A multiscale approach for coupled phenomena in fcc materials at high temperatures

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ABSTRACT

The initiation of surface cracks in single-crystal superalloy components exposed to an oxidizing environmental under thermomechanical loading conditions is known to be linked to the presence of internal 10–20 μm-diameter casting defects (i.e. porosities), and to the localization of inelastic strain. In this work, the effects of oxidation on both the local degradation of the superalloy microstructure and the initiation of surface cracks from such defects is investigated. The approach relies on a rate-dependent crystallographic theory to describe the viscoplastic behaviour of the single crystal, and on a coupled oxidation–deformation framework to incorporate explicitly the effects of micro-structural degradation due to oxidation. Predictions of the formation of surface cracks under constant far-field loading, linked to the nucleation and coalescence of microcracks from internal porosities, are obtained from a recently proposed mechanistic anisotropic void growth model. Coupled oxidation–deformation finite-element analyses in compact tension fracture specimens show that environmental effects reduce the time to crack initiation from blunt notches owing to an increase in the accumulated inelastic deformation in the vicinity of the porosities.

§1. INTRODUCTION

High-temperature components in gas turbines used for aerospace and power generation are generally subjected to severe and complex thermomechanical loading and harsh environmental conditions. Ni-based single-crystal superalloys have become widely used in such applications on account of their high-temperature strength. Such alloys contain a high volume fraction (typically 68%) of 0.5–1 μm size (γ’) precipitates embedded in a soft Ni-based solid solution (γ) matrix.

An important contributing factor to failure in these materials is the localization of the inelastic deformation caused by the heterogeneity of the material microstructure. In two-phase single crystals, heterogeneities exist at both the microscopic

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and the mesoscopic levels (figure 1). At the microscale (see figure 1 (c)), such heterogeneities result from the presence of the $\gamma'$ precipitates, while at the mesoscale (see figure 1 (b)), the presence of casting porosities introduces an additional degree of heterogeneity in the local microstructure (Douglas 1999, Busso et al. 2001b). A further source of heterogeneity at this length scale may arise from the
microstructural degradation caused by local oxidation, which becomes significant for uncoated Ni-based superalloys at temperatures in excess of about 800°C (e.g., see Martinez-Esnaola et al. (1997) and Andrieu and Pineau (1999)).

The effects of oxidation-induced degradation on crack growth behaviour in coated superalloys has been investigated under cyclic thermal loading conditions (Nusier and Newaz 1998, Sfar et al. 2002). Such studies are usually based on either elastic or rate-independent stress analyses and do not consider the local change in chemical composition due to diffusion and interdiffusion processes. It is only recently that time-dependent effects on the material constitutive behaviour have been coupled to the local phase composition resulting from oxidation processes (for example Busso et al. (2001a)).

To account for the effects of oxidation on the deformation and fracture behaviour of the material, a coupled oxidation–deformation formulation that incorporates both time- and spatial-dependent behaviour on different length scales is required. In this work, a multiscale continuum mechanics framework, implemented within the finite-element (FE) method, is presented to describe the effects of microstructure, kinetic processes and thermomechanical loads on the initiation of surface cracks exposed to an oxidizing environment. The approach relies on a rate-dependent crystallographic framework to describe the macroscopic (i.e. homogenized) constitutive behaviour of the two-phase single crystal. A damage model based on the initiation of microcracks from the surface of existing internal voids at the mesoscale is also incorporated within the proposed framework. The effects of microstructural changes on the crack tip stresses and strains, resulting from the local diffusion and interdiffusion processes linked to surface oxidation, as well as the subsequent local damage responsible for surface crack initiation, are studied numerically.

The structure of the paper is as follows. In §2, the crystallographic theory for the Ni-based superalloy single crystal is introduced. In §3, a brief overview of oxidation in Ni–Cr–Al alloys is given, followed by a description of the FE implementation of the oxidation and diffusion problem. In §4 the micromechanics void growth formulation used to predict the triaxiality-dependent critical void volume associated with the initiation of microcracks (i.e. damage) from the voids surface is presented. Details of the FE model of a compact tension (CT) specimen are given in §5, and then the results of the coupled oxidation–deformation-damage simulations of the CT specimen undergoing oxidation are presented and discussed.

§ 2. Multiscale crystallographic formulation

2.1. Slip system flow and evolutionary equations

The average macroscopic stress–strain behaviour of the two-phase ($\gamma + \gamma'$) superalloy single crystal of interest, namely CMSX4, is described by a multiscale rate-dependent crystallographic formulation which extends the dislocation-mechanics-based crystallographic model of Busso (1990) and Busso and McClintock (1996). The flow rule relies on a stress-dependent activation energy expressed in terms of two internal variables per slip system $\alpha$: a macroscopically average slip resistance $S^\alpha$ and an internal or back stress $B^\alpha$. Thus

$$\dot{\gamma}^\alpha = \dot{\gamma}_0 \exp \left[ - \frac{F_0}{k \theta} \left( 1 - \left( \frac{|\tau^\alpha - B^\alpha| - S^\alpha \mu/\mu_0}{\tau_0 \mu/\mu_0} \right)^p \right) \right] \text{sgn}(\tau^\alpha - B^\alpha),$$

(1)
where \( k \) is the Boltzmann constant, \( \tau^\alpha \) is the resolved shear stress on the slip system, \( \theta \) is the absolute temperature, \( \mu \) and \( \mu_0 \) are the shear moduli at \( \theta \) and 0 K, respectively, and \( F_0, \vec{\tau}_0, p, q \) and \( \gamma_0 \) are material constants.

