A study of intergranular fracture in an aluminium alloy due to hydrogen embrittlement

E. Pouilliera, A.-F. Gourgues a, D. Tanguyb, E.P. Busso a,*

Abstract

This work concerns a study of the effects of plasticity on the mechanism of intergranular cracking assisted by hydrogen induced embrittlement in an aluminium alloy. Here, tensile specimens charged with hydrogen were used to investigate quantitatively the effect of plastic deformation on the mechanism of intergranular crack initiation at the scale of the individual grains. An experimental procedure was set up to monitor the evolution of surface strain fields on in situ tested SEM notched specimens using digital image correlation techniques. In addition, measurements of the associated crystal orientation evolution at the micron scale were carried out using electron backscatter diffraction (EBSD). These measurements were then compared with finite element predictions of the local strain fields on the observed regions of the in situ specimen. The numerical predictions were obtained using a dislocation mechanics-based crystal plasticity model to describe the constitutive behaviour of each individual grain. The crystallographic grain orientations of the region of interest were discretised for the finite element analyses from EBSD maps. From this study, it was found that intergranular cracking due to hydrogen embrittlement in the Al alloy is locally triggered by high tensile grain boundary tractions, here estimated to be $170 \pm 35\, \text{MPa}$. As importantly, the results also revealed that the conditions needed for grain boundary microcracks to initiate are greatly affected by the deformation of neighbouring grains: i.e. it was established that boundaries between two “hard” grains, inside a neighbourhood of “softer” deformed grains, are the first to fail.

1. Introduction

Aluminium alloys, strengthened by elements in solid solution, can be sensitive to intergranular stress corrosion cracking (ISCC) in some specific microstructural states (Hollinsworth and Hunsicker, 1987). Initially, the mechanism thought to lead to ISCC was understood to be stress assisted localised dissolution (Dix, 1940). It was supported by microstructural observations showing that Mg-rich ($\text{Al}_3\text{Mg}_2\beta$-phase) precipitates form at grain boundaries during ageing. Here, locally enhanced Mg concentration was thought to create an electrochemical potential inhomogeneity that induced the anodic dissolution of the precipitate and, in so-doing, creating a preferential crack path. This understanding prevailed till the early seventies (Gest and Troiano, 1974) when the crucial role of hydrogen in the fracture process was identified. Then, it was found that when a certain amount of hydrogen is absorbed from the environment into materials such as steels, it can then diffuse along grain boundaries and eventually lead to their embrittlement.
Initially, the effect of hydrogen contamination in Al alloys was ignored due to the vanishingly small solubility of hydrogen when the metal is in contact with the H₂ gas. Nevertheless, it was shown that a hydrogen flux can develop through Al–Zn–Mg alloys (Gest and Troiano, 1974) even with a free corrosion potential. It was shown unambiguously later on (Gruhl, 1984) that hydrogen can diffuse over depths of some millimeters toward regions of high stress triaxialities (e.g. notch root region), be trapped at grain boundaries and induce intergranular fracture without any contribution of corrosion processes which may be active at the crack tip. The precise nature of “brittle” fracture, i.e. decohesion vs intense localised plasticity, is still debated nowadays. In the specific case of Al–Zn–Mg, TEM studies by Scamans et al. (1976) and Scamans (1978) show that it is a true intergranular decohesion mechanism (see also Malis and Chaturvedi, 1982). No evidence of grain boundary voids, associated with a ductile-type failure mechanism, has been linked to hydrogen embrittlement in Al alloys.

The β intergranular precipitates play multiple roles in the stress corrosion cracking (SCC) of Al–Mg alloys. It was shown (Jones et al., 2001, 2004) that they dissolve actively during SCC and that enough hydrogen must have been produced to embrittle the grain boundary in between successive β particles (Jones and Danielson, 2003). Furthermore, pitting tests (Tanguy et al., 2002) followed by a tensile deformation in air showed that the anodic dissolution of β particles at triple junctions released enough hydrogen to embrittle precipitate-free grain boundary facets. Moreover, the observation of hydrogen bubbles in Al–8%Mg (Ben Ali et al., 2011) suggests that β-precipitates must indeed be preferential sites for hydrogen trapping. Such understanding and the results of the numerical simulation of the intergranular fracture process (Malis and Chaturvedi, 1982) revealed that intergranular fracture is mostly dominated by the damage of the inter-precipitate regions. The role of plasticity in ISCC is known to be important even though it has only been studied macroscopically. For instance, Tanguy et al. (2002) showed that pre-straining of an Al–5%Mg alloy increases its sensitivity to ISCC under both tensile and fatigue loading conditions.

