A three-dimensional computational model for intergranular cracking

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Abstract

A three dimensional mechanical model for intergranular crack propagation is presented. The model follows the spirit of existing percolation-like models but offers the inclusion of mechanical effects. This is necessary in order to account more accurately for the crack driving force and the effect of crack bridging ligaments, observed experimentally to be formed by fracture resistant boundaries. The model uses a regular representation of the material's microstructure and a categorisation of grain boundaries as beneficial and detrimental to fracture. This categorisation makes the model applicable to assessing material's resistance to intergranular stress corrosion cracking. The model mechanical behaviour is consistent with experimental observations and demonstrates its capability of simulating the development of bridges in the crack wake as well as crack coalescence. Results show that increasing the fraction of resistant boundaries increases the degree of crack tip shielding developed. This is expected to increase the resistance to stress corrosion crack propagation. The model offers a significant reduction of the computational resources usually needed to simulate intergranular propagation.

Keywords: *Stress Corrosion Cracking; Microstructure; Grain boundaries; Fracture; Crack bridging; Finite element analysis; Stainless steels.*

1. Introduction

Intergranular stress corrosion cracking (IGSCC) is a mode of crack propagation in susceptible stainless alloys, such as nickel-based alloys and austenitic stainless steels used in structural components in the power generation industry. Uncertainties in short crack behaviour contribute substantially to the hazard in assessing the lifetime of such components. One source of uncertainty is the material's microstructure, which determines the resistance to IGSCC to a large extent. For the materials mentioned, a great deal of experimental evidence is now available which shows that the random grain boundaries form paths of low resistance for intergranular corrosion cracks to follow, while the special grain boundaries, such as twin boundaries, have improved resistance to corrosion and cracking. One of the first reported results in this respect, due to Palumbo and Aust [1], was for pure nickel. Later, Lin et al. [2], Pan et al. [3] and Alexandreanu et al. [4] provided evidence for the nickel-based alloys 600 and X-750. In addition to nickel-based alloys, Bruemmer and Was [5], and Gertsman and Bruemmer [6] demonstrated these properties of the grain boundaries for type 304 and type 316 stainless steels. These findings justify to a great extent a classification of the grain boundaries in a microstructure into two categories – “resistant” and “susceptible” to intergranular corrosion and cracking. This is a useful assumption for fully sensitised stainless steels, in which the grain boundary structure controls the degree of carbide precipitation. This locally reduces the grain boundary chromium content, and thus lowers its corrosion resistance. In other intergranular corrosion systems, and partial sensitisation of stainless steels, a spectrum of boundary properties develops. The

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concept of grain boundary engineering, introduced by Watanabe [7], had a primary goal to improve the microstructure resistance to intergranular failure by increasing the fraction of special boundaries. As a consequence, the resistance to IGSCC is also expected to improve. The binary classification of the grain boundaries allows for a percolation approach in studying the overall resistance of a microstructure to IGSCC and estimating the impact of grain boundary engineering. For example, by percolation simulations the size of the susceptible boundary paths for a given microstructure and the threshold fraction of resistant boundaries required to interrupt the percolating network can be determined [8]. Palumbo et al. [9] proposed a geometric model for assessing IGSCC susceptibility that was based on the probability of assembling a critical length of susceptible boundary path. This model used a two-dimensional (2D) hexagonal representation of the microstructure and a binary classification of the grain boundaries. Since then, a number of improved 2D hexagonal percolation-like models have been proposed (e.g. Gertsman and Tangri [10], Shuh et al. [11], Lehockey et al. [12]). The improvements reflect the increase in understanding of grain boundary character distribution and the classification of the boundaries in a microstructure (e.g. Gertsman et al. [13] and Randle [14]), as well as the role of topological constraints on the grain boundary networks (e.g. Kumar et al. [15] and Minich et al. [16]).

While the improved percolation-like models address important features of the microstructure, a common drawback for all of them is that they do not account for the mechanical component of IGSCC, namely the crack driving force, which can vary during crack evolution. It should be mentioned that some of the percolation-like models consider the grain boundary orientation with respect to the applied load in the decision for further percolation, but the account for the mechanical component is limited to this. The true development of the crack driving force is clearly of importance. For example, crack branching, observed experimentally [1]-[6] and in model simulations [9]-[12], would reduce the driving force for the main crack. In microstructures with certain fractions of susceptible boundaries this may even lead to crack arrest when a percolation path would be still available. Another factor for crack driving force reduction is found in experimental observations by Marrow et al. [17] and Engelberg et al. [18] showing that the non-sensitised special boundaries encourage crack bridging ligament formation. The non-sensitised boundaries responsible for bridging are associated with twin growth [19]. These bridging ligaments are expected to retard crack propagation and, together with the effect of branching, possibly lead to crack arrest. This cannot be assessed on the basis of the percolation-like models. Jivkov et al. [20] proposed a mechanical, finite element based, model for studies of IGSCC in 2D hexagonal microstructures, capable of simulating bridging ligament formation and thus mimicking the real three dimensional behaviour. The model results demonstrated the significant impact of the mechanical load, branching and bridging on the developed crack lengths and critical thresholds of resistant boundary fractions when compared to percolation-like models predictions. Essential to the mechanical model are the failure properties of the susceptible and the resistant boundaries. Experiments have shown that susceptible boundaries fail by stress corrosion at crack opening displacements of the order of several nanometres without noticeable development of inelastic deformation (e.g. Thomas and Bruemmer [21] and [6]), and the bridges formed by resistant boundaries develop significant plasticity prior to rupture [17], [18]. These observations set limits to the model parameters representing the failure properties of the resistant and susceptible boundaries. Parametric studies reported by Jivkov et al. [22] can be used for experimental identification of these

parameters by comparing fractured and bridged areas, as well as crack opening displacements and bridge elongations.

