , we get after some simplification

$$\dot{\epsilon}(t) = k \int_0^{A_{\max}} dA_0 A_0 \rho(A_0, t) + k \int_0^{A_{\max}} dA_0 \int_{t_{\min}}^t dt_0 \rho(A_0, t_0) \frac{\partial}{\partial t} \alpha(t; A_0, t_0), \quad (10)$$

where the lower limit of integration over  $t_0$  is

$$t_{\min} = \max [0, t_b(A_0, t)]. \quad (11)$$

Here  $t_b(A_0, t)$  is the (mean) time of birth of a loop that nucleates with area  $A_0$  and shrinks to zero area at time  $t$ , i.e., it is the solution of

$$\alpha(t; A_0, t_0 = t_b) = 0, \quad (12)$$

regarded as an equation for  $t_0$ .

Another useful representation for the strain rate can be obtained by changing variables from  $t_0$  to  $\alpha$  itself, by formally inverting the relation

$$\alpha = \alpha(t; A_0, t_0), \quad (13)$$

to write  $t_0$  as a function of  $\alpha$ ,  $t$  and  $A_0$ . We then obtain

$$\dot{\epsilon}(t) = k \int_0^{A_{\max}} dA_0 A_0 \rho(A_0, t) + k \int_0^{A_{\max}} dA_0 \int_{A_{\min}}^{A_0} d\alpha \rho(A_0, t_0(\alpha)) (\alpha_t / \alpha_{t_0}), \quad (14)$$

$$\text{where } \alpha_t \equiv (\partial \alpha / \partial t)_{t_0}, \quad \alpha_{t_0} \equiv (\partial \alpha / \partial t_0), \quad (15)$$

$$\text{and } A_{\min} = \max [0, \alpha(t; A_0, 0)]. \quad (16)$$

The partial derivatives in (14) must of course be regarded as functions of  $\alpha$ ,  $t$  and  $A_0$ , on eliminating  $t_0$  (after differentiation) with the help of (13). Equations (10) and (14) represent two related ways of looking at the same problem. In the former case we focus attention on specific loops and then trace their time evolution. In the latter picture, we fix our attention on a given instant of time and scan through loops with different areas. It is now appropriate to discuss briefly certain special cases of (10) that obtain under *further* assumptions about the nucleation and evolution of the loops, as there appears to be some confusion in this regard in the literature.

#### 2.4. Special cases

First, if all loops are assumed to nucleate with the same area (again denoted by  $A_0$ ) at a rate  $\rho(t)$ , (10) reduces to

$$\dot{\epsilon}(t) = k A_0 \rho(t) + k \int_{t_{\min}}^t dt_0 \rho(t_0) \alpha_t(t; t_0), \quad (17)$$

$$\text{and therefore } \epsilon(t) = k \int_{t_{\min}}^t dt_0 \rho(t_0) \alpha(t; t_0). \quad (18)$$

Next, suppose the lifetime of each loop is independent of its time of nucleation, i.e.,  $\alpha(t; t_0) = \alpha(t - t_0; 0) \equiv \alpha(t - t_0)$ , with  $\alpha(0) = A_0$  and  $\alpha(\tau) = 0$ . Since  $t_{\min} = \max(0, t - \tau)$  in this case, we obtain

$$\dot{\epsilon}(t) = \begin{cases} k \rho(0) \alpha(t) + k \int_0^t dt_0 \dot{\rho}(t_0) \alpha(t - t_0), & 0 \leq t \leq \tau, \\ k \int_{t-\tau}^t dt_0 \dot{\rho}(t_0) \alpha(t - t_0), & t > \tau. \end{cases} \quad (19)$$

Finally, if we make a third assumption that  $\rho(t) = \text{constant} = \rho$ , we have simply

$$\dot{\epsilon}(t) = \begin{cases} k\rho a(t), & 0 \leq t \leq \tau, \\ 0, & t > \tau. \end{cases} \quad (20)$$

