BRITTLE INTERGRANULAR FAILURE IN 2D MICROSTRUCTURES: EXPERIMENTS AND COMPUTER SIMULATIONS

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Abstract—Brittle intergranular fracture (BIF) is a common mode of failure for monolithic ceramics and intermetallics, as well as for some refractory metals and metals exposed to environmental corrosion, stress corrosion cracking or high temperature creep. As interest in applications for these materials grows, research programs have been developed to characterize and predict their fracture behavior. In order to experimentally quantify the effects of microstructure on local BIF, systems which have a minimum number of variables which influence fracture must be used. Evaluation of materials with two dimensional (2D) microstructures can considerably reduce the complexity of the system. In addition, providing a biaxial stress state in the 2D microstructure ensures that all boundaries experience exclusively Mode I loading prior to failure. Biaxial elastic loading of this simplified microstructure allows the calculation of (a) local stress and strain fields (and their concentrations) prior to failure, as well as (b) prediction of grain boundary strength criteria, and (c) prediction of intergranular crack paths. This can be achieved by conducting computer simulations of the experimentally observed fracture phenomena in polycrystalline specimens having a given texture and microgeometry. Since the grain boundary strength criterion is easily controllable in such simulations, it can be inferred by a comparison with actual experimental results. The latter is complemented by results on fracture of materials with very weak grain boundaries, thus providing a clear perspective on evolution of the failure process for varying degrees of embrittlement. Copyright © 1996 Acta Metallurgica Inc.

1. INTRODUCTION

In brittle ceramics, intermetallics and refractory metals, failure usually occurs via brittle intergranular fracture (BIF). Cracks initiate at structural or chemical heterogeneities and propagate along adjacent grain facets in a manner which is a complex function of the local triaxial stresses, facet orientations and grain boundary strengths. Increases in bulk toughness have been achieved in these materials through observation of local microstructural interactions that occur during fracture. For example, crack bridging and crack deflection have been observed to increase the energy necessary to propagate a crack [1]. Consequently, tougher ceramics have been designed with microstructures prone to experiencing increased bridging or kinking during fracture.

An inherent problem with studying local initiation and in situ crack propagation of brittle materials is that the site where failure occurs is not known a priori. It is virtually impossible to recognize and observe fracture at a scale where individual microstructural events dominate the crack path. Introducing a controlled flaw through Knoop or Vickers indentation overcomes this difficulty but deactivates the nucleation sites intrinsic to the material. Also, current methods for studying in situ crack propagation only allow the surface of a specimen to be observed. Components of a propagating crack which exist below the surface of observation are not detected. Experiments of this type yield limited information about local BIF.

A class of brittle materials with two-dimensional (2D) microstructures provides a unique opportunity to observe and quantify local microstructural effects on intergranular crack propagation. By fracturing materials with 2D microstructures, through thickness crack initiation and propagation can be observed and measured directly on the surface of a specimen. In addition, knowledge of local grain boundary toughness and inclination relative to the advancing crack tip provides insight into how the crack interacts with the microstructure during fracture.
The criteria necessary to define an approximately 2D microstructure were described in earlier work [2]. A specimen must be very thin in one dimension compared to the other two (i.e. a sheet or foil.) Also, the average size of the grains in the sheet must be significantly larger than the thickness of the sheet. Finally, the grain boundaries must be approximately orthogonal to the plane of the sheet. These criteria are illustrated in Fig. 1.

Through thermomechanical processing, an approximately 2D microstructure may be produced in metal sheets. Many examples of these 2D microstructures can be found in the literature [2–7]. Grah et al. [2] developed and described 2D microstructures in 3.25% Si-Fe sheets by cold rolling followed by annealing to induce primary recrystallization.

Wetting by liquid metals has been shown to cause grain boundary embrittlement in metals [8,9]. Specifically, liquid gallium strongly embrittles aluminum at temperatures above the aluminum–gallium eutectic at 26.6°C [10–15]. Liquid gallium which contacts aluminum beneath the oxide spreads along the oxide–metal interface and through grain boundaries by a combination of eutectic formation and wetting. This high spread rate is facilitated by a low eutectic at 26.6°C [10–15]. Liquid gallium which contacts aluminum beneath the oxide spreads along the oxide–metal interface and through grain boundaries by a combination of eutectic formation and wetting. This high spread rate is facilitated by a low eutectic at 26.6°C [10–15]. A liquid eutectic film remains on penetrated boundaries and reduces cohesion sufficiently that intergranular fracture occurs before plastic flow under applied stress.

