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HIGH TEMPERATURE FAILURE MECHANISMS
IN CERAMICS

by

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ABSTRACT

A statistical model of high temperature failure in ceramics, by the viscous or diffusive growth of cavities, has been developed. General expressions for the creep strain associated with cavitation and for the failure time have been derived, and shown to be in good agreement with available data. Specific models of cavity growth along grain facets, by diffusive and viscous mechanisms, have then been generated, and coupled with the statistical model: to provide specific predictions of the creep strain and the failure time. The predictions are in reasonable accord with experimental results.

HIGH TEMPERATURE FAILURE MECHANISMS IN CERAMICS

1. INTRODUCTION

There are adequate observations^{1,2} (Fig. 1) to indicate that high temperature fracture in ceramics frequently involves the nucleation, growth and coalescence of grain boundary located microcracks or cavities. Close scrutiny (Fig. 1) indicates that cavities (i.e., non-negligible surface separations) are more prevalent than microcracks*: implying that the fracture process has either anelastic or viscoelastic characteristics. The intent of the present paper is to examine models of cavity growth in ceramic polycrystals as a basis for both the definition of critical experiments and the elucidation of the material parameters (or microstructural characteristics) that influence the creep and failure processes.

Models of high temperature failure in ceramics must recognize the various microstructures that typically occur, particularly the presence and dispersion of second phases (amorphous or crystalline) at grain boundaries. Three types of microstructure, and the concomitant modes of cavity growth, will be distinguished in the present paper.

The first, but least common, microstructure is an equiaxed grain

*A temperature/stress regime may exist where microcracks form and propagate, due to a grain boundary sliding process. This regime is confined to situations where sliding is not controlled by diffusion (otherwise the diffusive flux would also modify the crack tip stress field and generate a crack-like cavity) and where boundary phases exhibit a high viscosity (low viscosity boundary phases would again yield cavities by crack tip relaxation). The microcracking regime is not, therefore, considered to be sufficiently extensive to merit specific consideration as a cavity propagation mechanism. The microcracking mechanism will be the subject of a subsequent paper.

structure with no second phase at grain boundaries (i.e., a single phase material). For this case, cavities are presumed to nucleate primarily near grain triple points³--a consequence of local stress concentrations developed due to boundary sliding*--and then to propagate along grain facets, primarily by atom migration along grain boundaries and along the cavity surface (Fig. 2a).

The second microstructure consists of grains completely encompassed by a very thin (~ 5 -100 Å thick) amorphous second phase, typical of many ceramics (especially covalent materials) fabricated by hot pressing or sintering⁴. (This condition arises of course when the dihedral angle between the amorphous and crystalline phases is small.) Again cavity nucleation is considered to occur primarily near triple points, and to extend along grain interfaces (Fig. 2b). However, the cavity growth in this instance is expected to be strongly influenced by the presence of the viscous amorphous phase: involving viscous deformation, solution/precipitation and atom migration.

For the third microstructure, the amorphous phase is considered to be confined to channels along three grain junctions or to be isolated at four grain junctions. In this instance, the cavities extend along grain boundaries with the amorphous phase contained near the cavity tip (Fig. 2c). The amorphous phase participates in the cavity extension by providing a rapid atom migration path and by modifying the

* Where specific cavity growth models are used in this paper, the time constant for grain boundary sliding is considered to be smaller than the time constants for the other deformation processes. This behavior is expected to be typical of nearly-planar boundaries without large ledges. Alternate models could be developed for conditions under which boundary sliding is the rate limiting process.

equilibrium condition at the cavity tip.

Useful high temperature failure models for ceramics, based on the above microstructures, must recognize the wide distribution of grain sizes that exist in a given material, and the anisotropy of the surface energies, as well as the range of morphologies, dispersions and dimensions of the second phase. These effects lead to preferred locations for cavity nucleation and growth; requiring these phenomena to be treated as heterogeneous or statistical. Pertinent models should therefore consider the successive growth of cavities of different size at adjacent or remote locations in the material. These cavities ultimately coalesce to generate the crack that produces failure (Fig. 3). A generalized statistical model of this type of process can be constructed without specific prior knowledge of the cavity growth mechanism, leading to expressions for the two important time dependent properties: the creep strain (due to cavitation) and the failure time. Some general relations (for the creep strain and the failure time) are deduced in the first part of the paper.

The scale parameters invoked in the statistical models are dependent upon the specific mechanisms of cavity nucleation and propagation. The second part of the paper is thus concerned with the development of cavity growth relations for each of the three principal microstructures. Then, in combination with the statistical results, the influence of the primary microstructure-controlled parameters (on the creep strain and failure time) can be deduced for each microstructure. The resultant predictions are discussed in the final section of this paper.

2. STATISTICAL MODELS

The present statistical models are developed in accord with the following sequence of events (Fig. 3, 4). Cavities continually evolve in the material by nucleation (where required) and growth (Fig. 3). The cavities propagate across grain facets, to become facet-sized cavities, and then stabilize (Fig. 4). Where cavities form on contiguous boundaries, coalescence will occur (Fig. 3). This process will continue until sufficient contiguous boundaries have cavitated that a macrocrack develops which is capable of self-extending to failure (Fig. 3). Thereafter, conventional approaches concerned with the slow growth of macrocracks can be used to predict the remaining life^{5,6}. A critical feature of the models is thus the stabilization of facet-sized cavities. This assumption is consistent with observation (Fig. 1) and will be rationalized in Section 3.