The evolutionary behaviour of the overall slip resistance for the slip system \( \alpha \) is expressed as

\[
\dot{S}^\alpha = \sum_{\beta=1}^{N} \delta^\alpha_\beta \left[ h_S - d_D (S^\beta - S^\beta_0) \right] |\dot{\gamma}^\beta|,
\]  

(2)

where \( h_S \) and \( d_D \) are the static and dynamic recovery coefficients, respectively, \( S^\beta_0 \) is the initial slip resistance, \( N \) is the total number of slip systems and \( \delta^\alpha_\beta \) is the cross-hardening or interaction matrix. The multiscale nature of the formulation is incorporated in equation (2) by making an explicit link between the evolution of the overall resistance to slip with the volume fraction \( v_f \) and mean size \( l \) of the \( \gamma' \) precipitate population at the microscale. Thus, by defining functional forms for \( d_D \) and \( S^\beta_0 \) in equation (2) in terms of the characteristics of the \( \gamma' \) precipitates, one obtains (see Busso (2003) for further details)

\[
d_D = \dot{d}_D \left\{ \frac{l}{l_m}, v_f, \theta \right\},
\]  

(3)

\[
S^\beta_0 = \dot{S}^\beta_0 \left\{ \frac{l}{l_m}, v_f \right\},
\]  

(4)

where \( l \) has been normalized with a mean reference value \( l_m \).

The second internal slip system variable, the back stress, evolves following a standard hardening-dynamic recovery form:

\[
\dot{B}^\alpha = h_B \dot{\gamma}^\alpha - \dot{r}_D (S^\alpha) B^\alpha |\dot{\gamma}^\alpha|,
\]  

(5)

where \( h_B \) is the hardening coefficient and \( \dot{r}_D \) a dynamic recovery function which depends explicitly on the slip resistance (for example Busso and McClintock (1996)).

Thus, through equations (1)–(5), the crystallographic model is able to account for the changes in the macroscopic stress–strain response resulting from changes in precipitate size and volume fraction linked to local kinetic processes.

The calibration of the single-crystal model was carried out under the assumption that 12 octahedral (\{111\} (011)) and six cubic (\{100\} (011)) slip systems can be potentially active. The reference data to calibrate the model for the initial precipitate volume fraction of \( v_f = 68\% \) were obtained from uniaxial monotonic, cyclic and creep tests (Busso 2003). For lower volume fractions \( (v_f < 68\%) \), it was necessary to determine equations (3) and (4) from reference data generated numerically from FE analyses of three-dimensional (3D) periodic unit cells of representative volume elements (RVEs). For the simplest case of regularly arranged precipitates, an RVE contains a single discrete precipitate embedded in a pure single-crystal \( \gamma \) matrix (Busso et al. 2000, Meissonnier et al. 2001). In that work, a non-local crystallographic theory was used to describe the behaviour of the \( \gamma \) phase so that the effects of all the relevant length scales (e.g. spacing between precipitates, precipitate size and mean dislocation spacing) can be accounted for. The values of the calibrated single-crystal model parameters are given in table 1.

Predictions of the monotonic (001) uniaxial behaviour of CMSX4 at 950°C for different strain rates are shown in figure 2 (a). In all cases, self-hardening is assumed.
so that $\delta^\alpha_\beta = \delta^\alpha_\beta$, the Kronecker delta, in equation (2). Consistent good agreement between the model predictions and the experimental data was found over a wide range of strain rates. The effect of the precipitate volume fraction on the uniaxial $(001)$ single-crystal response obtained from the unit-cell analyses is shown in figure 2 ($b$). The mechanical behaviour of the pure $\gamma$ phase used in the unit-cell analysis is also included in figure 2 ($b$) for reference.

As will be discussed in §3, the diffusion and interdiffusion processes linked to surface oxidation result in the local dissolution of the $\gamma'$ precipitates, leaving behind a pure $\gamma$ phase (Bouhanek et al. 1997), that is a region where $v_f = 0\%$. Thus, within the multiscale modelling framework presented here, the upper- and lower-bound responses of the superalloy will be given by the $v_f = 68\%$ curve and the pure $\gamma$ phase curve in figure 2 ($b$), respectively.