Thus only a *transient* occurs in this case, the strain saturating to a constant value once  $t$  exceeds the lifetime of a loop. What happens physically is that a steady state is reached in which the loss of loops in a given area interval by shrinkage is balanced exactly by the entry of other loops from the area interval immediately above the former range. It should be emphasised strongly that this result is valid only in the very special case defined by the three simplifying assumptions made above. In particular, we may no longer expect such a result if the lifetime  $\tau$  depends on the time of nucleation,  $t_0$ . We shall find, below, that the rate theory model (Krishan and Ray 1975) does incorporate such a dependence. It is therefore incorrect to conclude (Lewthwaite 1976) that this model leads to strictly transient creep.\*

### 3. Creep due to shrinkage of vacancy dislocation loops

#### 3.1 Expression for the deviatoric strain rate

The physics underlying the model has already been adequately described in Krishan and Ray (1975). In accordance with the justification given there, we assume that all the loops nucleate with the same area  $A_0$  at a constant rate of production  $\rho = K\epsilon_f/bA_0$ ; here  $K$  is the dose rate,  $\epsilon_f$  is the fraction of vacancies (produced in cascades) that condense into loops, and  $b$  is the magnitude of the Burgers vector of these loops. Under these conditions, (14) reduces to

$$\dot{\epsilon}(t) = k\rho A_0 + k\rho \int_{A_{\min}}^A da(a_t/a_{t_0}). \quad (21)$$

We assume that a homogeneous, uniaxial stress  $\sigma$  is applied along the  $x_3$  axis. The orientation  $\Omega(\theta, \phi)$  of a loop is specified by the direction of its Burgers vector  $\mathbf{b}$ , which is either parallel or antiparallel to the unit normal  $\hat{n}$  of the plane of the loop (see figure 1 of Krishan and Ray 1975). The rate of formation of loops with orientations in the range  $(\Omega, \Omega + d\Omega)$  is given by  $\rho d\Omega/2\pi$ . Introducing tensor indices and identifying  $k$  with  $-b$  (see below), the contribution to the strain rate from loops in the orientation range  $(\Omega, \Omega + d\Omega)$  is

$$d\dot{\epsilon}_{ij}(t, \Omega) = -(b\rho n_i n_j d\Omega/2\pi) [A_0 + \int_{A_{\min}}^{A_0} da a_i(\xi)/a_{i_0}(\xi)], \quad (22)$$

where  $\xi = \xi_0 \cos^2\theta$ ,  $\xi_0 \equiv (\sigma b^3/k_B T)$ . (23)

\*There is an error in the formalism of Krishan and Ray (1975) arising from an incorrect interpretation of the constitutive relation. This has been pointed out by Weiner (1976). The present formalism corrects the error and supersedes the earlier work.

The explicit dependence of  $\alpha$  on the parameter  $\xi$  indicated in (22) is meant to emphasise its orientation dependence. (We have anticipated the precise form of this dependence in defining  $\xi$ .) Note also that the factors  $n_i, n_j$ , etc., take care of the tensorial nature of the strain, so that the constant  $k$  is a scalar. In the present case  $k$  must be equal to  $-b$ , the minus sign arising because the formation of vacancy loops causes a negative strain (although, as we shall see, the creep rate will be positive). The creep rate is the deviatoric part of the strain rate tensor, and is defined as

$$\dot{\epsilon}_{ij}^d \equiv \dot{\epsilon}_{ij} - \frac{1}{3} \delta_{ij} \left( \sum_{l=1}^3 \dot{\epsilon}_{ll} \right). \quad (24)$$

When the expression in (22) is integrated over  $d\Omega$  to include loops of all orientations, the first term on the right in that equation does not contribute to  $\dot{\epsilon}_{ij}^d$ . Let us define

$$J \equiv \alpha_t(\xi)/\alpha_{t_0}(\xi), \quad (25)$$

$J$  being understood to be a function of  $\alpha, t$  and  $\Omega$  (or  $\xi$ ). We then find

$$\dot{\epsilon}_{ij}^d(t) = - (b\rho/2\pi) \int d\Omega (n_i n_j - \frac{1}{3}) \int_{A_{\min}}^{A_0} J d\alpha \quad (26)$$

for the creep rate. It should be remembered that  $A_{\min}$  (see (16)) depends on  $\Omega$ , through the implicit  $\xi$  dependence of  $\alpha$ .