An important parameter in this approach to cavitation is the time t_p taken for a cavity to propagate across a grain facet. This time is statistically distributed because of the variability of grain boundaries with regard to second phase content and composition, boundary energy, diffusivity, etc. It is required for present purposes that a distribution function be assigned to this propagation time; namely, a function that can be expected to accurately describe the grain boundary variability typical of ceramics. This decision is facilitated by noting that only a small fraction of the boundaries usually cavitate prior to failure: these being the boundaries with the greatest cavitation

susceptibility. When such conditions pertain,* a distribution that characterizes the large extreme of cavitation susceptibilities should apply. There are only three extreme value distributions⁷ and therefore the choice of functions can be considerably restricted by applying the extreme value condition. Initially, a distribution based on the second type of extreme value distribution is selected, because this distribution has previously been found to describe extreme value characteristics of ceramic microstructures.⁸ An additional rationale for the chosen distribution is provided in the following section. The assumed cumulative probability $\Phi(t_p)$ of observing facet sized cavities at time t_p is,

$$\Phi(t_p) = 1 - \exp \left[- \left(\frac{t_p}{t_o} \right)^k \right] \quad (1)$$

and for $t_p < t_o$,

$$\Phi(t_p) = (t_p/t_o)^k$$

where k is the shape parameter and t_o is the scale parameter. The scale parameter t_o will depend on the specific mechanism of cavity growth, and will be a function of the stress level, and of viscosity (diffusivity), boundary energy, etc. Appropriate relations for t_o will be derived in the following section, and inserted into the general relations derived below.

The number N of cavitated boundaries at time t , derived from Eqn. (1) is,

*At very low stresses and very high temperatures the fraction of boundaries that cavitate can be very large (e.g., Fig. 1a). Then, the underlying distribution function must be used instead of the extreme value distribution.

$$N = \rho A_b [1 - \exp(-(t/t_o)^k)] \quad (2)$$

where ρ is the number of available sites per unit area of boundary and A_b is the boundary area. Therefore, for a material with a grain diameter, d ,

$$\rho A_b \sim \frac{6V}{\pi \langle d^3 \rangle} \quad (3)$$

where V is the sample volume; and hence,

$$N = \frac{6V}{\pi \langle d^3 \rangle} \left[1 - \exp \left(- \left(\frac{t}{t_o} \right)^k \right) \right] \quad (4)$$

or,

$$N = \frac{6V}{\pi \langle d^3 \rangle} \left(\frac{t}{t_o} \right)^k \quad (5)$$

The rate of cavity development, obtained by differentiation ($t \ll t_o$) of Eqn. (5), is thus,

$$\frac{dN}{dt} = \frac{6kVt^{k-1}}{\pi \langle d^3 \rangle t_o^k} \quad (6)$$

The time dependence of the number of cavitated grain boundaries now becomes an input to expressions for the creep strain and for the failure time.

2.1 The Creep Strain

The external strain ϵ_∞ associated with the presence of a grain boundary cavity in a polycrystalline body, subjected to an applied stress σ_∞ , depends on the state of stress relaxation around the cavity.

We will consider that the strain that derives from non-interacting* cavities is approximately proportional to that associated with a crack distribution of similar size. This approximation is most pertinent when the cavity density is low and hence, applies in the early stages of cavitation creep. The strain** in a body containing N circular cracks of radii a, is⁹

$$\epsilon_{\infty} = \frac{\sigma_{\infty}}{E} \left[1 - \frac{16}{45} \frac{(1-\nu^2)(10-3\nu)}{(2-\nu)} NV\langle a^3 \rangle \right]^{-1}$$

$$\frac{\sigma_{\infty}}{E} \left[1 - \xi \frac{N}{V} \langle a^3 \rangle \right]^{-1} \quad (7)$$

where E is Young's modulus and ν is the effective Poisson's ratio. Since the strain varies rapidly with a, i.e., as a^3 , each cavity only contributes effectively to the measured strain as it approaches its fully-developed, facet-sized condition. The crack diameter, 2a in Eqn. (7) may thus be approximately replaced by the facet size ℓ . The strain-rate at constant stress can now be obtained by differentiation of Eqn. (7). For small strains ($Na^3/V \ll 1$), we obtain

$$\dot{\epsilon}_{\infty} = \xi \sigma_{\infty} \langle \ell^3 \rangle \dot{N} / 8EV \quad (8)$$

This result applies if the average size of the cavitate boundaries does not vary with time. Substituting for $\dot{N}$ from Eqn. (6) the strain-

* Non-interacting in the present context refers to non-interacting zones of stress relaxation (see Fig. 10) around individual cavities--see section 4.1 for further discussion.

** The elastic strain can be regarded as being converted into anelastic or viscoelastic strain by mass transport in the zones near the cavity tips (see section 4.1).

rate becomes;

$$\dot{\epsilon}_{\infty} = 3\sigma_{\infty}(\xi\beta k) t^{k-1}/4t_o^k E \quad (9)$$

where β is a constant relating $\langle d^3 \rangle$ to $\langle \ell^3 \rangle$.

It will be established in the following sections that all cavity growth mechanisms yield values of t_o that are stress dependent and proportional to an Arrhenius factor; such that,

$$t_o = \zeta \sigma_{\infty}^{-n} \exp(Q/RT) \quad (10)$$

where Q is the activation energy for the cavity growth process, RT has its usual meaning, n is the exponent of the stress dependence ($1 \leq n \leq 3$) and ζ is the parameter that contains the remaining cavity growth variables. Inserting t_o from eqn. (10) into eqn. (9) gives,

$$\dot{\epsilon}_{\infty} = \left[3\sigma_{\infty}^{(kn+1)} (\xi\beta k) t^{k-1}/4\zeta^k E \right] \exp(-kQ/RT) \quad (11)$$

This result reveals the possibility that the apparent activation energy Q_* will differ from that for the cavity growth mechanism by an amount that depends on the magnitude of k , i.e.;

$$Q_* = kQ \quad (12)$$

The time dependence incorporates the magnitude of the scale parameter k : a parameter that must be obtained empirically from observations of the cavity density as a function of time. It is interesting to note that steady state conditions can obtain if $k \sim 1$. The stress dependence is also affected by k and, in general, a non-linear stress dependence can be anticipated. Non-linear creep-strain rates are typical of materials that develop cavities.^{1,10} Specific results are discussed in section 4.

