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Probabilistic Assessment of Induced Intergranular Stresses in Polycrystalline Materials

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ABSTRACT

Predicting various ageing mechanisms and material-degradation modes in polycrystalline materials under macroscopic mechanical loading requires the knowledge of stresses induced between the crystal grains. These stresses depend on the external loading, material properties, and the exact configuration of all the grains in the aggregate (including their sizes, morphologies, and lattice orientations). A simple perturbative model has been developed in which each grain boundary is characterized by its orientation, and the crystallographic orientations of surrounding grains, while the more distant neighborhood is modeled by homogeneous and isotropic material. Such setting allows us to solve the constitutive (elastic) equations analytically, while the effect of realistic anisotropic neighborhood is treated by adding Gaussian fluctuations to the local stresses predicted by the model. Comparison with the results of numerical finite element simulations demonstrates that while we cannot accurately estimate the induced (local) stresses on individual grain boundaries, we can very well reproduce their distributions on the grain boundaries of a chosen type corresponding to the same grain-boundary strength. This means that while the model is not able to predict the actual crack-initiation sites within the aggregate, it could still be used to compute the probability for cracking.

1 INTRODUCTION

Metallic alloys possess some remarkable properties, such as stiffness, tolerance to high temperatures, toughness, and strength. These qualities make them an attractive choice for structural materials in many industries, including the nuclear technology, where they are commonly used in reactor internals and primary coolant circuits. However, various ageing mechanisms and material-degradation modes can compromise the structural integrity of metallic components, thereby shortening their service life.

One of the most significant and harmful examples of such mechanisms is *intergranular stress-corrosion cracking* (IGSCC), which can affect various materials commonly encountered in nuclear reactors.[1] These include austenitic stainless steel (found in reactor pressure vessels and fuel claddings), Ni-based alloys (used in steam generators and welds), Zr-alloys, high-strength Al-alloys, ferritic steels, and more. IGSCC corresponds to the formation and propagation of local, non-ductile cracks along the boundaries of crystal grains. The process itself is quite

[1] In the presence of intense neutron fields inside the reactor, where irradiation effects such as irradiation embrittlement begin to play a role, a particular form of IGSCC known as *irradiation-assisted stress-corrosion cracking* (IASCC) becomes the primary cause of damage.

intricate, involving various physical mechanisms. It also requires specific critical conditions: the material must be susceptible to IGSCC, and exposed to a mechanical loading of sufficient magnitude (typically $\gtrsim 0.5$ yield stress). At the same time, it should be in contact with a corrosive agent that weakens the grain boundaries (GBs). Under these circumstances, IGSCC can occur even in materials that are otherwise corrosion-resistant.

The process unfolds in several distinct phases. The initial two phases, incubation (when a passive oxide layer gets formed on the grain boundaries, preceding the appearance of the first microcracks) and initiation (when the film ruptures, leading to intergranular oxidation), are relatively lengthy processes. In contrast, the final stage, crack propagation, where the cracks actually grow, only transpires in the last 10-20% of the component's service life. This underscores the critical importance of accurately predicting the initiation process (microcrack formation). It not only helps prevent structural failures, making engineering components safer, but also prolongs their service life, reducing the costs associated with frequent inspections and replacements.

Microcracks typically originate at the material's surface, which is in contact with the corrosive medium. Once a critical density of microcracks is reached, they begin to coalesce into macroscopic cracks, which become visible under optical microscopy. These cracks then advance into the material's depth, following an exponential growth pattern over time. Whether a microcrack forms on a specific GB depends on two factors: the induced stress on the GB and its inherent strength. In these terms, the effects of loading can be decoupled from environmental factors like corrosion and irradiation (influencing the GB strength). This allows us to focus on the stress aspect of crack initiation by estimating the stress field induced in the material as a response to applied external loading.

The most precise approach to this problem is provided by numerical simulations, such as those using the finite element (FE) method. However, they are not only computationally intensive but also often impractical because they require the detailed knowledge about microstructure of the entire aggregate (including orientations of crystal lattices and GBs, and the identification of possible defects) to compute stresses on individual GBs. In many cases, we are more interested in estimating the probability of cracking in a macroscopic, manufactured component rather than predicting the precise location of a crack in a specific piece. With this in mind, we developed a semi-analytical model [1] to calculate the distributions of intergranular normal stresses (INS) based on applied stress, material properties, and GB type, and hence treat the problem statistically. Numerical simulations are used to validate the results obtained from this simplified model.

