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Grain boundary planes: New dimensions in the grain boundary character distribution

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Abstract

The five parameter grain boundary character distribution quantifies the relative areas of different types of grain boundaries, distinguished by their lattice misorientation and grain boundary plane orientation. The viewpoint presented in this paper is that this distribution is a sensitive metric of polycrystalline structure that can be related to macroscopic properties. To demonstrate the influence of the grain boundary character distribution on macroscopic properties, the stored elastic energy is calculated in model microstructures.

Keywords: Grain boundaries; Microstructure; Texture

1. Introduction

The term grain boundary character distribution has traditionally been associated with the distribution of grain boundary lattice misorientations. In particular, in the field of grain boundary engineering, increases in the relative areas of boundaries with specific coincidence site lattice (CSL) misorientations have been correlated with exceptional macroscopic properties [1–3]. However, the three parameters that specify the lattice misorientation (or the type of CSL boundary) are not sufficient to determine the degree of coincidence in the grain boundary plane. To characterize the coincidence at the boundary plane, it is also necessary to specify the grain boundary plane orientation. In other words, five independent parameters are required to determine the density of coincident sites in the grain boundary plane. In this paper, the term grain boundary character distribution is used to refer to the relative areas of grain boundaries with specific lattice misorientations and grain boundary plane orientations; we express the viewpoint that this five-dimensional distribution is a sensitive microstructural metric that can be related to macroscopic materials properties.

There is considerable evidence that the orientation of the grain boundary plane is as important as the lattice misorientation. Perhaps the most well-known example is the $\Sigma 3$ misorientation in crystals with the face centered cubic crystal structure (the misorientation that results from a 60° rotation about a common $\langle 1\,1\,1\rangle$ axis). When the grain boundary plane of this bicrystal is oriented so that the crystals on either side are terminated by $\{1\,1\,1\}$ planes, the boundary is known as a coherent twin or stacking fault. These boundaries are typically very straight and where they form triple junctions, their dihedral angles are close to 180°. These two characteristics imply that small rotations of the grain boundary plane orientation are energetically costly and that the boundaries have very low energy. On the other hand, when the $\Sigma 3$ boundary is not comprised of $\{1\,1\,1\}$ planes, it is less straight and has smaller dihedral angles, suggesting that the energy is closer to the average. However, because of the experimental challenges associated with measuring the orientations of grain boundaries in opaque samples, grain boundary plane orientations are not usually characterized.

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Automated orientation mapping using electron backscattered diffraction patterns has made it possible to measure grain boundary plane orientations from serial sections of samples and, more recently, the development of stereological procedures for the analysis of orientation maps from single section planes have made the measurement of grain boundary plane distributions relatively routine [4–6]. Because it is now possible to measure these distributions, it is also possible to use them as a metric for macroscopic materials properties. The purpose of this paper is to demonstrate that the macroscopic properties of polycrystals depend on the grain boundary character distribution, even when other microstructural properties, such as texture, are constant. The remainder of the paper is organized in the following fashion. In Section 2, the procedures for measuring the five parameter grain boundary character distribution are briefly described. Next, the differences between the three parameter grain boundary distribution (misorientation alone) and the five parameter grain boundary character distribution are illustrated. In Section 4, the thermoelastic properties of model polycrystals with a constant topology and grain orientation texture are computed to illustrate the influence of the average misorientation and grain boundary plane orientation on the stored elastic energy.

2. Measuring the five parameter grain boundary character distribution

Crystal orientation maps on planar sections can be obtained using electron backscattered diffraction (EBSD) mapping. The orientation maps provide four of the five parameters necessary to classify each boundary: the three lattice misorientation parameters (Δg) and one of the two parameters needed to describe the orientation of the grain boundary plane ($\mathbf{n}$). To extract the full five parameter distribution, we use either serial sectioning [5] or a stereological analysis of the data from a planar section [6]. The main requirement for the stereological procedure is that a sufficient number of grain boundary traces (lines of intersection between a grain boundary and the surface) be characterized with respect to their lattice misorientation and orientation within the section plane. While the actual plane orientation for each trace is never known, it must be in the zone of the trace and if enough traces are observed from symmetrically indistinguishable bicrystals, then the probability that certain grain boundary planes appear in the microstructure can be defined. The number of grain boundary traces needed to generate a grain boundary character distribution depends on the crystal symmetry, the resolution of the five angular parameters, and the desired accuracy. In the original description of this technique, it was found that if the angular parameters in a cubic system are resolved at 10°, then 50,000 grain boundary traces are sufficient to determine grain boundary character distribution with an acceptable level of accuracy [6].

