What does it mean to be special? The significance and application of the Brandon criterion

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Abstract We review the application of Brandon's criterion to identifying so-called ''special'' grain boundaries. The underlying principles of the Brandon criterion, and others that have followed it, are explained, and the choices of particular parameters within the criteria are considered in the light of experimental information in the literature. It is suggested that varying choices of the parameters may be appropriate for differing applications. An experimental study of the stability of CSL-related triple junctions is used to evaluate the applicability of Brandon-like criteria to these microstructural features.

Introduction

In 1966, David Brandon published a paper in which he set out to estimate the fraction of all grain boundaries that might be described by the Coincident-Site Lattice (CSL) model, assuming that certain deviations from the exact CSL misorientation could be accommodated by arrays of dislocations [1]. Although this paper was arguably not particularly successful in its original goal [2], it has become a classic of the literature on grain boundaries because it introduced a simple formula for the maximum angle of deviation from an exact coincident-site lattice, $\Delta\theta_{\text{max}}$, that could be sustained by a

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dislocation array, and this formula has become widely used as a criterion for determining whether a grain boundary has ''special properties.'' Although the 1966 paper has been extremely well-cited, most of the citations are related to the categorization of individual boundaries based upon their crystallographic parameters, which is an application that was never envisaged in the original work. Our purpose in this paper is to draw attention to some of the underlying concepts of the Brandon criterion, particularly attending to those issues that affect the use of the criterion in separating "special" from "general" grain boundaries.

Special grain boundaries may exhibit properties that deviate from those of ''random'' or ''general'' largeangle boundaries, such as:

- low interfacial energy;
- highly anisotropic interfacial energy;
- low susceptibility to segregation;
- low mobility, or conversely very high mobility;
- low susceptibility to grain boundary corrosion and stress-corrosion cracking;
- low solute diffusivity;
- low propensity for heterogeneous nucleation of second phases;
- low point-defect sink strength
- low electrical resistivity;

... and many others. In recent years the field of ''grain boundary engineering'' has grown up around the idea that it is possible, through careful processing, to increase the fraction of special boundaries above the level present in a random polycrystalline material [3], and thereby improve the properties of the material. Because grain boundary engineering depends critically

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upon identifying those boundaries that have special properties, the Brandon criterion (along with other related criteria described below) has become an important indicator. It is typically expressed as

$$
\Delta\theta_{\text{max}} = 15^{\circ}/\sqrt{\Sigma} \tag{1}
$$

where Σ is the ratio of the primitive CSL unit cell volume to the primitive crystal unit cell volume. Σ -values are usually considered to be significant from 1 (small-angle grain boundaries) up to some limit, which is usually chosen to be 25, following Brandon.

A number of other criteria of specialness have been suggested, and they all follow the same form as the Brandon criterion, which can be expressed as

$$
\Delta\theta_{\text{max}} = \theta_0 \cdot \Sigma^{-n}; 1 \le \Sigma \le \Sigma_{\text{max}} \tag{2}
$$

All published criteria use 15[°] as the value of θ_0 , and various users apply differing values of Σ_{max} , according to their own preference. The main differences among the various criteria lie in the value of n. Brandon used $n = \frac{1}{2}$ [1]; Pumphrey has suggested $n = 2/3$ [4]; Palumbo et al. prescribe $n = 5/6$ [5]; and Ishida and McLean give the most restrictive condition, with $n = 1$ [6]. We proceed here, to examine the theoretical underpinnings of this type of criterion of specialness, and to examine the evidence that supports particular choices for the various parameters, in an attempt to provide for better-informed usage, applicable to particular materials properties, concerns or conditions.