Cracks are three-dimensional (3D) features however, and although the proposed mechanical model for 2D simulations is found to be consistent in terms of mechanics, and can qualitatively reproduce experimentally observed behaviour, its results cannot be directly translated to three dimensions [22]. A few 3D percolation models have been discussed in the past, such as Lim and Watanabe [23], Lehockey et al. [24], and Frary and Schuh [25]. All these use a regular representation of the microstructure with a unit cell, called truncated octahedron. With six squares and eight regular hexagons for boundaries, the truncated octahedron offers the closest to reality coordination with the neighbouring grains and allows regular tessellation of the 3D space. A computational mechanical model for 3D IGSCC propagation is proposed in this work, based on the regular representation of the microstructure with truncated octahedra. Unlike the percolation-like models, this mechanical model allows assessment of the influence of branching and bridging on the crack driving force. Using finite element calculations of the stress after each change in geometry due to crack advance, the model offers a more accurate simulation of crack evolution, because the crack advance depends upon the actual mechanical conditions at the crack tip. Selected simulation results for crack propagation and cracks coalescence are presented. These are obtained for fractions of resistant boundaries that have been experimentally determined in an as-received and grain boundary engineered austenitic stainless steel [18]. In the range of the fractions allowed and for the boundary conditions considered, crack arrest is found to be improbable in the model, but the effect of crack bridging on the crack driving force demonstrated is expected to reduce the kinetics of crack propagation.

At present the model does not account for the kinetics of crack propagation and should be viewed as an advance from the percolation-like models by including mechanical effects. In this sense it is a tool for assessing the relative microstructure resistance to intergranular fracture. Applied to IGSCC the model is relevant only to fully sensitised materials, similarly to percolation-like models. It should be appreciated, however, that the proposed model allows for the incorporation of the dependence of propagation kinetics on the local mechanical fields and this is a topic of ongoing work.

2. Model description

The computational framework for simulating intergranular stress corrosion crack propagation proposed in this work includes a model of the microstructure and a strategy for crack advance. The microstructure model has two levels of abstraction, physical and computational, which are described in two subsections that follow. A third subsection is devoted to the strategies for crack advance adopted.

2.1. Physical assumptions

2.1.1. Grain geometry

The truncated octahedron, also known as a tetrakaidecahedron or mecon, see Figure 1(a), is used as a unit cell that substitutes one grain of the microstructure. Among all the polyhedra filling 3D space compactly, this polyhedron offers a coordination number (number of neighbouring grains) closest to those observed in reality (see e.g. Wells et

al. [8]). With this grain geometry assumption, a region of a polycrystalline solid is approximated by an assembly of identical cells. A major geometry parameter is the unit cell size, D , given by the edge of the cell bounding cube. This can be thought as representing the average grain size in the microstructure.

2.1.2 Grain boundaries character

Grain boundaries (the common faces in the assembly of cells) are classified as members of either of two sets. The first set includes the special (low-angle, low-energy) grain boundaries, which are generally accepted as resistant to corrosion. These non-sensitised boundaries, which are observed experimentally to cause crack bridging, are associated with twin growth [19], and their frequency is linked to the degree of twinning. The second set consist of the random (high-angle, high-energy) grain boundaries, which are accepted as susceptible to corrosion. A major microstructure parameter is the fraction of resistant boundaries, f , given by the ratio between the number of resistant boundaries and the total number of boundaries. For given f , the distribution of resistant boundaries is randomly assigned. The authors are aware that a more precise description of the grain boundary network would require the consideration of crystallographic first-order constraints at triple junctions and second-order constraints at quadruple points [25]. However, a random distribution of two simple boundary types suffices for the aims of this work to demonstrate the capabilities of the mechanical model.

2.1.3 Material failure and grain boundaries properties

Crack propagation along grain boundaries is assumed to be the only mechanism of material failure. The present model does not consider the kinetics of the stress corrosion mechanism. Hence, the real time scale of crack growth is not determined and crack velocities are not calculated. Instead, susceptible boundaries are assumed to fail immediately when encountered by a crack, if their failure criterion is exceeded. The experimental evidence [6], [21], that during IGSCC the susceptible boundaries fail at nanometre-range opening displacements with insignificant inelastic deformations suggests that the failure strain of the susceptible boundaries, denoted by ε_{sf} , is below the macroscopic yield strain of the material ε_y . In contrast, resistant boundaries can yield and rupture well into the inelastic region [17], [18]. Correspondingly, the model allows for failure of resistant boundaries after a significant amount of accumulated inelastic strain. This failure strain of the resistant boundaries is denoted by ε_{rf} and is taken to be a fraction of the material ultimate tensile strain ε_u .

2.2. Computational concepts

Within the finite element method framework, perhaps the most straightforward model for intergranular crack propagation simulations in an assembly of cells is based on the use of continuum and interface elements. Each cell is tessellated into continuum elements (e.g. tetrahedral, triangular prisms and parallelepipeds) and is connected to the neighbouring cells with some interface elements, representing the grain boundaries. Wei and Anand [26] have used such a modelling strategy for an irregular assembly of grains, but their work has been limited to about 50 grains due to the requirement for computer resources involved in the simulations. With the regular assembly used here, the representation of one truncated octahedron requires at least 72 tetrahedral elements with 39 nodes if linear interpolation of the displacements is used. The kinematic unknowns,

i.e. the nodal displacements, are 117 in this case. In addition, each grain would bring 36 triangular interface elements to the total number of interface elements used in the model (grains at the boundary of the assembly will contribute less). For an array of cells required to reproduce microstructural behaviour sought (i.e. 30 x 30 x 30 grains, to simulate cracks extending at least 10 grain sizes), the size of the problem would rapidly become impractical with current resources. Therefore, at the computational level of modelling a discrete representation of the assembly is suggested. This is viewed as a good first approximation, while the continuum approach is left for the future as more computational power becomes available.