Hydrogen induced embrittlement (HIE) processes can be studied experimentally using local strain measurements (Sachtleber et al., 2002). However, even though as more accurate local measurement techniques become available, it still remains difficult to measure intragranular strains in fine grained materials. For such cases, large focal lenses and microdiffraction tomography (Ludwig et al., 2009) techniques are not suitable. Fortunately, plastic strain fields can nowadays be more easily measured at the micrometer scale thanks to techniques which rely on fine grids or speckles spaced out at distances considerably smaller than the grain size. Examples are those based on local pitting corrosion, photolithography (Schoeter and McDowell, 2003; Liu and Fischer, 1997), micro-lithography (Allais et al., 1994; Biery et al., 2001), and digital image correlation techniques (DIC) (Allais et al., 1994; Tatschl and Kolednik, 2003; Hild et al., 2002; Sachtleber et al., 2002).

Even when the measurement of local strains is possible in small grained materials, numerical techniques are needed to identify the local stress fields. Intragranular stresses are known to play a significant role in intergranular fracture and result from complex phenomena (e.g. strain incompatibilities between adjacent grains) in a variety of materials such as Zn (Parisot et al., 2000), TiAl (Simkin et al., 1999) and duplex stainless steels (Marrow, 1996). The mechanical response of a polycrystalline aggregate can in principle be predicted by using either mean-field homogenisation or finite element (FE) techniques. However, when the spatial local distributions of strain and stress heterogeneities at the level of each individual grain are needed, then full-field computations are required. The finite element method has been used to model the intragranular response of polycrystalline materials under various macroscopic loading conditions, such as homogeneous (Becker and Panchangadeeswaran, 1995) and fretting (Goh et al., 2006) loading, and to study crack initiation and growth using methods such as those based on cohesive zone models (e.g. Clayton and McDowell, 2004). Crystal plasticity constitutive formulations have been based on semi-phenomenological (Asaro, 1983; Barbe et al., 2001a), and dislocations mechanics based local (Busso and McClintock, 1996; Harder, 1999) and non-local (Busso et al., 2000; Cheong et al., 2004) approaches. They are also able to account in some cases for, e.g., statistical effects of grain boundaries and lattice misorientations on local stress and lattice energy density distributions (Bieler et al., 2009; Clayton and McDowell, 2003), and twinning as a deformation mechanism (Staroselsky and Anand, 1998).

The main objective of this work is to understand the effect of local grain plasticity on the mechanism of intergranular crack initiation due to hydrogen induced embrittlement in an Al–5Mg alloy. To that purpose, notched specimens were locally charged with hydrogen and then tested under tension in situ within a scanning electron microscope. A combination of in situ 2D strain field and EBSD measurements, and finite element simulations of the observed specimen regions using a dislocation mechanics-based crystal plasticity model was relied upon to understand the local conditions responsible for intergranular crack initiation.

### 2. Experimental investigations

#### 2.1. Material

The Al–Mg alloy studied, known as AA 5083, was thermomechanically treated to obtain equiaxed grains. Its chemical composition is given in Table 1. Charging Al alloys with hydrogen is problematic because of the barrier effect of the oxide

**Table 1**

<table>
<thead>
<tr>
<th>Element</th>
<th>Si</th>
<th>Fe</th>
<th>Cu</th>
<th>Mn</th>
<th>Mg</th>
<th>Zn</th>
<th>Ti</th>
<th>Cr</th>
<th>Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>wt.%</td>
<td>0.40</td>
<td>0.40</td>
<td>0.10</td>
<td>0.70</td>
<td>4.50</td>
<td>0.25</td>
<td>0.15</td>
<td>0.25</td>
<td>Balance</td>
</tr>
</tbody>
</table>
layer. For a discussion of these issues see, e.g., Jones and Danielson (2003), Buckley and Birnbaum (2002), Ben Ali et al. (2011). The presence of the $\beta$-phase at the grain boundary was necessary for the charging procedure to induce embrittlement over a significant depth. The procedure consists of a cathodic polarisation at a current density of $-1 \, \text{mA/cm}^2$, during 12 h in a deaerated solution containing $\text{AlCl}_3$ with a concentration of 120 g/l (with addition of $\text{H}_2\text{SO}_4$ and saturated with boric acid) and a pH 2. Embrittlement was over at least one grain boundary facet when $\beta$ precipitates were present and less than 1 $\mu$m when they were not present. Note that longer charging times may damage the surface and not lead to an embrittlement. In order to prevent hydrogen desorption, specimens were stored in liquid nitrogen until the mechanical tests were performed. Preliminary uniaxial tensile tests, conducted up to a 10% elongation, confirmed that HIE was effective up to a depth of 5 $\mu$m and that intergranular cracking was observed on grain boundaries perpendicular to the tensile direction (see Fig. 1) after a few percent of plastic strain. Embrittlement was fully reversible by heat treatment.