The value of θ_0

The value of θ_0 is almost universally chosen to be 15^o, corresponding to the largest angle for which the dislocation-wall model of a small-angle boundary is thought to apply, based upon the observation that the energy of a grain boundary rises to that of a general high-angle boundary at this misorientation [7]. Most of the energy of such a boundary, of course, is stored in a strain-field, extending into the adjacent grains by a distance roughly equal to the dislocation spacing [8], but many of the interesting and useful properties of grain boundaries may be controlled more by the core structure of the boundary, or they may be more sensitive to the strain field than the energy is. For example, solute diffusion along the axis of a tilt boundary clearly occurs via pipe diffusion along the cores of the boundary dislocations, as beautifully demonstrated by the pioneering work of Turnbull and Hoffman [9]. In

this work, the grain boundary diffusivity is found to increase with the calculated density of grain boundary dislocations, up to 28°, considerably higher than the traditional small-angle limit of 15° , because the behavior in question is related to the core structure of the grain boundary, which appears to be preserved to larger angles than the strain-field that defines the grain boundary energy. In a similar vein, Nakamichi [10, 11] has measured the resistivity of grain boundaries in aluminum for currents perpendicular to the boundary, and finds that small-angle behavior extrapolates quite well up to misorientations of 22° . This is apparently a result of electron scattering from discrete regions of low density in the grain boundary [12], which continue to show steadily increasing density with misorientation, even after the strain fields of the dislocations are effectively cancelled out. Conversely, superconducting currents across grain boundaries in YBa₂Cu₃O_{7- δ} decrease very sharply before a misorientation of 8° , but also appear to continue to fall continuously up to misorientations of about 25° [13]. This would appear to be a result of the sensitivity of magnetic vortex pinning to localized strain, and the small value of θ_0 for large supercurrents would appear to explain the lack of observable impact of special behavior with respect to superconductivity, in large-angle grain boundaries [14]. These examples all illustrate that, irrespective of extension to special large-angle grain boundaries, the appropriate value of θ_0 depends upon the property of interest and should be chosen accordingly, rather than relying on the canonical value of 15°.

Extension of small-angle behavior to CSL-related boundaries

The basis of the Brandon criterion rests in the notion that a boundary that deviates in its misorientation by a small angle, $\Delta\theta$, from a CSL-misorientation, will have a structure consisting of an array of secondary dislocations, with Burgers vectors defined by the DSC lattice, superimposed upon the structure of the perfect CSL boundary, just as a small-angle boundary is made up of an array of primary dislocations superimposed on the perfect crystal (*i.e.* $\Sigma = 1$) structure.

For the purpose of a simple model, we can calculate the spacing of the dislocations in the boundary as

$$
S = \frac{b/2}{\sin(\Delta \theta/2)}
$$
 (3)

where b is the Burgers vector of the dislocations in the array. For small angles of deviation, this simplifies to

$$
S \approx \frac{b}{\Delta \theta} \tag{4}
$$

For a small-angle grain boundary in an FCC material, this can be written as

$$
S \approx \frac{a_0/\sqrt{2}}{\theta} \tag{5}
$$

where a_0 is the lattice parameter. The minimum acceptable dislocation spacing is then taken to be

$$
S_{\min} \approx \frac{a_0}{\theta_0 \sqrt{2}}\tag{6}
$$

where θ_0 is the largest angle for which "small-angle" behavior" is expected, typically 15°, as described above. For larger angles, we usually rationalize that the dislocations are too closely spaced to behave as distinct line defects, and they ''lose their physically distinct identity.''

It is often assumed that all of the properties of coincidence-related large-angle boundaries can be deduced by analogy with the corresponding properties of small-angle grain boundaries. Although the structures are in many respects similar, there can be some distinct differences, too. The strain energy of such a boundary is often assumed to be superimposed upon the ''core'' energy of the CSL-related boundary, as supported by many observations of the strain-fields of secondary dislocations [15, 16] so the energy variation can perhaps be considered to be cusped at the CSL misorientation. Effects that depend upon the cores of dislocations, such as diffusion or electron scattering, however, may not follow the behavior of small-angle grain boundaries, because the cores of DSC dislocations can be denser than the surrounding material, while the cores of primary dislocations are always less dense [17].