The analysis yields a normalized five-dimensional function, $\lambda(\Delta g, \mathbf{n})$, which is measured in units of multiples of a random distribution (MRD); values greater than one indicate planes observed more frequently than expected in a random distribution. It is convenient to select a specific misorientation according to the axis–angle convention and then plot the distribution grain boundary planes on a stereographic projection.

3. Use of the grain boundary plane in the characterization of special boundaries

For the past several decades, grain boundaries have been most frequently classified according to the imaginary coincidence of the interpenetrating lattices of the two adjacent crystals. In this scheme, boundaries are assigned a "Σ" value that is the reciprocal of the number fraction of coincident sites. It seems intuitive that boundaries between grains whose lattices exhibit partial coincidence would have lower energies and special properties and, as a result, the concept has enjoyed widespread acceptance. Interest in this concept was sparked by Aust and Rutter [7], who reported that boundaries with low Σ CSL misorientations (a relatively high degree of lattice coincidence) in Sn-doped Pb migrate at a rate much greater than general boundaries. Since that time, it has been reported that a range of macroscopic properties improve as the density of boundaries with low Σ CSL misorientations increases [2,3]. However, lattice coincidence is likely to have physical significance only when it occurs in the boundary plane. Thus, while CSL boundaries are defined by misorientation, the condition of high coincidence in the grain boundary is more restrictive; for a specific CSL misorientation, high coincidence at the interface occurs only at a few specific grain boundary plane orientations.

Measurements of the relative grain boundary energy of symmetric bicrystals conclusively demonstrate that low Σ CSL boundaries only have a low energy for certain symmetric boundary planes, such as occurs at the symmetric twin. For example, studies of $\langle 1\,1\,0\rangle$ symmetric tilt boundaries in Al, NiO, and MgO show that the energies of low Σ CSL boundaries depend on the orientation of the grain boundary plane [8–10]. For example, rotations of 70.53° and 109.47° about [1 1 0] are both Σ3 misorientations; however, a high degree of coincidence in the intergranular region occurs only for symmetric {1 1 1} boundary planes. This configuration is realized by a 70.53° rotation, but not by the symmetric 109.47° rotation. In the case of Al (see Fig. 1), the energy of 109.47° Σ3 rotation is approximately four times greater than the 70.53° Σ3 rotation [8]. The situation is similar for MgO and NiO [9,10]. Therefore, we surmise that the boundary plane has a significant influence on the energy and that any special properties that might be associated with a CSL misorientation are also influenced by $\mathbf{n}$.

When grain boundaries are distinguished on the basis of misorientation alone, the special characteristics of bound-

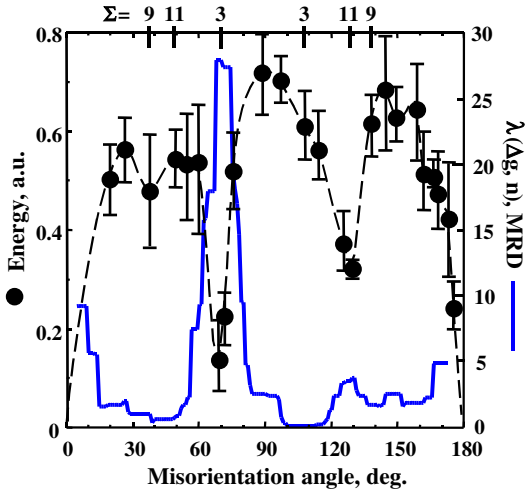

Fig. 1. Comparison of measured energies [8] (dashed line) to populations [11] (solid line) for symmetric [1 1 0] tilt boundaries. Both lines are simple interpolations between the data points. The misorientation angle is the angle between the [1 1 0] directions in the two crystals.

aries with specific grain boundary planes is easily overlooked. For example, a study of the five parameter grain boundary character distribution for commercially pure Al shows that the Σ3 misorientation occurs with roughly twice the frequency expected in a random distribution [11]. However, when the Σ3 boundary population is plotted as a function of the grain boundary plane orientation, there is a significant peak at the position of the coherent twin (see Fig. 2). In other words, when the area of coherent twins is compared to all possible boundaries in the five parameter space of grain boundary types, its area is more than 25 times that expected in a random distribution. In a metal with extensive twinning, such as brass, the area of twins is thousands of times that expected in a random distribution [12]. So, although the twin density in Al is small in comparison to brass, its relatively high population indicates that it has special characteristics.