It is re-emphasized that the above results can only be expected to apply at relatively small cavity densities and small creep strains. For larger strains where the relaxation fields around neighboring cavities appreciably overlap (see Fig. 10), much of the external strain derives from the average 'thickening' or separation that occurs at grain boundaries. The best analytic estimates of the strain might then be obtained from solutions for the symmetric growth of cavity arrays,¹¹ as evaluated for each specific cavity growth mechanism.

2.2 The Failure Time

The failure time under creep conditions can be considered to comprise of two components: the time t_1 taken to develop a macrocrack, through the coalescence of grain-facet sized cavities, followed by the time t_m taken for the macrocrack to propagate to a critical size. The latter is treated in the conventional manner by using crack growth rate $\dot{c}$, stress intensity factor K relations pertinent to diffusive growth.⁶ The former which involves statistical considerations, is examined in the following analysis. The time t_1 is often expected to be the major contribution to the total failure time, particularly at low stress levels. The relations derived for t_1 can thus be used, in many cases, as approximate estimates of the total failure time.

The development of a statistical model for the failure time requires that a critical cavity size, c , be defined at which a coalesced array of cavities constitutes a macrocrack. This is, in general, a rather nebulous concept, because the critical size depends on the microstructure, and on the size of the crack tip 'process zone' (i.e., the enhanced cavitation zone created by the crack). The latter depends,

in turn, on the level of the applied stress or, more likely, on the stress intensity factor K . The simplest approach capable of yielding useful results is to assume that a macrocrack has developed when K attains a specified level, viz, when K reaches a certain fraction f of the critical stress intensity factor K_c . The critical size c for a penny-shaped crack can then be written as;

$$c = \pi f^2 K_c^2 / 4 \sigma_\infty^2 \quad (13)$$

The formation of a microcrack of size c will be determined by statistical considerations, through the distribution of times, t_p , for the development of facet-length cavities (eqn. (1)). Analysis of this process can be conducted without resorting to numerical methods if it is assumed that t_p is not appreciably influenced by the prior existence of cavities on adjacent facets, i.e., if interaction effects are neglected. Then, the probability P of forming contiguous facet-sized cavities of sufficient extent to produce a macrocrack of length c (Fig. 3) can be obtained from McClintock's result,¹²

$$P = \frac{A_T^{1/2} \ell_{np}}{\ell^2 (0.5 \ell_{np} - 1)} \exp[(c/\ell) \ell_{np}] \quad (14)$$

where p is the probability that a given facet has cavitated at time t and A_T is the total grain boundary area subjected to the stress σ_∞ . Noting that p is just the cumulative probability of cavity development, then for small p (the case of present interest) combining eqns. (1), (13) and (14) gives the probability $P(t_i)$ of macrocrack formation at time t_i as;

$$P(t_i) = \left(\frac{2A_T}{\ell^2} \right) (t_i/t_o)^{k[0.5 + \pi f^2 K_c^2 / 4 \sigma_\infty^2 \ell]} \quad (15)$$

At a specific probability level, e.g., the median level ($P = 0.5$), the macrocrack incubation time can be obtained directly from eqn. (15) as;

$$\ln(t_i/t_o) = \frac{\ln(\ell^2/4A_T)}{k(0.5 + \pi f^2 K_c^2 / 4\sigma_\infty^2 \ell)} \quad (16)$$

For most conditions of interest in ceramics $K_c^2 \gg \sigma_\infty^2 \ell$, whereupon eqn. (16) reduces to;

$$\ln t_i = \ln t_o - \frac{4\sigma_\infty^2 \ell}{\pi k f^2 K_c^2} \ln(4A_T/\ell^2) \quad (17)$$

The t_o term again depends on the specific cavity extension mechanism.

However, invoking the general requirement that t_o be proportional to an Arrhenius and a stress term (eqn. 10), eqn. (17) can be written as;

$$\ln[t_i \exp(Q/RT)] = \ln \zeta - n \ln \sigma_\infty - \sigma_\infty^2 (4\ell/\pi k f^2 K_c^2) \ln(4A_T/\ell^2) \quad (18a)$$

or

$$\ln \theta_r = A - n \ln \sigma_\infty - C \sigma_\infty^2 \quad (18b)$$

where θ_r is the Orr-Sherby-Dorn rupture parameter,¹³ C is a parameter that depends on the grain size and toughness and, marginally, on the sample size (through A_T/ℓ^2), and A and n are constants that depend on the details of the cavity propagation process.

The appearance of θ_r is satisfying because it implies that all of the temperature effects, correlated by other investigators¹⁴ through this parameter, will also apply to the present model. The general shape of the failure time, stress curve predicted by eqn. (18b) is plotted in Fig. 5; the failure time is observed to exhibit the asymptotic

behavior at both small and large times. The general utility of the predicted failure time relation can be explored by plotting the logarithm of the rupture parameter θ_r as a function of σ_∞^2 , and treating A, n and C as adjustable parameters (with n being confined to the range, $1 \leq n \leq 3$). The results of Walles¹⁵ on Al_2O_3 and SiC fibers (the only comprehensive data presently available) taken from the correlation developed by Charles¹³ are plotted in Fig. 6a, b. The correlation is very good. Such a correlation does not, of course, substantiate the validity of the model, because alternate models^{14,15} can provide correlations of nearly equal quality. It does, however, permit the model to be regarded as a serious candidate, and encourages further development. Detailed tests of the model will emerge from correlations of the parameters A, C and n with actual material properties (e.g., K_c , ℓ , n) pertinent to the relevant cavity growth mechanism. Further discussion of this issue will be continued in a subsequent section.