The appropriate values for Σ

While it is commonly assumed that special properties are associated with low values of Σ , the converse is not necessarily true: not all low values of Σ produce special properties. For example, low rates of diffusion-induced grain boundary migration (DIGM) are observed for symmetric tilt boundaries in copper, corresponding to $\Sigma = 5$ and $\Sigma = 17$, but not for $\Sigma = 13$ [18]. The existence of ''preferred'' and ''non-preferred'' boundaries has been discussed in terms of a structural unit model, by Sutton and Vitek [19]. While all of the boundaries that

fall into the ''preferred'' category are symmetric CSLrelated boundaries, not all of these boundaries are preferred.

Another complication arises from the propensity of low stacking-fault energy fcc metals to form twins, and twin intersections resulting in increased incidences of boundaries with $\Sigma = 3^N$. It is not clear whether a $\Sigma = 81$ boundary has any special properties beyond its increased incidence in certain fcc metals, but it is probably true that a $\Sigma = 27$ boundary shows at least some moderate level of specialness.

It is clear that any arbitrary cut-off, allowing consideration of all values of Σ up to some limit, does not correctly identify all special boundaries: it allows some non-special ones to be included, and excludes some that might have an effect on the properties of a material. The inclusion of non-special boundaries is not a significant problem, unless these are specifically increased in the boundary population through processing, but there is not yet any method that can address a single type of CSL, such as $\Sigma = 13$.

The value of *n* and the assumptions underlying the form of the equation

Brandon noted that the Burgers vector strength of primitive dsc dislocations associated with CSL boundprimitive asc alsociations associated with CSL boundaries varies as $a_0/\sqrt{2\Sigma}$, as illustrated for [100] rotations in Fig. 1. This actually applies only to some of the

Fig. 1 Illustrating the essential relationships of the CSL size and the dsc-lattice size to the value of Σ , for CSLs formed by rotations about [100] in fcc crystals. The height of the CSL perpendicular to the page is the same for all of these cases. The lateral dimension of the CS L varies as $\Sigma^{1/2}$, while the dsc-lattice size (fine mesh) varies as $\Sigma^{-1/2}$

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Burgers vectors, as we shall discuss below. It is specifically true for Burgers vectors that are perpendicular to the rotation axis associated with the CSL disorientation, which we shall call the primary rotation axis. These are sometimes referred to as b_1 or b_2 Burgers vectors: b_3 Burgers vectors have a component parallel to the primary rotation axis.

Using this variation of the Burgers vector with Σ , we can write the dislocation for CSL-related boundaries as

$$
S = \frac{a_0}{\Delta \theta \sqrt{2\Sigma}}\tag{7}
$$

and we have

$$
S_{\min} = \frac{a_0}{\Delta \theta_{\max} \sqrt{2\Sigma}}\tag{8}
$$

or

$$
\Delta\theta_{\text{max}} = \frac{a_0}{S_{\text{min}}\sqrt{2\Sigma}}\tag{9}
$$

Substitution of the fixed value of S_{min} from Eq. 6 recovers the Brandon criterion, showing that the one of its basic assumptions is that all dislocations lose their physically distinct identity at a fixed spacing, irrespective of their Burgers vector, or indeed the lattice parameter of the material.

We can now investigate the effects of different assumptions about the critical separation at which grain boundary dislocations lose their identity. A simple model can be derived if it is assumed that the critical dislocation spacing is proportional to the Burgers vector, rather than a constant length. For this case, smaller-strength dsc dislocations are allowed to be closer together than for the Brandon case, before they lose their identity. This model, however, produces a fixed value of $\Delta\theta_{\text{max}} = \theta_0$, which obviously does not accord with the collected observations from the grain boundary engineering community. A different approach derives from taking note of the fact that the boundary periodicity is determined by the size of the CSL, and that this would be the expected Peierls energy periodicity for the dsc dislocations: when the dislocations are closer together than this, they essentially occupy the same Peierls valley, and should be considered to have overlapped. This case has a critical separation rather larger than Brandon's. We can then derive a criterion in which the critical separation is derive a criterion in which the critical separation is
given by the CSL size, or $S_{\text{min}} = a_0 \sqrt{\Sigma}$. Substituting this expression into Eq. 9, we obtain an expression of the same form as the Ishida–McLean criterion, with $n = 1$ [6]. Other criteria, such as Pumphrey's [4] or that of Palumbo et al. [5] do not correspond to any clearly identifiable assumption about the critical separation of grain boundary dislocations, but rather reflect a consideration of possible statistical variations in the populations of different Burgers vectors of the interfacial dislocations.

