Modelling stress-corrosion microcracking in polycrystalline materials by the Boundary Element Method

I. Benedetti[1,a], V. Gulizzi[1,b] and A. Milazzo[1,c]

[1]Dipartimento di Ingegneria

Università degli Studi di Palermo, Viale delle Scienze, Edificio 8, 90128, Palermo, Italy

[a]ivano.benedetti@unipa.it, [b]vincenzo.gulizzi@unipa.it, [c]alberto.milazzo@unipa.it

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Abstract. The boundary element method is employed in this study in conjunction with the finite element method to build a multi-physics hybrid numerical model for the computational study of stress corrosion cracking related to hydrogen diffusion in polycrystalline microstructures. More specifically a boundary integral representation is used to represent the micro-mechanics of the aggregate while an explicit finite element method is used to model inter-granular hydrogen diffusion. The inter-granular interaction between contiguous grains is represented through cohesive laws, whose physical parameters depend on the concentration of inter-granular hydrogen, diffusing along the interfaces according to the Fick's second law. The model couples the effectiveness of the polycrystalline boundary element micro-mechanics model with the generality of the finite element representation of the inter-granular diffusion process. Few numerical tests are reported, to demonstrate the potential of the proposed technique.

Introduction

Stress corrosion cracking (SCC) is an example of environmentally assisted cracking in which the complex interaction between materials, chemically aggressive species and sustained loading activate processes that may lead to the premature failure of mechanical components, at critical loads lower than those observed in non-aggressive environments.

In several instances, SCC is related to the absorption and diffusion of hydrogen within the material, which, enhanced by local stress conditions, is one of the main SCC mechanisms. Hydrogen embrittlement may be responsible for the mechanical degradation of the inter-granular interfaces in polycrystalline materials, which include the majority of metals employed in engineering applications. In temperature and loading conditions typical of several structural applications, inter-granular interfaces are indeed the regions more susceptible to hydrogen diffusion [1].

Such considerations have led to the development of modeling strategies for SCC based on the use of cohesive zone modeling, as a theoretical/computational framework suitable for the representation of fracture processes along propagation paths a priori known; SCC has been effectively modeled using cohesive traction-separation laws whose parameters are degraded by the local concentration of hydrogen [2].

In this contribution, we discuss a cohesive grain-boundary model for hydrogen-assisted inter-granular stress corrosion cracking at the grain-scale in 3D polycrystalline aggregates, where the primary variables are inter-granular displacements and tractions, thus providing a tool particularly suitable for the study of inter-granular processes, sometimes dominant in polycrystalline materials, see e.g. Refs.[3-13]. The model is based on: *a*) a Voronoi representation of artificial polycrystalline micro-structures; *b*) a boundary integral representation of the micro-mechanics of individual grains, numerically addressed by the BEM; *c*) the employment of cohesive laws embodying hydrogen concentration dependent parameters, for modelling the inter-granular interfaces; *d*) a finite element model of the hydrogen diffusion along the interfaces; *e*) a suitable hybrid incremental/iterative solver for the solution of the micro-mechanical SCC problem. Next sections illustrate the key elements of the developed formulation.

Hybrid BEM-FEM modelling for polycrystalline hydrogen related SCC

As mentioned, the proposed hybrid micro-mechanical formulation is based on the combination of several items. Such items are individually described in the following paragraphs.

Artificial micro-morphologies. The model is built starting from a suitable morphological and crystallographic representation of the material microstructure. Here, three-dimensional (3D) Voronoi tessellations are employed as a first order approximation of real microstructures; each crystal is assigned random crystallographic orientation in the 3D space.

Boundary integral representation of grains micro-mechanics. Each grain within the aggregate is modeled as a linear elastic anisotropic domain with arbitrary spatial orientation and the Boundary Element Method (BEM) for 3D anisotropic elasticity is employed as numerical formulation [14]. The boundary integral equation employed for a generic individual grain $\mathcal{G}_k$ reads