2 NUMERICAL SIMULATIONS

Numerical simulations were conducted using the FE solver ABAQUS [2] within the small strain regime. The constitutive elastic equations were numerically solved for a Voronoi aggregate (depicted in Fig. 1) with 4000 grains and 31255 GBs consisting of 1039562 quadratic tetrahedral finite elements (C3D10). Periodic boundary conditions were enforced in all three spatial directions.

Preserving the topology of the grains, a crystallographic orientation can be independently assigned to each grain. Normally, these orientations are uniformly distributed throughout the aggregate and uncorrelated with the orientations of the GBs. Such a configuration is employed when computing the INS distribution on random GBs. However, when considering the INS distribution on a specific GB type, appropriate crystallographic orientations are imposed on the grains surrounding the largest available GBs. Due to topological constraints, this process was feasible for 1636 GBs containing 159461 finite elements, which represented approximately 17% of the total GB area.

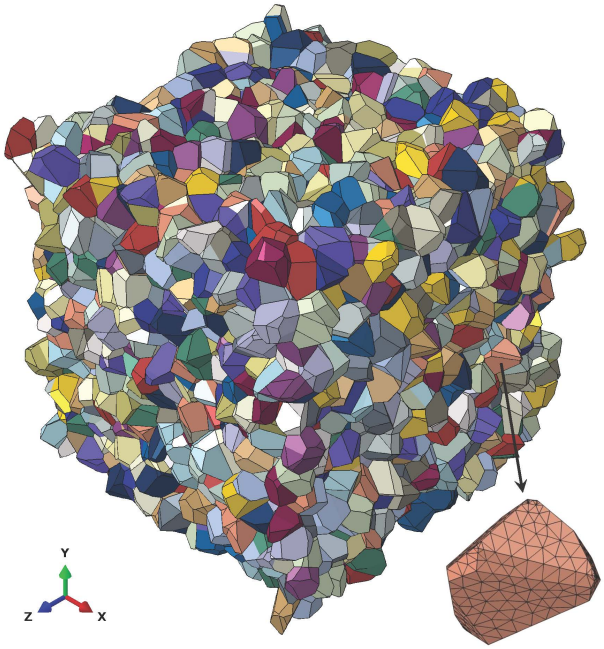

Figure 1: 3D periodic Voronoi aggregate with 4000 grains used in our numerical study. Different grains are denoted by different colors, which correspond to the orientations of their crystal lattices. The finite element mesh is shown for one selected grain.

3 PERTURBATIVE TREATMENT OF INDUCED STRESSES

Polycrystalline aggregates consist of randomly shaped grains with arbitrary orientations of their crystal lattices, see Fig. 2. When an external loading ($\boldsymbol{\Sigma}$) is applied to the aggregate, it induces stress at each GB. The size of the stress component along the GB-normal direction (σ_{nn}) depends on the material properties and the specific arrangement of all the grains. However, their contributions can be ordered by their relative importance. The primary factor is the orientation of the GB relative to the external loading, followed by the crystallographic orientations of both adjacent grains, and subsequently, the orientations of more distant grains, and so forth.

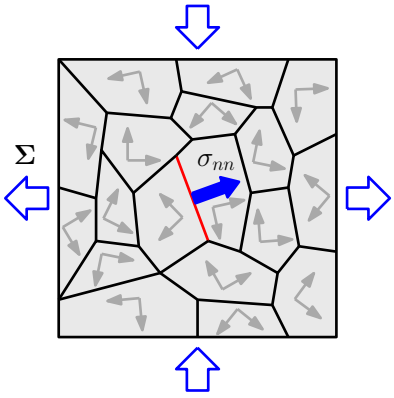

Figure 2: Schematic depiction of a polycrystalline aggregate under uniform external loading $\boldsymbol{\Sigma}$, inducing normal stress σ_{nn} at the examined GB.

The essence of our perturbative approach is to model all the grains in the neighborhood as isotropic [3]. In the lowest order of perturbation $(k = 1)$, all grains surrounding the GB are considered a part of the neighborhood. In the subsequent, next-to-leading order $(k = 2)$, only the bicrystal pair of grains enclosing the GB is not treated as isotropic. With each higher order, an additional layer of grains is introduced, whose crystallographic orientations are appropriately

taken into account.