### 2.2. Numerical integration procedure

The proposed crystallographic model was implemented numerically into the FE method using a hyperelastic and large-strain kinematics framework. It relies on the classical multiplicative decomposition of the total deformation gradient, $F = F^e F^p$, where $F^e$ is the component which accounts for elastic deformation and rigid-body rotations and $F^p$ is the inelastic component associated with pure slip. $F^p$ is calculated from the inelastic velocity gradient:

$$ L^p = F^p F^{p^{-1}} = \sum_{\alpha=1}^{n} \dot{\gamma}^\alpha (m^\alpha \otimes n^\alpha), $$

where $m^\alpha$ and $n^\alpha$ are the slip direction and the slip plane normal vectors, respectively, for the slip system $\alpha$. The numerical implementation scheme relies on a fully implicit algorithm to update the stress tensor and the internal slip system variables. The crystallographic model was implemented numerically as a user-defined material subroutine in a commercial FE code (ABAQUS 2001). Further details of the numerical implementation have been given by Busso et al. (2000) and Meissonnier et al. (2001).
3. Oxidation model

3.1. Oxidation of two-phase Ni–Cr–Al alloys

Isothermal oxidation of Ni–Cr–Al alloys at temperatures higher than about 900°C generally results in the formation of Al₂O₃ as the primary oxide. Other oxides (e.g. NiO, Cr₂O₃ and Ni(Cr, Al)₂O₄) may develop at the early transient stages of oxidation. The first oxide to form on an Ni–Cr–Al alloy is a mixture of NiO and Ni(Cr, Al)₂O₄ owing to the internal oxidation of Ni and Cr (Giggins and Pettit 1971). Next, Cr₂O₃ and Al₂O₃ form below the NiO–Ni(Cr, Al)₂O₄ scale as a result of the interdiffusion of Al and Cr towards the oxide, and the diffusion of oxygen into the alloy. Then either one or a mixture of these oxides will form, depending on the Cr and Al content of the material (Giggins and Pettit 1971).
The material under examination in the present study, the Ni-based superalloy CMSX4, has a low Cr content (7.5 at.%) and a high Al content (19.7 at.%). Thus, following the early stages of oxidation, a continuous and stable Al₂O₃ layer forms (Bouhanek et al. 1997, Gross et al. 1998). As shown schematically in figure 3 (a), Al₂O₃ forms at the expense of a loss of Al from the alloy. Furthermore, the rate of growth of the oxide is controlled by the rate at which Al interdiffuses within the superalloy and the oxide itself. Experimental measurements on CMSX4 revealed that the growth rate of the oxide follows a parabolic law (Bouhanek et al. 1997).

The Al flux from the metal into the oxide can be expressed as (Nesbitt 1982)

\[ J_{\text{Al}} = -\lambda \left(1 - v_{\text{Al}} C_{\text{Al}}^1 \right) \left( \frac{k_p}{t} \right)^{1/2}, \]  

where \( k_p \) is the parabolic rate constant, \( t \) the time, \( C_{\text{Al}}^1 \) the Al concentration at the metal–oxide interface (i.e. \( \gamma \) region oxide) and \( v_{\text{Al}} \) the partial molar volume of Al. The value of the parameter \( \lambda \) in equation (7) can be obtained from the published literature (Sekerka et al. 1975, Nesbitt 1984a,b, 1989, Bouhanek et al. 1997).

The degradation of the superalloy due to Al–Cr–Ni diffusion can be illustrated with the aid of the schematic ternary Ni–Cr–Al phase diagram shown in figure 3 (b).

Figure 3. Schematic diagrams of (a) the oxidation–diffusion processes at the crack faces, (b) Ni–Cr–Al ternary phase diagram and (c) Al concentration profiles at two different times.
Note that care must be taken when predicting microstructure evolutions under inherently non-equilibrium conditions from equilibrium phase diagrams. In this work, the non-equilibrium conditions associated with the diffusion processes across the $\gamma-(\gamma+\gamma')$ interface have been approximated as a quasi-equilibrium problem since, experimentally, no discernible transition region (i.e. gradient in the precipitate volume fraction) (Bouhanek et al. 1997) can be seen. Thus, points a and b in figure 3(b), represent the compositions of the $\gamma$ and $\gamma'$ phases in equilibrium with each other. Furthermore, point c in figure 3(b), which lies on the so-called tie-line, a–b, represents the initial superalloy composition. As the Al is supplied to form the oxide, a non-equilibrium state thus exists in the $\gamma+\gamma'$ region of the alloy, leading to the dissolution of the $\gamma'$ precipitates. The precipitates continue to dissolve as oxidation progresses, leading to the development of two distinct regions: a pure $\gamma$ region near the surface, and the unaffected $\gamma+\gamma'$ region within the bulk of the crystal. An illustration of the development of these two distinct regions is given in figure 3(c), which shows typical Al concentration profiles (from the oxide–$\gamma$ region interface) at two different times for an Al–Cr–Ni alloy. Here, the interface between the two regions translates away from the oxidizing surface with time, leading to a thickening of the $\gamma$ region. The corresponding diffusion path in the ternary phase diagram’s single-phase $\gamma$ region would be c–d (see figure 3(b)).

In addition, Ni, like Al, diffuses from the two-phase region towards the single-phase $\gamma$ region, owing to the dissolution of the precipitates, while Cr diffuses away from the moving oxide–$\gamma$ region interface and tends to accumulate near this interface as it moves. In this study, Ni and Cr diffusions will not be modelled as Al diffusion dominates the mechanism of oxide scale formation.