$$\tilde{c}_{ij}^{k}(\boldsymbol{x})\tilde{u}_{j}^{k}(\boldsymbol{x})+\int_{B_C\cup B_{NC}}\tilde{T}_{ij}^{k}(\boldsymbol{x},\boldsymbol{y})\tilde{u}_{j}^{k}(\boldsymbol{y})dB^{k}(\boldsymbol{y})=\int_{B_C\cup B_{NC}}\tilde{U}_{ij}^{k}(\boldsymbol{x},\boldsymbol{y})\tilde{t}_{j}^{k}(\boldsymbol{y})dB^{k}(\boldsymbol{y}) \tag{1}$$

where $\tilde{u}_j^k$ and $\tilde{t}_j^k$ are components of displacements and tractions of points belonging to the surface of the grain $\mathcal{G}_k$, the tilde refers to quantities expressed in *local reference systems* set on the grain surfaces, $\tilde{U}_{ij}^k$ and $\tilde{T}_{ij}^k$ are the 3D displacement and traction anisotropic fundamental solutions, computed here using the spherical harmonics expression proposed in Ref. [15] and $\tilde{c}_{ij}^k$ are coefficient stemming from the limiting boundary collocation procedure [14].

Eq.1 is used for each grain, by direct collocation at the nodes of a suitably prepared grain-boundary mesh, which takes into account the specific features of the polycrystalline morphology [6,9]; the integrals are evaluated over the grain boundary, which generally includes *contact* interfaces B_C and external *non-contact* surfaces B_{NC}, over which suitable *boundary conditions* are assigned. The collocation of Eq.1 at the nodes of all the grains and the subsequent integration, performed within the framework of the BEM, produce the system of algebraic equation that, complemented by the set of boundary conditions and *interface equations*, which take into account the hydrogen-induced mechanical degradation, will allow the incremental-iterative solution of the polycrystalline SCC problem.

Interface cohesive model. The aggregate inter-granular interfaces are modeled through cohesive laws including a damage parameter, which allows tracking the irreversible degradation of the grain boundary, and a parameter accounting for the effect of the hydrogen concentration on the strength of the interface itself. Interfaces are assumed initially pristine, so that mechanical continuity holds at the beginning of the analysis. When, at an interface point, the local stress state fulfills the threshold condition

$$t_{eff}=\left[\left\langle\tilde{t}_n\right\rangle^2+\left(\frac{\beta}{\alpha}\tilde{t}_t\right)^2\right]^{\frac{1}{2}}\geq\hat{T}_{\max}(\phi)=(1-\gamma\,\phi)T_{\max} \tag{2}$$

then, a cohesive law is introduced to link the inter-granular tractions with the local displacement jumps. In Eq.2, the *effective traction* t_{eff} is expressed in terms of direct and shear tractions $\tilde{t}_n$ and $\tilde{t}_t$; α and β are cohesive parameters that give different weights to mode I and mode II loadings, $T_{\max}$ is the *interface cohesive strength* of an interface in absence of hydrogen relative concentration ϕ, whose presence is then accounted by introducing a local strength degradation through the term $(1-\gamma\,\phi)$, in which γ is a material parameter to be determined, either experimentally or through first principle calculations [2,16]. When Eq.2 is fulfilled, the traction-separation law

$$\tilde{\mathbf{t}}\left(\delta\tilde{\mathbf{u}},d^*,\phi\right)=\tilde{T}_{\max}(\phi)\mathbf{K}\left(d^*\right)\cdot\delta\tilde{\mathbf{u}} \tag{3}$$

is introduced at the interface, where $d^*\in[0,1]$ is an irreversible damage parameter, whose evolution takes place during local *loading*, such that the interface is pristine for $d^*=0$, while it is failed for $d^*=1$.

Upon failure, Eqs.(3) are replaced by the laws of *frictional contact mechanics*, see e.g. [7], that address *separation*, *stick* and *slip* of the micro-crack surfaces. From Eq.3 it is evident as hydrogen diffusion *weakens* the interface through a reduction of the cohesive strength; it is worth noting however how the presence of local stress is needed to induce local damage.

