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Grain boundary misorientation distributions

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Abstract

This review describes the theoretical basis, experimental considerations, data analysis and applications of studies of grain misorientations in cubic materials. Recently, there have been considerable advances in experimental techniques, allowing huge sample populations of misorientations to be collected. To handle such amounts of data, and to balance sacrificing detail for convenience, poses challenges.

1. Introduction

A 'grain boundary disorientation' is the crystallographic orientation relationship between two neighbouring grains of the same phase. Its importance in materials science and related solid-state disciplines is that the total disorientation across a grain boundary is a primary factor controlling its properties, for example, energy, diffusivity and mobility. During the last five years or so, there have been significant developments in practical techniques and it has become possible to obtain vast quantities of data from sample populations comprising hundreds or thousands of misorientations.

This review will encompass a brief description of the theoretical framework for describing misorientations, followed by a relevant description of state-of-the-art methodologies for data collection, data representation, and data analysis. The review will confine itself to studies in cubic materials, and will mention only briefly extended application of misorientation analysis, namely as a starting-point for the Rodrigues vector formulation and the coincidence site lattice (CSL). This is not to say that these two applications are of little importance, but rather that there is insufficient space here to describe them adequately.

2. Theoretical framework

The orientation of a grain can be defined by an 'orientation matrix', g. The misorientation between two neighbouring grains, called grain 1 and grain 2, which share a common reference frame is given by:

$$M = g_1^{-1} g_2 \tag{1}$$

where M is the matrix which embodies the misorientation between g_2 and g_1, and g_1 is arbitrarily chosen to be the reference orientation.

Although the matrix needs to be used for calculation, the most useful, popular and graphic orientation descriptor is the 'angle and axis of misorientation' or 'angle/axis pair' which can be easily extracted from the elements of M. In the cubic crystal system there are 24 'crystallographically-related solutions' of the misorientation. Although they are entirely interchangeable, it is conventional to quote the angle/axis pair containing the lowest angle, termed the 'disorientation', to describe a misorientation [1]. This allows low angle boundaries (usually defined as $<15°$) to be immediately recognised. For example, an annealing twin boundary is usually described by its disorientation, $60°/\langle 111\rangle$, although another crystallographically-related solution, e.g. $70.5°/\langle 110\rangle$, would be equally valid. This almost universal use of the disorientation entails certain disadvantages and will be discussed further in Section 5.1.

To describe a grain boundary misorientation fully, the concept of degrees of freedom can be invoked: one degree of freedom is required to describe the misorientation angle, and two each are required to describe the axis of misorientation and grain boundary plane, respectively. The indices of the grain boundary plane can be expressed in the coordinate systems of both grains, and the misorientation

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matrix transforms between the two. In some circumstances it may be pertinent to decompose the full misorientation into tilt and twist components, or to describe the misorientation using the 'interface/plane scheme'. Details of these aspects of misorientations formalism can be found elsewhere [2–4].

Most experimental work on misorientations in polycrystals neglects the grain boundary plane as a variable and records only the angle/axis pair. This is because there are considerable experimental challenges involved in obtaining the plane indices, whereas orientation measurement has become routine with use of modern techniques (Section 3). However, the boundary plane orientation has a major effect on boundary properties [5]. In the authors' view it is valuable to measure grain boundary plane orientations or at least to take into account the missing information [6,7].

Grain boundaries do not of course exist in isolation in polycrystals; grain boundaries meet at 'triple junctions', whose geometry is defined by the misorientations of the three constituent boundaries in the triple junction. The connectivity of the entire grain boundary network is therefore interdependent, which is discussed further in Section 5.2

3. Experimental methodology

In recent years dramatic advances have taken place in electron microscopy-base techniques to measure orientations, from which misorientations are calculated according to Eq. (1). In the scanning electron microscope (SEM) automatic solve algorithms for electron back-scatter diffraction (EBSD) patterns can be coupled to beam or stage scanning of the specimen to produce an 'orientation map' [*3]. The orientation changes associated with grain boundaries depicts them in the map. For example, Fig. 1 shows an orientation map from a nickel-based superalloy with the grain boundary network enhanced by delineation. Grain boundary statistics, based on misorientation, can be extracted from such maps. The statistic usually quoted is the length (i.e. projected area) of certain grain boundary types, such as low angle boundaries, twins or other CSLs, which can be measured and outputted as a proportion of the total grain boundary length. Recently it has been ascertained that there are appreciable errors associated with calculation of misorientation axes from EBSD data when the misorientation magnitude is $<5^\circ$ and so caution should be exercised for these cases [**8].