Measurements of the grain boundary energy as a function of all five grain boundary parameters have shown that there is an inverse correlation between the grain boundary energy and the grain boundary character distribution [13].

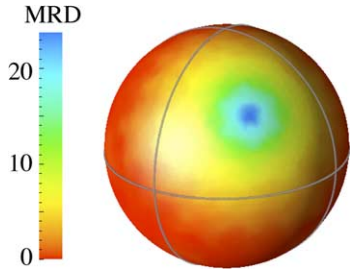

Fig. 2. Distribution of grain boundary planes for Σ3 boundaries in commercially pure aluminum. This is the distribution of all boundaries with a 60° misorientation about [1 1 1]. The data are plotted on the surface of a sphere in the crystal reference frame. The gray lines intersect at the positions of the ⟨1 0 0⟩ poles.

Therefore, we interpret the high population of coherent twins in Al as an indicator that it has a very low energy and this is consistent with the data in Fig. 1. It should be noted that the preference for the {1 1 1} twist-type boundary is not only a characteristic of the Σ3 bicrystal: for all misorientations about [1 1 1], the peak in the boundary population is at the orientation of the pure twist boundary [11]. In summary, because the high population and low energy of the Σ3 boundary are associated only with a unique grain boundary plane orientation, the five parameter description of the grain boundary character distribution is more accurate than the three parameter misorientation distribution for assessing the fractional content of boundaries with special characteristics.

4. The sensitivity of thermoelastic properties to the grain boundary character distribution

To illustrate the sensitivity of the thermoelastic properties of polycrystals to the five-dimensional grain boundary character distribution, we have used a finite element simulation to compute the local elastic energy density in specimens with model microstructures. The microstructures are all generated from the same 100 randomly oriented grains. In all cases, the orientations of the grains with respect to the sample normal are preserved. To generate the microstructures in 3(a), (b), and (d), the crystals were simply rearranged to achieve a relatively low, intermediate, and high average misorientation. The grain boundaries in Fig. 3(a)–(d) are colored with respect to their misorientation. Note that since only the arrangement of the crystals is changed, the orientation texture and the distribution of grain boundary planes are preserved. To alter the grain boundary plane distribution without changing the other microstructural parameters, the crystal axes of all 100 grains in Fig. 3(c) were rotated by 70°. Note that this transformation does not alter the distribution of misorientations and, because of this, Fig. 3(b) and (c) are identical.

To compute the thermal stresses and stored elastic energy, we used the object oriented finite element program, OOF [14]. The grains were assumed to have linear coefficients of thermal expansion with orthorhombic symmetry. The thermal expansion coefficients along the three axes were given the following values: $1.0\times10^{-5}\,K^{-1}$, $0.99\times10^{-5}\,K^{-1}$, and $1.01\times10^{-5}\,K^{-1}$. For simplicity, the elastic properties were assumed to be isotropic (Young's modulus = 500 GPa and Poisson's ratio = 0.33). Further, it was assumed that the length of the sample was 20 μm, that it was initially at mechanical equilibrium, and that it was then cooled by 500 °C. After assigning properties and orientations to each grain, the microstructures were discretized using ~68,000 triangular finite elements. The OOF software package (OOF, version 1.1.17, National Institute of Standards and Technology, Gaithersburg, MD) was used to calculate the responses of the discretized microstructures to thermal loads. In these simulations, the stress and strain at each point in the microstructure is calculated.