### 3.2. Solution of the one-dimensional diffusion problem

A one-dimensional (1D) diffusion model linked to the surface oxidation of the superalloy previously discussed was developed to provide an accurate reference solution for the more complex two-dimensional and three-dimensional diffusion problem required in the coupled kinetics–constitutive continuum mechanics framework. The relevant diffusion and mass conservation equations which control the problem were implemented numerically using finite-difference (FD) techniques. The numerical model relied on the following assumptions (Lee et al. 1987).

1. $\text{Al}_2\text{O}_3$ is formed exclusively.
2. The driving force for Al results only from the Al concentration gradient.
3. The diffusion coefficient is independent of Al concentration.
4. The $\gamma-(\gamma+\gamma')$ interface is planar.
5. Diffusion occurs only in the $\gamma$ region.

Numerical modelling of atomic diffusion in systems containing a moving interface requires the solution of Fick’s second law constrained by mass balance at the moving interface. In one dimension,

$$\frac{\partial C_{\text{Al}}}{\partial t} = D_{\text{Al}} \frac{\partial^2 C_{\text{Al}}}{\partial x^2},$$

where $C_{\text{Al}}$ is the Al concentration, $D_{\text{Al}}$ the diffusion coefficient, $t$ the time and $x$ the position from the oxide–metal interface.
The mass balance equation at the $\gamma$–($\gamma' + \gamma''$) interface can be written as

$$\left( C_{\text{Al}}^{\gamma'} - C_{\text{Al}}^{\gamma + \gamma''} \right) \frac{d \xi}{dt} = J^{\gamma},$$

(9)

where $C_{\text{Al}}^{\gamma'}$ and $C_{\text{Al}}^{\gamma + \gamma''}$ are the aluminium concentrations at the $\gamma$ and $\gamma' + \gamma''$ sides, respectively of the $\gamma$–($\gamma' + \gamma''$) interface, $\xi$ is the position of the interface with respect to the oxide–metal interface and $J^{\gamma}$ is the Al flux at the pure $\gamma$ region side of the $\gamma$–($\gamma' + \gamma''$) interface (see figure 3 (c)). The latter can be obtained from Fick’s first law:

$$J^{\gamma} = -D_{\text{Al}} \frac{\partial C_{\text{Al}}}{\partial x}.$$  

(10)

All the material parameters required for the diffusion–oxidation model, namely $\lambda$, $k_p$, $v_{\text{Al}}$, $D_{\text{Al}}$, $C_{\text{Al}}^{\gamma'}$, $C_{\text{Al}}^{\gamma + \gamma''}$ and $C_{\text{Al}}^{i}$, were obtained from the work of Taylor and Floyd (1952), Sekerka et al. (1975), Nesbitt (1984a,b, 1989) and Bouhanek et al. (1997) and are summarized in table 2.

The diffusion problem defined by the set of equations (8)–(10), subject to the flux boundary condition given by equation (7), was solved numerically using an explicit FD method and a flexible regular grid of points across the $\gamma$ region via an iterative procedure. The grid position was updated on the basis of the position of the interface, and the concentrations adjusted using the Murray–Landis transformation (Lee et al. 1987).

The predicted Al concentration profiles (from the oxide–$\gamma$ region interface) and the interface positions at different times are shown in figures 4 (a) and (b), respectively. It can be seen that the $\gamma$ region thickness is predicted to be approximately 80 $\mu$m after exposure for 500 h at 850°C (see figure 4), which is consistent with observations in similar Ni–Cr–Al alloys (Nesbitt 1982).

3.3. Finite-element implementation of the two-dimensional diffusion–oxidation problem

In order to enable the oxidation–diffusion processes described in §3.1 to be coupled with the multiscale crystallographic model of §2, it is necessary to incorporate the former into an FE framework. In this study, the oxide scale will not be explicitly included in the FE analysis as, under the conditions examined, its thickness can be considered to be negligibly small when compared with a relevant geometric length scale (e.g. 100 $\mu$m notch radius) and will thus have a small effect on the mechanical response at the mesoscale. Therefore, only Al diffusion and the subsequent growth of the pure $\gamma$ region next to the oxide will be modelled. In this case, the only parameters which need to be prescribed in the FE mass diffusion analysis are the Al flux boundary condition, given by equation (7), and the surface diffusivity.

Owing to the inability of standard FE methods to deal with the discontinuity in Al concentration at the $\gamma$–($\gamma + \gamma'$) interface (namely ($C_{\text{Al}}^{\gamma'} - C_{\text{Al}}^{\gamma}$) in figure 3 (c)),

Table 2. Diffusion–oxidation parameters.