Marya and Wyon [17] have shown that the catastrophic embrittlement caused by gallium in aluminum can be reduced through subsequent anneals. During anneals, they determined that gallium in the grain boundary eutectic film diffuses into the adjacent grains. After sufficient annealing times at elevated temperature, complete ductility is restored to the aluminum metal.

Gallium embrittled aluminum sheets with 2D microstructures provide a good experimental model for brittle intergranular fracture in two dimensions. The ability to tailor the scale of the microstructure, extent of embrittlement and area of embrittlement provides an opportunity to observe and quantify the effects of microstructure on initiation and local brittle intergranular fracture. Selective embrittlement of aluminum sheets facilitates the application of biaxial stresses during fracture tests. All of these capabilities also provide a system which is nicely amenable to modelling by 2D numerical analysis.

The above mentioned simulations rely on fine mesh finite-difference-type approximations of continuum media, i.e. crystals in the present study. Any crystal anisotropy can be modeled, so that the mechanics of deformation and fracture of a given polycrystal can be simulated in analogy to existing models of strength of atomic lattices with defects, see e.g. Ref. [18]. Basic ideas of this research were outlined in Ref. [19].

2. EXPERIMENTAL PROCEDURES

2.1. Two-dimensional microstructures

The aluminum used in this investigation was annealed foil, 254 µm thick, (98.5% including 1% Si and Fe) from Alfa Chemical Company. The foil was cut into 76.2 mm × 76.2 mm sheets and annealed at 225°C for 16 h and 400°C for 2 h to remove cold work.

Annealed strips were clamped by 75 mm wide grips and strained various amounts by tensile elongation in a Syntec-MTS tensile loading rig at a strain rate of 0.1/min. Three specimens were strained 3, 5, 7, 9 and 12%. Additional specimens were strained 12% for embrittlement and biaxial loading.

After the strain was introduced, specimens were annealed at 600°C for 2 h to ensure complete primary recrystallization. Six holes were punched in the sheet in a hexagonal pattern to provide anchors in the loading rig. Specimens were chemically etched within an aqueous solution of 5% NaOH and 4%NaF. The caustic etch was carried out at approximately 95°C for 20 s to remove the oxide and reduce specimen thickness to ~240 µm. Samples were rinsed in hot water and a solution of 70% HNO₃ and 30% H₂O to remove the smut. Microstructure was observed through grain contrast under oblique lighting. Grain boundary orthogonality was verified by graphographing both sides of an etched specimen. One of the negatives was flipped over and then the negatives of both sides were aligned so that the facets were superimposed. The resulting mismatch between boundary projections was then observed as a measure of orthogonality. Orthogonality was further verified by observing the angle of fractured facets on optical and scanning electron microscope.

2.2. Liquid metal embrittlement of aluminum

Specimens were embrittled by wetting them with 99.999% pure gallium. Gallium was applied in a circular patch 20–24 mm in diameter in the center of a specimen. Wetting was facilitated by scratching the surface area lightly with a razor blade surrounded with liquid gallium so that the oxide was displaced. Excess gallium was allowed to remain on the wetted surface at 50°C for 10 min.
After exposure, excess gallium was wiped off with a paper towel. Specimens were then immersed in water for 20 s. The water reacts with aluminum in the liquid gallium on the surface. Hydrogen gas produced in the reaction quickly lifts any remaining gallium from the surface. After embrittlement, specimens were annealed to induce partial de-embrittlement. De-embrittlement parameters were determined by preparing and fracturing an array of specimens with annealing times from 1 to 60 min and annealing temperatures of 100 and 150°C. Biaxially loaded specimens with optimized properties were de-embrittled for 10 min at 150°C or 45 min at 100°C.

2.3. Biaxial fracture

The biaxial loading rig is shown schematically in Fig. 2. A variable speed, motor-driven screw forces a push plate and teflon ring up against the fixed specimen. When the embrittled specimen is properly attached in the rig and loaded, stresses are transferred elastically and equibiaxially to the embrittled region by the surrounding ductile regions. A miniature (2200 N) load cell between the push plate and the teflon ring measures the applied load. The load is converted to biaxial stress through elastic calibrations. Signals from the load cell were routed to a Macintosh computer via an analog input board, converted to biaxial stress and saved as data files along with time and date files. Load resolution is better than 50 g (~0.05 MPa).