3. CAVITY GROWTH MODELS

Most ceramic materials contain pre-existing voids (located primarily at triple points), even in nominally fully-dense materials produced by hot-pressing. For present purposes, it is thus assumed that the cavitation time t_p for a grain facet is simply the time taken to propagate a small pre-existing triple point void across the facet. In instances where there are no pre-existing voids, a nucleation stress must be exceeded; this may modify the following results, primarily by introducing a threshold stress.

The models to be developed involve the growth of cavities by mass transport (viscous flow or diffusion) with the material displaced from the cavity being accommodated on the cavitating facet and on the neigh-

boring facets (Fig. 4). This accommodation can be achieved in the presence of freely sliding grain boundaries by relaxing the stress in the adjoining grains, as depicted in Fig. 4. Such an accommodation mechanism only permits small 'thickening' displacements and small cavity surface separations. It thus has merit under conditions which encourage crack-like cavity formation. This condition seems to be prevalent in ceramic materials, as noted earlier. Note, however, that substantial additional accommodation can occur if the material is undergoing a general diffusive creep strain, upon which the cavitation creep is superimposed.

An approach of this type has been adopted because it permits simple models to be developed for the formation of isolated boundary cavities. The models involve mass transport on highly susceptible boundaries, in a manner consistent with the grain structure of the material, without requiring cavitation along the more resistant neighboring boundaries. The approach is thus entirely compatible with the statistical models developed in the previous section, which invoke the individual development of facet-sized cavities. Usually, it will not be sufficient that only one susceptible boundary be involved, because the displaced matter deposited on the boundary will stabilize the cavity before it extends across the facet. Although this situation may obtain in certain cases, a more favorable situation for cavitation pertains when two contiguous boundaries with comparable mass transport susceptibilities are involved (Fig. 4). Then the displaced material can be accommodated along the non-cavitating facet. This is the situation emphasized in the present analysis. However, it is recognized that the probability p^* of finding two susceptible boundaries

at contiguous locations is much smaller than the underlying probability distribution, p (i.e., $P \propto p^2$). In fact, there must be a coupling between the statistical aspects discussed in section 2 and the cavity propagation models to be presented below. These effects are neglected in the present analysis, in order to provide simple solutions suitable for preliminary interpretation of the creep strain and failure time results. More complex models that take full account of all neighboring boundaries can be developed later, using numerical techniques.

3.1 Continuous Second Phase

In a material containing a continuous second phase, cavitation is expected to occur primarily within the second phase by viscous deformation.* We will consider in the present analysis that the ease of boundary sliding permits the deposition of material by viscous flow to occur along the cavitating boundaries with spatial uniformity, so that the boundary thickening is independent of location along the boundary (Fig. 7). When the second phase completely wets the major phase, the stress σ_{tip} at the cavity tip is simply related to the grain separation 2δ by;

$$\sigma_{tip} = 2\gamma_{lv}/\delta \quad (19)$$

where γ_{lv} is the surface energy of the viscous phase. The average velocity of motion v_l of a viscous phase at a location x along a boundary of uniform separation is, at steady state;¹⁶

$$v_l = - \frac{\delta^2}{3\eta} \left(\frac{d\sigma}{dx} \right) \quad (20)$$

*The incidence of solution/reprecipitation can introduce a second mode of cavity extension, concurrent with viscous separation. It involves the net migration of atoms from the cavitating boundary, by surface diffusion in series with diffusion through the viscous phase. No specific attempts are made to derive models for this process.

where η is the viscosity of the viscous phase and σ is the stress at location x . This is a two-dimensional result which is used, at this stage, to maintain simplicity. Conservation of matter requires that;

$$\frac{\delta dv_{\ell}}{dx} = \frac{d\delta}{dt} \quad (21)$$

By combining eqns. (20) and (21), the governing differential equation is obtained as;

$$\frac{d^2 \sigma(x)}{dx^2} + \frac{3\eta}{\delta^3} \frac{d\delta}{dt} = 0 \quad (22)$$

The boundary conditions pertinent to the present problem (Figs. 4, 8) are, the stress at the cavity tip (eqn. 19), and the zero flux ($d\sigma/dx = 0$) at the triple point remote from the cavity. Hence for a cavity of length a (Fig. 4)

$$\sigma(x) = \frac{1}{\delta} \left[2\gamma_{\ell v} + \frac{3\eta \dot{\delta} x}{2\delta^2} (4\ell - 2a - x) \right] \quad (0 < x < 2\ell) \quad (23)$$

$$\sigma(x) \approx 0 \quad (-a < x < 0)$$

The average stress over the separating facets in the direction of grain motion (Fig. 4) is thus;

$$\begin{aligned} \langle \sigma \rangle &= \frac{(3/4)}{2\ell} \int_0^{2\ell-a} \sigma(x) dx \\ &= \frac{3(2-a/\ell)}{4\delta} \left[\gamma_{\ell v} + \eta \frac{\dot{\delta} \ell^2}{2\delta^2} (2-a/\ell)^2 \right] \end{aligned} \quad (24)$$