In the absence of plastic deformations, we basically need to solve the generalized Hooke's law for the two grains on either side of the GB. Due to the linearity of elasticity, the induced opening mode stress σ_{nn} takes the form:

$$\sigma_{zz} = \sum_{i,j=1}^{3} a_{ij}\,\Sigma_{ij} = a_{xx}\,\Sigma_{xx} + a_{yy}\,\Sigma_{yy} + a_{zz}\,\Sigma_{zz} + 2\,(a_{xy}\,\Sigma_{xy} + a_{xz}\,\Sigma_{xz} + a_{yz}\,\Sigma_{yz})\,. \qquad (1)$$

In practice, the neighborhood in not isotropic. This affects the INS distribution, making it wider. In our model, the effect of an inhomogeneous neighborhood is imitated by adding random fluctuations to σ_{nn}. These fluctuations follow a normal distribution and tend to increase with the growing anisotropy of the material. Since fluctuations are uncorrelated to the estimated values of σ_{nn}, their combined distribution corresponds to the convolution of a modeled INS distribution with a Gaussian of appropriate width.

3.1 Grain-boundary type

Each GB type is characterized by five macroscopic parameters which specify the relationship between the orientation of the GB plane and the orientations of the crystal lattices in both adjacent grains. These parameters can represent the three Euler angles needed to relate the crystallographic orientations of both grains, along with the polar and azimuthal angles of the GB normal expressed in the coordinate system of one of the grains. Alternatively, two degrees of freedom per each grain are required to define the GB with unit-length vectors (a, b, c) and (d, e, f) that are perpendicular to the GB plane. In addition, the twist angle $\Delta\omega$ about the GB normal is used to unambiguously determine the relative orientations between both grains. Interpreting the values of Euler angles needed for these transformations as RGB coordinates allows us to assign a specific pair of colors to each GB, as shown in Fig. 3.

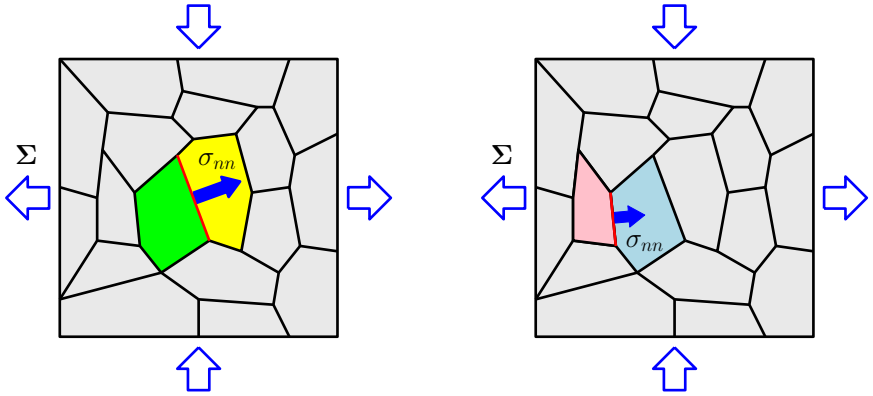

Figure 3: Illustration of two different GBs in the aggregate along with their respective GB types. The color of each grain represents the orientation of its crystal lattice relative to the GB orientation. In general, three Euler angles are required to rotate the crystal axes of a grain to the local coordinate system of the GB. The values of these angles determine the RGB coordinates, thus uniquely defining the colors of both adjacent grains for each GB.

GBs of the same type are expected to exhibit identical GB strength, regardless of their orientation relative to external loading. This implies that a common critical stress σ_c is applicable to all GBs of a given type, indicating microcrack formation at sites where the induced normal stress σ_{nn} exceeds this threshold. On the other hand, due to the different (although uniformly distributed) orientations of GBs within a specific type, the induced stresses experienced by individual GBs also vary, demanding an analysis of their respective distributions. Since numerical studies [4] indicate that the exact value of $\Delta\omega$ does not significantly affect the INS distributions, we can assign a random twist angle to each GB in the aggregate and refer to them collectively as the $[abc]$-$[def]$ GB type.