The rig was designed so that crack propagation could be observed and recorded in the embrittled region as a specimen was loaded. Figure 3 is a schematic representation of the loading rig, measuring apparatus and recording apparatus. A Javelin CCD camera sends images to a high resolution 14" monitor. The CCD camera has a resolution of 480 lines while the monitor is rated at 450+ lines of vertical resolution. A manual zoom lens on the video camera provides effective magnifications of between 2.2× and 40×. The video signal is also sent to a VCR so that crack propagation can be recorded in real time. A special time and date video titler is incorporated into the system so that the time fracture events which occur on the screen can be correlated with remote stress and time files that are recorded simultaneously during a fracture test. Micrographs of fracture are taken by photographing images on the monitor with a 35 mm SLR camera. After testing, fracture surfaces of selected specimens are observed on an SEM.

Just prior to mounting in the rig, specimens are coated on the observed surface with a light layer of flat white enamel paint. This paint greatly improves the contrast of the growing cracks against the specimen surface.

Each embrittled specimen was loaded and fractured within 2 h from the time of embrittlement. At the start of the experiment, data acquisition of the time and the applied biaxial stress was begun simultaneously with the VCR recording of the experiment. Initially, magnification was kept at ~10× so that the entire range of possible initiation sites could be observed.

After fracture, crack initiation and propagation were studied by stereological analysis of the fracture movies and 35 mm photographs in conjunction with the biaxial stress-time data files. Microstructures were analyzed on SEM by observing the sides of specimens not coated with white paint.

2.4. Determination of facet misorientations

Grain orientations were measured in selected specimens at the National Institute of Standards and Technology in Gaithersburg, Maryland. The Kikuchi surface electron backscattering technique was used.
employed. Fractured specimens were etched and photographed, and grain boundary maps of the embrittled regions were produced. The crystallographic orientation of grains was measured relative to the straining direction and the sheet normal.

The misorientation matrix of adjacent grains in the specimens was calculated. A program was written to compare the misorientation matrices to that of each CSL relationship from \( \Sigma = 1 \) to \( \Sigma = 49 \). The CSL matrices were obtained from a list provided by Grimmer [20]. Boundaries with CSL relationships were determined by calculating the deviation of boundary misorientation from each CSL misorientation and comparing that deviation to Brandon’s criterion [21].

3. COMPUTER SIMULATIONS
3.1. The finite mesh spring network model
3.1.1. Preliminaries. The presence of many crystals in the specimen and their inherent anisotropy precludes any simple analytical solution of the crack propagation problem. We thus adopt a method analogous to those used in modelling of failure of atomic lattices and composite materials, see e.g. Refs [18, 22]. Such investigations are based, in principle, on simulations of breakdown of a lattice of springs, a spring network, in which bonds are progressively being taken out as they exceed the strength criterion. The process of crack elimination represents thus a growth of a crack or a field of cracks.

In the case of atomic lattices, nodes represent atoms and springs their interactions—usually in a linear approximation. In the case of composites, the spring network represents a discretization of continuum phases with the springs adjusted locally according to the specific elasticity tensors of all the phases. In the following, we follow the latter picture in the sense that a polycrystal is a composite of a number of anisotropic phases (i.e. particular crystals).

Thus, we first need to develop a spring network model of anisotropic elasticity. This is done, in the framework of infinitesimal strains, by introducing a regular (equilateral) lattice (Fig. 4) made of linear elastic normal (central force) and angular springs. This lattice may be covered by hexagonal unit cells centered at its nodes, whereby each cell consists of six angular springs and halflengths of the normal springs that actually belong to this cell. These halflengths are assigned spring constants \( c_n^s \), while the former ones are assigned spring constants \( c_n^e \), where \( n \) is the index of a given spring, \( n = 1, 2, \ldots, 6 \). The unit vectors \( \hat{x} \) and angles \( \theta \) with the coordinate axis \( x \), of all the \( e \) springs are given for the first three springs as

\[
\begin{align*}
\theta^1 &= 0^\circ & \hat{r}_1 &= 1 & \hat{r}_2 &= 0 \\
\theta^2 &= 60^\circ & \hat{r}_1 &= \frac{1}{2} & \hat{r}_2 &= \frac{\sqrt{3}}{2} \\
\theta^3 &= 120^\circ & \hat{r}_1 &= -\frac{1}{2} & \hat{r}_2 &= \frac{\sqrt{3}}{2}.
\end{align*}
\]  

(1)

The other three springs \( (n = 4,5,6) \) must, by the requirement of symmetry with respect to the center of the unit cell, have the same properties as \( n = 1,2,3 \), respectively. All the \( s \) springs are of length \( \ell \), that is, the spacing of the triangular mesh is \( 2\ell \). Thus, the area of the hexagon cell \( A = 2\sqrt{3}\ell^2 \).