This average stress can now be equated to the stress that acts on the grains adjoining the cavitating boundary. The stress acting on these

grains is the applied stress, as relaxed by the boundary separation needed to accomodate the cavity growth. The relaxation occurs over a length approximately equal to the length of the adjacent sliding boundaries, ℓ , in the relaxing grain (Fig. 4). The relaxed stress σ_R is thus of the order;

$$\sigma_R \approx \sigma_\infty - 2E(\delta - \delta_0)/\ell \quad (25)$$

where $2\delta_0$ is the initial thickness of the viscous phase. However, the volume of the viscous phase must be conserved at the separating boundaries. The separation δ is thus related to the initial thickness by;

$$\delta = 2\delta_0/(2-a/\ell) \quad (26)$$

The relaxed stress is thus;

$$\sigma_R = \sigma_\infty - \frac{2E\delta_0 a}{\ell^2(2-a/\ell)} \quad (27)$$

and the separation rate $\dot{\delta}$ is

$$\dot{\delta} = \frac{2(\delta_0/\ell)\dot{a}}{(2-a/\ell)^2} \quad (28)$$

Equating σ_R in eqn. (27) to $\langle \sigma \rangle$ in eqn (24) and substituting for $\dot{\delta}$ from eqn. (28) and δ from eqn. (26), we obtain an expression for the cavity growth rate;

$$\dot{a} = \frac{32\delta_0^2}{3\eta(2-a/\ell)^4 \ell} \left[\sigma_\infty - \frac{2E\delta_0 a}{\ell^2(2-a/\ell)} - \frac{3(2-a/\ell)^2 \gamma_{lv}}{4\delta_0} \right] \quad (29)$$

The important features of eqn. (29) are the requirement that the applied stress exceed $3\gamma_{lv}/2\delta_0$ in order for cavitation to proceed,

the inverse dependence of the growth rate on the viscosity, and the tendency for the growth rate to increase at small a/ℓ but to decrease at large a/ℓ (i.e., as $a/\ell \rightarrow 2$).

The materials of greatest practical interest are those with a reasonable strength: materials for which δ_o/ℓ is very small (e.g., $\lesssim 10^{-3}$). For such materials, provided that the stress exceeds $\sim 2E (\delta_o/\ell)$, the cavity growth will continue to accelerate to a length approaching one facet length, ℓ , and then quickly stabilize. Under these conditions, the stress relaxation is negligible and an approximate time t_p for the formation of facet-sized cavities can be obtained from eqn. (29) as

$$t_p \approx \frac{3\eta\ell}{32\delta_o} \int_0^\ell \frac{(2-a/\ell)^4 da}{4\sigma_\infty\delta_o - 3\gamma_{lv}(2-a/\ell)^2} \quad (30)$$

Integration of eqn. (30) in the form of a series gives;

$$t_p = \frac{3\eta\ell^2}{128\sigma_\infty\delta_o^2} \left[\frac{31}{5} + \frac{127}{14} \left(\frac{\sigma_o}{\sigma_\infty} \right) + \frac{511}{36} \left(\frac{\sigma_o}{\sigma_\infty} \right)^2 + \frac{2031}{88} \left(\frac{\sigma_o}{\sigma_\infty} \right)^3 + \dots \right] \quad (31)$$

where σ_o is the threshold stress for cavity growth ($\equiv 3\gamma_{lv}/2\delta_o$).

Generally, the stress levels of interest are significantly in excess of σ_o . For example, if $\sigma_\infty > 3\sigma_o$, the series in eqn. (21) converges rapidly. The solution for small σ_o/σ_∞ is thus a useful one for examining trends in the propagation time;

$$t_p \approx 0.15(\eta/\sigma_\infty)(\ell/\delta_o)^2 \\ \equiv 0.15(\eta_o/\sigma_\infty)(\ell/\delta_o)^2 \exp(Q_\eta/RT) \quad (32)$$

where η_0 is a constant and Q_η is the activation energy for viscous flow. This solution for t_p will be used in conjunction with the statistical results to provide preliminary expressions for the creep strain and the failure time in the following section. More detailed results will require that both the stress relaxation and the behavior at stress levels close to the threshold be evaluated. This will be the subject of subsequent publications.

Finally, we note that since the cavity propagation rate depends strongly on the crack length a/ℓ , most of the time t_p is expended while the cavity is small. The time t_p may thus be regarded as a time when the influence of the cavity is first established, i.e., for $t \lesssim t_p$ the cavity is too small to appreciably influence the creep strain or the failure sequence.

3.2 Single phase materials

A similar approach to that described above will be used to obtain propagation times for single phase materials. In this instance, the cavity growth will be dictated by the diffusion of atoms along the cavity surface and through the grain boundary.¹⁷ For convenience, the same uniform grain relaxation condition will be used, so that the boundary 'thickening' δ will be independent of location x along the boundaries subject to diffusive flow. Then, the governing differential equation for the atom migration along the boundary is;^{17,18}

$$\frac{d^2\sigma(x)}{dx^2} + \frac{kT\dot{\delta}}{\Omega D_b \delta_b} = 0 \quad (33)$$

Superimposing the boundary conditions, $d\sigma/dx = 0$ at $x = 2\ell - a$, and

$\sigma = \sigma_{\text{tip}}$ at $x = 0$, we obtain;

$$\sigma(x) = \sigma_{\text{tip}} + \frac{kT\dot{\sigma}x}{2\Omega D_b \delta_b} (4\ell - 2a - x) \quad (0 < x < 2\ell) \quad (33a)$$

and

$$\langle \sigma \rangle = \frac{3(2\ell - a)}{8} \left[\sigma_{\text{tip}} + \frac{kT\dot{\sigma}(2\ell - a)^3}{3\Omega D_b \delta_b} \right] \quad (33b)$$

where $D_b \delta_b$ is the boundary diffusion parameter and Ω is the atomic volume.