In practice, it turns out that we need retain terms only upto first order in  $\xi$  in the above formula, because  $\sigma b^3/k_B T \ll 1$  ( $b \simeq 2 \text{ \AA}$  for the loops concerned; for temperatures above  $350^\circ\text{C}$  and stress levels upto  $\sim 10 \text{ MNm}^{-2}$ , this linear approximation introduces a relative error of  $\sim 1\%$  in the creep rate.) Expanding the integral over  $\alpha$  in (26) in powers of  $\xi$ , and noting that the  $O(\xi^0)$  term vanishes on integration over  $d\Omega$ , we obtain to  $O(\xi)$  the result

$$\begin{aligned} \dot{\epsilon}_{ij}^d = & - (b\rho \xi_0/2\pi) \int d\Omega (n_i n_j - \frac{1}{3}) \cos^2 \theta \left[ \int_{A_{\min}}^{A_0} d\alpha (\partial J/\partial \xi) \right. \\ & \left. - (\partial A_{\min}/\partial \xi) J(\alpha = A_{\min}) \right]_{\xi=0}. \end{aligned} \quad (27)$$

The second term in the square brackets vanishes identically when  $A_{\min}=0$ , that is, when  $t$  is equal to or greater than the lifetime of a loop that nucleates at  $t_0=0$ . We define this to be the non-transient region (see below). We shall henceforth restrict our attention to the non-transient regime and therefore drop the transient term in (27). Changing variables to the loop radius  $r$  ( $\alpha=\pi r^2$ ) we obtain

$$\dot{\epsilon}_{ij}^d = (8\pi b\rho \xi_0/45) \int_0^{r_0} (\partial J/\partial \xi)_{\xi=0} r dr, \quad (28)$$

with  $\pi r_0^2 = A_0$ . It is easily verified that  $\dot{\epsilon}_{11}^d = \dot{\epsilon}_{22}^d = -(\frac{1}{2}) \dot{\epsilon}_{33}^d$  as required, and that the off-diagonal terms of  $\dot{\epsilon}_{ij}^d$  vanish identically. The quantity  $J$  is given by (25); it is evident that  $\alpha(\xi)$  can be replaced by  $r(\xi)$  in that equation. Note that the dose rate dependence of the creep rate is contained in both  $\rho$  and  $J$ .

### 3.2. Creep in a rate theory model

The task now is to evaluate the derivatives (with respect to  $t$  and  $t_0$ ) of  $r(\xi)$  required to obtain  $J$ . For this we need an explicit form for the most probable radius  $r(t; A_0, t_0)$ . The rate theory model (Bullough *et al* 1975; see also Krishan and Nandedkar 1979) provides a framework for calculating this quantity. We emphasise the following:

Equation (28) is a general result for the model under consideration, essentially following from (1) and (2) alone. Once we adopt rate theory, however, we inherit all the assumptions contained therein, direct or implied. Since rate theory has been well discussed in the literature cited, we refrain from listing these here. In the usual notation, rate theory gives

$$\frac{\partial}{\partial t} r(t; t_0) = -\frac{1}{b} (Z_I D_I C_I - D_V C_V) - \frac{1}{b} D_V C_V^e P(r) e^{\xi}. \quad (29)$$

Here  $D$  and  $C$  with appropriate suffixes (V for vacancies and I for interstitials) are the diffusion constants and the time-dependent concentrations, and  $Z_I$  is the dislocation bias factor for preferential interstitial migration to dislocations. The equilibrium vacancy concentration is  $C_V^e = \exp(-E_f/k_B T)$ ,  $E_f$  being the vacancy formation energy. Further,

$$P(r) = \exp [b^2 (\gamma_{SF} + F_{el}(r))/k_B T], \quad (30)$$

where  $\gamma_{SF}$  is the stacking fault energy and

$$F_{el}(r) = \mu b^2 \ln \left( 1 + \frac{r}{b} \right) / 4\pi (1 - \nu) (r + b) \quad (31)$$