Any \( \beta \) spring acts between two contiguous \( e \) springs that are incident onto the same node and thus represents a constraint on the rotation of the normal springs. Also by the requirement of symmetry, only three of the \( \beta \) springs (say \( n = 1, 2, 3 \)) are independent.

It is seen that three models can be defined: (i) an \( e \)-model with \( \beta \)-springs absent; (ii) a \( \beta \)-model with \( e \)-springs absent; and (iii) an \( e\beta \)-model with all the springs present. In the following we derive these three models in detail, and then show how the general model reduces to the well known spring network model of isotropic elasticity.

3.1.2. Central force interactions: the \( e \)-model. The relation of elastic properties between the continuum and the discrete model will be derived from the equivalence of energy of both models. For the continuum model, the elastic (strain) energy \( E \) of the hexagon is

\[
E = \frac{A}{2} C_{ijkl} \epsilon_{ij} \epsilon_{kl}.
\]  

(2)
where $C_{ijkl}$ is the fourth order stiffness tensor, due to $\alpha$-springs only, and $e_k$ is the linear strain tensor assumed to be uniform within the unit cell.

Now, let $OL$ represent the $n$th spring of stiffness $\alpha_e$, with $O$ being at the cell center and $L$ being on the cell boundary (Fig. 4). $N$ is directed along the axis of the spring and $T$ tangential to it. Displacements of $L$ along $N$ and $T$ are $u_N$ and $u_T$, respectively.

The force $P$ in the spring $\alpha_e$ is related to $u_T$ by

$$P = \sigma_e u_T$$

so that the strain energy in the spring is

$$E_e = \frac{1}{2} P u_T = \frac{1}{2} \alpha_e N_T N_T e_N e_N$$

and the total energy of all six $\alpha$ springs is

$$E = \sum_{n=1}^{6} E_e = \frac{1}{2} \sum_{n=1}^{6} \alpha_e N_T N_T e_N e_N.$$

Comparing expressions (4) and (5), the elastic moduli of a material represented by the discrete spring model are found as

$$C_{ijkl} = \frac{1}{2\sqrt{3}} \sum_{n=1}^{6} \alpha_e N_T N_T e_N e_N.$$

3.1.3. Angular interactions: The $\beta$-model. We now consider the unit cell with six $\beta$-springs present. With reference to Fig. 4(b) we see that $\Delta \theta$ is the angle change in the location of an $\alpha$ spring when the point $L$ moves to the point $L'$, i.e. between $I$ and $I'$. It is seen that

$$1 \times u = 1 \times l' = 1 \cdot \Delta \theta$$

which leads to

$$\Delta \theta = \frac{1}{i} x u = \frac{1}{i} e_{ijk}l'u_i = \frac{1}{i} e_{ijk}l'u_i$$

where $e_{ijk}$ is the permutation tensor and $i,j,k = 1,2$. The angle change between two contiguous $\alpha$ springs is measured by $\Delta \theta = \theta^{n+1} - \theta^{n}$. Substituting equation (7) into (8), we get for the strain energy stored in $\beta^n$

$$E^n = \frac{1}{2} \beta^n e_{ijk} e_{ijk} (l_i l_j - l_i l_j)^2$$

and, noting the $e-6$ identity, the total energy in a cell with six angular springs is

$$E = \sum_{n=1}^{6} E^n = \sum_{n=1}^{6} \frac{1}{2} \beta^n e_{ijk} e_{ijk} (l_i l_j - l_i l_j)^2$$

where superscript $(n+1)$ takes the value of 1 when $n = 6$. Equation (10) is really an expression of the strain energy of the unit cell of discrete model. In order to get the expression for elastic moduli, the symmetry with respect to $i$ and $j$, $k$ and $l$, as well as $ij$ and $kl$ should be satisfied, so that upon appropriate permutations equation (10) can be rewritten as follows

$$E = \frac{1}{2} \sum_{n=1}^{6} \left\{ (\beta^n + \beta^{n-1}) e_{ijkl} e_{ijkl} - (\beta^n + \beta^{n-1}) e_{ijkl} e_{ijkl} + e_{ijkl} e_{ijkl} e_{ijkl} e_{ijkl} + e_{ijkl} e_{ijkl} e_{ijkl} e_{ijkl} \right\}$$

By comparing the above with the energy of a unit cell

$$E = \frac{A}{2} C_{ijkl} e_{ijkl}$$
the elastic constants of the equivalent continuum model can be inferred as

\[ C_{ij} = \frac{1}{2\sqrt{3}} \sum_{n=1}^{b} (\beta^n + \beta^{-n-1}) \delta_{i,j} \rho_{i} \rho_{j} \rho_{k} \rho_{l} \]

- \left( \beta^n + \beta^{-n-1} \right) \rho_{i} \rho_{j} \rho_{k} \rho_{l}

- \beta \delta_{i,j} \rho_{i} \rho_{j}^{-1} \rho_{k} \rho_{l}^{-1} \rho_{t} \rho_{k}^{-1} \rho_{l}^{-1} \rho_{t}^{-1}

- \beta \delta_{i,j} \rho_{i} \rho_{j}^{-1} \rho_{k} \rho_{l}^{-1} \rho_{t} \rho_{k}^{-1} \rho_{l}^{-1} \rho_{t}^{-1}.