2.2. Room temperature tensile tests and strain field measurements

Experimental and numerical investigations were performed using two different types of 1 mm-thick specimens, see Fig. 2. The smooth specimen was relied upon to calibrate the crystal plasticity model, and the specimen with two symmetrical notches to localise a priori the deformation and intergranular fracture events in a region small enough to be suitable for the local strain and grain orientation measurements. Note that in Fig. 2, the plotted stress is the axial component on the specimens, calculated as the applied axial load divided by the smallest specimen cross-section, rather than the Mises equivalent stress. Consequently, one can see the strengthening effect of the notch due to the higher local triaxiality in the notch region with respect to that in the smooth specimen.

After the surface preparation, the contour of the specimen zone to be studied was defined by microindentation marks, see Fig. 3(a). These marks were also used as references for optical extensometry in order to confirm the average measured strain obtained with the digital image correlation (DIC) technique.

The use of both EBSD analysis and DIC techniques required a specific surface finishing. After machining, specimens were recrystallised at 460 $^\circ$C for one hour and water-quenched to the keep Mg in solid solution. Then a mechanical polish was performed down to 1 $\mu$m in order to remove any surface damage. The specimens were then tempered at 150 $^\circ$C for 7 days in order to precipitate the Al–Mg $\beta$-phase and relieve any residual stresses which may have been introduced due to the mechanical polishing. A final polishing step from 1 $\mu$m down to 1/4 $\mu$m was carried out before polishing chemically the specimen with colloidal silica to optimise the surface condition for the EBSD scanning (see Fig. 4(a)). After the EBSD mapping, the region of interest was precharged with hydrogen following the procedure described previously. To be able to measure the strain heterogeneities inside the grains, the region of interest on the specimen surface had to be patterned with a specific speckle. However, to obtain accurate results by DIC at this scale, surface heterogeneities should be less than 200 nm in size. Slight corrosion pits, i.e. the smallest white features in Fig. 4(b), observed after hydrogen charging, were kept and employed as surface heterogeneities for contrast enhancement purposes.

A 450 $\times$ 300 $\mu$m area was scanned with EBSD, the result of which can be seen in Fig. 5. EBSD maps were colour coded according to the orientation of the tensile direction (x) of the specimen with respect to the crystal axes, as depicted in

![Fig. 1. Optical micrograph of intergranular fracture after 10% axial strain along the x-axis.](image-url)
In the present study, a ‘grain’ was defined from the EBSD maps as a crystal of at least 6 µm in size, surrounded by boundaries with misorientation angles of at least 5°. In Fig. 5, the mean grain size was found to be 27.0 µm with a standard deviation of 8.6 µm. The distribution of misorientations between grains was found to be in the range of 10–60°, without any evidence of texture observed. The selected region in Fig. 5 is composed of 458 textureless grains with a total boundary length of ~25 cm. This region was assumed to be representative and sufficiently large to conduct the study. Even though 10 specimens were tested, only the results of one of them will be reported as they were found to be representative of all the other ones obtained in this study.

Tensile specimens were mounted on a mini-tensile testing machine in a Leo SEM 1450VP vacuum chamber. The machine has a dimension of 502 × 327 × 250 mm, a total screw travel of 150 mm, and a maximum load capacity of 5000 N. A computer integrated stepping motor control was used to carry out the tensile test at an imposed displacement rate of 0.36 mm/min. The microindented decorated specimen surface was imaged under both secondary electron (SE) and backscatter electron (BSE) imaging modes and a set of 8-bit grey scale digital images was obtained. The specimen was then loaded up to a
A series of digital images within the microindentation decorated surface were acquired in situ at several applied strain levels in order to record the surface strain field evolution and to identify intergranular crack initiation events.

Fig. 4. Optimum surface preparation for (a) EBSD (before hydrogen charging), and (b) DIC (after hydrogen charging) with a superimposed correlation grid of 32 × 32 pixels.