The tip stress σ_{tip} depends on the mode of cavity growth: equilibrium or non-equilibrium.^{17,18} For the problem of present interest, very little cavity growth could occur in the equilibrium mode because the relatively large volume of material displaced from the cavity would quickly relax the stress at the cavitating boundary. We will thus consider conditions under which the cavity growth is dominated by the non-equilibrium mode. A different approach would be required for situations in which the equilibrium mode prevails.

The non-equilibrium mode dominates when the surface diffusivity is appreciably slower than the boundary diffusivity: a behavior which could occur in certain ceramics.¹³ The tip stress is,¹⁸

$$\sigma_{\text{tip}} = (2 - \gamma_b / \gamma_s)^{1/2} a^{1/3} \left(\frac{kT \gamma_s^2}{\Omega^{4/3} D_s} \right)^{1/3} \quad (34)$$

where γ_b is the boundary energy, γ_s the surface energy, and D_s the surface diffusivity. The cavity width w is,^{18,19}

$$w = \gamma_s (2 - \gamma_b / \gamma_s) / \sigma_{\text{tip}} \quad (35)$$

and hence, the boundary thickening required to accomodate the displaced atoms is;

$$\begin{aligned} \delta &\approx wa / (2\ell - a) \\ &\equiv \gamma_s (2 - \gamma_b / \gamma_s) a / (2\ell - a) \sigma_{\text{tip}} \end{aligned} \quad (36)$$

and the thickening rate is;

$$\dot{\delta} = \frac{2\ell (2 - \gamma_b / \gamma_s)}{(2\ell - a)^2 \sigma_{\text{tip}}} \dot{\gamma}_s \quad (37)$$

Inserting δ from eqn. (37) and σ_{tip} from eqn. (34) into eqn. (33b) and equating $\langle \sigma \rangle$ to σ_R from eqn. (27), an expression for the cavity growth can be obtained as;

$$\begin{aligned} \dot{a}^{2/3} &\left[\frac{\ell^3 (2 - a/\ell)}{2} \left(\frac{(kT)^{1/3}}{D_b \delta_b \gamma_b^{1/3} \Omega^{5/9}} \right) \right] + \frac{3}{8} \dot{a}^{1/3} \\ &- \left[\frac{\sigma_\infty (2 - a/\ell) - 2E(\delta_o/\ell)(a/\ell)}{(2 - a/\ell)^2 (2 - \gamma_b / \gamma_s)^{1/2}} \right] \left[\left(\frac{D_s}{kT} \right)^{1/3} \frac{\Omega^{4/9}}{\gamma_s^{2/3}} \right] \end{aligned} \quad (38)$$

This very complex expression becomes tractable only under the condition that the surface diffusivity is very low, so that narrow cavities develop. Then, eqn. (38) reduces to

$$\dot{a} = \frac{19\sigma_\infty^3}{\ell^3 \gamma_s^2 (2 - a/\ell)^3} \left(\frac{D_s}{kT} \right) \frac{\Omega^{4/3}}{(2 - \gamma_b / \gamma_s)^{3/2}} \quad (39)$$

The equivalent expression for the propagation time is;

$$t_p = \frac{0.2\ell^3 \gamma_s^2 (2-\gamma_b/\gamma_s)^{3/2}}{\sigma_\infty^3 \Omega^{4/3} (D_s/kT)}$$

$$\equiv 0.2\ell^3 (\gamma_s^2/\sigma_\infty^3) [(2-\gamma_b/\gamma_s)^{3/2}/\Omega^{4/3} D_o] kT \exp[Q_s/dT] \quad (40)$$

where D_o is the diffusion content and Q_s is the activation energy for surface diffusion.

3.3 Isolated second phase

When a material contains an isolated second phase, the cavity will tend to propagate along grain boundaries of the major phase, but with the second phase contained in a region near the cavity tip (Fig. 2c). The presence of the viscous phase has several important influences; the force equilibrium at the cavity tip is now dictated by solid, liquid surface energies: the atom migration rate within the cavity is enhanced in the region of the liquid; the pressure within the liquid can modify the chemical potential gradient. These influences can be complex, and are examined in a separate publication. An approximate measure of the cavity growth rate can be obtained in situations where the liquid cavitates (Fig. 8), such that the pressure in the liquid remains at approximately zero. Then, it is simply required that γ_s in eqn. (40) be replaced by $\gamma_{s\ell}$ (the surface energy in the presence of the liquid) and $D_s \Omega^{1/3}$ by $D_\ell c_\ell \xi$ (where D_ℓ is the diffusivity of the solid in the liquid, c_ℓ is the concentration of the solid in the liquid and ξ is the average liquid film thickness on the cavity surfaces),²⁰ to give;

$$t_p = \frac{0.2 \ell^3 \gamma_{sl}^2 (2 - \gamma_b / \gamma_{sl})^{3/2}}{\sigma_\infty^3 \Omega (D_\ell c_\ell \xi / kT)}$$

4. DISCUSSION

4.1 Creep strain

The derivation of statistical relations for the creep strain requires that the assumptions of the statistical model and the cavity propagation model be consistent. Further discussion of the models is needed to establish pertinent consistency requirements. The present statistical model was developed by using the elastic relation for a body containing cracks to obtain the component of strain that can be attributed to the cavities. The strain thus derives from the elastic opening of the cracks, and evidently introduces the usual zones of large stress concentration near the crack tips (Fig. 9). Conversely, the cavity propagation times were obtained for the uniform separation of the boundaries: with relaxations, (accommodated by the contiguous sliding boundaries), being confined to the adjoining grains. Thus, the cavity opening displacement is not transferred to the external surfaces. An elastic opening (Fig. 9) must clearly be superimposed to permit consistency with the statistical model. A detailed cavity growth model that includes an elastic displacement field superimposed on the uniform separation should, therefore, be constructed to ensure compatibility. However, the influence of the elastic displacement on the cavity propagation rates is probably secondary in most cases, because the displacements (which nominally decrease rapidly (as $r^{1/2}$) with distance from the cavity tip) are substantially dispersed by mass transport along the

cavitating boundaries.³ (The trend is for the elastic displacement to counteract the relaxation term in the cavity growth relations.) The cavity propagation times are thus regarded as suitable initial approximations for use with the statistical creep strain model.