(13)

Here superscript \((n - 1)\) takes the value of 1 when \(n = 1\).

3.1.4. The general \(\alpha\beta\)-model. By superposition of equations (6) and (13), the elastic moduli of a cell of continuum are expressed in terms of spring constants of the general anisotropic \(\alpha\beta\)-model as follows

\[ C_{ij} = \frac{1}{2\sqrt{3}} \sum_{n=1}^{b} \frac{1}{\beta^n + \beta^{-n-1}} \delta_{i,j} \rho_{i} \rho_{j} \rho_{k} \rho_{l} \rho_{m} \rho_{n} \rho_{o} \rho_{p} \rho_{q} \rho_{r} \rho_{s} \rho_{t} \rho_{u} \rho_{v} \rho_{w} \rho_{x} \rho_{y} \rho_{z} \]

- \left( \beta^n + \beta^{-n-1} \right) \rho_{i} \rho_{j} \rho_{k} \rho_{l} \rho_{m} \rho_{n} \rho_{o} \rho_{p} \rho_{q} \rho_{r} \rho_{s} \rho_{t} \rho_{u} \rho_{v} \rho_{w} \rho_{x} \rho_{y} \rho_{z}

- \beta \delta_{i,j} \rho_{i} \rho_{j}^{-1} \rho_{k} \rho_{l}^{-1} \rho_{m} \rho_{n} \rho_{o} \rho_{p} \rho_{q} \rho_{r} \rho_{s} \rho_{t} \rho_{u} \rho_{v} \rho_{w} \rho_{x} \rho_{y} \rho_{z}

- \beta \delta_{i,j} \rho_{i} \rho_{j}^{-1} \rho_{k} \rho_{l}^{-1} \rho_{m} \rho_{n} \rho_{o} \rho_{p} \rho_{q} \rho_{r} \rho_{s} \rho_{t} \rho_{u} \rho_{v} \rho_{w} \rho_{x} \rho_{y} \rho_{z}.

(14)

This provides the basis for a spring network representation of an anisotropic material; it is also a generalization of the Kirkwood model [23] of an isotropic material that is discussed below.

3.1.5. The isotropic material: Kirkwood model. For the sake of completeness we also provide in this section the special case of an isotropic material, which is originally due to Kirkwood [23]. We recover this case by assigning the same \(\alpha\) to all the normal and the
same $\beta$ to all the angular springs, so that equation (14) simplifies to

$$C_{\text{ang}} = \frac{\alpha}{2\sqrt{3}} \left( \sum_{i=1}^{\infty} \frac{a_i}{r_i^3} \right)$$

$$\times \frac{\beta}{2\sqrt{3}} \sum_{i=1}^{\infty} \left( 2\delta_i a_i r_i^{-1} + \frac{a_i}{r_i^3} + \frac{1}{r_i^3} \right)$$

$$- \delta_i a_i \frac{a_i}{r_i^3} + \frac{1}{r_i^3}$$

whereby we find

$$C_{1111} = C_{2222} = \frac{1}{2\sqrt{3}} \left( \frac{9}{4} \alpha + \frac{19}{4\beta} \right)$$

$$C_{1122} = C_{2211} = \frac{1}{2\sqrt{3}} \left( \frac{3}{4} \alpha + \frac{19}{4\beta} \right)$$

$$C_{1212} = \frac{1}{2\sqrt{3}} \left( \frac{3}{4} \alpha + \frac{19}{2\beta^2} \right)$$

It is also observed that the condition

$$C_{123} = \frac{1}{2}(C_{1111} - C_{1122})$$

is satisfied, so that there are only two independent elastic moduli.