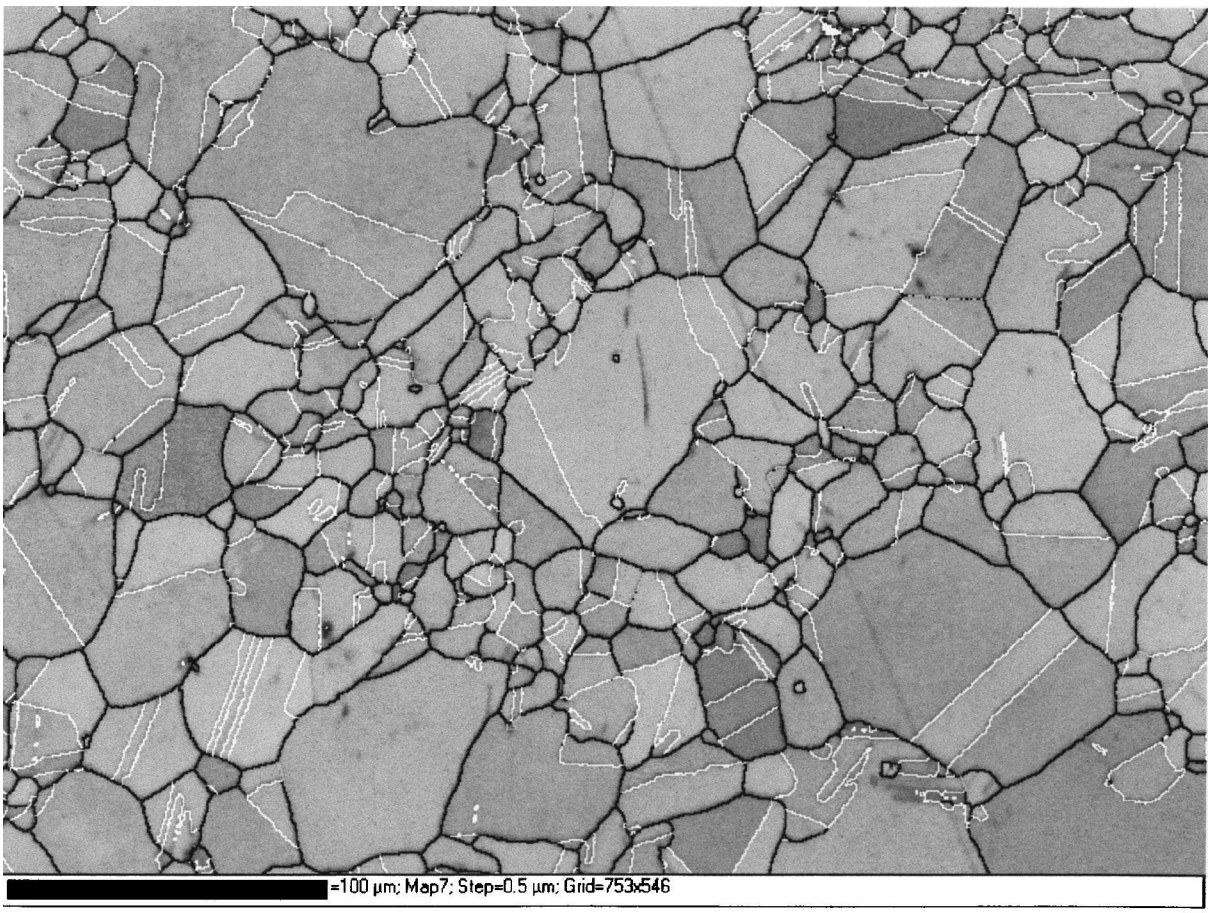

Fig. 1. Orientation map from a nickel-based superalloy, having high-angle grain boundaries delineated in black and twin-related boundaries delineated in white.