Although the elastic displacements may have only a secondary influence on the cavity propagation times, they must have a very appreciable effect on the mass transport that occurs along boundaries neighboring the cavitating boundary. The large elastic stresses that develop adjacent to the cavity tip act as a strong driving force for local mass transport,^{3,18} even along boundaries that are resistant to diffusion (Fig. 9). The mass transport relaxes the stress as a function of time,³ to produce a stress relaxation zone (Fig. 10), and converts the elastic strain into an anelastic and/or viscoelastic strain. In general, therefore, the measured creep strain, according to the present model, will have elastic, anelastic, and viscoelastic components: the latter relating to diffusion in the relaxation zone along boundaries adjacent to the cavity. This is typical of creep results obtained on ceramics that exhibit cavitation.^{10,21}

Expressions for the expected creep strain can now be derived by correlating the propagation time results with the statistical relations. The procedure is illustrated for the continuous second phase microstructure, because comparative experimental data are available for this type of microstructure.¹⁰ A similar procedure can be adopted for the other microstructures.

Commencing with the expression for the cavity propagation time (Eqn. 32)

$$t_p = 0.15(\eta_o/\sigma_\infty)(l/\delta_o)^2 \exp(Q_\eta/RT)$$

an assessment can be made concerning the expected t_p distribution. The primary variability in t_p undoubtedly derives from local variations in the relative film thickness, $\Delta (= \delta_o/\ell)$, rather than variations in the viscosity. Hence, assuming a second extreme value form for the distribution in Δ as a typical extreme value relation that pertains to microstructural features,⁸ the probability that the relative film thickness will exceed Δ is;

$$\Phi(\Delta) = 1 - \exp \left[- \left(\frac{\Delta_o}{\Delta} \right)^\lambda \right] \quad (42)$$

where λ is the shape parameter and Δ_o is the scale parameter ($\Delta_o \equiv (\delta_o/\ell)_o$). Inserting Δ from eqn. (32), the probability that a cavity will form at a time less than t becomes;

$$\Phi(t) = 1 - \exp \left[- \left(\frac{6.7\sigma_\infty t \Delta_o^2}{\eta_o \exp(Q_\eta/RT)} \right)^{\lambda/2} \right] \quad (43)$$

This relation is identical to eqn. (1) with

$$t_o = 0.15\eta_o \exp(Q_\eta/RT)/\sigma_\infty \Delta_o^2 \quad (44)$$

and

$$k = \lambda/2$$

This provides the principal justification for the choice of eqn. (1).

Inserting t_o from eqn. (45) into eqn. (9) the creep strain rate becomes:

$$\begin{aligned}\dot{\epsilon}_{\infty} &= 0.75 (0.15)^{-k} \xi \beta k (\delta_o / \ell)_o^{2k} \eta_o^{-k} \sigma_{\infty}^{(k+1)} t_o^{(k-1)} E^{-1} \exp(-kQ_{\eta}/RT) \\ &\equiv \omega \sigma_{\infty}^{(k+1)} t_o^{(k-1)} E^{-1} \exp(-kQ_{\eta}/RT) .\end{aligned}\quad (46)$$

Note that the stress and time dependencies, the activation energy, and the grain size dependence all depend on the magnitude of the shape parameter k of the distribution. No fundamental information concerning k is available at present. However, some recent creep data obtained by Lange¹⁰ can be used as an initial test of the utility of the statistical approach. Lange has measured density changes as well as creep rates. The change in density $\Delta\rho$ due to the presence of N cavities of average length $\langle \ell \rangle$ is

$$\Delta\rho = \pi N \langle \ell^2 w \rangle \quad (47)$$

where w is the average cavity width. The rate of change of density with time is thus;

$$\begin{aligned}\Delta\rho &= \pi \dot{N} \langle \ell^2 w \rangle \\ \dot{\epsilon} &= 6kV \left(\frac{\langle \ell^2 w \rangle}{\langle d^3 \rangle} \right) t_o^{-k} t^{k-1}\end{aligned}\quad (48)$$

The rate dependence of the density thus affords a direct measure of the shape parameter k . In Lange's experiments $\Delta\rho$ varied approximately linearly with time suggesting that $k \sim 1$. Hence, from Eqn. (46) the present model would predict a steady state creep strain rate;

$$\dot{\epsilon}_{\infty} = \omega \sigma_{\infty}^2 E^{-1} \exp(-Q_{\eta}/RT) \quad (49)$$

The predicted stress exponent of 2 is in close agreement with the observations.¹⁰

This correlation provides sufficient encouragement for further, critical study of the statistical model. In particular, the shape parameter k and its origins require investigation; as well as detailed studies of the anelastic and elastic components of the creep strain vis-a-vis the cavity propagation.

Finally, we note that the creep strain for the two other microstructures will exhibit a stronger dependence of the stress, through the σ_∞^3 term in eqns. (40,41). For example, for the single phase material;

$$t_o = kT \exp(Q_s/kT) \sigma_\infty^{-3} q_o [k^3 \gamma_s^2 (2-\gamma_b/\gamma_s)^{3/2} D_o^{-1}] \quad (50)$$

where q_o is the scale parameter that contains the variabilities in facet size, surface energies and diffusivities. The creep strain-rate is thus;

$$\dot{\epsilon}_\infty = \omega_* \sigma_\infty^{(3k+1)} t^{(k-1)} E^{-1} kT \exp(-kQ_s/kT) \quad (51)$$

where ω_* is the term equivalent to ω in eqn. (46).