<table>
<thead>
<tr>
<th>$v_{\text{Al}}$ (at.%)$^{-1}$</th>
<th>$\lambda$ (mm$^2$ mg$^{-1}$)</th>
<th>$k_p$ (mg$^2$ mm$^{-4}$ s$^{-1}$)</th>
<th>$D_{\text{Al}}$ (mm$^2$ s$^{-1}$)</th>
<th>$C_{\text{Al}}^{\gamma}$ (at.%)</th>
<th>$C_{\text{Al}}^{\gamma + \gamma''}$ (at.%)</th>
<th>$C_{\text{Al}}^{i}$ (at.%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>81.8</td>
<td>0.125</td>
<td>$8 \times 10^{-10}$</td>
<td>$3.47 \times 10^{-9}$</td>
<td>12.5</td>
<td>19.7</td>
<td>7.0</td>
</tr>
</tbody>
</table>
the position of this interface will not be explicitly solved for in the FE mass diffusion problem as was the case in the FD calculations (e.g. via equation (9)). Instead, the interface will be implicitly introduced by selecting the value of $D_{\text{Al}}$ depending on whether the material point is within the $\gamma$ or the $\gamma + \gamma'$ region. If the concentration of Al is below the upper solubility limit of Al in $\gamma$ (point $b$ and $C_{\text{Al}}^\gamma$ in figures 3(b) and (c)), then the material point is designated to be in the $\gamma$ region and the appropriate diffusion coefficient is assigned. Otherwise, $D_{\text{Al}}$ is taken to be zero and no diffusion is assumed to take place at this material point. In this way, the dissolution of the $\gamma^0$ precipitates at the $\gamma-(\gamma + \gamma')$ interface can be accounted for by determining the position of the interface as the location where the upper solubility limit has been reached.

To assess the validity of the FE approach, FE mass diffusion analyses were carried out to compare the results with the 1D FD predictions. The good agreement between both solutions may be seen in figures 4(a) and (b), which show the position of the $\gamma-(\gamma + \gamma')$ interface as a function of time, and the Al concentration profiles
after 160 and 500 h at 850°C, respectively. It can be seen that the absence of a true discontinuity in the Al concentration has no real implications in the numerical FE results if a fine enough local FE mesh is used.

§ 4. MECHANISMS OF CRACK INITIATION

Observations of fracture surfaces in superalloy uniaxial creep specimens loaded along the [001] crystallographic direction (Douglas 1999, Dennis 2000) revealed multiple [001] facets resulting from the propagation of microcracks which appeared to have initiated at casting porosities. Figure 5(a) illustrates such a mechanism of crack propagation from a blunt notch. A typical cracked porosity is given in figure 5(b), and a fracture surface showing facets cracks and initiation sites in figure 5(c) (shown by the white arrows). Under increasing localized deformation at the notch root, microcracks propagate and coalesce, leading to the growth of the mesoscopic crack and to the eventual fracture of the specimen (Douglas 1999, Dennis 2000).

To formulate a void growth model and a criterion for microcrack initiation, the multiscale rate-dependent crystallographic formulation presented in §2 was used to quantify the behaviour of an RVE or unit cell of the single crystal containing an initially spherical void (Busso et al. 2001b). An extensive FE parametric study was undertaken to determine the effect of stress state, temperature, relative crystallographic orientation and characteristic length scale (here defined as the ratio of the distance from a free surface to the void diameter) on the void growth rate. A micromechanics-based void growth formulation was afterwards developed using the results of this parametric investigation. This is outlined next.

Figure 5. (a) Schematic diagrams of the crack propagation mechanism from a blunt notch by microcrack coalescence, (b) typical cracked porosity and (c) square facets originating from porosity sites (indicated by the white arrows) observed at 950°C under a [010] creep load (Dennis 2000).
4.1. Void growth formulation

The void growth formulation is an extension of that used by Budiansky et al. (1982) for isotropic materials. The total volumetric void growth rate $\dot{V}$ is assumed to be controlled by slip and defined as

$$\frac{\dot{V}}{V} = \left[ a_1 \left( \frac{\dot{V}_{\text{oct}}}{V} \right)^r + a_2 \left( \frac{\dot{V}_{\text{cub}}}{V} \right)^r \right]^{1/r}, \quad (11)$$

where $V$ is the current void volume, $\dot{V}_{\text{oct}}$ and $\dot{V}_{\text{cub}}$ are the contributions to the void growth rate from slip on the octahedral and cubic slip systems, respectively, and $r$, $a_1$ and $a_2$ are parameters calibrated numerically from the FE void growth calculations.

Expressions for $\dot{V}_{\text{oct}}$ and $\dot{V}_{\text{cub}}$ are

$$\frac{\dot{V}_{\text{oct}}}{V\dot{\varepsilon}_{\text{oct}}} = \hat{a}_{\text{oct}}(n) \left| \frac{\sigma_{\text{m}}}{\sigma_c} \right|^{w_{\text{oct}}}, \quad (12)$$

$$\frac{\dot{V}_{\text{cub}}}{V\dot{\varepsilon}_{\text{cub}}} = \hat{a}_{\text{cub}}(n) \left| \frac{\sigma_{\text{m}}}{\sigma_c} \right|^{w_{\text{cub}}}, \quad (13)$$

where $\dot{\varepsilon}_{\text{oct}}$ and $\dot{\varepsilon}_{\text{cub}}$ are the equivalent inelastic strain rates due to octahedral and cubic slip respectively, $\sigma_{\text{m}}$ is the mean stress and $\sigma_c$ is an equivalent stress defined as

$$\sigma_c = \left( \frac{3}{2} S_{ij} S_{ij} \right)^{1/2}, \quad (14)$$

where $S_{ij}$ are the deviatoric components of the stress tensor. In equations (12) and (13), $w_{\text{oct}}$, $w_{\text{cub}}$, $\hat{a}_{\text{oct}}$ and $\hat{a}_{\text{cub}}$ are functions calibrated from the FE unit cell void calculations (Dennis 2000, Busso et al. 2001b) and $n$ is the instantaneous material strain rate sensitivity defined as

$$n = \frac{\partial (\log \dot{\varepsilon})}{\partial (\log \sigma_c)}. \quad (15)$$

Here, $\dot{\varepsilon}$ is the equivalent strain rate.