The Burgers vectors of grain boundary dislocations

All of the discussion of the form of the equation, above, relates specifically to grain boundary dislocations of the b_1 and b_2 types, which are perpendicular to the primary rotation axis, and can accommodate deviations (secondary rotations, away from the exact CSL) that are also about the same axis. For cases other than [100] and [111] rotations, the b_1 and b_2 moduli are different, as shown in Fig. 2, but the overall variation is still similar, and the form of the criterion is still only affected by one's assumptions about the critical dislocation separation.

If the secondary rotation is about an axis other than the primary rotation axis, then its accommodation requires the inclusion of b_3 dislocations. The strengths of these Burgers vectors do not vary in the same way as the b_1 and b_2 types, as shown in Fig. 2. They are rather asymptotic to the plane spacing perpendicular to the primary rotation axis, as Σ increases. As pointed out by Balluffi and Schober [20], for large values of Σ , these dislocations also correspond to the ''plane matching'' dislocations first described by Pumphrey [21]. Because these dislocations retain large Burgers vectors, their separation is larger than for the b_1 and b_2 types for similar deviation angles. Thus, irrespective of our assumption about the critical dislocation spacing, we

Fig. 2 The variation of dsc Burgers vector magnitudes with Σ -value, for various rotation axes in fcc crystals. The b_1 and b_2 magnitudes vary as $\Sigma^{-1/2}$. The b_3 magnitudes asymptotically approach the plane spacing along the rotation axis

should expect that larger deviation angles can be accommodated when the secondary rotation axis is not the same as the primary rotation axis, and calls for b_3 dislocations in the boundary. If we follow the Ishida–McLean assumption, that the minimum dislocation spacing is given by the CSL dimension, then these dislocations can also be spaced relatively closely together: the relevant CSL dimension is the one parallel to the primary rotation axis, which is a constant, equal to the lattice period, for all values of Σ . The greatest allowable deviation from the exact CSL will thus always be for the case where the secondary rotation axis is perpendicular to the primary axis. This deviation will be relatively large, and not a strong function of Σ , irrespective of the assumption about the closest approach distance for the grain boundary dislocations.

Another concern about the dislocations that accommodate deviations from an exact CSL misorientation is that they need not be primitive dsc dislocations. Lattice dislocations are observed on some boundary planes, while dsc dislocations are preferred on others, in the observations of Bollmann et al for example [22]. Ichinose and Ishida have observed a dislocation in a Σ 11 boundary in gold, with two times the primitive dsc Burgers vector [23], and this appears to relate to the energy associated with a relatively large step component of the primitive defect [24]. In each of these examples the dislocations have larger-than-dsc Burgers vectors, and would thus have larger spacings for a given value of $\Delta\theta$. This would extend the range of special behavior beyond the value predicted by any of the criteria currently in use. In other cases, dsc dislocations may dissociate into smaller Burgers vectors [17, 25] and this would correspondingly reduce the acceptable range of misorientation to be associated with a CSL.

Are all special boundaries CSL-related?

While it is established that many, but not all CSLrelated grain boundaries have special properties, it is not clear whether there are special boundaries that are not related to CSL's. Plane-matching, or coincident axial direction (CAD) boundaries may be a case in point, in which periodicity is preserved in only one direction. Although it has been pointed out that these boundaries can be described by large Σ -value CSL's [20] it is not clear whether this is especially useful in the context of grain boundary engineering, where these boundaries are not routinely identified. The migration of plane-matching boundaries, however, has been found to produce distinctive microstructures [26].