In the discrete model, each cell is represented as a geometrical point and its connections to the neighbouring cells, which are the grain boundaries in reality, are represented by linear structural members. The idea is illustrated in Figure 1(b). The finite element model consists of nodes, placed in the centres of the grains, and beam-type finite elements for structural members. Thus, the grains deformability is transferred to the deformability of the adjacent beam elements. The discrete representation leads to a significant reduction of the nodes and elements in the model and hence of the computational effort. The number of beam elements contributed by each grain is now 7, and the kinematic unknowns contributed are 6 – three translational and three rotational degrees of freedom per grain. The discrete model assumptions are described in the following subsections.

2.2.1 *Beam elements geometry*

Essential to the discrete model is the question of how well it represents the solid behaviour. The correspondence model-solid can be controlled by the selection of elements cross sections and elastic properties. The regular geometry used introduces preferential directions in the assembly. This makes it generally impossible to choose cross sections and properties so that the discrete assembly behaves as a continuum solid for all possible modes of deformation at once. For every particular deformation mode, however, such a selection is possible. In the model there are two distinguishable element types – longer, representing the square boundaries, and shorter, representing the hexagonal boundaries. In this work all beam elements are chosen to have circular cross sections with areas $A_4 = 0.106D^2$ and $A_6 = 0.358D^2$, for the beams representing square and hexagonal boundaries, respectively. With this selection the assembly studied subject to tensile deformation in the elastic range behaves like a solid under the same deformation while all elements have identical modulus of elasticity and Poisson's ratio.

2.2.2 *Solid mechanical properties*

The macroscopic mechanical properties are taken to be those of type 304 stainless steel. The constitutive relationship chosen is elastic-plastic with linear isotropic hardening, which for the purposes of this work describes well the real metal behaviour. The parameters involved in the constitutive description are Young's modulus, $E = 206$ GPa, Poisson's ratio, $\nu = 0.3$, yield (proof) strength, $\sigma_y = 205$ MPa, yield strain $\varepsilon_y = 0.002$, ultimate tensile strength, $\sigma_u = 515$ MPa and ultimate tensile strain (elongation) $\varepsilon_u = 0.4$.

2.2.3 *Beam elements mechanical properties*

Beam elements are ascribed the same mechanical properties as for the solid. In the course of crack propagation, however, the beams representing susceptible boundaries would fail upon reaching their prescribed failure strain ε_{sf} , which is a fraction of the

material yield strain ε_y , while the beams representing resistant boundaries would fail upon reaching their prescribed failure strain ε_{rf} , which is a fraction of the material ultimate tensile strain ε_u . This behaviour of the beam elements is shown schematically in Figure 2. The average strain, i.e. the strain in the centre of the cross section in beam elements is used in the calculations.

2.3. Crack advance strategy

The crack advance strategy uses finite element solutions for equilibrium of the evolving assembly with the applied loads. After a change of assembly connectivity due to grain boundary failure, an equilibrium solution provides the strains at all intact grain boundaries (i.e. at midpoints of the beam elements). These are necessary to decide upon further crack propagation in the next step. Every grain boundary, resistant or susceptible, with resolved strain above its failure strain is a candidate for failure in any step of the simulations. In a “single-failure” strategy only one boundary is allowed to fail at a time. As every failure event leads to redistribution of load that cannot be judged in advance, the single-failure strategy ensures that the recalculated equilibrium will deliver the correct strains. Strictly speaking, while the crack is advancing the mechanical equilibrium found by finite elements is physically unstable and turns into physical equilibrium only at crack arrest. The objective of the simulations presented here was to demonstrate the development of crack bridging and shielding and therefore equilibrium is not strictly required.

Figure 3 illustrates the approach for crack advance, on a two-dimensional mesh for clarity. The current crack is shown with a thick white line. All boundaries that are in contact with the crack surface are shown with thick dark grey lines. The search for candidates for failure is performed only among these boundaries, as they are considered to be in contact with the corrosive environment. For each grey boundary, the difference between its resolved strain by finite element solution, ε , and its failure strain is calculated. This difference is $\delta = (\varepsilon - \varepsilon_{sf})$ for the susceptible boundaries and $\delta = (\varepsilon - \varepsilon_{rf})$ for the resistant boundaries. Boundaries with $\delta > 0$ form the set of candidates for failure at the current solution step. If this set is empty, the crack is arrested and the assembly is in equilibrium with the applied loads. Otherwise, the boundary with largest δ is accepted as critical and the corresponding beam element is removed from the structure and the simulation continues. Departure from the single-failure strategy is made in simulations of coalescence of two surface cracks, presented later. In this case the entire set of candidates for failure boundaries is removed at the same step, in order to allow both cracks to grow as long as the driving force is sufficient. This presents a less accurate determination of strain fields and generally leads to an increased crack branching. The procedure was adopted to allow crack coalescence to be achieved in reasonable time, for the purpose of demonstrating the model capabilities. A more advanced procedure, which fails only a fraction of candidate boundaries, is being developed. This will enable equilibrium conditions to be approached and is a necessary step for the development of any kinetic model for environment assisted cracking.

The crack advance strategies, described above, are implemented by an in-house computer programme. The programme is responsible for creating the initial model and consecutively calling the external equilibrium solver, ABAQUS [27], deciding upon crack advance from the equilibrium solution and then changing the model if required.