The $\alpha$ and $\beta$ constants are related to the planar bulk and shear moduli as

$$\kappa = \frac{1}{2\sqrt{3}} \left( \frac{3}{4} \alpha + \frac{19}{4\beta} \right)$$

$$\mu = \frac{1}{2\sqrt{3}} \left( \frac{9}{4} \alpha + \frac{19}{4\beta} \right)$$

where $\mu$ is the shear modulus and $\kappa$ is the two-dimensional bulk modulus. It is noted here that the angular springs have no effect on the $\kappa$, i.e. the presence of angular springs does not change the area of material. We refer to Ref. [24] for a detailed discussion of relations between planar, plane stress, plane strain and 3D isotropic elasticity. Suffice it to note here that the conventional 3D Poisson's ratio $\nu_{3D}$ is calculated in terms of the planar one ($\nu$) as

$$\nu/(1+\nu),$$

whereby $\nu$ is seen to range from $-1$ to $+1$, in contradistinction to $\nu_{3D}$ which ranges from $-1$ to $1/2$. The formula for a planar Poisson's ratio is [24]

$$\nu = \frac{\kappa - \mu}{\kappa + \mu}$$

which, in view of equation (18), becomes

$$\nu = \frac{\alpha r_1^2 - 3\beta}{3\alpha r_1^2 + 3\beta}$$

From equation (20) there follows the full range of Poisson's ratio which can be covered with this model. It has two limiting cases

$$\nu = \frac{1}{3}$$ if $\frac{\beta}{\alpha} \rightarrow 0$ \quad $\alpha$-model

$$\nu = -1$$ if $\frac{\beta}{\alpha} \rightarrow \infty$ \quad $\beta$-model.
Note that for materials of Poisson’s ratio above 1/3 one can use either the model from Keating [25] or another one of Day et al. [26]. The first one also uses an angular spring contribution, but of a different kind than that in the Kirkwood model. The second one uses an equilateral lattice of three types of central force springs, where each type has a honeycomb pattern.

3.2. Simulation of a polycrystal

3.2.1. Computer image analysis. An image of the polycrystalline specimen was scanned into an HP-computer workstation (715/750) and saved into a file with special format compatible with that of a “Starbase” graphics software. The entire domain of the polycrystal’s image was then colored in such a way that each crystal was given a different color associated with a numerical index. The purpose is to distinguish between different crystals so that we can assign properties (stiffness and strength) to all the springs of the spring-network according to which crystal they belong to.

3.2.2. Analytical calculations. In order to assign spring stiffnesses to any node of the spring network mesh, the anisotropic 3D stiffness tensor $C_{ijkl}$ for each crystal must be found according to its transformation (rotation) matrix $a_{ij}$ ($i,j = 1,2,3$); the latter was provided from the experiment. Thus, we start from the stiffness matrix $C_{kl}$ (i.e. in a matrix form) of an anisotropic aluminum crystal which is given as

$$C_{ijkl} = \begin{bmatrix}
1 & 0 & 0 & 0 \\
0 & 1 & 0 & 0 \\
0 & 0 & 1 & 0 \\
0 & 0 & 0 & 1
\end{bmatrix} \begin{bmatrix}
\frac{1}{\sqrt{3}} & \frac{1}{16\sqrt{3}} & \frac{1}{16\sqrt{3}} & \frac{1}{4} \\
0 & \frac{9}{16\sqrt{3}} & \frac{9}{16\sqrt{3}} & \frac{9}{4} \\
0 & \frac{1}{16} & \frac{1}{16} & 0 \\
0 & \frac{3}{16} & \frac{3}{16} & 0
\end{bmatrix}$

$$

$$

$$

$$

$$

While there was no ambiguity concerning the spring constant of any $a$-bond that entirely belonged to any given crystal domain, additional care had to be taken of the bonds that straddled the boundary of two crystals. The effective stiffnesses were assigned according to a series rule: $a = (1/2a_a + 1/2a_b)^{-1}$. Assignment of $\beta$-springs presented no such ambiguities.

3.2.3. Fracture simulation. The computer simulation of damage formation is now carried out by subjecting the fine mesh anisotropic $\alpha\beta$-model of the elastic polycrystal to the same loading conditions as applied in the experiment. The actual procedure involves the following steps.

(i) In order to account for the fact that the grain boundary regions are, due to the gallium penetration, substantially weaker than the crystals themselves, the strength of any $a$-bond crossing the boundary had to be reduced by a factor with respect to the strength of all the other $a$-bonds. This factor was taken here as 0.01. The strength of any boundary-crossing $a$-spring is taken as the weighted average of the respective strengths of two length portions of that spring belonging to both contiguous crystals.
(ii) Loading of our rectangle-shaped lattice (i.e., fine mesh such as the one of Fig. 5) was carried out by subjecting its boundary to kinematic boundary conditions

\[ u_i = \varepsilon_{ij} x_j \]  

where \( \varepsilon_i \) is the macroscopic strain corresponding to biaxial loading

\[ \varepsilon_{11} = \varepsilon_{22} = \varepsilon \quad \varepsilon_{12} = 0. \]  

(iii) The increasing loading conditions were simulated by raising \( \varepsilon \) by small increments \( \Delta \varepsilon \) in every next run of the simulation.