Fig. 5. Inverse pole figure map of the region studied by EBSD. The y-axis represents the specimen tensile axis and the zone delineated in the black box was that modelled for the FE calculations.

total axial strain of 5%. A series of digital images within the microindentation decorated surface were acquired in situ at several applied strain levels in order to record the surface strain field evolution and to identify intergranular crack initiation events.
An optimised digital image correlation technique was used to compute surface strain fields from the recorded images using the software CorrelIQ4GD (Hild et al., 1999, 2002). The field of view of the digital images was approximately $850 \times 600$, $300 \times 225$, and $150 \times 112$ $\mu m^2$ for the regions (c)-(d)-(e) identified in Fig. 3, respectively. These three scales of correlation provided a range of fine-scale strain field measurements as well as the boundary conditions to be applied to the crystal aggregate region used in the modelling part of the study. The spatial resolution of the system is $32 \times 32$ pixels, with an average displacement resolution of at least a pixel size. The pixel size, which determines the minimum spatial resolution for each magnification, is therefore 0.5, 0.12 and 0.07 $\mu m$ for the large, medium and low magnification images, respectively. With this method and with the level of applied macroscopic strains (0.5% up to 3%), the relative computed error in the measured strains is less than 3%. The use of three scales of correlation allowed a link between microscopic and macroscopic scales. The local strain fields were obtained from the DIC data by strain averaging over the smallest area investigated, viz. (e) in Fig. 3.

After the test, the deformed specimens were removed from the mini-tensile test stage and analysed again by EBSD to identify the grain orientations after deformation. Due to the surface roughening caused by plastic deformation, the specimens were polished to improve the quality of EBSD patterns by removing a layer of approximately 0.2 $\mu m$.

3. Model of the polycrystal region

The dislocation-mechanics based rate-dependent crystal plasticity model for FCC materials proposed by Cheong and Busso (2006) was used in the FE simulations of the in situ test to compute the strain and stress fields. This formulation will be outlined next.

3.1. Crystal plasticity model

The rate-dependent single crystal constitutive model is formulated following finite-strain kinematics. It relies in the classical multiplicative decomposition of the total deformation gradient $F$,

$$ F = F^p F^e $$

into a plastic, $F^p$, associated with crystallographic slip, and an elastic component, $F^e$, which accounts for the elastic stretching and rotation of the crystal lattice. The velocity gradient of plastic slip is given by

$$ L^p = F^p F^p \cdot 1 = \sum_{s=1}^{N} \gamma_s m_s \otimes n^s $$

Here, the sum extends over $N$ active slip systems, $\gamma_s$ represents the crystallographic slip rate on a generic slip system $(s)$, and $m_s \otimes n^s$ the dyadic product between the slip direction ($m^s$) and associated slip plane normal ($n^s$).

The slip rate, $\dot{\gamma}_s$, is assumed to be dominated by the thermally activated glide of dislocations over obstacles (i.e. mainly forest dislocations as the heat treatment used in the alloy of interest did not lead to full intergranular precipitation of Mg (Tanguy et al., 2002)). The slip rate is related to the resolved shear stress, $\tau^s$, through the exponential function proposed by Busso and McClintock (1996) and Busso et al. (2000),

$$ \dot{\gamma}_s = \gamma_0 \exp \left\{ -\frac{F_a}{k T} \left[ 1 - \left( \frac{\tau^s - S_c^p \mu^p / \mu_0}{\tau} \right)^p \right]^q \right\} \text{sgn}(\tau^s) $$

which accounts for the absolute temperature ($\theta$, $K$) and the stress dependence of the activation energy. In Eq. (3), $F_a$ represents the Helmholtz free energy of activation at 0 K, $k$ the Boltzmann constant, $\gamma_0$ a reference slip rate and, $\tau$, the maximum glide resistance at which dislocations can be mobilised without thermal activation. Furthermore, $\mu$ and $\mu_0$ are the shear moduli at $\theta$ and 0 K respectively. The exponents $p$ and $q$ describe the shape of the energy barrier vs. stress profile associated with interactions between dislocations and obstacles. The main contributions to the slip resistance to plastic flow, $S_c^p$, are due to the friction stress induced by the Mg atoms in solid solution, $S_m^p$, and to the athermal slip resistance $S_t^p$. As the slip resistance contributions are assumed to be additive, then

$$ S_c^p = S_m^p + S_t^p $$

Since only a very small part of the Mg atoms precipitate during the heat treatment, the concentration of Mg in solid solution is set to be equal to the average concentration in the material. Thus the friction stress due to Mg atoms in solid solution is calculated based on the atomic size and concentration of Mg in the alloy (see Table 1), as proposed by Saada (1968). The athermal slip resistance $S_t^p$ is expressed as:

$$ S_t^p = 2\mu b \sqrt{\sum_{s=1}^{N} \frac{h_{c,s}^p \rho_0^p}{\rho_s^p}} $$

where the overall dislocation density for a given slip system $s$, $\rho_s^p$, is obtained from a discretisation of the dislocation structure into pure edge and pure screw types, of densities $\rho_s^e$ and $\rho_s^s$, respectively. Thus,
\[ \rho_s^e = \rho_s^e + \rho_s^f \]  
In Eq. (5), \( \lambda \) is a statistical constant and \( h^{xy} \) represents the dislocation interaction matrix defined as