is the elastic energy of a loop of radius  $r$ ;  $\mu$  is the shear modulus and  $\nu$  the Poisson ratio. The function  $P(r)$  has a complicated dependence on  $r$ , but, as has been shown *numerically* by Krishan and Ray (1975), it can be replaced without much loss of accuracy by a suitable linear function of  $r$  over the range of interest. (This introduces a maximum relative error of  $\sim 5\%$  in the effective range of variation of  $r$ , i.e., between 4 Å and 20 Å. The choice  $r_0 = 20$  Å is based on experimental observation, while the cut-off at the lower end is very reasonably taken to be 4 Å, the Burgers vector itself having a magnitude of 2 Å.) We therefore write  $P(r) \simeq pr + q$ ; from the numerical values given in the reference cited,  $p \approx -5 \times 10^6 \text{ cm}^{-1}$  and  $q$  varies between 2 and 3, depending on the temperature. The vacancy shrinkage rate can now be written as

$$\partial r / \partial t = - [f(t) + ar], \quad (32)$$

$$\text{where } f(t) = B(t) + q D_v C_v^e (1 + \xi)/b, \quad (33)$$

$$\text{with } B(t) = (Z_I D_I C_I - D_v C_v)/b \quad (34)$$

$$\text{and } a = D_v C_v^e p (1 + \xi)/b, \quad (35)$$

with only terms linear in  $\xi$  being retained.

The time dependence of  $B(t)$  is crucial in determining the creep rate. This is found in rate theory by solving a complicated set of coupled equations. It will be very convenient to relate  $B(t)$  to a physical parameter, the loop life-time (see below). This also makes it convenient to deduce the dependence of the creep rate on external variables such as the dose rate and temperature. (As in Krishan and Ray 1975, we shall work with reference to neutron irradiation of M316 stainless steel at a dose rate of  $10^{-6}$  dpa/sec). The numerical values obtained for  $B(t)$  permit one to approximate  $B(t)$  by a piecewise linear function of  $t$  over the lifetime of the loops, under the quasi-steady state conditions of interest to us. We shall now proceed taking  $f(t)$  to be a known function of time (i.e, input information).

Integration of (32) over a time span  $(t_0, t)$  gives  $r$  explicitly as a function of  $t_0$  and  $t$ . Since  $f(t)$  is independent of  $t_0$ , we have

$$\partial r(t; t_0)/\partial t_0 = [f(t_0) + ar_0] \exp [-a(t-t_0)]. \quad (36)$$

In order to carry out the integration in (26), we must write  $t_0$  as a function of  $r$ . For this, we formally introduce the variable  $\tau(r, t) \equiv (t-t_0)$ , which has the significance that it represents the 'age' of a loop having a radius  $r$  at time  $t$ . The quantity  $\tau(0, t)$  is the loop *lifetime*, and will be denoted simply by  $\tau$ . An exact expression for  $\tau(r, t)$  can be found in principle. However, in the present case, the piecewise linearity of  $B(t)$  with a small slope leads to the satisfactory approximation

$$\tau(r, t) \simeq -(1/a) \ln [(f(t) + ar)/(f(t) + ar_0)]. \quad (37)$$

This can be further approximated (on expanding the logarithm) by

$$\tau(r, t) \approx \tau(0, t) \left(1 - \frac{r}{r_0}\right). \quad (38)$$

Note that the loop lifetime is then given by

$$\tau = r_0/[f(t) + ar_0]. \quad (39)$$

We turn now to the numerical estimation of the creep rate proper.