3. Formulation of Problems

The solid region that is modelled via tessellation of the truncated-octahedral structure is shown in Figure 4(a) for reference. One assembly of grains has been used throughout the simulations reported here. With respect to a fixed coordinate system (X_1, X_2, X_3) , the assembly fills the region $\{-7.5D \leq X_1 \leq 7.5D, -20D \leq X_2 \leq 20D, 0 \leq X_3 \leq 25D\}$. The region contains 30000 grains, which form 198310 internal grain boundaries, modelled by 30000 nodes and 198310 beam elements in the finite element model. This structural model is nearly the maximum possible that can be simulated on a personal computer with 2GB of memory. For a given fraction of resistant boundaries f , all boundaries are classified as either resistant or susceptible in a random manner. The mechanical properties of the beam elements are given in the previous section. The load is symmetric and applied via prescribed displacements: $u_1 = -0.00375D$ along the boundary plane $X_1 = -7.5D$ and $u_1 = 0.00375D$ along the boundary plane $X_1 = 7.5D$. Displacements $u_3 = 0$ are prescribed along the boundary plane $X_3 = 25D$, while zero stresses are prescribed for all other boundary conditions. These boundary conditions introduce a homogeneous strain in the assembly $\varepsilon_\infty = 5 \times 10^{-4}$, equivalent to a homogeneous stress $\sigma_\infty \approx 0.5\sigma_y$, at the beginning of the simulations.

Simulations have been performed for either a single pre-crack or two pre-cracks introduced on the surface $X_3 = 0$. The single pre-crack is introduced into the structure by removing three beam elements intersecting the plane rectangle at $X_1 = 0$ given by $\{-0.5D \leq X_2 \leq 0.5D, 0 \leq X_3 \leq 0.5D\}$. This corresponds to an initial semi-circular crack of radius $0.5D$ placed at the origin of the coordinate system with surface perpendicular to the X_1 axis. The position of the single crack is shown in Figure 4(b) with a thick white segment. In view of the applied load, the single crack is expected to grow predominantly parallel to the X_2 axis on the surface and parallel to the X_3 axis in depth. The two pre-cracks for the coalescence studies are introduced in the same way but at different positions on the plane $X_3 = 0$, shown with thick dark grey segments in Figure 4(b). As for the single pre-crack, the two pre-cracks correspond to two initial semi-circular cracks with surfaces perpendicular to the X_1 axis. The expectation here is that the growing cracks will not remain parallel to the X_2 axis in the $X_3 = 0$ plane, but rather will develop curved paths as they eventually coalesce.

To compare the finite element results with analytical theory, the theoretical solution of the stress intensity factor for a semi-elliptical surface crack is used (see e.g. Tada et al. [28]). This stress intensity factor is a function of the position along the crack front, given by an angle φ , measured from the larger semi-axis a , and depends on the remote load, given by σ_∞ , and the dimensions of the two semi-axes a and b ($a > b$). Figure 5(a) illustrates a semi-elliptical crack, where the crack size in X_3 direction is assumed to be the larger semi-axis. The formula for the stress intensity factor is:

$$K_I = \frac{\sigma_\infty \sqrt{\pi b}}{E(m)} (1 - m^2 \cos^2 \varphi)^{1/4} \quad (1)$$

where m is the axes mismatch parameter given by

$$m = \sqrt{1 - \frac{b^2}{a^2}} \quad (2)$$

and $E(m)$ is the complete elliptic integral of the second kind, defined with

$$E(m) = \int_0^{\pi/2} \sqrt{1 - m^2 \sin^2 \theta} d\theta \quad (3)$$

To account for the finite geometry considered in Figure 4, correction factors have been applied to the value obtained by Eq. (1). These correction factors have been taken from 2D solutions that can be found in Ref. [28]. The correction factor for determining the stress intensity factor at point P in Fig. 5a is the one used for an edge crack of size a in a finite plate of thickness $W = 25D$ (see Figure 4) and is given as a function of the parameter $\alpha = a / W$ with:

$$F_3(\alpha) = 1.12 - 0.231\alpha + 10.55\alpha^2 - 21.72\alpha^3 + 30.39\alpha^4 \quad (4)$$

Eq. (4) gives an error of $\pm 0.5\%$ for $\alpha \leq 0.6$. The correction factor for determining the stress intensity factor at point Q in Figure 5(a) is the one used for a central crack of size $2b$ in a finite plate of thickness $W = 40D$ (see Figure 4) and is given as a function of the parameter $\alpha = 2b / W$ with:

$$F_2(\alpha) = \left(1 - 0.025\alpha^2 + 0.06\alpha^4\right) \sqrt{\sec\left(\frac{\alpha\pi}{2}\right)} \quad (5)$$

Eq. (5) gives an error of $\pm 0.2\%$ for any α . The finite geometry in the loading direction, X_1 , has not been accounted for.

Computationally, two stress intensity factors, considered as measures of the crack driving force, have been determined in a standard (approximate) way via the calculated stresses ahead of the crack front in X_2 and X_3 directions, respectively. An illustration of a developed crack, projected onto the X_2 - X_3 plane, is shown in Figure 5(b) in light grey, bounded by the crack front which is shown by a thick black curve. The dotted line shows the corresponding semi-elliptic crack of the same semi-axes that has been used for the comparisons. The rectangles ahead of the crack front in the two directions considered show the regions that are accounted for in determining the stress intensity factors. These rectangular regions are the projections onto the X_2 - X_3 plane of parallelepipeds that extend in X_1 -direction over the real crack thickness, i.e. several grains corresponding to the current crack geometry out of plane. The estimated plastic zones are dashed ($\sim$ one grain), while the rest of the rectangular regions are taken to span

over five grains in the corresponding direction. The calculation of the stress intensity factor in the X_3 direction is given as an example, which is to be compared to the value given by Eq. (1) at $\varphi = 0$ multiplied by the correction factor given by Eq. (4). The stress in the direction of load application, σ_{11} , (out of the plane of the figure) is the only stress taken into account in calculating the stress intensity factor. For any given distance from the crack front, r_3 , in steps of one grain diameter, a stress σ_{11} has been calculated by averaging the stresses in all grains with the corresponding X_3 -coordinate along the line shown in the figure and in thickness of the parallelepiped in X_1 -direction. Thus, an approximate relationship $\sigma_{11}(r_3)$ has been established. The function

$$\kappa(r_3) = \sigma_{11}(r_3) \sqrt{2\pi r_3} \quad (6)$$

possesses nearly linear behaviour in the considered range of r_3 and by extrapolating this linear relationship to $r_3 = 0$, the stress intensity factor at the crack front is found, i.e.

$$\sigma_{11}(r_3) \sqrt{2\pi r_3} \xrightarrow{r_3 \rightarrow 0} K_I^{FEM}. \quad (7)$$

The stress intensity factor in X_2 direction is calculated in the same manner and is compared to the value given by Eq. (1) at $\varphi = \pi/2$ multiplied by the correction factor given by Eq. (5). It should be noted, that the stress intensity factors thus determined, are not used in the criterion for grain boundary failure. Failures are decided solely on the base of local strain fields, found with finite element calculations.

