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May 31 1994

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re: Contract #N00014-91-C-0067

Dear Dr. Vasudevan:

Please find attached one copy of the quarterly (March-May) technical report in accordance with the contract above.

Work during this last quarter has centered on ferroelectrics.

Yours sincerely

Dr. M.S. Duesbery
Vice-President
Fairfax Materials Research, Inc.

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Computer Modelling of Cyclic Deformation of High-Temperature Materials

TECHNICAL PROGRESS REPORT

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May 31, 1994

Period of performance
March 1, 1994 through May 31, 1994

I. Introduction and Program Objective

Current methods of lifetime assessment leave much to be desired. Typically, the expected life of a full-scale component exposed to a complex environment is based upon empirical interpretations of measurements performed on microscopic samples in controlled laboratory conditions. Extrapolation to the service component is accomplished by scaling laws which, if used at all, are empirical; little or no attention is paid to synergistic interactions between the different components of the real environment. With the increasingly hostile conditions which must be faced in modern aerospace applications, improvement in lifetime estimation is mandated by both cost and safety considerations.

This program aims at improving current methods of lifetime assessment by building in the characteristics of the micro-mechanisms known to be responsible for damage and failure. The broad approach entails the integration and, where necessary, augmentation of the micro-scale research results currently available in the literature into a macro-scale model with predictive capability.

In more detail, the program will develop a set of hierarchically structured models at different length scales, from atomic to macroscopic, at each level taking as parametric input the results of the model at the next smaller scale. In this way the known microscopic properties can be transported by systematic procedures to the unknown macro-scale region. It may not be possible to eliminate empiricism completely, because some of the quantities involved cannot yet be estimated to the required degree of precision. In this case the aim will be at least to eliminate functional empiricism. Restriction of empiricism to the choice of parameters to be input to known functional forms permits some confidence in extrapolation procedures and has the advantage that the models can readily be updated as better estimates of the parameters become available.

II. Program Organization

The program has been organized into specific tasks and subtasks as follows.

Task 100. Lifetimes of metallic dispersed-phase composites

Most service materials fall into the category of dispersion-hardened metallic composites. This task will consider the problem of dispersion hardened materials in general, but with two specific materials, NiAl and $MoSi_2$/SiC in mind.

Task 110. Identification and modelling of micromechanisms

The purpose of this task is to determine what micromechanisms are operative in the high-temperature deformation of dispersion-hardened materials. In the general case this will be done by a literature search. For specific materials, the micromechanisms will be determined from the experimental program at NRL. Once identified, each of these micromechanisms will be modelled, in order to determine what are the critical parameters which determine its effect on plastic flow and values for these parameters. Also to be determined is whether the modelled critical values are dependent on quantities which must be obtained from a smaller scale model.

Task 111. Equiaxed dispersoids

This task will consider dispersions of the type encountered in NiAl-like materials. That is, the dispersoids are considered to be small compared to the grain size. The term 'equiaxed' is used because the particles are roughly of the same size in all three dimensions. However, this is not a requirement for this task. Rather, it is necessary that the particles not be too large in the dimension normal to the slip plane, so that they can be surmounted with relative ease by cross-slip and/or climb without the generation of appreciable back-stress.

Task 112. Anisotropic dispersoids

This task covers the case of dispersoids which are elongated in the direction normal to the slip plane. An example is SiC fibers in $MoSi_2$. In this case, plastic flow around the dispersoids takes place by a combination of glide and climb, but is a protracted process during which large stresses acting in opposition to the applied load are developed.

Task 113. Grain boundary effects

This task will examine the role of grain boundary processes in high-temperature deformation.

Task 120. Macroscopic stochastic model for creep

In real materials it is likely that more than one mechanism will be operative, either in parallel or in series. The information gained in task 110 is not sufficient to describe this situation. Once the critical parameters for individual mechanisms have been determined, it is necessary to combine them in a macroscale stochastic model. This will be done by determining critical stresses and activation enthalpies as a function of local geometry and using these values in a finite-temperature simulation of creep through a random array of dispersoids. Careful attention must be paid to possible interactions between mechanisms.