### 3.3. Non-transient creep rate

The creep rate can be obtained by using (28). For this we need to evaluate the quantity

$$J = - [(f(t) + ar)/(f(t) + ar_0)] \exp [a(t-t_0)] \quad (40)$$

where we have used (32) and (36) for the two derivatives of  $r$ . Elimination of  $t_0$  gives

$$J = g [f(t) + ar + g]^{-1} - 1 = (g/h) - 1, \quad (41)$$

$$\text{where } g \equiv (1/a) (\partial B/\partial t) [\exp(-a\tau(r, t)) - 1], \quad (42)$$

$$\text{and } h \equiv f(t) + ar + g. \quad (43)$$

We now have all the quantities we need and the creep rate can be calculated by substituting the first derivative of  $J$  with respect to  $\xi$  in (28) and performing the integral. For the numerical estimate we are interested in, the quantity  $(\partial J/\partial \xi)$  appearing within the integral can be brought out and replaced by its average value  $(\partial J/\partial \xi)_{av}$  corresponding to an intermediate value of  $r$ , say,  $r = r' = (\frac{1}{2})r_0$ : this presumes a nearly linear dependence of  $(\partial J/\partial \xi)$  on  $r$  which will be justified later and is consistent with the other approximations to be made. The creep rate is then

$$\dot{\epsilon}_{ss}^d = - (4\pi\rho b r_0^2 \xi_0/45) (\partial J/\partial \xi)_{av}. \quad (44)$$

The problem of calculating  $\dot{\epsilon}_{ss}^d$  reduces to evaluating  $(\partial J/\partial \xi)_{av}$ . Now the numerical values of the parameters involved in  $a$  (see (35)) are as follows:  $D_v C_v^e = \exp[-(E_m + E_f)/k_B T]$  with  $E_m + E_f = 2.9$  eV and  $b = 2$  Å. The vacancy dislocation loop lifetimes have been reported earlier; they are  $\sim 10^7$  sec,  $10^6$  sec and  $10^4$  sec at  $300^\circ\text{C}$ ,  $400^\circ\text{C}$  and  $500^\circ\text{C}$  respectively for a dose of 10 dpa. It follows then that  $|a| \tau \ll 1$ . Utilising this and evaluating an average value for  $g$  at  $r'$ , we find from (42) and (38) that

$$g \approx - (\partial B/\partial t) \tau(r', t) = - (\tau/2) (\partial B/\partial t). \quad (45)$$

In a similar manner we evaluate an average  $(\partial r/\partial t)_{av}$  from (32). Using (38) and (39) we then obtain (the subscript 'av' is to be understood wherever appropriate in the rest of this section)

$$\partial r/\partial t \simeq - [f(t) + ar] \simeq - r'/\tau(r', t) \simeq r_0/\tau. \quad (46)$$

It follows from (33) and (46) that

$$\partial B/\partial t = (\partial/\partial t) [f(t) + ar'] = - (r_0/\tau^2) (\partial\tau/\partial t), \quad (47)$$

$$\text{and } (\partial/\partial \xi) [f(t) + ar'] = D_v C_v^e (q + pr')/b \simeq - (r_0/\tau^2) (\partial\tau/\partial \xi). \quad (48)$$

Taking the derivative of  $J$  with respect to  $\xi$  in (41), setting  $r = r'$  and making use of the above relations, we get

$$\left(\frac{\partial J}{\partial \xi}\right)_{av} = -\frac{1}{2} \left( \frac{1}{h} - \frac{g}{h^2} + \frac{1}{h^2} \frac{r_0}{\tau} \right) \left( \frac{\partial B}{\partial t} \right) \left( \frac{\partial \tau}{\partial \xi} \right). \quad (49)$$

Since all the parameters ( $h$ ,  $g$  and  $\partial B/\partial t$ ) can now be expressed in terms of  $r_0$  or  $\tau$  and its derivatives with respect to  $t$  or  $\xi$ , we can estimate the creep rate. However, a further simplification can be made in the above results. From figure 3 of Krishan and Ray (1975) we obtain the estimate that at 400°C the vacancy loop lifetime  $\tau$  changes from approximately  $10^5$  sec to  $1.6 \times 10^6$  sec in 20 dpa, the transient part being neglected. Assuming a linear variation of  $\tau$  with  $t$  we obtain the estimate

$$\partial\tau/\partial t \approx 8 \times 10^{-2}. \quad (50)$$

It follows from (45) and (46) that  $g \approx (-r_0/2\tau) (\partial\tau/\partial t)$ , while  $f(t) + ar' \approx r_0/\tau$ . Using (50) we may write  $h \approx f(t) + ar' \approx r_0/\tau$ . In a similar manner it can be shown that  $g/h^2 \approx -(1/2h) (\partial\tau/\partial t)$ , and hence may be neglected in comparison to  $(1/h)$ . Making use of these facts in (49) we obtain finally the simple expression

$$(\partial J/\partial \xi)_{av} = (1/\tau) (\partial\tau/\partial t) (\partial\tau/\partial \xi). \quad (51)$$