A frequently asked question is 'how many misorientations are needed to represent accurately the entire distribution?' One approach is to test continually the randomness of the distribution until an asymptotic result is discerned. This procedure defines the minimal data required [**9].

Although automated EBSD offers a tremendous advance in terms of the size of sample populations there is a danger that more subtle aspects of the data, such as how close a boundary misorientation is to a CSL and how the misorientation varies along the boundary, will be overlooked.

4. Data output and analysis

The most popular outputs of grain boundary misorientation data are as follows:

- Angle alone
- Angle/axis pair
- Angle/axis combined
- Continuous functions

The disorientation angle alone is quite a common representation, and it is often referred to as the 'misorientation' [10]. Only one of three independent variables is specified rather than all three but, although it is not the true misorientation, in many cases it is an acceptable approximation [11]. The usefulness of the angle distribution is enhanced by comparing it with the distribution obtained for random misorientation pairs in cubic crystals, often called the 'Mackenzie plot' [12,13]. Fig. 2a shows a recent experimental example where boundaries between equiaxed grains have the form of the Mackenzie plot, i.e. a random distribution, and boundaries between columnar grains have a non-random misorientation angle distribution [**14].

The misorientation axis in the angle/axis pair is usually displayed in a single unit triangle of the stereogram, accompanied separately by the angle distribution, e.g. [15].

This has the disadvantage that the pairing of individual angles and axes is not obvious. Similarly to the case for misorientation angles, there is a distribution of disorientation axes for the random case [12] as shown in Fig. 2b, against which an experimental distribution can be evaluated.

The Rodrigues vector is based on a function which combines the angle and axis, and is plotted in Rodrigues–Frank space [16]. Since this approach has many advantages for displaying misorientations, it is surprising that it has not been adopted more widely by the materials community. A CSL analysis also relies on a combination of the angle and axis. The value of the CSL notation is that there is some evidence of a relationship between CSL misorientations and properties, particularly for the case of the annealing twin [17]. Misorientations can also be expressed as a continuous function called a 'misorientation distribution function', MODF or MDF, which is described in detail elsewhere [*3].