(iv) At every step we use a conjugate gradient method [27] to determine the state of equilibrium of the lattice, and next, we search for any grain boundary \( z \)-bond(s) whose (central) force exceeds the local fracture criterion. The latter is formulated, in general, as

\[ F \leq F_a, \]  

where \( F_a \) depends on the relative anisotropy of two contiguous crystals. If equation (27) is violated, the given bond is being removed from the lattice as well as its associated four angular springs—thus representing a crack—and the macroscopic strain \( \varepsilon \) is increased according to step (ii).

(v) The increase of \( \varepsilon \) by \( \Delta \varepsilon \) is conducted by first unloading the entire lattice, and then reloading it by strain \( \varepsilon + \Delta \varepsilon \).

It is possible that more than one bond meets the fracture criterion at any given step, in which case all such bonds have to be removed at the same time. This process is continued until the lattice is completely cracked.

Admittedly, the drawback of our spring-network-type approach is the inability to resolve the true stress concentrations right at the grain boundary corners and at the crack tips. Problems of that nature, however, are very difficult to resolve with other existing numerical methods too. This consideration, coupled with the possibility of an easy assignment of any crystal anisotropy on the mesh as well as the successes of this approach in simulations of atomic lattice fracture and damage of composites [28], motivates the choice of our simulation/solution scheme.

(The fracture step is performed along with the image analysis step in one FORTRAN code having some 3000 lines.)

4. RESULTS

4.1. Two-dimensionality of aluminum sheet specimens

The two dimensionality of the aluminum specimens was measured relative to the three criteria...
described earlier. Obviously, using sheets with length and width to thickness ratios well over 100 ensures that the first criterion is satisfied. By varying the amount of strain before the recrystallization anneal, a full range of average grain sizes could be induced in the sheets. The relationship between strain and grain size is shown in Fig. 6. As strain is varied between 3 and 12%, average grain size varies between 2 and 6.5 mm. These thicknesses correspond to grain size to thickness ratios of 8:27. For all strains, these ratios are significantly greater than one, the second criteria for two dimensionality. The final criterion for two dimensionality, grain boundary orthogonality, was analyzed by taking photos of both sides of a specimen. In Fig. 7, both sides of an aluminum specimen strained at 10% and annealed have been superimposed. The back and front side negatives have been overlaid in the print so that the boundary orthogonality can be measured. As evident in Fig. 7, very little mismatch exists between the front and back.

Fig. 11. SEM micrographs of the crack blunting events in Fig. 10. Note these photos are inverted relative to those in Fig. 10 since they were taken from the back, unpainted side of the specimen.
facet images. In fact, within the resolution of the micrographs, it was impossible to measure any mismatch through the thickness. This implies that the grains are highly orthogonal.

These results indicate that approximately 2D microstructures can be produced in aluminum sheet by the strain annealing technique.

4.2. Gallium embrittlement

Gallium caused catastrophic embrittlement of the sheet specimens after the initial treatment. Specimens loaded without de-embrittlement fractured quickly and completely under applied biaxial loads of 2-3 MPa. The de-embrittlement treatment was established to increase the cohesion of the boundaries enough so that crack initiation and propagation could be observed and recorded. At the same time, the brittle, intergranular mode of fracture was retained. By varying the de-embrittlement time at a fixed temperature, a full range of fracture behavior could be induced. This range of behavior is illustrated in Fig. 8, where gallium embrittled specimens have been de-embrittled for various times at 100 °. The fracture behavior goes through a relatively sharp transition at around 60 min from BIF to completely ductile behavior. Specimens de-embrittled for 45 min at 100 ° fractured through BIF but possessed sufficient cohesion so that initiation and fracture could be observed and recorded. Similarly, specimens annealed for 10 min at 150 ° possessed the same desirable attributes.

Spreading gallium over a circular region in the specimens produced a centered, uniform region of embrittlement. Figure 9 shows the side opposite to where the gallium was applied on a specimen. The gallium which penetrated through the grain boundaries from the other side was discolored by the wash in water. As evident in Fig. 9, the diffusion of gallium was constrained to a circular region in the center of the specimen. Within that region, the grain boundary embrittlement was quite consistent. Since no gradients in gallium concentration existed laterally on the specimens, we can infer that all the grains within the embrittled region experienced the same extent of embrittlement.