Therefore the creep rate is given by

$$\dot{\epsilon}_{ss}^d = -(4\pi\rho br_0^2 \xi_0/45) [(1/\tau) (\partial\tau/\partial t) (\partial\tau/\partial \xi)]. \quad (52)$$

Let us now establish our earlier statement regarding the linear dependence of  $(\partial J/\partial \xi)$  on  $r$ . It will be sufficient to show this dependence for  $J$  in (41). It has already been argued that  $g$  can be neglected in the denominator of the first term on the right. In the numerator, it is directly proportional to  $\tau(r, t)$  to the same degree of accuracy as (45). But it has already been shown in (38) that  $\tau(r, t)$  is virtually linear in  $r$ , and this happens to be the dominant  $r$ -dependent term in  $J$ .

We return to the task of numerical estimation. The value of  $(\partial\tau/\partial \xi)$  is obtained from (48). At 400°C,  $D_v C_v^e = 1.1 \times 10^{22} \text{ sec}^{-1}$ ;  $r' = 10 \text{ \AA}$  and  $q + pr' \approx 2.5$ .

$$(\partial\tau/\partial \xi) \approx -8.8 \times 10^4 \text{ sec}, \quad (53)$$

so that  $(\partial J/\partial \xi)_{av} \approx -8.8 \times 10^{-3}$ . (54)

The value of  $\rho$ , the loop production rate for the typical reactor dose rate of  $10^{-6}$  dpa/sec, is about  $3.3 \times 10^{13}$  loops/cm<sup>3</sup> (see Bullough *et al* 1975). The numerical factor outside the square brackets in (52) is  $6 \times 10^{-11} \text{ sec}^{-1}$  for a load of  $10^{-2}$  MNm<sup>-2</sup> (or 1 kg/mm<sup>2</sup>; this is the order of magnitude of the typical load that develops in a fuel pin in a reactor.) At about 10 dpa where  $\tau \approx 8 \times 10^5$  sec, we get a value of  $\dot{\epsilon}_{ss}^d \approx 5 \times 10^{-13} \text{ sec}^{-1}$  at 400°C. Detailed calculations using (28) directly have also been done at other temperatures and show that in the temperature range 400°C to 450°C, creep rates in the range  $10^{-13}$  to  $10^{-12} \text{ sec}^{-1}$  are possible and decrease by an order of magnitude at 300°C and 500°C. Calculations have also been done for cyclotron irradiation at a dose rate of  $10^{-3}$  dpa/sec and show similar features, the creep rate being two orders of magnitude higher than in the reactor case, with a maximum around 500°C.

### 3.4. Temperature, dose rate and dose dependence of the creep rate

Let us first consider the temperature dependence of the creep rate given by (52). The dominant dependence occurs in the terms contained in the square brackets. With

the help of (48) for  $(\partial\tau/\partial\xi)$ , we see that this reduces to analysing the  $T$ -dependence of  $D_v C_v^e \tau (\partial\tau/\partial t)$ . At low temperatures, the Arrhenius-type variation of  $D_v C_v^e$  with  $T$  results in a rapid decrease of the creep rate. On the other hand, at high temperatures  $(\partial\tau/\partial t)$  tends to zero: thermal emission is the dominant mechanism for loop shrinkage, and irradiation has little effect on  $\tau$ ; the lifetime therefore tends to a constant value. This is also evident from the results of computer calculations plotted in figure 3 of Krishan and Ray (1975) and figure 7 of Bullough *et al* (1975). Thus the creep rate is small at both temperature extremes, and has a maximum at some intermediate temperature. This turns out to be in the range 400°C–450°C for the case we have been concerned with in the preceding section.