Hydrogen inter-granular diffusion modelling. In this work, only inter-granular diffusion of hydrogen is considered, which applies to large grains at room temperature. Hydrogen diffusion is modeled introducing the *relative concentration function* $\phi(\boldsymbol{\eta}, t)$, which represents the ratio between the local absolute concentration and the saturation concentration of hydrogen at the considered interface point, identified by the local coordinates $\boldsymbol{\eta}$ over the considered interface, which, due to the employed Voronoi representation, is a flat convex polygon. The relative concentration evolves with time in accordance with the Fick's second law

$$\frac{\partial \phi}{\partial t} = \frac{\partial}{\partial \eta_k}\left(D \frac{\partial \phi}{\partial \eta_k} \right) \tag{4}$$

where summation with respect to the repeated subscripts is assumed and D is the effective diffusivity. The previous law holds over the inter-granular interfaces and must be coupled with the consistent initial and boundary conditions. The diffusion process is numerically addressed employing a standard finite element formulation for diffusion problems, whose details can be found in Ref.[11].

Discretization and numerical solution. The presented formulation requires only meshing of the grain surfaces. Upon meshing and collocation, discretization and integration of Eqs.(1) [9], the following system is obtained

$$\begin{bmatrix} \mathbf{A}_1 & \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \ddots & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \mathbf{A}_{N_g} \\ \{\ldots & \boldsymbol{\Psi}\left[\boldsymbol{d}^*, \boldsymbol{\phi}(t)\right] & \ldots\} \end{bmatrix} \begin{bmatrix} \mathbf{x}_1 \\ \vdots \\ \mathbf{x}_{N_g} \end{bmatrix} = \begin{bmatrix} \mathbf{y}_1(BCs) \\ \vdots \\ \mathbf{y}_{N_g}(BCs) \\ \boldsymbol{\psi} \end{bmatrix} \tag{5}$$

where the blocks $\mathbf{A}_k$ contain columns of the BE matrices corresponding to the unknown displacements and tractions of the k-th grain, contained in the vector $\mathbf{x}_k$, while $\mathbf{y}_k$ derive from the applications of the known boundary conditions (BCs). The block $\boldsymbol{\Psi}\left[\boldsymbol{d}^*, \boldsymbol{\phi}(t)\right]$ implements the evolving coefficients of the interface equations taking into account the presence of hydrogen concentration, which is generally evolving with time according to the finite element system

$$\mathbf{M}\dot{\boldsymbol{\phi}} + \mathbf{K}\boldsymbol{\phi} + \mathbf{Q} = \mathbf{0} \tag{6}$$

where the overdot represents differentiation with respect to time. The overall phenomenon is modeled as a quasi-static process from the mechanical point of view, i.e. no inertial effects ever arise. Time can be assumed as primary variable and any contingent loading parameter, expressing progressive mechanical loading, can be expressed as a consistent function of time. Systems (5-6) must be solved simultaneously and their solution gives the evolution of inter-granular hydrogen concentration, as well as the evolution of the mechanical interface state. To track the evolution of a polycrystalline microstructure, a suitable *incremental/iterative algorithm* must be employed, whose detailed features can be found in Ref.[11].

Computational tests

Some results obtained from the described formulation are given here. Fig.(1a) shows a pseudo-3d morphology subjected to a uniaxial strain rate and a concentrated source of hydrogen. The corresponding macroscopic stress-strain curves as functions of the material parameters γ and D are reported in Fig.(1b), which highlights the detrimental effect of the hydrogen diffusion on the mechanical strength of the considered microstructures. As shown in Fig.(2), the presence of hydrogen also affects the evolution of the crack paths, favoring a more pronounced localization with respect to the case of no hydrogen diffusion.

Fig.(3a) shows a fully 3D 200-grain polycrystalline morphology subjected to a uniaxial strain rate and a concentrated source of hydrogen. As shown in Fig.(3b), even though the region in contact with the source of hydrogen is small compared to the whole morphology surface, the diffusion of aggressive species strongly affects the mechanical strength of the microstructure. Figure (4*a*) shows the damage distribution in case of no hydrogen diffusion at $t = 558$ s, whereas Figs.(4*b*,4*d*) and Figs. (4*c*,4*e*) show the hydrogen and damage distribution, respectively, for two selected pairs of the material parameters γ and D at the same time instant.

Also in this case, it is possible to notice that the presence of a localized source of hydrogen drives the crack initiation and induces a localization of the damage distribution.
The present results are intended as an illustration of the potential of the technique in the study of micro-cracking of polycrystalline components, including MEMS, in chemically aggressive environments. More comprehensive tests are presented and discussed in Ref.[11].