\[ h^{xy} = \omega_1 + (1 - \omega_2)\delta^{xy} \]  
where \( \omega_1 \) and \( \omega_2 \) are two cross-hardening constants and \( \delta^{xy} \) the Kronecker delta. The evolutionary equations of the individual dislocation densities account for the competing dislocation storage-dynamic recovery processes in FCC metals and are expressed as (Cheong et al., 2004; Cheong and Busso, 2006),

\[ \dot{\rho}_s^e = \frac{C_e}{b^2} \left[ K_s \sum_{j=1}^{N} \rho_s^e - 2d_e \rho_s^e \right] [\gamma^e] \]  
\[ \dot{\rho}_s^f = \frac{C_f}{b^2} \left[ K_s \sum_{j=1}^{N} \rho_s^f - \rho_s^e \left( \pi d_u^2 K_i \sum_{j=1}^{N} \rho_s^e + 2d_e \right) \right] [\gamma^e] \]  
Here, the parameters \( C_e \) and \( C_f \) describe the relative contributions to the overall slip from edge and screw dislocations, while \( K_e \) and \( K_f \) are mobility constants associated with their respective mean free paths. Recovery processes are associated with the parameters \( d_e \) and \( d_s \), which represent critical annihilation distances between dislocations of opposite Burgers vectors for both edge and screw types. The theory is completed by considering the stress–strain relation,

\[ T = \varepsilon : E \]  
where \( T \) is the lattice–based second Piola–Kirchhoff stress tensor, \( \varepsilon \) is the anisotropic elasticity tensor, while \( E = [F^T F - I]/2 \) is the Green–Lagrange strain tensor, with \( I \), the second-order identity tensor. Furthermore, \( T \) can be expressed in terms of the Cauchy stress tensor, \( \sigma \), as

\[ T = (\det F) F^{-1} \sigma F^{-T} \]  
Details of the numerical implementation of the above constitutive theory into the finite element method can be seen in Busso et al. (2000) for an implicit implementation and in Abrivard (2009) for an explicit one.

The values of the model’s parameters are given in Table 2. They are inspired by those calibrated for pure aluminium in Cheong and Busso (2006), except for the additional term which accounts for the Mg solid solution effect. A comparison between the uniaxial stress–strain tensile curve obtained from the smooth specimen and that predicted using a 100 grain aggregate and the single crystal model is shown in Fig. 2.

3.2. FE mesh and boundary conditions of the region studied by EBSD

To mesh automatically the region of interest, namely Region (b) in Fig. 3, an arbitrary grain colour map was first plotted. To this aim, grains were defined as sets of at least 10 pixels in size with a misorientation between neighbouring pixels of at most 5°. From this map, triple junctions were first identified using an in-house developed image processing tool. Then, grain boundaries were revealed using both triple junctions and colour gradient detection. The grain boundary discretisation was carried out by dividing them into small straight segments while taking their local curvatures into account (Fig. 6(b)). Finally, grains were meshed using 6-node generalised plane strain triangular elements to allow a proper geometric description of grain boundaries. Note that generalised plane strain boundary conditions imply that the out-of-plane strain (\( \varepsilon_z \)) is uniform on the \( (x,y) \) plane. For the free out-of-plane \( z \) surfaces of interest in the specimen, it implies that \( \varepsilon_z \neq 0 \) and that, on average, \( \sigma_z = 0 \). Such assumption constitutes the best 2D approximation to a full 3D model of the polycrystal. Other approaches relying on square meshing based on EBSD scan step or on correlation microgrids were not deemed to be appropriate as regular meshing can result on an over estimate of the strains and stresses at grain boundaries (e.g., see Barbe et al. (2001a)). Here, a mesh composed of 15,230 elements was found to be suitable to account for the spatial resolution of the EBSD measurements, the description of grain boundary morphologies, and to yield acceptable computational times.