A post-processing computer programme has been developed to implement the computational procedure described above for determining the stress intensity factors. The programme takes the finite element results after each step of the simulations and delivers the required stress intensity factors, as well as the current crack geometry including the number of bridges along the crack surface. The bridges can be in the elastic regime or yielding. In addition, the programme calculates the shielding effect of the bridges (total and that due only to yielding bridges) via the shielding stress intensity factor that the bridges create with respect to the corresponding crack fronts. The stress intensity factor, K_{br} , due to all bridges is an algebraic sum of the contributions from every bridge, given by the formula:

$$K_{br} = F_i(\alpha) P \sqrt{\frac{\pi}{2r}} \quad (8)$$

where P is the force in the bridge parallel to the loading (crack opening) direction, r is the distance to the corresponding crack front and $F_i(\alpha)$ is given by either Eq. (4) for stress intensity sought in X_3 direction or by Eq. (5) for stress intensity sought in X_2 direction. Eq. (8) is the solution for a crack loaded in Mode I by a couple of symmetric

concentrated forces of magnitude P placed on the crack surfaces at distance r from the tip [28].

The programme has been checked with developed cracks of nearly semi-elliptic shape with no bridges to compare with theoretical values given by Eq. (1) in combination with Eq. (4) or Eq. (5). These checks have been performed for a range of crack sizes from one grain to eight grains in depth. Results of the computational and theoretical approaches are almost identical, with an error less than 1%, which gives confidence in the stress intensity factors procedure as well as in the mechanical model adopted in this work.

4. Results and discussion

In the framework of the proposed model, the microstructure effects on stress corrosion crack propagation could be studied by a series of simulations with variable strengths of the susceptible and the resistant boundaries, and a variable fraction of the random distribution of resistant and susceptible boundaries. The results can be used to assess to what extent the observed resistant boundaries control crack growth and also how this changes with the strength and fraction of susceptible and resistant boundaries. The rather detailed parametric study, presented in Ref. [22] for 2D hexagonal microstructures, aimed at answering these questions. In the 3D cases studied here one crack propagation simulation for given fraction, distribution and properties of the grain boundaries required typically one week of computation time on a personal computer. Therefore, the results presented in this work do not cover the influences of all model parameters, and an extensive series of simulations is left for the future using more computational power. The aim of the 3D solutions conducted so far has been to demonstrate that the assumptions necessary for crack bridging in the 2D model are supported by the more realistic 3D implementation of the same mechanism.

4.1. Single crack propagation

Simulations performed with a single crack use failure strains of the susceptible and the resistant boundaries, fixed to the values $\varepsilon_{sf} = 0.1\varepsilon_y$ and $\varepsilon_{rf} = 10\varepsilon_y$, respectively. Seven different fractions of resistant boundaries, $f = 0.20, 0.25, 0.30, 0.35, 0.40, 0.45, 0.50$, have been considered. These enclose the experimentally determined fractions in microstructures of Type 304 austenitic stainless steels, for “as received” materials and “grain boundary engineered” materials [17]. For each fraction, five random distributions of the boundaries in the assembly have been simulated. Initial simulations showed that crack arrest was not achievable for the considered microstructures at the initial applied load. Therefore, artificial termination of the computations after 200 steps was adopted in the results presented.

Figure 6 illustrates crack development in a microstructure with $f = 0.35$. The crack configuration after 50 (a), 100 (b) and 200 (c) steps (failed boundaries) is shown. The plane X_2 - X_3 is drawn parallel to the initial crack plane. Holes in the crack surface designate bridges formed during the evolution of the crack. Ruptured grain boundaries with different orientation are shown with different shades of grey. To demonstrate the crack deviation from the original crack plane, a top-down view of the crack at step (c) is shown in Figure 6 (d).

The total crack area, including ruptured boundaries and bridges, projected on the initial plane is denoted by A_{crack} and is taken as the independent parameter in the following figures. This crack area is measured in units D^2 , noting that D is the grain size. In general, the crack's geometry in the X_2 - X_3 plane was observed to be maintained very close to a semi-circular shape during evolution. An amount of growth in X_2 -direction was balanced by subsequent growth in X_3 -direction, so that with reference to Figure 5, the substituting semi-elliptic crack maintains the relation $a \approx b$ for its semi-axes. This is demonstrated in Figure 7 and further confirmed by the calculated crack driving forces using the methodology described by Eq. (7) in the two directions, giving nearly identical development. In the absence of bridges, these crack driving forces coincide with the analytical predictions for stress intensity factors for semi-circular crack, given by Eq. (1) corrected for the finite geometry with Eq. (4) or Eq. (5).

Figure 8 shows an example of the development of the stress intensity factor (crack driving force) with crack area in a microstructure with $f = 0.35$. The stress intensity factor is determined via Eq. (7) in the X_3 -direction and normalised as shown in the figure. In addition, the theoretical value for a semi-circular crack of same semi-axes as the real crack is given, normalised accordingly. As mentioned already, the stress intensity in X_2 -direction follows closely the same curve as the one shown in the figure and the theoretical value is the same in both directions. Depicted with K_{sh} in the figure is the microstructure shielding effect, i.e. the difference between the crack driving force for an ideal plane semi-circular crack without bridges and the crack driving force determined in the real microstructure.