4.2. Crack initiation criterion and macroscopic damage

In this work, it is assumed that microcracks such as that shown in figure 5 (b) initiate when a critical value of the equivalent inelastic strain is reached at the void surface. For convenience, this condition can be equivalently expressed in terms of a corresponding critical void volume. The unit-cell FE studies (Dennis 2000, Busso et al. 2001b) revealed that, for a fixed critical microstrain, the corresponding critical void volume depends on the global triaxiality $\sigma_{\text{m}}/\sigma_c$. Then, the normalized critical void volume may be expressed as

$$\frac{V_c}{V_0} = 1 + b \exp \left( c \frac{\sigma_{\text{m}}}{\sigma_c} \right), \quad (16)$$

where $V_c$ and $V_0$ are the critical and initial void volumes, respectively. Thus, the criterion for crack initiation (at the microscale) is given when $V \geq V_c$, where $V$ is obtained explicitly by integrating equation (11). The parameters $b$ and $c$ in equation (16) were calibrated from notched bars loaded under creep conditions in conjunction with 3D FE simulations of the actual tests. For further details, see Dennis (2000) and Busso et al. (2001b).
Figure 6 shows the average uniaxial equivalent or failure strain obtained by integrating up to the moment when the volume \( V = V_c \), normalized by the material ductility in a [001] uniaxial tensile test, as a function of the stress triaxiality and imposed strain rate. Note that either or both far-field [001] and [010] loadings are considered when integrating equations (11)–(16) and that, owing to the normalization, the curves are independent of the initial void volume. It may be seen from the figure that the triaxiality dependence of the failure strain is almost independent of strain rate over the range examined and that, for \( \sigma_m / \sigma_c > 1.0 \), the failure strain is weakly dependent on triaxiality. The response of the conventional isotropic failure model proposed by Yatomi et al. (2002), based on the well-known Rice–Tracey (1969) void growth model for isotropic elastoplastic materials, has also been included in the figure for comparison. It can be seen that the responses of the current formulation and the isotropic formulation are similar at low triaxialities \( (\sigma_m / \sigma_c \leq 0.5) \) while they differ at higher triaxialities.

Following the above definition of the void growth rate and critical normalized void volume, one can define a corresponding macroscopic damage variable \( D \):

\[
D = \frac{V - V_0}{V_c - V_0}.
\]  

This definition of damage ensures that \( D = 0 \) for \( V = V_0 \) and \( D = 1 \) for \( V = V_c \).

The above void growth formulation, initiation criterion and corresponding damage variable have been implemented numerically into the FE user-defined material subroutine for the CMSX4 alloy. In the absence of any experimental data, this calibration will also be assumed to hold for the \( \gamma \) region.
§ 5. Application to an oxidizing compact tension fracture specimen

5.1. Finite-element model of the compact tension specimen

To investigate the interaction between environmental effects and the local conditions for crack propagation in a real component, the fracture behaviour of a typical CT fracture specimen containing either a blunt notch or a sharp crack was considered. The dimensions of the CT specimen are shown in figure 7. As indicated in figure 7(a), the plane of the specimen is normal to the [001] crystallographic axis and the crack lies on the (010) plane. Because of symmetry, only half the specimen needs to be modelled (see figure 7(b)). The local FE mesh refinement used to account for the notch shape and sharp crack are shown in figures 7(c) and (d), respectively. Figure 7(c) contains a semicircular notch of 0.1 mm radius and a crack-length-to-specimen-width ratio $a/W$ of 0.51, and figure 7(d) shows a sharp crack with an $a/W$ ratio of 0.55. For the latter, the crack is positioned 1 mm ahead of the notch, giving an overall initial crack length of 14.3 mm. This case represents the effect of a sharp fatigue or creep crack initiating from a blunt notch. The small increase in crack length leads to a 10% increase in the stress intensity factor $K$ at the same load. Note that the dimensions $h_1$ in Figure 7(a) are the same in both cases. The FE meshes are composed of 630 and 670 plane-strain elements for the notch and sharp crack CT specimens, respectively. Furthermore, the smallest element size, chosen to be of the order of a typical initial void size, is approximately 15 μm. In the mass diffusion analysis, four-node linear elements are used while, in the stress analysis, they are quadratic plane strain hybrid elements with reduced integration and linear pressure.