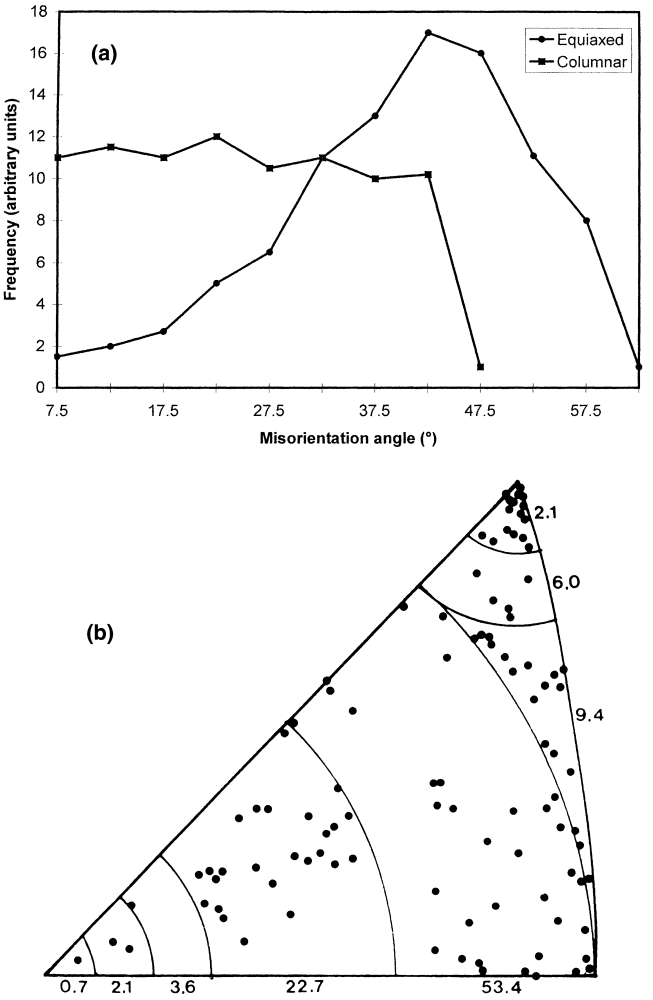

Fig. 2. (a) Disorientation angle distributions ('Mackenzie plots') for two types of grains, equiaxed and columnar, in nickel (adapted from Ref. [**14] with permission from the Minerals, Metals and Materials Society (TMS) and ASM International). The equiaxed grains have the form of a random distribution. (b) Disorientation axis distribution from grain boundaries in a nickel alloy, displayed in a single unit triangle of the stereogram. The numbers refer to the proportions of axes residing in each delineated region for a random distribution.

5. Applications of misorientation

The main objective of grain boundary misorientation-related investigations is usually either to seek some relationship with a particular material property, or to gain topological information about aspects of the grain boundary network in a polycrystal.

5.1. Grain boundary properties

The grain boundary misorientation is essentially a geometric measurement and makes little direct contact

with physical properties of the grain boundary, which are dominated by effects relating to the free volume at the grain boundary and its defect structure as described by a dislocation model. Only grain boundaries having certain misorientations, i.e. low angle types and CSLs, can be effectively described using a dislocation model. For simple cases (e.g. a symmetrical tilt boundary) a change in the density of the same kind of dislocations changes the rotation angle whereas changing the nature of grain boundary dislocations accommodates a change in the grain boundary plane or rotation axis [18]. Generally, the free volume is governed by the grain boundary plane orientation in addition to the misorientation [2]. In short, knowledge of the grain boundary misorientation provides some indication of grain boundary properties such as mobility, energy, diffusivity and general susceptibility to intergranular degradation.

Examples of misorientation distributions in polycrystals are well represented in the literature [19–21] particularly for face-centred cubic (fcc) metals and alloys, and many investigations are linked to grain boundary properties [22–26]. Bicrystals, which have been fabricated with specific misorientations, are also subjects of investigation [27–29]. Frequently, the misorientation is related to the nearest CSL. However, there is evidence that misorientations other than those close to a CSL may be associated with so-called 'special' grain boundary properties. An example is the misorientation 45°/⟨100⟩ [30]. A further example relates to comparison of cracked and uncracked grain boundaries in austenitic stainless steel [31].

Certain misorientations have traditionally been associated with grain boundary mobility maxima. These are 30–40°/⟨111⟩ for fcc metals and ~30°/⟨110⟩ for body-centred cubic (bcc) metals, although all misorientations on ⟨110⟩ are thought to have a mobility advantage [32,33]. The presence of high mobility boundaries has significant implications for microstructural control via grain growth and texture evolution. However, it has recently been realised that adoption of the disorientation convention preselects only one misorientation axis from the possible 24 crystallographically-related solutions, and this solution may not be the most significant with respect to physical properties [**33,**34]. For example, Fig. 3 shows the distribution of misorientations in a bcc steel, where the crystallographically-related solution having misorientation axis nearest to ⟨110⟩ is selected. Table 1 shows an illustration of all 24 solutions for a misorientation between a recrystallised and unrecrystallised grain in a bcc interstitial-free steel, accompanied by the angle of each misorientation axis solution from ⟨110⟩. It is apparent from solution number 24 that this misorientation axis is less than 3° from ⟨110⟩.

5.2. Grain boundary topology

An important extension to the study of grain boundary misorientations in polycrystals is the physical connectivity, or topology, of particular grain boundary types in the microstructure [35]. Each time some grain boundary degradation phenomenon, such as a crack, arrives at a triple junction, the crack must 'decide' along which grain boundary, if either, it can propagate [36,37]. As discussed above, the grain boundary misorientation gives a first-order indication of which grain boundaries are degradation-susceptible, and clearly their distribution in the microstructure affects the 'decision tree'. If a simple binary criterion