In 2D hexagonal microstructures, this shielding effect has been shown to be a complex combination of the contributions from bridges and branches that depend on the fractions, and to depend on the failure properties of the susceptible and resistant boundaries [22]. In the 3D model, at least for the crack extensions studied, the degree of crack branching out of the initial plane is minor (see Figure 6(d)) and correspondingly its contribution to the shielding effect is found to be insignificant. Nearly the entire shielding effect of the microstructure is due to the bridges. Figure 9 demonstrates the development of the stress intensity factor created by the bridges (crack bridging force) with respect to the crack front in the X_3 -direction for three different microstructures, $f = 0.25, 0.35, 0.45$. The crack bridging forces are calculated via Eq. (8) and normalised as in Figure 8. The difference between the bridges' contribution to the total shielding effect in the 2D and the 3D cases exhibits the importance of studying the three-dimensional behaviour of cracks in the microstructure.

The effect of the resistant boundaries' fraction on the shielding effect of bridges is demonstrated in Figure 10, where the results for K_{sh} for cracks of size $A_{\text{crack}} = 40D^2$ are plotted. It should be noted, that this crack size, developed close to the termination points of the simulations, corresponds to crack depths of 6-8 grains in different cases. These are short cracks with limited crack openings, which did not allow the bridges to start yielding. There were very few exceptions to this, but in all cases the crack sizes were not sufficient to observe bridge failure. Hence, the shielding effect demonstrated in the figure is principally due to elastically strained bridges. The figure shows a nearly linear dependence of this shielding effect on the fractions of susceptible boundaries. This is expected, because while the bridges are still in elastic regime and no failures occur, the shielding effect should be proportional to the total area of bridges (or their number in the present regular model), which is proportional to the fraction of resistant boundaries. The effect of yielding bridges and hence of resistant boundaries failure properties could

be studied only with larger models and more extensive crack growth. Yielding and failure of bridges has been shown in the 2D model [22].

The grain size does not enter the model explicitly. However, the effect of the grain size on the shielding stress intensity, K_{sh} , for a given crack size, can be estimated by rescaling the normalised data shown in Figure 9 for selected grain sizes. This can be done, because for the studied crack extensions the problem is still nearly linear, i.e. no failures or significant plasticity of bridges has developed. Hence, a microstructure with smaller, say twice as small, grains is expected to develop about twice as many bridges with approximately the same total area as a microstructure with given grain size. Such a rescaling is used in Figure 11, where the (non-normalised) shielding stress intensity factor is plotted as a function of the physical crack surface area, given in mm^2 . Two grain sizes, $D = 20 \mu\text{m}$ and $D = 40 \mu\text{m}$, are used. Two points on the two curves, corresponding to one and the same simulation result are depicted to demonstrate the rescaling. As can be read from the figure, the gain in shielding effect due to grain refinement can be very significant. The magnitude of the grain size effect should be regarded with caution, however, because the crack sizes and the number of bridges developed are small. If the simulated case of a crack which has propagated about 8 grains in depth is identified with a microstructure with grain size $20 \mu\text{m}$, then the crack depth in a microstructure with grain size $40 \mu\text{m}$ will be about 4 grains only. It can be said that for such short cracks the figure gives a good estimate for the grain size effect on shielding, but with further crack growth the gain in shielding effect is expected to decrease. This is due to a limit on the maximum size of the crack bridging zone, in the wake of the crack tip, due to the failure of bridges when the crack opening exceeds the bridge failure strain. This has been observed in 2D simulations [29].

4.2. Coalescence of pair of cracks

An example of the growth and coalescence of two cracks is shown in Figure 12 for the fraction of resistant boundaries $f = 0.35$ at three selected stages of the simulations. As in Figure 6 the plane $X_2 - X_3$ is set parallel to the initial cracks planes and ruptured boundaries of different orientation are shown with different shades of grey. Generally, after initial growth as single cracks, the two cracks “sense” the presence of each other and their paths curve out of their original planes to eventually coalesce. Bridges develop along the crack surfaces, but are not as visible as in Figure 6 because of the larger extent of out of plane branching. This is demonstrated in Figure 12(d) with the top-down view on the two cracks at the end of simulations. The much larger extent of crack branching out of the original crack plane, compared to the single crack behaviour in Figure 6, is due to the multiple-failure strategy adopted for crack coalescence simulations. As described in Section 2.3, according to this strategy all boundaries strained above their failure strains are allowed to fail at a time. This may overestimate the realistic degree of branching, and as discussed previously, a refined strategy for multiple-failure will be sought in future work. This would be advantageous not only for crack coalescence simulations, but also for single crack simulations in 3D, as it could reduce significantly the computational time to reach a prescribed crack size. Nevertheless, the simulations performed so far demonstrate that the model proposed is capable of reproducing crack coalescence. Future work will focus on the effect of special boundary fraction on crack coalescence behaviour.

4.3. Closing discussion

The simulations presented show that the 3D model, which is formulated in response to experimental observations, is capable of qualitatively reproducing crack advance, bridge formation and crack coalescence. They also show that for the fractions of resistant boundaries studied at the applied load, crack arrest is improbable. Due to the insufficient number of simulations with random distributions of boundaries for a given fraction, a deduction about the probability of arrest cannot be made. For the random distributions studied, crack arrest has not been achieved for any of the resistant boundaries fractions considered. The effect of bridges is to decrease the crack driving force, but their number is insufficient to lead to arrest. One may speculate at this stage, that if crack arrest occurs in an assembly with the studied fractions of resistant boundaries it will be due to a rare distribution of resistant boundaries along the front of the propagating crack rather than due to the effect of bridges. The bridges, however, are expected to have impact via their shielding effect on the crack growth kinetics, which will be studied in future using this modelling methodology. Grain size is predicted to have a significant effect on the amount of shielding developed for the short crack extension studied. With the increase of crack size this effect is expected to decrease and become strongly dependent on the failure properties of the resistant boundaries, a factor not considered in this work. A reduction in the failure strain of resistant boundaries with grain size may enable earlier failures of bridges, which may change the trends expected in microstructures with different grain sizes.