<table>
<thead>
<tr>
<th>Table 2</th>
<th>Single crystal model parameters.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Elastic constants (GPa)</td>
<td>Flow rule (Eq. (3))</td>
</tr>
<tr>
<td>( C_{11} = 112.9 )</td>
<td>( \tau_0 = 3 \times 10^{-10} )</td>
</tr>
<tr>
<td>( C_{22} = 66.4 )</td>
<td>( \tau = 8.0 \text{ MPa} )</td>
</tr>
<tr>
<td>( C_{44} = 27.9 )</td>
<td>( S_{xy}^e = 42.7 \text{ MPa} )</td>
</tr>
<tr>
<td>( \mu = 49 )</td>
<td>( \gamma_0 = 1.73 \times 10^{-6} \text{ s}^{-1} )</td>
</tr>
<tr>
<td>( \mu = 45 )</td>
<td>( p = 0.141 )</td>
</tr>
<tr>
<td>( q = 1.10 )</td>
<td>( \varepsilon = 0.3 )</td>
</tr>
</tbody>
</table>
When comparing numerical and experimental results, the influence of the neighbouring grains on the simulated microstructure must be taken into account, since they can affect the mechanical behaviour from as far away as 5–10 times the average grain size (Doumalin et al., 2003). In order to take neighbouring grains into account, the measured displacements at the fringes of the meshed region, whose locations are defined by the microindentations shown in Fig. 3, were interpolated and applied as boundary conditions.

4. Results and discussion

4.1. Strain field measurements and intergranular cracking observations

Fig. 7 shows regions oriented 90° and 45° to the loading direction where deformation is localised, as also reported elsewhere at similar scales (Barbe et al., 2001a,b). The approximate width of the more highly deformed regions is 80 μm (i.e. 3 times the average grain size). Microcracks were found within these deformed regions, which suggests that crack initiation requires a certain minimum average level of deformation. Cracks were only seen to nucleate at grain boundaries oriented...
normal to the tensile direction. Thus, intergranular crack initiation seems to be related to both average strain level and grain boundary orientation.

The strain field was then studied at a finer scale (see upper half of Region (d) in Fig. 3) to investigate the heterogeneity of the deformation within the more highly deformed regions. The measured strain distributions at medium magnifications are shown in Fig. 8 for macroscopic applied strains of (a) 0.5%, (b) 1.6%, (c) 2.6%. In Fig. 8(d), a SEM micrograph taken after 4.5% strain is shown for comparison. Here, white arrows point out strongly deformed regions and the black ones cracked grain boundaries. These results reveal that microcracks appear only after a small amount of macroscopic strain, between grains which are relatively less deformed compared to their neighbours (less than 1% in Fig. 8(a)). To confirm these results, the level of lattice rotation due to plastic deformation was investigated within each individual grain. Fig. 9 shows the crystal orientation evolution of Grains 1, 2 and 3 identified in Fig. 8 before and after deformation. It can be seen that Grains 2 and 3 undergo a very small amount of rotation, whereas Grain 1 rotates considerably more towards the [101] orientation. These EBSD results are in good agreement with the fact that Grains 2 and 3 are less deformed than Grain 1, as revealed by Fig. 8.

These observations also reveal that grain boundaries between undeformed or only slightly deformed grains do not necessarily crack. This implies that, in order to describe the cracking phenomena correctly, grains neighbouring the failed grain boundaries need to be taken into account. The DIC results also showed that grains surrounding the failed grain boundaries deformed more than those where cracks were found. As it will be discussed below, the numerically predicted strain fields at smaller scales confirm this experimental observation.

In the region where detailed EBSD analyses were conducted (see Fig. 5), 12 grain boundaries were identified as having failed during the test. Furthermore, 11 out of the 12 were found to be partly located in the vicinity of highly deformed regions, and between grains which had not undergone a great deal of deformation. This is illustrated in Fig. 10 which gives an schematics of (a) the corresponding grain configuration and (b) an idealised configuration to be used in the model presented below. Thus, the following preliminary conclusions can be made: micro-cracks seem to initiate at grain boundaries (i) contained within a strongly deformed region, (ii) oriented normal to the loading direction, and (iii) located at the boundary between two “hard” grains which are, in turn, contained within a larger “softer” region.

Fig. 8. Measured strain distributions at medium magnification along the x-direction for macroscopic applied strains of (a) 0.5%, (b) 1.6% and (c) 2.6%, and (d) SEM image taken after 4.5% applied strain. White arrows indicate strongly deformed regions and the black ones cracked grain boundaries.
A failure criterion for intergranular cracking can also be proposed by considering that the grains at either side of the failed grain boundary develop high normal tractions. Typically, the critical value of these tractions were seen to be attained where the local inelastic strain reaches approximately 0.33%. Such low value of plastic deformation indicate that those grain boundaries fail in a brittle manner. It is assumed here that crack initiation is controlled by a critical normal grain boundary traction, henceforth denoted $\sigma_d$.