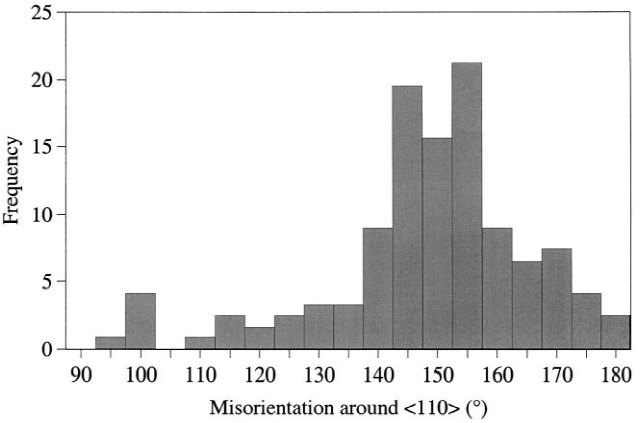

Fig. 3. Frequency distribution of misorientations, displaying the solution having misorientation axis closest to ⟨110⟩, for a bcc steel (adapted from Ref. [**33]).

Table 1
Twenty four crystallographically-related solutions of a misorientation expressed as angle and axis (the deviation of each axis solution from ⟨110⟩ is also listed)

Variant number	Misorientation angle (°)	Misorientation axis (direction cosines)			Deviation from ⟨110⟩ axis (°)
1	25.55	−0.8493	0.0292	0.5271	13.28
2	92.08	−0.0699	−0.9517	0.299	27.83
3	179.26	0.1166	−0.9752	0.188	34.66
4	93.52	−0.2954	0.9529	−0.0691	28.03
5	69.35	0.9787	0.1529	0.1368	36.85
6	158.35	0.9929	0.1187	−0.0065	38.19
7	112.33	−0.9901	−0.0937	0.1047	39.27
8	105.23	−0.1614	0.1729	0.9716	35.97
9	166.61	−0.0065	−0.1891	−0.9819	34.1
10	78.93	−0.2161	−0.2017	−0.9553	34.07
11	117.34	0.3965	0.7528	0.5254	25.33
12	125.84	−0.5913	−0.7149	−0.3731	22.53
13	99.96	0.586	−0.679	−0.4422	26.56
14	141.19	−0.6749	0.5582	0.4827	29.32
15	100.93	0.4308	0.5903	−0.6826	25.83
16	140.41	−0.5527	−0.477	0.6834	29.07
17	126.67	−0.7196	0.379	−0.5819	23.04
18	116.46	0.5355	−0.3907	0.7487	24.76
19	165.27	0.6122	0.7784	−0.1385	10.47
20	170.02	0.0781	−0.8255	−0.5589	11.78
21	174.23	0.6951	0.2155	0.6859	12.45
22	164.21	0.7794	−0.613	0.1294	10.08
23	171.07	−0.0872	0.5585	−0.8249	11.98
24	155.14	0.7015	−0.0515	−0.7108	2.98

which categorises boundaries according to either 'special' misorientations or random high angle misorientations is adopted, from a materials manufacturing point of view it seems important to avoid a connected path of random high angle misorientations, which are susceptible to degradation, percolating through the microstructure. Clustering of related misorientations in the microstructure is often observed [**14]. This is in part a consequence of the following relationship between three misorientations at a triple junction:

$$M_1 M_2 M_3 = I \quad (2)$$

where M_{1-3} are the misorientation matrices of three boundaries at a common junction and I is the identity matrix. Furthermore, the equilibrium relationship between three boundaries comprising a triple junction provides an opportunity to relate misorientation to grain boundary energy [*38,39] or mobility [40,41].

5.3. Conclusions

This short review has described the theoretical basis, experimental considerations, data analysis and applications of studies of grain misorientations in cubic materials. We have demonstrated that distribution of grain boundary misorientations can be measured routinely, as part of a microstructure/microtexture assessment, and provides an insight into grain boundary structure–property linkages.

The principal feature which has implications for misorientation-related investigations in the last half-decade is improvement of experimental techniques, whose sophistication now enables huge sample populations to be obtained and plotted as orientation maps of the microstructure. To handle these vast amounts of data, and to balance sacrificing detail for convenience, poses challenges for the next few years. Finally, misorientation connectivity in the three-dimensional microstructure, and its relationship to those material properties which are of importance in service, is becoming a more meaningful focus of research attention than misorientation statistics alone.

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* of special interest;
** of outstanding interest.

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