3.2 Isotropic Model

In the first-order approximation, all grains are treated as isotropic (see Fig. 4). The induced stress tensor exactly matches the external stress tensor, resulting in:

$$a_{zz} = 1 \quad \text{and} \quad a_{ij} = 0\,; \quad i,j \neq z\,. \tag{2}$$

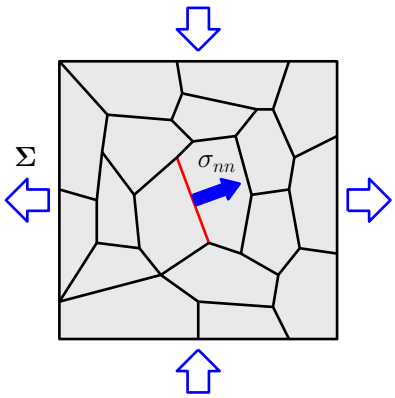

Figure 4: First-order approximation: isotropic model $(k = 1)$.

Since grains are all equivalent in this case, there is only a single GB type and stresses correspond to the INS distribution on random GBs. To facilitate a comparison with our analytical model, where stress remains constant within each grain and on its surface, we have generated a single value of normal stress per GB also in numerical simulations. This value is obtained as a weighted average across all finite elements that belong to it. The results of this analysis are shown in Fig. 5.

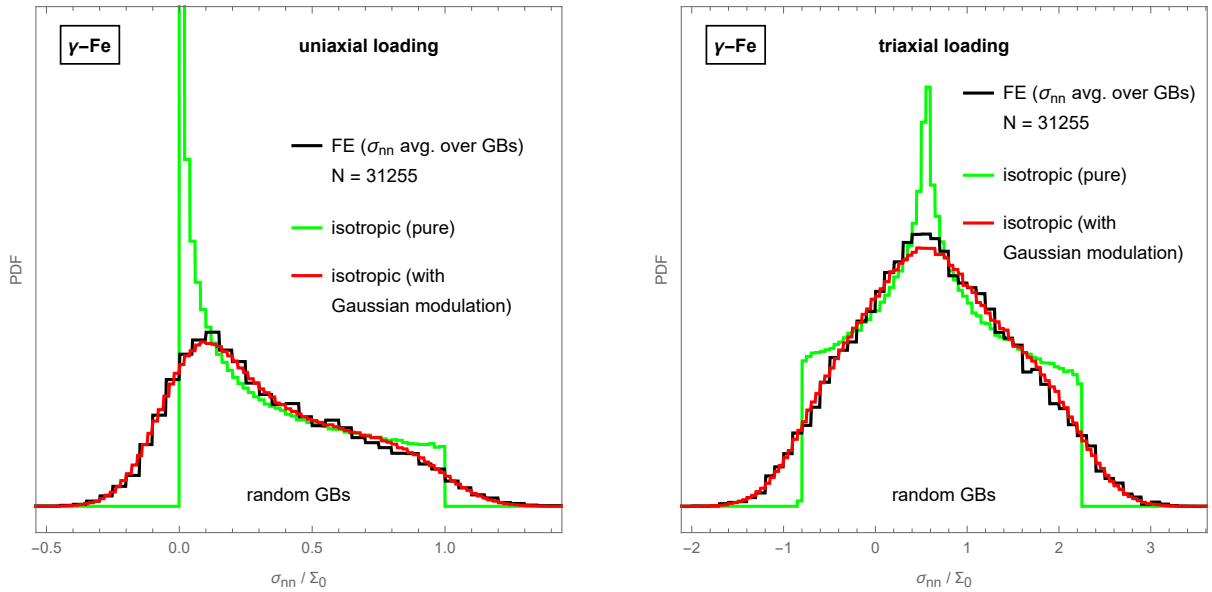

Figure 5: INS distributions on random GBs in a polycrystalline aggregate with randomly shaped and oriented grains (zero crystallographic texture), exhibiting γ-Fe properties. Their *probability density functions* (PDFs) are shown for two different macroscopic loadings: the *uniaxial* tensile loading of strength Σ_0, and the *triaxial* loading with strengths $2.25\,\Sigma_0$, $0.56\,\Sigma_0$, and $-0.80\,\Sigma_0$ along three orthogonal directions. A comparison is made between the results of a numerical (FE) study (*black*), a pure isotropic model (*green*), and its convolution with a Gaussian distribution (*red*).