4.2 The failure time

An expression for the failure time is derived for the continuous second phase microstructure. It is recalled that the relation will be pertinent to situations in which there is little interaction between the cavities that create the macrocrack, i.e., t_p is not greatly affected by the existence of prior cavities. This simplification must be emphasized in the use of the failure time relations. However, if it

is established from the simplified noninteraction models have merit, numerical techniques can subsequently be used to incorporate interaction effects.

Combining eqns. (18), (10) and (45) the failure time for a material with a continuous second phase becomes;

$$\ln \theta_r = \ln \left[0.3 \eta_o (\ell / \delta_o)_o^2 \right] - \ln \sigma_\infty - \sigma_\infty^2 (4\ell / \pi k f^2 K_c^2) \ln (4A / \ell^2) \quad (52)$$

Correlation of eqn. (52) with the test data (Figs. 6a,b) is relatively straightforward. A detailed comparison is conducted for the Al_2O_3 fibers. The pertinent dimensional information is; area tested,¹⁵ $A \sim 1.2 \times 10^{-5} m^2$, grain diameter²² $d \sim 5 \mu m$. From eqns. (52) and (18b) and Fig. 7a the following relations can be obtained;

$$n = 1 \quad (53a)$$

$$(4\ell / \pi k f^2 K_c^2) \ln (4A / \ell^2) = 3 \times 10^{-16} m^3 Pa^{-2} \quad (53b)$$

$$\log \left[0.3 \eta_o (\ell / \delta_o)_o^2 \right] = -5.5 \quad (53c)$$

The n value of unity suggests that the continuous second phase microstructure may be relevant to the Al_2O_3 fibers: encouraging further comparison. From eqn. (53b), and the given A and d dimensions, $k(fK_c)^2 \sim 2 \times 10^{11} Pa^2 m^{-3}$. Hence for $k \sim 1$, as observed for the creep tests on Si_3N_4 , we obtain $fK_c \sim 0.5 MPa \sqrt{m}$: a reasonable value for high temperature macrocrack propagation in fine-grained Al_2O_3 .²³ From eqn. (53c), $\eta_o (\ell / \delta_o)_o^2 \sim 10^{-6}$ poise. Assuming a film thickness of $\sim 5 nm$ (a typical value for sintered ceramics), yields $\eta_o \sim 10^{-1}$ poise. In conjunction with the activation energy, the resultant viscosity is plotted in Fig. 11, where it is compared with the viscosity of SiO_2 :²⁴

a common source of second phases. The comparison is close enough to provide some credence in the present model.

5. CONCLUSIONS

Statistical models for the creep strain and the failure time in the presence of grain boundary cavitation have been developed. The cavities form by the viscous flow of a boundary phase or by diffusive processes in the presence of both an external stress and grain boundary sliding displacements. Individual cavities coalesce, where they form on contiguous boundaries, and eventually, form a macrocrack that propagates to failure.

The creep strains predicted by the models are non-linear in stress, in accord with experimental observation. The models also predict rupture times that exhibit trends consistent with available data. The statistical approach thus appears a credible one for ceramic materials. However, substantial model development and extensive experimental observation of cavity distributions, grain boundary phases, grain boundary diffusivities, etc., are needed to establish an acceptable comprehension of cavitation creep in these materials.

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FIGURE CAPTIONS

- Fig. 1: a) Optical micrograph of an alumina tested in flexure at 1550°C, showing the cavities that develop on the surface (arrowed).
b) A transmission electron micrograph of a cavity in alumina (courtesy D. R. Clarke).
- Fig. 2: Schematics of cavities in the three microstructures considered in the paper (a) single phase material (b) continuous second phase (c) isolated second phase.
- Fig. 3: A schematic indicating the sequence of cavity formation leading to the development of a macrocrack.
- Fig. 4: The propagation of a cavity along a boundary accommodated by sliding of the adjacent boundaries.
- Fig. 5: A schematic indicating the effects of the applied stress σ_{∞} on the rupture parameter θ_r predicted by the statistical model.
- Fig. 6: A correlation of rupture data with the generalized statistical model; the parameters are indicated on the figures (a) Al_2O_3 fibers (b) SiC fibers.
- Fig. 7: The stress distribution within the viscous phase contained between a triple point and a cavity.
- Fig. 8: Scanning electron micrographs of a fracture surface of Al_2O_3 tested at 1550°C. The skeletal structure on the grain facets is the viscous phase; the cavitation sites are circumvented by the viscous phase.
- Fig. 9: A schematic indicating the elastic crack opening that occurs in the presence of a boundary cavity, and the relaxation that occurs by mass transport along adjacent boundaries.
- Fig. 10: A schematic of the relaxation zones around cavities indicating the condition for 'cavity interaction'.
- Fig. 11: A comparison of the viscosity of the boundary phase inferred from an analysis of failure time data for alumina with the viscosity of SiO_2 .

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Figure 1

(a) SINGLE PHASE MATERIAL

(b) CONTINUOUS SECOND PHASE

(c) ISOLATED SECOND PHASE

XBL 793-5996

Figure 2

XBL793-5995

Figure 3

XBL 793-5998

Figure 4

XBL 793-6000

Figure 5

XBL793-6002

Figure 6a

XBL793-6003

Figure 6b

XBB 792-1658

Figure 8

XBL 793-6001

Figure 7

XBL 793- 5997

Figure 9

XBL 793-5994

Figure 10

XBL 793-5999

Figure 11

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