Figure 7. (a) CT specimen geometry (all dimensions in millimetres), (b) FE mesh and (c), (d) focused FE mesh for (c) the notch and (d) the sharp crack.
To solve the overall oxidation–deformation problem, sequentially coupled mass diffusion stress analyses were performed. During the first stage of the analysis, the nodal Al concentrations are stored as a function of time. The mechanical properties at each integration point are then chosen based on the current values of the local concentrations. If the concentration is above the upper solubility limit of Al in γ, the material properties are assigned to be those of γ + γ’; otherwise they are defined as those of pure γ (see figure 2(b)). The use of a sequential analysis, although significantly more efficient computationally than a fully coupled analysis, is unable to account for the changes in the surface geometry due to deformation and the effect of pressure on the rate of diffusion. As will be seen, the levels of deformation are relatively small and are not expected to have a significant influence on the diffusion process. Moreover, data on the effects of stress on diffusivities are not available for this alloy. Nevertheless, as will be shown, the levels of hydrostatic pressure around the notch or sharp crack are relatively small to give rise to stress-assisted diffusion of Al.

The FE analyses are conducted at a constant temperature of 950°C. In the mass diffusion analysis, the Al flux normal to the crack faces (see figures 7(c) and (d)) is prescribed via equation (7). A constant load P is applied to the specimen (see figure 7(a)) over typically 1000 h. Several analyses have been carried out for loads of 2, 3 and 4 kN, corresponding to stress intensity factors for the notched specimen of $K = 10, 15$ and 20 MPa m$^{1/2}$, respectively. In the next section, the majority of the results to be shown will be those for the 4 kN load case, with a few selected results for the lower loads.

5.2. Results and discussion

The results of the coupled oxidation–deformation-damage FE analyses will be discussed next. Firstly, the notch analyses with and without oxidation will be shown in terms of stresses, strains and damage at the mesoscale. This will include the effects of oxidation on the hydrostatic and opening stresses, and on the localization of inelastic deformation and damage. Finally, the same effects linked to a sharp crack rather than a blunt notch and the implications of these findings in relation to time to crack initiation will be discussed.

5.2.1. Blunt notch with no oxidation

The profile of the normal stresses $\sigma_{010}$ as a function of distance from the notch tip at different times is shown in figure 8. It can be observed that the peak stress drops by more than 50% in the first 50 h, from 1650 to 800 MPa, owing to stress relaxation. The stress decreases by a further 30% over the next 950 h, which is close to the steady-state stress distribution. The position of the peak stress also moves from 20 μm from the notch root at $t = 0$, to approximately 260 μm at 1000 h.

In figure 9, contours of the accumulated inelastic strain around the notch region due to octahedral slip $\varepsilon_{oct}^p$ and cubic slip $\varepsilon_{oct}^p$ are presented. The contours are plotted on the deformed configuration and the maximum notch opening is approximately 7 μm. Here, the uniaxial equivalent strain measures $\varepsilon_{oct}^p$ and $\varepsilon_{cubic}^p$ are calculated from

$$\varepsilon_{oct, cub}^p = \left[\frac{2}{3} (F_{oct, cub}^p - 1) : (F_{oct, cub}^p - 1)\right]^{1/2}, \quad (18)$$
where $\mathbf{F}_{\text{oct}}$ and $\mathbf{F}_{\text{cub}}$ are obtained by integrating equation (6) for the octahedral and cubic slip families, respectively, and $\mathbf{I}$ is the second-order identity tensor.

It can be seen in figure 9 that the peak strains at the notch root are highly localized (over distances less than 250 µm) and range between 1% and approximately 10%. The strain patterns are typical of glide on these systems: the peak octahedral strains lie on $\{111\}$ planes (intersecting the $(001)$ plane at $\pm 45^\circ$ in figure 9(a)) while the peak cubic strains lie on $\{100\}$ planes ($90^\circ$ in figure 9(b)). It is also worth noting that, although octahedral slip systems are dominant under $(001)$ uniaxial loading, there is a significant contribution to the inelastic strain from cubic slip since the resolved shear stress on these slip systems is no longer zero due to the presence of the notch.

Figure 10 shows contours of damage as defined by equation (17) at two different times. The size of the damaged region increases with time as the critical strain for crack initiation is reached in elements further ahead of the notch. Regions where damage is equal to 1 are taken to have failed. The damage patterns suggest...
that damage is strongly influenced by the accumulated inelastic strains (see figure 9) and that crack growth will not occur straight ahead of the notch but rather at a $\pm 45-60^\circ$ angle. This is consistent with experimental observations on fatigue tests done on CMSX4 notched bars (Boubidi, 2000). It was also found that the peak hydrostatic stress $\sigma_m$ occurs directly ahead of the notch tip while the peak inelastic strains are at an angle to the notch, as shown in figure 9. However, the magnitude of the triaxiality $\sigma_m/\sigma_e$ in the notch region is in the range $0.6 < \sigma_m/\sigma_e < 2.5$. As can be seen in figure 6, over this triaxiality range the failure strain is relatively independent of triaxiality. Therefore the magnitude and extent of damage are expected to depend more strongly on the localization of inelastic strain in this region.