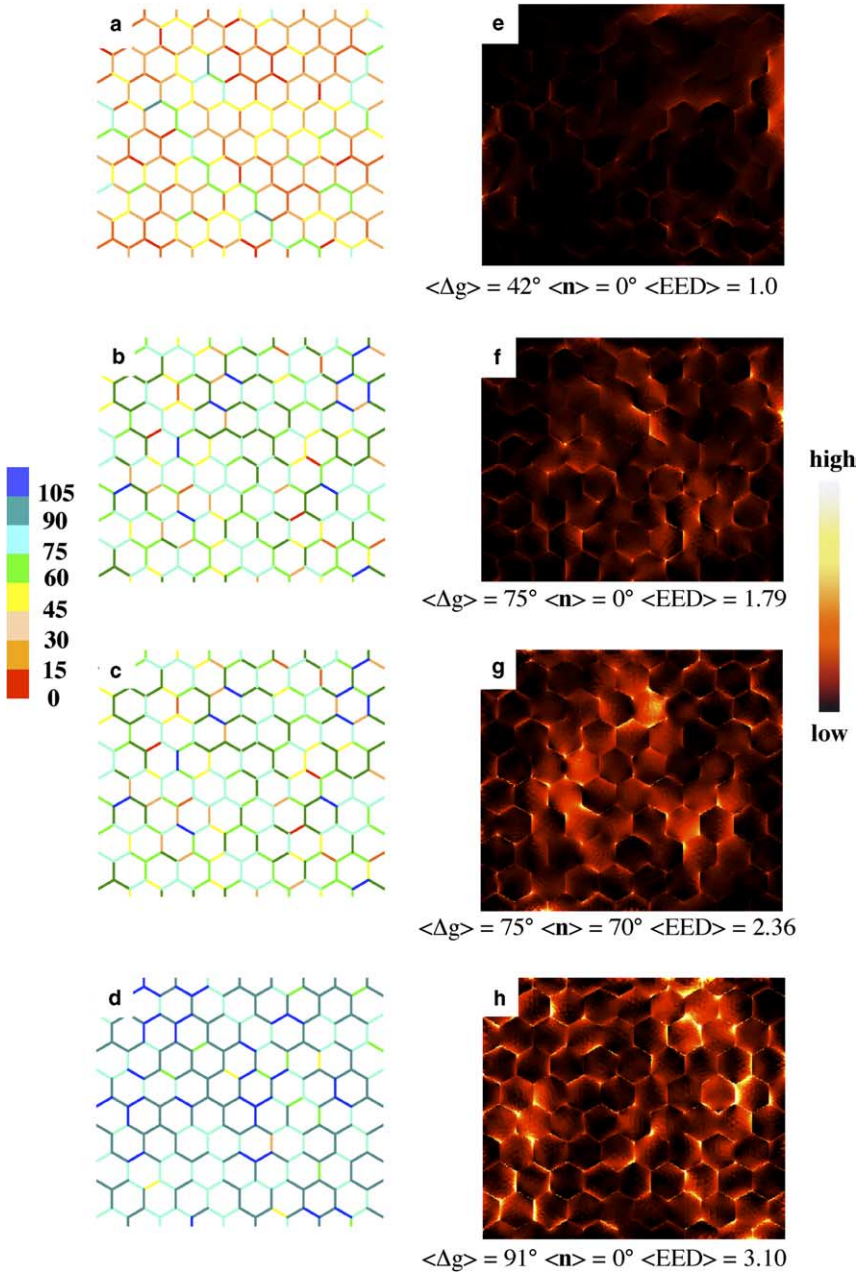

Fig. 3. Calculated elastic energy densities in microstructures with the same orientation distribution, but different grain boundary character distributions, after quenching. The average elastic energy densities (⟨EED⟩) are presented in relative units. (a–d) Grain boundary maps, colored by minimum misorientation angle. The lowest misorientations are red and the highest are blue. (e) EED in the starting microstructure, with a low average misorientation. (f) The EED after shuffling the grains to alter the misorientation distribution, ⟨Δ*g*⟩, while preserving the orientation distribution and boundary plane distribution. (g) After rotating all of the crystal axes by 70° with respect to the sample normal to alter the distribution of boundary planes, ⟨**n**⟩, while preserving the misorientation distribution and the orientation distribution with respect to the normal direction. (h) EED after shuffling the grains to increase the average misorientation.

All of the simulations in this work were carried out using plane-stress and free boundary conditions. The calculated elastic energy densities are plotted in Fig. 3(e)–(h), with lighter colors indicating a greater stored energy. Because the assumed properties and the thermal load were constant in each microstructure, the units have been normalized for a simplified comparison. The average energy densities are computed by averaging the stored elastic energy in each element in the microstructure.

The results clearly demonstrate that the stored elastic energy depends on both the distribution of misorientations and on the grain boundary plane distribution. In general, as the average misorientation increases, so does the stored elastic energy; as the average misorientation increases from 42° to 91°, the stored energy increases by a factor of three. In brittle materials, the fracture strength is inversely proportional to the average elastic energy density [15]. It should be noted that in addition to the average, the

maximum values also increase. The extreme values are important because, under mechanical loads, failure is most likely to initiate at the locations of maximum stored energy.