A question of growing importance concerns the possible existence of grain boundaries that have special properties, but are not associated in any way with a CSL, even to the extent of plane-matching in a single direction. There is some evidence that boundaries that are parallel to a low-index plane in at least one of the adjacent crystals may occur with greater-than-random probability [27], showing that the grain boundary plane orientation may have greater significance than previously recognized. Another type of special boundary may be associated with misorientations that represent symmetry planes in misorientation-space. These include cases like $45^{\circ}/[100]$; $90^{\circ}/[110]$ and $30^{\circ}/[111]$. None of these produce CSLs, but Neumann's principle [28] requires that they produce extrema of energy with respect to misorientation.

Experimental

A singular data-point on the utility of criteria of interfacial specialness has been obtained during studies of triple-junction behavior and properties. For the purpose of these studies, we have designed a set of triple junctions with fully-specified misorientations and symmetric tilt grain boundary planes that ought to produce energetically-stable configurations [29]. These tricrystal designs take full account of the effect of boundary inclination upon the boundary energy. One of the proposed configurations is a Σ 5: Σ 5: Σ 25 tricrystal, as illustrated in Fig. 3. We have grown two copper tricrystals of this nominal design by a seeded, vertical Bridgman growth technique, with the triple junction

Fig. 3 Schematic crystallographic details of the tricrystal design used for the experiments

parallel to the growth direction of the specimen. This gives us a long length of tricrystal which can be sliced at different locations, allowing for reproducible experiments to be performed. The details of the tricrystal growth process are described in a forthcoming paper.

Results

The misorientations of the tricrystals have been checked by Laue X-ray diffraction, and also by electron backscatter diffraction techniques. The precision of individual orientation measurements was estimated to be better than 1° , and repeated measurements of misorientations with the EBSP system confirmed this by reproducing within 0.8°. Errors in the actual misorientations result from lack of precision in setting the seed crystals, and the two tricrystals were found to embody grain boundaries with the misorientations shown in Table 1. In Table 1, the rotation axes are rounded to the nearest integer, although the precise measured values are used in calculating the reported angular deviations from the ideal CSL misorientations using conventional matrix methodology. Tricrystal 2 is clearly closer to the intended misorientations than Tricrystal 1.

The intended dihedral angles were checked by slicing the tricrystals perpendicular to their length (i.e. perpendicular to the nominal triple junction direction) and performing routine metallography. Typical results for Tricrystal 1 are shown in Fig. 4, and for Tricrystal 2 in Fig. 5. The measured dihedral angles for various slices of the two tricrystals are given in Table 2. Tricrystal 1 exhibits quite widely varying dihedral angles, with a range of as much as 26° of variation, even though the misorientations of the grain boundaries do not change.

Table 1 Designed "target" and "actual" misorientations obtained for two copper tricrystals

Grain boundary	Target misorientation	Measured misorientation	Deviation	
Tricrystal 1				
Σ 5	36.87°/[001]	$32.0^{\circ}/[1\ 1\ 25]$	5.2°	
Σ 5	36.87 $^{\circ}/[001]$	$41.7^{\circ}/[1\ 1\ 17]$	5.6°	
Σ 25	16.26°/[001]	$16.5^{\circ}/[1\ 1\ 13]$	1.5°	
Tricrystal 2				
Σ 5	36.87°/[001]	$35.2^{\circ}/[0 1 26]$	2.0°	
Σ 5	36.87°/[001]	$37.9^{\circ}/[001]$	1.2°	
Σ 25	16.26°/[001]	$17.0^{\circ}/[0 1 10]$	1.7°	