As was mentioned earlier (at the end of § 3.1), the dose rate dependence arises from two factors  $\rho$  and  $J$  (equivalently,  $\tau$ ). The first ( $\rho$ ) is directly proportional to  $K$  in our model. Regarding the second, Brailsford (1978) has examined the dependence of  $\tau$  on various external parameters under different limiting conditions. The result relevant to our discussion reads, with some changes in notation,

$$\tau = \tau_0 [1 + (K\eta\tau_0/S_I)]^{-1}, \quad (55)$$

where  $\tau_0$  is the ( $T$ -dependent) 'thermal lifetime' of the loops in the absence of irradiation.  $S_I$  is the time-dependent total sink strength for interstitials due to all the defects.  $\eta$  may be regarded as a constant for present purposes. Equations (52) and (55) yield, together with  $\rho = K\epsilon_f/\pi b r_0^2$ ,

$$\dot{\epsilon}_{ss}^d = (4\xi_0 \eta \epsilon_f / 45b\tau_0) D_v C_v^e P(\tau_0/2) (K\tau_0)^3 \cdot [1 + (K\eta\tau_0/S_I)]^{-3} S_I^{-2} [\partial S_I / \partial(Kt)]. \quad (56)$$

The  $K$ -dependence of the creep rate is explicit in this result if we make the plausible assumption\* that the derivative of  $S_I$  with respect to the dose  $Kt$  is itself independent of  $K$ . At high temperatures,  $(K\eta\tau_0/S_I) \ll 1$  because  $\tau_0$  decreases rapidly as  $T$  increases. Hence the creep rate varies as  $K^3$ , although its magnitude will be very small. On the other hand, at low temperatures  $(K\eta\tau_0/S_I) \gg 1$  (see Brailsford 1978), and the creep rate clearly tends to a value independent of  $K$ . At intermediate temperatures (the range in which the creep rate is significant), the dose rate dependence lies between these two extremes.

The variation of the creep rate with dose (or time) can be derived from the dose-dependence of  $\tau(\partial\tau/\partial t)$ . From figure 3 of Krishan and Ray (1975) and figure 7 of Bullough *et al* (1975), we see that at low temperatures ( $<500^\circ\text{C}$ ), the vacancy loop life-time increases linearly with dose, showing no saturation effects. It follows from the dependence of the creep rate on the loop lifetime (and its time derivative) that the non-transient creep rate will also linearly increase with time. This may be further seen from (55) if we consider the low temperature region where  $(K\eta\tau_0/S_I) \gg 1$ , so that  $\tau = (S_I/K\eta)$ , and the creep rate is proportional to  $S_I(\partial S_I / \partial(Kt))$ . As before\*, taking  $\partial S_I / \partial(Kt)$  to be a constant, the time or dose dependence of the creep rate will be the same as that of  $S_I$  which increases nearly linearly with dose, at least in the region of interest to us (till about 20 dpa). The steady state in the vacancy loop life-time does not appear at any value of dose at low temperatures. This feature, which is

\*Note that this is in keeping with the spirit in which dose is introduced as a variable replacing time, although, strictly speaking,  $\partial S_I / \partial(Kt)$  may have a weak dependence on  $K$ .

evident from (55), is discussed in greater detail by Krishan and Nandedkar (1979). In contrast, Lewthwaite (1976), Heald and Speight (1977), Weiner and Boltax (1977), and Boltax *et al* (1977) have all used a steady state condition which assumes  $\tau$  to be independent of time in the temperature range below 500°C. We have argued that this is incorrect, so that their conclusion that vacancy loops will not contribute to non-transient creep is not valid.

#### 4. Concluding remarks

We have developed a simple methodology for the calculation of the creep rate in a material arising from the kinetics of the defects in it. The result is expressed in terms of two quantities: the rate of production of the defect type concerned, and a probability distribution function characterising the time evolution of the pertinent physical property of the defect. In many instances, the assumption of a sharply-peaked distribution suffices to yield a satisfactory calculational scheme, besides providing a proper interpretation for the variables occurring in standard rate theory for the process under study.