4.3. BIF of biaxially loaded specimens with 2D microstructures

The sequence of photographs in Figs 10(a)-(d) illustrates the evolution of fracture in a biaxially loaded embrittled sheet. The range of remote stresses during the fracture process is 5-15 MPa. No plastic flow is apparent during crack growth except at specific crack bridges. Multiple crack initiation sites are evident. The growing cracks from these sites quickly interact to form a main crack which is generally linear. As is evident in the photos, the two approximately collinear cracks which form the main
Fig. 13. (a) Scanned image of the Al polycrystal sheet, and (b) experimentally observed crack pattern in the sheet.
(a)

(b)

Fig. 14(a–b) (Caption overleaf)
Fig. 14. Damage crack patterns from simulations for $N = 31, 61, 101, 201$. Note the very good agreement with Fig. 13(b).
crack do not join. Rather, the crack tips are blunted approximately two facet lengths from each other. Figures 11(a) and (b) show the back side (inverted) of this specimen in the regions where the crack tips were blunted. These SEM micrographs indicate that some plastic flow occurred in these regions, and the cracks did not propagate intergranularly. Obviously, the facets in the vicinity of the crack tips were not embrittled.

The crack tip blunting events which occurred in the embrittled specimen appear to be caused by insufficient embrittlement of those facets near the crack tip. This nonhomogeneous embrittlement may be explained in part by observing the possible CSL misorientations of the facets where the cracks were blunted. In Fig. 12, a map outlining all the grain boundaries has been overlaid over an optical micrograph of the back side of the embrittled specimen. All boundaries which satisfy a CSL misorientation are labeled. All non-labeled boundaries are random high angle boundaries. In the central region where both main cracks were blunted before they could join, the crack tips were separated by facets with two $\Sigma$1 and a $\Sigma$9 boundaries. Since these boundaries have been shown to possess lower internal elastic strain energy than random boundaries, they should experience less gallium embrittlement and quicker de-embrittlement than other random boundaries. Thus, their CSL orientations could very well explain the apparent lack of embrittlement and the resulting crack bridge.

4.4. Computer simulations

Computer simulations of fracture described in Section 3.2 were implemented here to reproduce the same failure patterns as observed experimentally. Computations were carried out based on the scanned image of the specimen [see Fig. 13(a)], and the anisotropic Kirkwood model, combined with a triangular lattice network of a globally rectangular $(N_x \times N_y)$ shape, where $N_x$ and $N_y$ are the total numbers of mesh spacings in the $x$ and $y$ directions, respectively. Due to the limitations of computer space and time we had to confine our simulations to four cases: $(N_x, N_y) = (31, 25), (61, 51), (101, 85), (201, 169)$. Results of the experiment are shown in Fig. 13(b), while those of computer simulations in Figs 14(a)-(d). It is seen that all the cracking patterns match the one obtained from the experiment very well—even for the lowest resolution mesh. However, with the increasing mesh refinement, the simulation results do not display a definite improvement, but, rather, each one of them displays its own departures from the actual experimentally observed crack pattern of Fig. 13(b). Several reasons are noted for this.

(i) The image of the specimen which we obtained from the experiments was not entirely complete—several crystal boundaries were not fully closed (and the numbering scheme of crystals was not totally reliable). Thus, the four meshes of Fig. 14 do not represent a sequence of progressive resolutions of the same well defined system.

(ii) While the grain boundaries should ideally be presented as zero thickness zones, this was not the case in the scanned image. Thus, some of the nodes of the spring network fell onto the grain boundaries, and their properties were then assigned properties to be the same as properties of one of the neighboring nodes.

(iii) The key aspect in the computer simulation concerned the assignment of strength to all the grain boundary points. Here it was done according to the criterion mentioned earlier in Section 3.2.3, that is, by taking stiffness and strength of any boundary-crossing spring as the weighted average of the two length portions of that spring belonging to both contiguous crystals. Since the grain boundary region is much weaker than that of the crystal itself, a reduction factor in strength equal to 0.01 was then adopted for all such bonds. This was, of course, a simplified way of modeling the actual laboratory system in which the distribution of gallium and its degree of penetration of the Al crystal boundaries was most likely nonuniform.

As pointed out in the Introduction, the ultimate objective of this study is to develop a model leading to the determination of the grain boundary strength criterion. Given the very good agreement of the simulated crack patterns with the one observed experimentally, the method presented here offers a powerful tool for such tasks. It was, to the best of the authors' knowledge, the first instance that crack formation in a polycrystal was thus simulated.

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