The limitation here is the relatively small number of GBs in the aggregate, resulting in low statistics. To address this issue, in Fig. 6 we compare the model to normal stresses across all finite elements in the aggregate that lie on GBs. An excellent agreement was found upon convolution with a Gaussian distribution (of slightly larger width than previously).

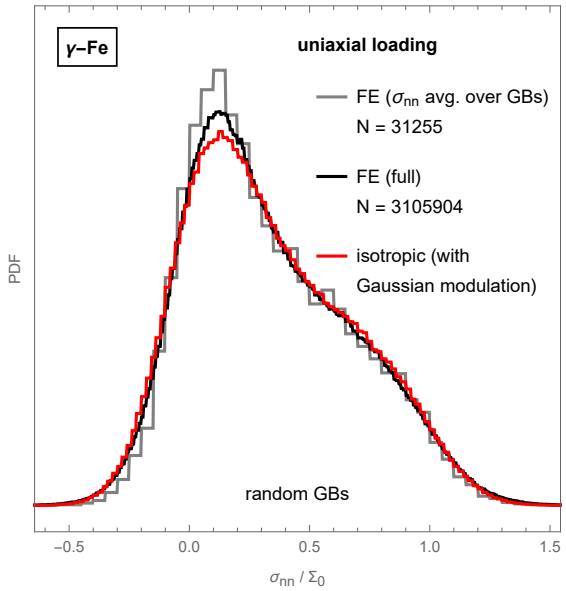

Figure 6: Comparison of three different methods for obtaining INS distributions on random GBs: 1) GB-averaged normal stresses corresponding to first producing a weighted average of normal stresses across all finite elements that belong to each GB, and then weighting these values with respect to GB size (*gray*); 2) Normal stresses on all finite elements, weighted by element size (*black*); and 3) Distribution generated by analytical isotropic model convoluted with Gaussian fluctuations. All three cases utilize properties of γ-Fe and uniaxial loading of strength Σ_0.

In summary, we have:

$$\textbf{isotropic case} + \textbf{Gaussian fluctuations} \approx \textbf{INS distribution on random GBs} \,, \tag{3}$$

where the width of Gaussian fluctuations is largely determined by the elastic anisotropy of the material and external loading. Similar conclusions have been reached in [3] (see Appendix F there).

3.3 Bicrystal Model

In the next-order approximation, we also consider the crystallographic orientations of the bicrystal pair enclosing the GB (see Fig. 7).

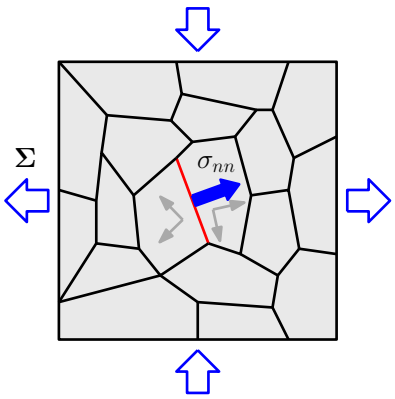

Figure 7: Second-order approximation: bicrystal model $(k = 2)$.

The system of coupled linear equations (generalized Hooke's law) for the two grains on either side of the GB is solved analytically by imposing the conditions of stress continuity and strain compatibility across the GB, and fitting the average strain of the bicrystal pair for a selected GB type and material to the FE results (for details, see [1]). As a result, $a_{ij} = f(\text{GB type, material properties})$, but it does not depend on the orientation of the GB.

Similar plots as in the isotropic case are shown in Figs. 8 and 9.