5. Conclusions

The work describes a model that has the potential to simulate intergranular stress corrosion crack propagation and crack coalescence in a realistic manner, including the phenomenon of crack bridging by resistant boundaries. The model accounts for the effects of external load magnitude and the failure properties of the susceptible and the resistant boundaries. The proposed discrete representation of the microstructure offers a substantial reduction of the computational resources needed to simulate crack propagation in grain assemblies of significant sizes. Although used for regular assemblies in the work, the discrete representation can be conveniently applied to more realistic irregular microstructures. Irregular assemblies are expected to have a homogenising effect on the geometrical and mechanical properties of the structural members of the discrete representation. In the present work, these were selected so that the regular (textured) assembly behaves like the substituted solid under the loading conditions considered.

In the model, crack propagation corresponds to a sequence of failure events and calculations for equilibrium. A boundary is in a critical state and is a candidate for failure if its resolved strain is larger than its failure strain – a model parameter. The single crack propagation simulations presented use a single-failure strategy for crack advance, i.e. at each simulations step the failure of one, the most critical, grain boundary is followed by equilibrium calculations to find the redistributed stress and strain fields. The simulations of coalescence of two cracks use an all-failure strategy, i.e. at a given step all critical boundaries are failed before next equilibrium is sought. While the former strategy is too restrictive and requires a substantial time for simulations, the later

is not precise in terms of the strain field development during crack propagation. A refined multiple-failure strategy, “intermediate” between the two used is to be developed to speed up the simulation process computationally.

In the model simulations performed, crack arrest was not observed for the resistant boundaries fractions of practical importance. However, a significant degree of crack tip shielding was developed. This demonstrates the increased capabilities of the proposed model, compared to the percolation-like models. The shielding was found to be primarily due to bridges formed by resistant boundaries and to depend on their fraction. This would be expected to reduce the crack propagation rate when crack tip strain-dependent kinetics is considered. Increasing the fraction of the resistant grain boundaries is therefore predicted to increase the resistance of short cracks to intergranular stress corrosion cracking. Decreasing the grain size is also predicted to increase the microstructure resistance, but with the note that this is significant for short cracks only. Further studies are needed to determine the influence of grain size over a larger interval of crack extensions.

Any further refinement of the model requires that it be related to experimental observations of intergranular stress corrosion crack extensions and distributions to tune the model parameters. The first step is the identification of the susceptible boundaries’ failure strain, which is essentially a local criterion for stress corrosion crack advance. This could be estimated from experimental results without noticeable bridging. The influence of the bridging ligaments could then be determined, comparing the local crack driving force in the model with the stress intensity predicted theoretically for a non-bridged crack of the same geometry and applied load. Another tuning parameter is the failure strain of the resistant boundaries, which may be connected to annealing twin thickness. This parameter could be estimated comparing experimental results showing failed bridging ligaments with model simulations.

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References

- [1] G. Palumbo, K.T. Aust, Structure-dependence of intergranular corrosion in high purity nickel, *Acta Metall. Mater.* 38 (1990) 2343-2352.
- [2] P. Lin, G. Palumbo, U. Erb, K.T. Aust, Influence of grain-boundary-character-distribution on sensitization and intergranular corrosion of alloy-600, *Scripta Metall. Mater.* 33 (1995) 1387-1392.
- [3] Y. Pan, B.L. Adams, T. Olson, N. Panayotou, Grain-boundary structure effects on intergranular stress corrosion cracking of alloy X-750, *Acta Mater.* 44 (1996) 4685-4695.
- [4] B. Alexandreanu, B. Capell, G.S. Was, Combined effect of special grain boundaries and grain boundary carbides on IGSCC of Ni-16Cr-9Fe-xC alloys, *Mater. Sci. Eng. A300* (2001) 94-104.
- [5] S.M. Bruemmer, G.S. Was, Microstructural and microchemical mechanisms controlling intergranular stress corrosion cracking in light-water-reactor systems, *J. Nuclear Mater.* 216 (1994) 348-363.
- [6] V.Y. Gertsman, S.M. Bruemmer, Study of grain boundary character along intergranular stress corrosion crack paths in austenitic alloys, *Acta Mater.* 49 (2001) 1589-1598.
- [7] T. Watanabe, An approach to grain-boundary design for strong and ductile polycrystals, *Res. Mechanica* 11 (1984) 47-84.