Task 130. Extension to cyclic deformation

The final step in task 100 is to extend the results to the case of cyclic deformation. Irreversibility is an intrinsic feature of the model in task 120. However, it is likely that other, as yet unrecognized, characteristics of cycled deformation will have to be considered.

Task 200. Lifetimes of piezoelectric ferroelectrics

Failure in cyclic loading of sensors and actuators formed from lead zirconate titanate (PZT) is a continuing problem. PZT is a ceramic and therefore differs from the materials considered in task 100 in that plastic deformation is not involved. This task will examine, modelling as necessary, the operation of PZT devices, in order to determine the factors governing lifetime limitation.

Task 300. Reporting

Running concurrently with tasks 100 and 200, this task will inform the Navy Program Manager and Contracting Officer of the technical and fiscal status of the program through R&D status reports.

III. Technical Progress

Task 200. Lifetimes of piezoelectric ferroelectrics

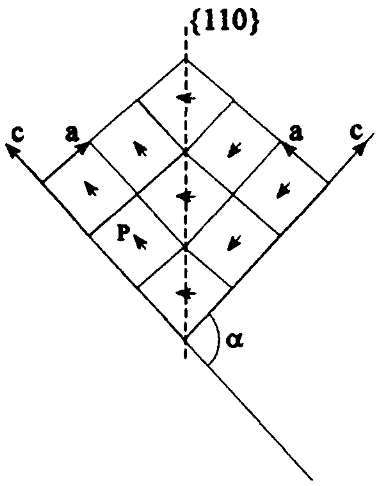

Figure 1 Structure of a {110} domain wall

Most of the work done so far on PZT has assumed that domain walls lie parallel to {100} planes, so that an elastic misfit is implied for the tetragonal structure, a misfit which can be relieved by dislocations if it is larger than some critical value. In fact it has been pointed out[1] that a domain structure which is essentially free of elastic strain can be obtained by taking the domain walls to lie along the undistorted {110} planes. The structure is shown schematically in Figure 1. The tetragonal unit cells on either side of the domain walls are shown, together with the directions of the **c** and **a** axes and the polarization vectors **P**. Evidently the domain wall can be described equivalently as a rotation twin boundary. From a continuum viewpoint, the domains fit together without deformation along the (dashed line) domain wall, with an angle

$$\alpha = \pi - 2\arctan(a/c) \tag{1}$$

between the characteristic vectors of each domain. This means that the polarization vectors are not orthogonal, so that there is a finite electrostatic energy. The force due to this electrostatic energy will tend to force the domains back towards orthogonality, and the equilibrium state is expected to have finite elastic and electrostatic energies, with a value of α lying between that given by (1) and $\pi/2$. It is perhaps more realistic to adopt a quasi-atomistic viewpoint and recognise that the cells intersected by the domain wall are not in their ground state, rather having an energy which can be approximated roughly as the elastic energy necessary to deform the tetragonal unit cell into this twinned state. It is not clear whether this twin-type domain structure will have an energy lower than the {100} wall. Attention will be paid to this topic in the next quarter; in the meantime, the current report will be concerned with {100} walls only.

Grain Boundary Fracture in PZT

It has been found previously (see 11/93 quarterly report) that there are forces G for the growth of pairs of microcracks, each of length (a-c), which are formed in grain boundaries due to the presence of misfit η across domain walls. These are of amount:

$$G = \frac{\pi\mu}{4(1-\nu)c(a^2-c^2)}\left[\frac{2\eta^2(a^2-c^2)^2}{(1-\nu)^4} + \frac{a^2b^2}{2K^2(k)} + \frac{2\eta(a^2-c^2)ab}{K(k)(1-\nu)}\right] \tag{2}$$

These microcracks will grow provided

$$G > 2\gamma_b, \tag{3}$$

where γ_b is the energy density necessary to form a crack along a grain boundary.