4.2. Crack initiation criteria

4.2.1. Effect of strain incompatibility across grain boundaries

As discussed above, grain boundary cracking seems to be driven by local grain boundary tractions arising from strain incompatibilities between “harder” and “softer” grains. Furthermore, cracking events could not be related to either a strong gradient in the Schmid factor (assuming grains with uniform lattice orientations and stresses, as in a Sachs assumption) or to a strong strain gradient across boundaries (as can be seen in Fig. 8). In addition, a local study of misorientation line profiles before and after deformation, across both “sound” and “cracked” grain boundaries, did not reveal any significant role of local misorientations either near (i.e. local lattice rotations due to plasticity) or across grain boundaries as it is sometimes observed in intergranular fracture (Gourgues, 2002). In can be said that our findings indicate that strain incompatibilities at grain boundaries are not sufficient to explain intergranular fracture in the present case. To address this issue and to obtain a detailed local description of both strain and stress fields, FE calculations were carried out and the corresponding results are shown next.

4.2.2. Numerical predictions of local stress and strain fields

Fig. 11(a) and (b) shows the measured and predicted strain fields in Region (e) of Fig. 3 after 0.45% applied macroscopic strain (which corresponds to Point A in Fig. 2), respectively. Fig. 11(c) is a SEM micrograph of Fig. 11(a)’s region and Fig. 11(d) gives the predicted Mises stresses at the time when the first grain boundary failure was observed (also at Fig. 2’s Point A).

It should be noted that at the locations 1, 2 and 3 indicated in Fig. 11, the high levels of axial strain measured by the DIC system are to a great extent different from the real values due to the distortion introduced by the opening of the cracks during the digital image correlation measurements. Note also that the spatial resolution of the scanning electron microscopy image was not high enough to detect cracks for applied strains lower than 0.1%.

Fig. 9. Comparison between the inverse pole figures for (a) and (d) Grains A, (b) and (e) Grain B, and (c) and (f) Grain C identified in Fig. 8(c) before deformation (a–c) and after (d–f) 4.5% applied strain.
Fig. 10. Schematics of the typical grain configuration observed around a cracked grain boundary (indicated by the dashed line): (a) polycrystalline region and (b) idealised configuration to be used in the simplified model. The dashed line delimits the boundary that is assumed to fail.

Fig. 11. (a) Measured and (b) predicted axial strain fields after 0.45% applied macroscopic strain, (c) micrograph of the same region as (a) and (b) at the end of the test (4.5% applied strain), and (d) predicted Mises stresses after 0.45% applied strain. (Loading direction is parallel to the x-axis).
Fig. 12 shows both the predicted and measured axial strains along the two dashed lines in Fig. 11(a) and (b). Note that the very high value of the measured axial strain at the crack location (distance = 18 \mu m in Fig. 12) is due to crack opening effects rather than to strain localisation. It can be seen that the model predictions along the vertical line are in satisfactory agreement generally with the measurements. However, the model overestimates the local axial strains along the horizontal line at a distance greater than 60 \mu m (that is, greater than 60 \mu m from Fig. 11(a) and (b)’s left border), as it is the case over a 4 \mu m long region close to the crack at distances of between 10 and 14 \mu m. The latter is understood to be due to the stress redistribution which follows the crack initiation event.

At \( d = 0 \) (both vertical and horizontal lines) and \( d = 60 \mu m \) (vertical line), the model predictions and experimental measurements are close as expected since the experimental measurements were prescribed as the finite element model’s boundary conditions. However, the predictions and measurements do not coincide for the horizontal line at \( d = 68 \mu m \) due to the fact that the size of the region studied by digital image correlation was smaller than that discretised with finite elements. Thus, here the boundary conditions were prescribed slightly further away in the numerical model. Nevertheless, the effect introduced by this extrapolation of the boundary conditions was found to be only local and it can be seen that the axial strain field is reasonably well predicted close to the boundary which is the first to fail. The predicted local stress fields around grain boundaries just before crack initiation will be discussed next.

4.2.3. Simplified stress based grain boundary failure criterion

A first approximation to the normal tractions needed to fracture a given grain boundary, \( \Sigma_{\text{gb}} \), can be obtained by solving an Eshelby problem consisting of a “hard” spherical elastic inclusion embedded in a “softer” infinite elasto-viscoplastic matrix (as illustrated in Fig. 10(b)). It will be assumed that the grain boundary belongs to two “hard” grains which behave elastically, very much as the grain boundary in Fig. 10(b)’s elastic inclusion. Under this assumption, it can be shown that the solution to this problem yields uniform stresses within the inclusion, and that the maximum principal stress is given by (Beremin, 1981),

\[
\sigma_{xx} = \Sigma_{xx} + k(\Sigma_{eq} - \sigma_y)
\]  