- [8] D.B. Wells, J. Stewart, A.W. Herbert, P.M. Scott, D.E. Williams, The use of percolation theory to predict the probability of failure of sensitized, austenitic stainless-steels by intergranular stress-corrosion cracking, *Corrosion* 45 (1989) 649-660.
- [9] G. Palumbo, P.J. King, K.T. Aust, U. Erb, P.C. Lichtenberger, Grain-boundary design and control for intergranular stress-corrosion resistance, *Scripta Metall. Mater.* 25 (1991) 1775-1780.
- [10] V.Y. Gertsman, K. Tangri, Modelling of intergranular damage propagation, *Acta Mater.* 45 (1997) 4107-4116.
- [11] C.A. Schuh, R.W. Minich, M. Kumar, Connectivity and percolation in simulated grain-boundary networks, *Philos. Mag.* 83 (2003) 711-726.
- [12] E.M. Lehockey, A.M. Brennenstuhl, I. Thompson, On the relationship between grain boundary connectivity, coincident site lattice boundaries, and intergranular stress corrosion cracking, *Corros. Sci.* 46 (2004) 2383-2404.
- [13] V.Y. Gertsman, M. Janecek, K. Tangri, Grain boundary ensembles in polycrystals, *Acta Mater.* 44 (1996) 2869-2882.
- [14] V. Randle, Mechanism of twinning-induced grain boundary engineering in low stacking-fault energy materials, *Acta Mater.* 47 (1999) 4187-4196.
- [15] M. Kumar, W.E. King, A.J. Schwartz, Modifications to the microstructural topology in f.c.c. materials through thermomechanical processing, *Acta Mater.* 48 (2000) 2081-2091.
- [16] R.W. Minich, C.A. Schuh, M. Kumar, Role of topological constraints on the statistical properties of grain boundary networks, *Phys. Rev. B* 66 (2002) 052101.
- [17] T.J. Marrow, L. Babout, B.J. Connolly, D. Engelberg, G. Johnson, J.-Y. Buffiere, P.J. Withers, R.C. Newman, High-resolution, in-situ, tomographic observations of stress corrosion cracking, In: *Environmentally Induced Cracking of Materials*, Shipilov S. (Ed.), Elsevier, 2005.
- [18] D.L. Engelberg, T.J. Marrow, R.C. Newman, L. Babout, Grain boundary engineering for crack bridging: A new model for Intergranular stress corrosion crack propagation, In: *Environmentally Induced Cracking of Materials*, Shipilov S. (Ed.), Elsevier, 2005.
- [19] T.J. Marrow, L. Babout, A.P. Jivkov, P. Wood, D. Engelberg, N.P.C. Stevens, P.J. Withers, R.C. Newman, Three dimensional observations and modelling of intergranular stress corrosion cracking in austenitic stainless steel, *J. Nucl. Mater.* (2006) in press.
- [20] A.P. Jivkov, N.P.C. Stevens, T.J. Marrow, The roles of microstructure and mechanics in intergranular stress corrosion cracking. In: *Simulation of electrochemical processes*, C.A. Brebbia, V.G. DeGiorgi, R.A. Adey (Eds.), WIT Press UK (2005) 217-226.
- [21] L.E. Thomas, S.M. Bruemmer, High-resolution characterization of intergranular attack and stress corrosion cracking of Alloy 600 in high-temperature primary water, *Corrosion* 56 (2000) 572-587.
- [22] A.P. Jivkov, N.P.C. Stevens, T.J. Marrow, A 2D mesoscale model for intergranular stress corrosion crack propagation. *Acta Mater.* (2006) in press.
- [23] L.C. Lim, T. Watanabe, Fracture-toughness and brittle-ductile transition controlled by grain-boundary character distribution (GBCD) in polycrystals, *Acta Metall. Mater.* 38 (1990) 2507-2516.
- [24] E.M. Lehockey, G. Palumbo, P. Lin, A.M. Brennenstuhl, On the relationship between grain boundary character distribution and intergranular corrosion, *Scripta Mater.* 36 (1997) 1211-1218.
- [25] M. Frary, C.A. Schuh, Connectivity and percolation behaviour of grain boundary networks in three dimensions, *Philos. Mag.* 85 (2005) 1123-1143.
- [26] Y.J. Wei, L. Anand, Grain-boundary sliding and separation in polycrystalline metals: application to nanocrystalline fcc metals, *J. Mech. Phys. Solids* 52 (2004) 2587-2616.
- [27] ABAQUS User's Manual, Version 6.4, Abaqus Inc., 2004.
- [28] H. Tada, P.C. Paris, G.R. Irwin, *The stress analysis of cracks handbook*, 3d Ed. ASME Pres, New York, 2000.
- [29] A.P. Jivkov, N.P.C. Stevens, T.J. Marrow, Meso-scale mechanical model for intergranular stress corrosion cracking and implications for microstructure engineering, *J. Press. Vessel Technol.-Trans. ASME* (2006) accepted.

Figure captions

Figure 1. The unit cell (truncated octahedron) substituting one grain in the model of microstructure: (a) solid representation at the physical level of modelling; (b) discrete representation at the computational level of modelling.

Figure 2. Schematic description of the deformation and failure behaviour of the susceptible (failing in a brittle manner) and the resistant (failing in a ductile manner) boundaries.

Figure 3. Illustration of the crack advance strategy. Candidates for failure boundaries are sought among the surface boundaries (thick dark grey lines) that are currently in contact with the crack surface (thick white lines).

Figure 4. Geometry of the problems studied: (a) the solid region tessellated into truncated-octahedral structure; (b) the location of the one or the two pre-cracks introduced on the surface.

Figure 5. Stress intensity factors calculation: (a) scheme for theoretical calculations with semi-elliptic surface crack; (b) scheme for calculations using stresses from finite element solution.

Figure 6. Crack morphology development in a microstructure with $f = 0.35$: (a) 50 simulation steps (50 failed boundaries); (b) 100 steps; (c) 200 steps; (d) top-down view of the crack at 200 steps.

Figure 7. In-plane crack shape evolution with crack area for $f = 0.35$.

Figure 8. Crack driving force evolution with crack area for $f = 0.35$.

Figure 9. Crack bridging force evolution with crack area for $f = 0.35$.

Figure 10. Microstructure shielding effect as a function of resistant boundaries fraction.

Figure 11. Grain size effect on microstructure shielding for $f = 0.35$. Figure obtained by rescaling Fig. 8 for the two grain sizes shown.

Figure 12. Development of a pair of cracks in a microstructure with $f = 0.35$: (a) 10 simulation steps; (b) 15 steps; (c) 20 steps; (d) top-down view of the crack at 20 steps.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 7. In-plane crack shape evolution with crack area for $f = 0.35$.

Figure 8

Figure 9

Figure 10

Figure 11

Figure 12