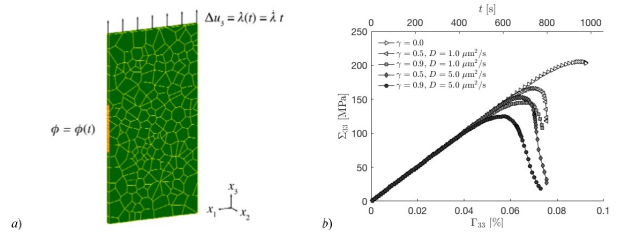

Fig.1: *a*) Pseudo-3D 200-grain polycrystalline morphology subjected to a prescribed strain rate ($\dot{\lambda} = 0.75 \cdot 10^{-4}$ μm/s) and a localized source of hydrogen; *b*) Computed macro stress-strain curves as functions of the material parameter γ and the effective diffusivity *D*.

Summary

In this study a hybrid computational framework for hydrogen assisted inter-granular stress corrosion cracking in polycrystalline microstructures has been described. The model is based on the conjoined use of a boundary element formulation for the polycrystalline micro-mechanics and of a finite element model for the inter-granular diffusion of hydrogen, whose interfacial concentration evolves according to the Fick's second law. The link between the polycrystalline mechanics and the diffusion process is provided by inter-granular cohesive interfaces embodying parameters that express mechanical degradation due to the hydrogen concentration evolution. The performed analyses provide a qualitatively sound representation of hydrogen assisted inter-granular stress corrosion cracking processes. The implemented framework may constitute a valuable tool in the study of polycrystalline materials and components subjected to aggressive environments.

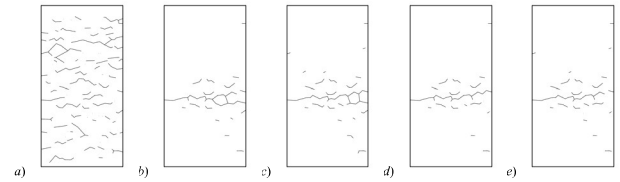

Fig.2: Crack paths at the last computed time steps for the case study shown in Fig.(1): (*a*) No hydrogen diffusion; (*b*) $\gamma = 0.9,\ D = 5.0\ \mu m^2/s$; (*c*) $\gamma = 0.5,\ D = 5.0\ \mu m^2/s$; (*d*) $\gamma = 0.9,\ D = 1.0\ \mu m^2/s$, (*e*) $\gamma = 0.5,\ D = 1.0\ \mu m^2/s$.

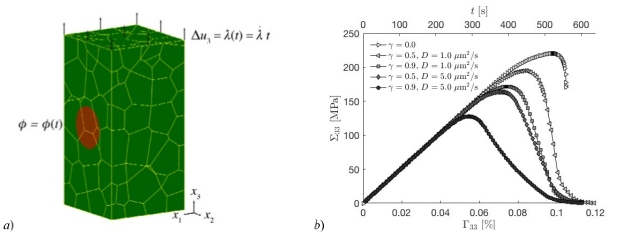

Fig.3: *a*) Fully 3d 200-grain polycrystalline morphology subjected to a prescribed strain rate ($\dot{\lambda} = 1.05 \cdot 10^{-1}$ μm/s) and a localized external source of the aggressive species; *b*) Computed macro stress-strain curves as functions of the material parameter γ and the effective diffusivity D.

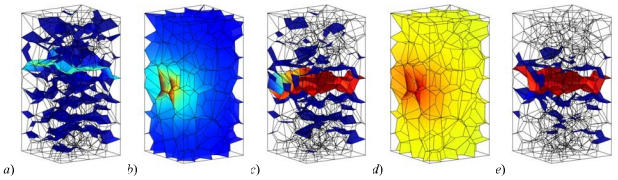

Fig.4: Contour plots evaluated at $t = 558$ s for the case study shown in Fig.(3): (*a*) Damage distribution in case of no hydrogen diffusion; (*b*) hydrogen relative concentration: $\gamma = 0.5, D = 1.0\ \mu m^2/s$; (*c*) damage distribution: $\gamma = 0.5, D = 1.0\ \mu m^2/s$; (*d*) hydrogen relative concentration: $\gamma = 0.9, D = 5.0\ \mu m^2/s$; (*e*) damage distribution: $\gamma = 0.9, D = 5.0\ \mu m^2/s$.

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