As noted before the form of this relation is such that G passes through a minimum at some value of c lying between 0 and a. Fracture can be complete only if G at this minimum value exceeds the critical value indicated above. The determination of position of this minimum involves the solution of a quintic equation. Rather than solve this equation we have resorted to graphical methods. Thus, we have plotted G as a function of the variable $x = c/a$ when, as previously shown, $b = 2\eta a$. We have assumed typical values; the tetragonality factor $\eta = .04$; $K(k) \approx \ln(a/b) \approx 10$ and we have taken Poisson's ratio ν as 0.3. The resulting expression is:

$$G(x) = \frac{\pi\mu}{4(1-\nu)}\frac{a}{x(1-x^2)}[.16(1-x^2)^2 + .0008 + .0048(1-x^2)]. \tag{4}$$

The result is shown in Figure 2. It may be seen therefrom that the minimum in G occurs when $c \approx .8\,a$ and is given by:

$$G_c = 0.494\frac{\pi\mu a}{4(1-\nu)}. \tag{5}$$

Since this is normally much larger than $2\gamma_b$ fracture will generally follow the formation of dislocation dipoles through the sliding of domain walls.

This leads to the view that a length of grain boundary which provides a termination for a sequence of parallel domain walls will be fractured between alternate walls. Fatigue can then be seen to result from the externally induced motion of domain walls of a sense such as to expand the fractured regions. Such motion can be produced by either electrical or mechanical stressing.

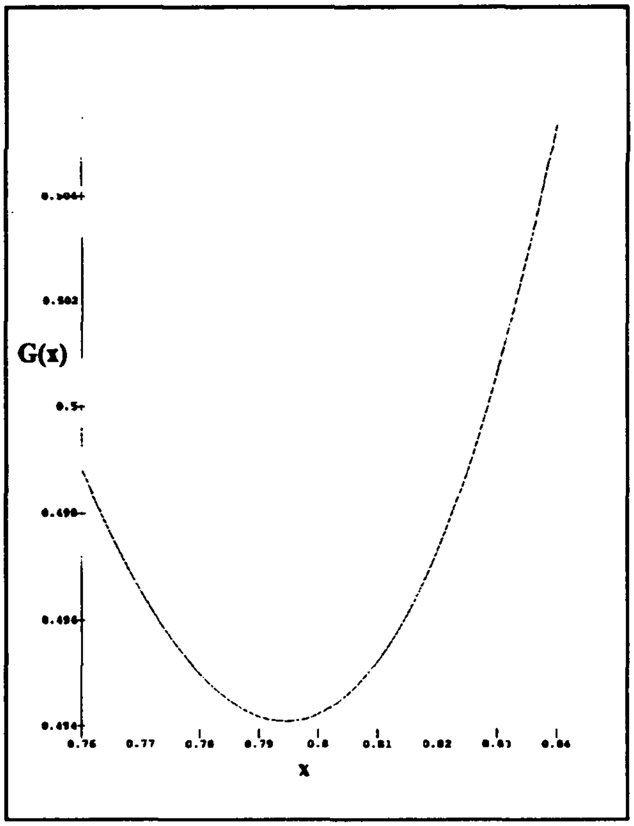

Figure 2 Solution of Equation (3)

Mobility of 90⁰ Domain Walls

The mobility of 90⁰ domain walls (assumed to lie on {100} planes) in the limit of pinning by dislocations has been treated in a previous report. In this report the mobility of unpinned walls will be discussed. A 90° domain wall is the interface between tetragonal crystal lattices oriented with c-axes at right angles to each other. An electic field is directed parallel to the c-axis and thus the direction but not the intensity of the electric field changes at a 90° domain wall. Movement of a wall in a direction normal to itself then necessarily requires a change in both lateral dimensions and in the direction of the local electric field in the volume swept as a consequence of the motion.