where \( \sigma_{xx} \) is the average axial stress in the polycrystal at the moment the first grain boundary fails. This value can be approximated from Fig. 2’s Point A, which yields \( \Sigma_{xx} = 120 \text{ MPa} \). Also, \( \sigma_y \) is the material yield stress (=85 MPa), and \( \Sigma_{eq} \) the average Mises stress in the inclusion. From the FE calculations, the average von Mises stress in Grains B and C (see Fig. 11(d)) was found to be \( \Sigma_{eq} = 140 \text{ MPa} \). Finally, \( k \) is a function of the inclusion shape – equal to 1 for the spherical particle assumed here (Pineau and François, 2001). With these values, Eq. (12) yields \( \sigma_{xx} = 175 \text{ MPa} \). This value will henceforth be considered to be a first estimate of the critical grain boundary traction, \( \sigma_{\text{gb}} \), required to fracture a grain boundary.

4.3. Critical grain boundary traction predicted by a full FE analysis of the crystal aggregate

The predictions obtained from the previous section’s analytical model were compared to the local normal tractions obtained from the numerical simulations of the crystal aggregate. Here, the normal grain boundary tractions were determined numerically by resolving the local stress tensor into a direction normal to the grain boundary.
The predicted normal tractions along some of the most highly stressed grain boundaries within the region studied by DIC, labelled 1–10 in Fig. 13(a), are shown in Fig. 13(b). Here, the tractions are expressed in terms of the distance along the grain boundary of interest from a triple point, \( l \), normalised by the overall grain boundaries length, \( L \). It can be seen that the normal grain boundary tractions tend to be higher in the vicinity of triple junctions, as has also been observed elsewhere (Masayuki et al., 2007). This is to a great extent due to the complex deformation constraints imposed by the material surrounding the triple junctions.

It is worth noting from Fig. 13(a) and (b) that, out of the 10 grain boundary regions along which their normal tractions were calculated, only those numbered 1–3 failed during this particular test. It should also be recalled that these three failed grain boundaries were already identified in Fig. 11(a). Thus the predicted tractions shown in Fig. 13(b) reveal that, at the onset of cracking, the tractions in Grain Boundaries 1–3 vary between an upper bound of 205 MPa to a lower bound of 135 MPa. Thus, writing the critical tractions in these grain boundaries in terms of their mean value, one finds 170 ± 35 MPa. This numerical prediction is close to the one obtained analytically in the previous section using a simplified model based on an extension of Eshelby’s theory, namely 175 MPa. This good correlation is encouraging as far as the suitability of the configuration considered in Fig. 10 to describe intergranular crack initiation due to hydrogen embrittlement is concerned.

5. Conclusions

An in situ tensile testing procedure was set up to investigate intergranular fracture due to hydrogen embrittlement at the free surface of an aluminium–magnesium alloy. Results obtained from a combination of the in situ tests, local EBSD analyses,
strain field measurements by digital image correlation techniques and finite element modelling have led to the following conclusions:

- Grain boundary cracking occurs on boundaries normally oriented to the applied tensile stress when the average local axial strain is as low as 0.45%. In addition, failed grain boundaries were found to be between grains that undergo very limited plastic deformation despite being embedded in large localised deformation regions.

- The critical local grain boundary tractions obtained from an analytical Eschey-type approach, which was based on a “hard” inclusion embedded in a “softer” matrix, of 175 MPa is consistent with the numerical prediction of 170 ± 35 MPa obtained from a finite element model of the polycrystal using crystal plasticity concepts.

The aluminium alloy investigated in the present study exhibits a high stacking fault energy, which prevents it from annealing twinning. This behaviour strongly limits the possibility of increasing the intrinsic grain boundary resistance to environmentally assisted cracking through the use of grain boundary engineering. The results of this study suggests that, in such cases, grain boundary cracking sensitivity can be limited or reduced provided that in-service mechanical loading conditions are well known. This approach, which could be referred to as “computer-assisted texture engineering”, should enable the tailoring of the crystallographic texture of a material so as to lower the probability of finding clusters of “harder grains” embedded in a softer surrounding.

To provide guidelines for an “optimal” targeted texture, a number of numerical simulations of polycrystal aggregates (obeying a given average texture) could be carried out in parallel and the probability of finding the above mentioned grain configuration could be estimated on a statistical basis. Moreover, in cases where clusters of “harder” grains were “frequently” found, the orientation of grain boundary planes within these clusters should also be optimised (by proper design of grain morphologies obtained after thermo-mechanical processing) to limit the normal tractions on such grain boundaries. In such an approach, crystal plasticity models appear as a key tool to help designing new metallic alloys resistant to grain boundary cracking.

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