We have applied the formalism to the evaluation of the contribution to irradiation-induced creep from a specific mechanism, namely, the shrinkage of vacancy dislocation loops at rates that depend on their orientation with respect to the direction of the applied stress. Besides obtaining numerical estimates to establish that the mechanism does lead to observable effects, we have pointed out also the precise circumstances under which the shrinkage of loops leads to *non-transient* creep, and derived a convenient expression for this component. Two other aspects of the evolution of the loops (also pointed out by Lewthwaite 1976) may be mentioned here, as we have not taken them into account in the present work. The first is the possibility of the nucleation of loops in preferred orientations in the external stress field. As already stated in Krishan and Ray (1975), this is rather unlikely for vacancy dislocation loops (in contrast to the case of interstitial dislocation loops): these loops nucleate suddenly and athermally in the vacancy-rich core regions of cascade damage. Further, the large local stresses produced by the interstitials occupying the periphery of the cascade region should effectively mask the applied stress and preclude any preferred direction of *nucleation*. Hence the effect of preferential nucleation may be neglected. The second aspect is the operation of the Heald-Speight-Wolfer mechanism: this is the stress-induced attraction exercised by dislocations on interstitial atoms, and leads to an orientation ( $\xi$ ) dependence in the function  $B(t)$  occurring in the rate equation for  $\partial r/\partial t$ . We remark that there is no difficulty at all in modifying our derivation to include such an additional dependence. It is similarly possible, too, to formally incorporate preferential nucleation of loops, although we have argued that this effect would be unimportant.

The application of our methodology to other mechanisms of irradiation-induced creep (for example, those listed in Gilbert 1971) will be taken up elsewhere.

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**References**

- Boltax A, Foster J P, Weiner R A and Biancherra A 1977 *J. Nucl. Mater.* **65** 1974
- Brailsford A D 1978 *J. Nucl. Mater.* **71** 227
- Bullough R 1975 in *Proc. Gatlinburg Conf. on Fundamental effects of radiation damage in metals* ed. M T Robinson and F W Young Jr. (Springfield, Va: US Department of Commerce)
- Bullough R, Eyre B L and Krishan K 1975 *Proc. R. Soc. London* **A346** 81
- Cundy M R, Von der Hardt P and Loelgen R H (eds) 1977 *J. Nucl. Mater.* **65** 157 *Proc. Conf. on Measurement of irradiation-enhanced creep in nuclear materials* (Petten; The Netherlands, May 1976)
- Gilbert E R 1971 *Reactor Tech.* **14** 258
- Gittus J 1975 *Creep, viscoelasticity, and creep fracture in solids* (London: Applied Science)
- Harries D R 1977 *J. Nucl. Mater.* **65** 157
- Heald P T and Speight M V 1974 *Philos. Mag.* **29** 1075
- Heald P T and Speight M V 1977 *J. Nucl. Mater.* **64** 139
- Hesketh R V 1972 in *Proc. BNES Conf. on Irradiation embrittlement and creep in fuel cladding and core components* (London) p 221
- Krishan K and Ray S K 1975 *J. Nucl. Mater.* **58** 285
- Krishan K and Nandedkar R V 1979 *Pramana* (to be published)
- Lewthwaite G W 1976 *J. Nucl. Mater.* **61** 313
- Piercy G R 1968 *J. Nucl. Mater.* **26** 18
- Weiner R A 1976 (private communication)
- Weiner R A and Boltax A 1977 *J. Nucl. Mater.* **66** 1
- Welch D O and Smoluchowski R 1972 *J. Phys. Chem. Solids* **33** 1115
- Wolfer W G and Ashkin M 1976 *J. Appl. Phys.* **47** 791
- Venkataraman G and Balakrishnan V 1977 in Indo-German Seminar on Radiation Damage (Kalpakkam: Reactor Research Centre, Department of Atomic Energy)