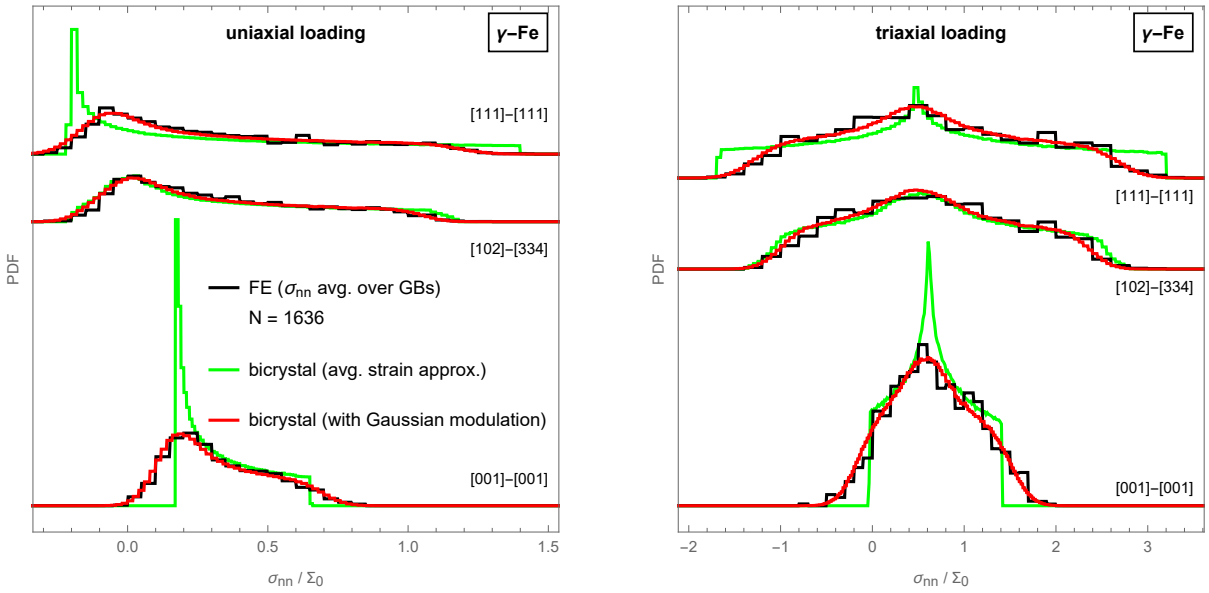

Figure 8: INS distributions for three representative GB types in a γ-Fe aggregate under two distinct uniform loadings. The PDFs correspond to the FE simulation results (*black*), a pure bicrystal model assuming average (macroscopic) elastic properties of the aggregate for the bicrystal pair of grains (*green*), and with the inclusion of normally distributed random fluctuations on top of the σ_{nn} estimates based on the GB-type specific boundary conditions for the bicrystal pair (*red*).

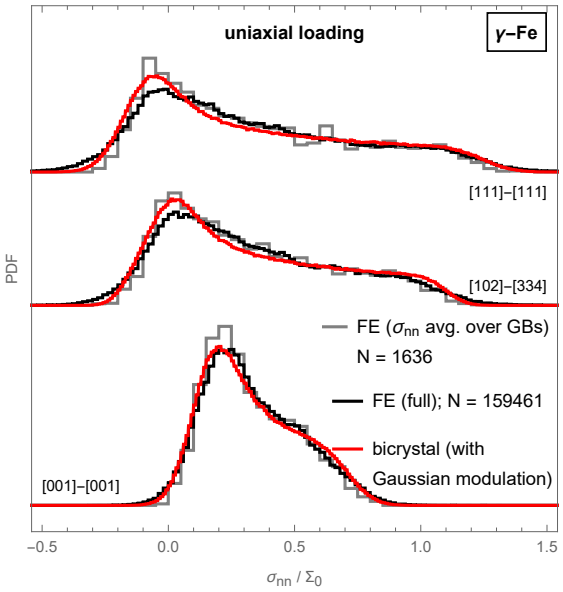

Figure 9: Numerical (*black*) and analytical (*red*) INS distributions for three distinct GB types in γ-Fe under uniaxial loading conditions. Additionally, the distribution of GB-averaged normal stresses (*gray*) is displayed for comparison.

Once again, good agreement can be observed for:

$$\textbf{bicrystal model} + \textbf{Gaussian fluctuations} \approx \textbf{INS distributions on individual GB types}\ , \qquad (4)$$

where the width of Gaussian fluctuation in this case depends on the effective GB stiffness and on the elastic anisotropy.

4 CONCLUSIONS

The perturbative model presented in this paper is designed for estimating GB-normal stresses in elastic polycrystalline materials, which represents the initial step in the probabilistic modeling of GB-damage initiation. The model offers the capability to quickly and efficiently generate INS distributions for arbitrary uniform loading, and for any chosen material and GB type.

Nonetheless, the method does have its limitations. It does not account for plastic deformations, assumes constant stress and strain within grains and on their boundaries, and provides estimates for stress distributions rather than the stresses on individual GBs.

ACKNOWLEDGMENTS

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