Fig. 4 Slices 2 and 3 from the first tricrystal, illustrating the variation of the dihedral angles formed by the grain boundaries. The approximate traces of the intended symmetrical tilt grain boundary planes are indicated

Discussion

It is obvious that Tricrystal 1 is rather less successful than Tricrystal 2, in meeting the design specifications. The reasons for the striking variability of dihedral angle at a fixed set of misorientations will be addressed in a separate paper. For the tricrystal that is closer to the target misorientations, the dihedral angles appear to "lock in" within a degree of the intended values indeed, closer than the experimental error of measurement––but the less precise adherence to the desired misorientations for Tricrystal 1 seems to result in much poorer control of the dihedral angles. This provides an opportunity to assess the effectiveness of the various criteria of specialness, described above, in predicting the behavior of these grain boundary junctions. Table 3 compares the measured $\Delta\theta$ values for the grain boundaries with the allowed deviations,

Fig. 5 Slices 1 and 2 from the second tricrystal, illustrating the stability of the dihedral angles formed by the grain boundaries. The approximate traces of the intended symmetrical tilt grain boundary planes are indicated

according to the various criteria. For this purpose, we have chosen to assume that θ_0 is 15°, consistent with

the role of grain boundary energy in determining the dihedral angles.

The Brandon criterion does not draw any distinction between our tricrystal specimens: all of the boundaries in both specimens satisfy this criterion. All three boundaries in our second specimen meet the Pumphrey criterion, while two out of the three in the first tricrystal fail this test. Increasing the magnitude of the exponent to -3/4 gives us a previously unutilized criterion which all three boundaries of Tricrystal 1 fail, but two of the three from Tricrystal 2 pass. Larger negative values of n make only the same level of distinction between the two tricrystals.

It is not yet clear whether a triple junction may have "special" properties in its own right, and it is also unclear whether it is necessary for all three boundaries to be special in their own right in order to allow this. At this point we may only conclude that the Brandon criterion does not provide useful information about the behaviors of triple junctions, though an exponent somewhere between –2/3 and –1 seems to give a criterion that may be predictive of a consistently-behaving junction.

Table 3 Comparison of the deviations of misorientation in our two tricrystals with the limits allowed by various criteria of specialness. A check-mark indicates that the experimental boundary satisfies the given criterion

			Brandon	Pumphrey			Palumbo et al. $n = -5/6$	Ishida-McLean
			$n = -1/2$	$n = -2/3$	$n = -3/4$	$n = -4/5$		$n=-1$
Allowed by Criterion	Σ 5 Σ 25	$\Delta\theta_{\rm max}$ $\Delta\theta_{\rm max}$	6.71° 3°	5.13° 1.75°	4.49° 1.34°	4.14° 1.14°	3.92° 1.03°	3.00° 0.60°
Tric. 1 "Bad"	Σ 5 Σ 5 Σ 25	$\Delta\theta = 5.6^{\circ}$ $\Delta\theta = 5.2^{\circ}$ $\Delta\theta = 1.5^{\circ}$	\Box \Box \Box	\Box				
Tric. 2 "Good"	Σ 5 Σ 5 Σ 25	$\Delta\theta = 1.2^{\circ}$ $\Delta\theta = 2.0^{\circ}$ $\Delta\theta = 1.7^{\circ}$	\Box \Box \Box	\Box П \Box	П П			\Box \Box

Conclusions

Careful consideration of the underlying principles of the Brandon criterion, and the various related criteria of specialness for grain boundaries, can provide useful insight into its proper use and application in the originally-unintended application of categorizing individual grain boundaries. The proper selection of parameters to be used in such criteria depends upon the specific property or behavior under consideration, and should minimally reflect an informed choice of θ_0 , based upon suitable information in the literature.

The Brandon criterion appears to be too permissive to predict the behavior of a triple junction, but it remains unclear whether any other criterion is more reliable, or indeed how such criteria should be applied to the three boundaries that meet at a junction, with respect to the number of boundaries that must meet the criterion in order for the junction to have special properties in its own right.

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