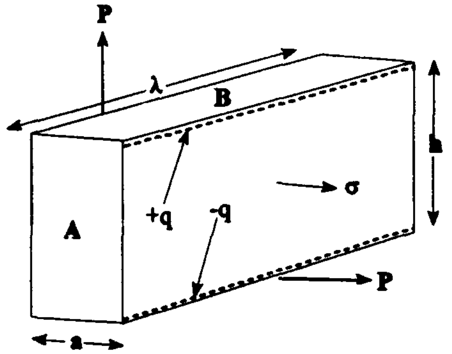

Figure 3 A "pillbox" excitation on a domain wall

The domain wall configuration (and hence its energy also) are invariant to translation through an integral number of lattice spacings. That is, the energy is periodic with the period of the lattice. In the absence of an atomistic calculation it is not possible to estimate the energy of a domain wall at a position lying between its periodic minima, but the experimental observation that the walls are straight suggests that this energy is relatively large. Therefore it seems likely that the situation is analogous to that of dislocation mobility in most crystalline solids - that the mechanism of mobility is the formation of a nucleus and its subsequent growth. In the case of dislocations, the nucleus is an advance of a small length of the line through a single atomic spacing, forming an excitation known as a kink pair. By analogy, the nucleus for a domain wall is "pillbox"-like small area of the wall advanced through a unit step into an adjacent domain. This is illustrated in Figure 3, in which an area of dimensions λ x h has been moved through a distance a from left to right from a domain with polarization P vertical into one with polarization horizontal. A normal stress σ is supposed to act on the wall. Because the normal component of the electric displacement must be continuous across the faces B, layers of depolarization charge density ±q (dashed lines in Figure 3) of uncertain magnitude develop on these interfaces. The enthalpy of the "pillbox" can be written as

$$H = 2aE_w(h+\lambda)+E_{es}-\sigma h\lambda a \tag{6}$$

where the first term is the energy of the sidewalls A and B (E_w is the wall energy per unit area), E_{es} is the electrostatic energy and the third term is the work done by the wall stress in forming the pillbox.

The electrostatic energy consists of three terms. First, the energy of the repolarized material within the pillbox, which is

$$E_I = \frac{1}{2}h\lambda a\frac{P^2}{\kappa\kappa_o} \tag{7}$$

where P is the remanent polarization per unit volume, κ is the dielectric constant and κ_o is the permittivity of free space. Second, the self-energy of the depolarizing dipole layer,

$$E_{II} = \frac{q^2}{\kappa\kappa_o}\int_0^{\lambda} d\lambda_1 \int_0^{\lambda} \frac{d\lambda_2}{\sqrt{h^2+(\lambda_2-\lambda_1)^2}} \tag{8}$$

and finally, the interaction energy between the depolarizing dipole layer and the remanent polarization.

$$E_{III} = -\frac{qh\lambda P}{\kappa\kappa_o} \tag{9}$$

The only one of these terms which requires any effort in evaluation is E_{II} in (8), which can be rewritten in parametric form

$$\begin{aligned} E_{II} &= \frac{q^2}{\kappa\kappa_o} I \\ I &= h\int_0^{\frac{\lambda}{h}} ds \int_{-s}^{\frac{\lambda}{h}-s} \frac{dt}{\sqrt{1+t^2}} \end{aligned} \tag{10}$$

The double integral I in (10) has been evaluated numerically, by gaussian integration, as a function of the aspect ratio of the pillbox, λ/h. The results are shown in Figure 4. Cursory examination does not suggest any approximate analytic form for I and therefore it is likely that the equations (6-9) will have to be solved numerically. This will be done during the next period of performance.

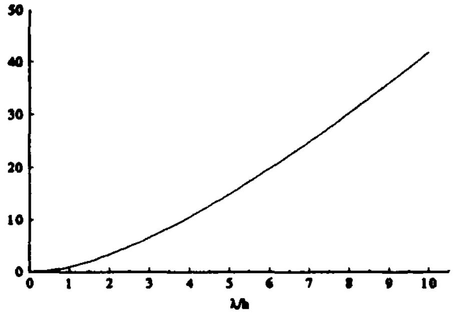

Figure 5 The depolarization energy integral

References

1. W. Cao and L.E. Cross, Phys. Rev. **B44** (1991)