A particularly interesting point is that changing the orientations of the grain boundary planes, while fixing all other aspects of the microstructure, influences the stored energy (see Fig. 3(f) and (g)). When the grain boundary planes are rotated by 70°, the stored energy increases by 30% and the maximum value also increases. The influence of the lattice misorientation and grain boundary plane on the stored elastic energy changes with orientation texture [16]. The results of additional simulations, not presented here, show that the stored elastic energy is more sensitive to the grain boundary character distribution in textured microstructures. The microstructures described here were random and their properties therefore exhibit the minimum sensitivity to the grain boundary character distribution.

This example was selected to illustrate that although the orientation texture and grain size are microstructural parameters that strongly affect properties, the grain boundary character distribution also has a significant influence, even when these other factors are held constant. Changes in the average misorientation and the distribution of grain boundary planes alter the internal thermal stresses and polycrystals with more stored elastic energy are expected to fail at lower loads than those without as much stress. The difference in the thermal stresses in these materials supports the viewpoint that the grain boundary character distribution is a more sensitive metric of a polycrystal's structure and properties than grain size distribution and orientation texture alone. It is noteworthy that conventional microstructural statistics, such as the grain size distribution and orientation texture, can be determined from the data that are used to measure the grain boundary character distribution.

Although the five parameter grain boundary character distribution considers only pair-wise interactions among crystals, it appears from the data in Fig. 3 that extreme values in the elastic energy density are correlated over length scales that are larger than the dimensions of a single grain. It has been recognized for some time that the connectivity of boundaries influences certain properties and, procedures for describing these higher order correlations have been developed [17]. The source of such correlations is probably rooted in the crystallographic constraint that governs the types of misorientations that can meet at triple junctions [18]. The impact of this constraint on the possible populations of special boundaries has been explored [19,20] and, more recently, the analysis has been extended to longer range correlations [21]. Future work will be directed at developing an explanation for the correlations among the general boundaries that lead to the patterns of stored energy apparent in Fig. 3.

5. Conclusions

Finite element calculations have been used to show that changes in the five parameter grain boundary character distribution alter the internal thermal stresses within polycrystals. The examples demonstrate that when the orientation texture and grain size are fixed, macroscopic properties can still be affected by changes in the distribution of misorientations and grain boundary plane orientations. This supports the viewpoint that the grain boundary character distribution is a sensitive metric of polycrystalline structure that can be related to macroscopic properties.

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References

[1] Watanabe T. Res Mech 1984;11:47.
[2] Palumbo G, Lehockey EM, Lin P. JOM 1998;50:40.
[3] Randle V. Acta Mater 2004;52:4067.
[4] Rohrer GS, Saylor DM, El Dasher BS, Adams BL, Rollett AD, Wynblatt P. Z Metall 2004;95:197.
[5] Saylor DM, Moraweic A, Rohrer GS. Acta Mater 2003;51:3663.
[6] Saylor DM, Adams BL, El Dasher BS, Rohrer GS. Metall Mater Trans 2004;35A:1981.
[7] Aust KT, Rutter JW. Trans TMS-AIME 1959;215:119.
[8] Hasson GC, Goux C. Scripta Metall 1971;5:889.
[9] Dhalenne G, Dechamps M, Revcolevschi A. J Am Ceram Soc 1982;65:C11.
[10] Kimura S, Yasuda E, Sakaki M. Yogyo-Kyokai-Shi 1986;94:795.
[11] Saylor DM, El Dasher BS, Rollett AD, Rohrer GS. Acta Mater 2004;52:3649.
[12] Kim CS, Hu Y, Rohrer GS, Randle V. Scripta Mater 2004;52:637.
[13] Saylor DM, Moraweic A, Rohrer GS. Acta Mater 2003;51:3675.
[14] Carter WC, Langer SA, Fuller Jr RE. The OOF Manual. Version 1.0.8.6. National Institute of Standards and Technology (NIST), 2001.
[15] Kim C-S, Massa TR, Rohrer GS. Int J Refract Met Hard Mater 2006;24:89.
[16] Saylor DM, Fuller Jr ER. Private communication.
[17] Adams B, Olson T. J Prog Mater Sci 1998;43:1.
[18] Miyazawa K, Iwasaki Y, Ito K, Ishida Y. Acta Crystallogr 1996;A52:787.
[19] Minich RW, Schuh CA, Kumar M. Phys Rev B 2002;66:052101.
[20] Schuh CA, Minich RW, Kumar M. Philos Mag 2003;83:711.
[21] Schuh CA, Frary M. Scripta Mater 2006;54:1023.