5.2.2. Blunt notch with oxidation

When oxidation is accounted for, the distribution of the normal stresses near the notch tip is affected by the formation of the pure $\gamma$-phase region and the associated change in material properties (see figure 2 (b)). The effect of oxidation on the notch tip stresses is shown in figure 11, where the stress profiles are those at 1000 h. At this time, the size of the $\gamma$ region ahead of the notch is approximately 110 $\mu$m. It can be seen that oxidation causes the $[010]$ stress component at the notch root to decrease by more than 30%, even though its peak value, located outside the $\gamma$ region, is only 5% lower. Further away from the notch surface, oxidation causes the stresses to increase as they are redistributed on to the higher strength $\gamma + \gamma'$ region.

Figure 12 shows the effect of oxidation on the hydrostatic stress contours around the notch root. Following the trend of the $[010]$ stress component in figure 11, the region of high hydrostatic stress ahead of the notch contracts due to oxidation, and the magnitude of the peak hydrostatic stress decreases by about 7% (from $430$ to $400$ MPa).

The corresponding evolution of damage with time is shown in figure 13. A comparison between figures 10 (a) and 13 (a) reveals that, at short times ($t \leq 50$ h), the damage contours are not significantly affected by oxidation, as the $\gamma$ region at this time is only a few microns in size. However, at longer times ($t = 1000$ h), damage extends further when oxidation effects are accounted for (see for example figures 10 (b) and 13 (b)).

5.2.3. Sharp crack with oxidation

Here, the effect of notch acuity on the strain fields and damage evolution will be discussed. The accumulated inelastic strain contours due to octahedral and cubic slip
Figure 11. Effect of oxidation on the distribution of the [010] stress versus the distance from the notch root.

Figure 12. Effect of oxidation after 1000 h at 950°C on the hydrostatic stress for an applied load of 4 kN.

Figure 13. Contours of damage after oxidation for (a) 45 h and (b) 1000 h at 950°C under an applied load of 4 kN.
systems for a sharp crack undergoing oxidation are shown in Figure 14. The presence of the sharp crack strongly affects the patterns of the strain distribution around its tip, being more localized than for the blunt notch case (see for example Figures 14(a) and (b)). For instance, the peak cubic strains localized very closely on the (100) plane directly ahead of the crack tip.

The effect of the sharp crack can also be seen in the local damage distributions (Figure 15). A comparison between Figures 13(b) and 15 reveals quite different patterns of damage. For the sharp crack geometry, the damage pattern appears to follow the strain pattern even more closely and the peak damage is now at an angle close to \(90^\circ\) relative to the crack. Furthermore, the crack opening at this time is about 5 \(\mu m\), a small fraction of the notch radius in the previous analysis.

5.3. Prediction of time to crack initiation

As discussed previously, when damage reaches unity \((D = 1)\), it corresponds to the initiation of a microcrack within an element. It is assumed that, once a microcrack initiates, it grows rapidly, leading to the failure of that element. Since the condition for microcrack initiation depends on element size (particularly in the case of a sharp crack, as a decrease in the crack tip element size will lead to a shorter initiation time), a length scale must be assigned to define the onset of crack initiation on the notch scale (mesoscale). In this work, the time to crack initiation is defined as the time when the fully damaged region \((D = 1)\) extends 50 \(\mu m\) ahead of the initial notch or crack. This length is chosen as half a typical size of the facet cracks shown in Figure 5(c).
With the above definition, it is now possible to predict the time to crack initiation for the notch and crack geometry with and without oxidation effects (figure 16). The effects of both oxidation and notch acuity appear to be significant, with that due to oxidation being most significant for the blunt notch. For an applied load of 4 kN, the ratio of the time to crack initiation without oxidation to that with is about three, while that same ratio for the sharp crack is close to one. However, at lower loads, when cracks take longer to initiate, they are exposed to the oxidizing environment for longer times, and the effect of oxidation becomes important for the sharp-crack case.

The strong dependence of the time to crack initiation on notch acuity may be observed by comparing figures 16(a) and (b). For an applied load of 4 kN, the time to initiation is approximately 50 times faster for the crack than for the notch (note that this effect cannot be explained by the slightly higher $K$ value for the former). This result suggests that, once crack initiation occurs near a notch, the

![Figure 16](image-url)

Figure 16. Effect of oxidation on the time to crack initiation versus the applied load for the (a) blunt notch and (b) the sharp crack. The arrows indicate that initiation did not occur within the calculation period.
crack growth rate will increase rapidly (i.e. up to 50 times). Under these conditions, the life of a component may be controlled primarily by the time to initiate a micro-crack rather than by the time for such a crack to grow to a critical size.

§6. Conclusions

Coupled oxidation–deformation FE analyses have been carried out to examine the development of mesoscale damage, linked to a microcrack initiation criterion, in CT specimens of an Ni-based single-crystal superalloy. A rate-dependent crystallographic formulation has been used to model the mechanical behaviour of the crystal with both octahedral and cubic slip systems potentially active. It has been shown that the time to initiate a crack in a notched specimen may be reduced by a factor of three owing to oxidation effects. Furthermore, an increase in notch acuity was found to lead to a significant reduction in the time to crack initiation. Experimental studies are ongoing to validate these numerical studies and to provide further insight into crack initiation and growth mechanisms in single-crystal superalloys.

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References


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