Grain orientation statistics of grain-clusters and the propensity of multiple-twinning during grain
boundary engineering

Tingguang Liu ^{a,b}, Shuang Xia ^{a,c,*}, Baoshun Wang ^d, Qin Bai ^a, Bangxin Zhou ^a, Cheng Su ^d

^a School of Materials Science and Engineering, Shanghai University, Shanghai 200072, China

^b National Center for Materials Service Safety, University of Science and Technology Beijing,
Beijing 100083, China

^c State Key Laboratory of Advanced Special Steel, Shanghai University, Shanghai 200072, China

^d Technical Department, Jiuli Hi-Tech Metals Co., Ltd., Huzhou 313008, China

* Corresponding author: S. Xia, E-mail: xiashuang14@sohu.com, Tel: +86-18019322802, Fax:
+86-021-56337934. Postal address: P.O. Box 269, 149 Yanchang Road, Shanghai, zip code
200072, P. R. China.

E-mail addresses: tgliu@outlook.com (T. Liu), wangbaoshun@jiuli.com (B. Wang),
baiqin31@shu.edu.cn (Q. Bai), zhoubx@shu.edu.cn (B. Zhou), sc@jiuli.com (C. Su)

Abstract

Large grain-cluster or so-called twin-related domain is a typical characteristic of the grain boundary (GB) engineered microstructure. Grain-cluster is formed via numerous twinning operations starting from single nucleus, and the process is referred to as multiple-twinning. This work investigated the orientation diversity within grain-clusters and the twinning ordering of multiple-twinning based on the statistics of grain-orientations in 30 large-sized grain-clusters from GB-engineered Ni-based alloy 690. The statistics show that the grain-cluster apparently has several dominant orientations. A few dominant orientations occupy most area and most grains in a

grain-cluster. Moreover most misorientations between these dominant orientations are of low-order $\Sigma 3^n$ -type ($n=1, 2$), and the 4 sub-dominant orientations are twinning variants of the first-dominant orientation in most cases. These statistical characteristics of grain-clusters reflect the general behavior of multiple-twinning: back-and-forth pattern and preferential orientations. The twinning operations produce not only higher (forward) but also lower (backward) generation orientations, and the backward probability is higher than the forward. The multiple-twinning shows a propensity to form or access to a few preferential orientations, and results in the formation of dominant orientations of the formed grain-cluster.

Keywords : EBSD; Grain boundary engineering; Multiple-twinning; Grain-cluster; Grain orientation.

1. Introduction

Twinning is an interesting phenomenon in some materials and can occur during annealing [1-3] or deformation [4], and annealing twin-boundaries or deformation twin-boundaries are formed, respectively. As a deformation mechanism, twinning has important influence on the mechanical properties of materials [5]. Additionally, the annealing twin-boundaries have special properties, such as stronger resistance to intergranular degradation, compared with random-boundaries [6-8]. Therefore, the twinning can be applied for materials design, and the concept of “grain boundary (GB) engineering” [9-15] was proposed to improve the GB-related properties via “grain boundary control or design” [9].

GB-engineering has been successfully applied to many structural materials, such as austenitic stainless steel [7], Ni-based alloys [11, 12], copper and its alloys [13]. After the treatment of GB-engineering, resistance to carbide precipitation [6], impurity segregation, intergranular

corrosion [7] and intergranular stress corrosion cracking can be improved resulting from the formation of a great number of special boundaries. The low- Σ CSL boundaries (Σ -value ≤ 29) are generally considered as the special boundaries [16], where the Σ -value denotes the reciprocal density of coinciding sites of two neighboring grains based on the CSL model (coincide site lattice). Twin-boundaries ($\Sigma 3$) and its variants ($\Sigma 3^n$, $n=2, 3$) take most fraction of the total special boundaries [17-20]. The proportion of twin-boundaries could be increased to more than 60% from about 40% by the treatment of GB-engineering [11, 12, 17-21], because the face-centered-cubic low stacking-fault-energy metals, such as Ni-based alloys, austenitic stainless steel and brass, are prone to twinning [1, 2] in the wake of migrating of the recrystallization front boundaries during annealing. The iterative process of twinning operations starting from a single nucleus is referred to as multiple-twinning [21-25], and the generated annealing twins can be reconstructed as a twinning tree, which is referred to as twin-chain [8, 11, 22, 26, 27]. The assembly of all grains in the tree is named grain-cluster or twin-related-domain [8, 11, 12, 24, 26]. All inner grains of the grain-cluster have $\Sigma 3^n$ ($n=1, 2, 3, \dots$) mutual misorientations, and all inner boundaries are types of $\Sigma 3^n$ but outer boundaries are crystallographically random. Large-sized grain-clusters and high proportion of twin-related boundaries are two prominent characteristics of the GB-engineered microstructure [6, 10-14, 16-18, 28-30]. Both of them are the consequences of multiple-twinning [21-25]. So, it is significant to investigate the behavior of multiple-twinning.

A grain may have a great number of twins but four twinning-orientations (variants) at most [25]. It was expected that the probabilities to form any one of the four variants are equal, but no paper confirmed this expectation or existence of preferential variant. The physical process of multiple-twinning is not entirely clear, because it is difficult to observe the process of

multiple-twinning in-situ, not mention mapping the grain-orientations simultaneously. Generally we study the multiple-twinning by statistical analysis of grain-clusters. Mason et al. [31] found that the distribution of the number of grains per grain-clusters can be fit to Weibull distribution. Cayron [24] got similar results about the grain number distribution. Meanwhile the lengths of the longest twin-chain in the grain-clusters were calculated in Ref. [24]. The number of grains in a grain-cluster corresponds to the number of twinning operations of the multiple-twinning to form the grain-cluster. Therefore, the potential number of twinning-operations per multiple-twinning is fit to the Weibull distribution. During recrystallization, a few nuclei perform multiple-twinning extensively but most nuclei just perform a few iterations of twinning.

Apart from the number of grains, the number of orientations is another significant parameter for grain-cluster statistics. Some grains may have the same orientation in a grain-cluster, and thus the number of orientations is less than the number of grains. In addition, the number of grains per orientation in a grain-cluster should be considered as well. The three parameters, i.e. the number of grains, the number of orientations and the numbers of grains per orientation in a grain-cluster, would synthetically reflect the physical process of multiple-twinning. In the present work, statistics of the three parameters of many large-sized grain-clusters from GB-engineered Ni-based alloy 690 were obtained, and several statistical characteristics of grain-clusters were found, and the physical process of multiple-twinning was explored subsequently. The results are significant to understand the mechanism of GB-engineering that form a high proportion of $\Sigma 3^n$ boundaries in materials.

2. Experiment

The experimental material used in this work is a Ni-based Alloy 690, with the measured

chemical composition (in wt%): 30.39 Cr, 8.88 Fe, 0.023 C, 0.002 S, 0.006 P, 0.07 Si, 0.23 Mn, 0.22 Al, 0.26 Ti, 0.02 Cu, and the balance Ni. The as-received metal was cold-rolled and annealed to get a uniform microstructure firstly, and then it was treated for GB-engineering. Cold working with about 5% deformation combined with annealing at 1100 °C for 1 h or at 1150 °C for 5 min are good procedures for GB-engineering to produce high proportion of twin-boundaries (>70%), as described previously [17, 18]. The specimens described in Ref. [17, 18] were used in the statistics of this work.

These specimens were mechanical-polished and finally electro-polished in an electrolyte containing 20% HClO₄ and 80% CH₃COOH (volume fraction) at room temperature with about 30 V direct current for 120 s for EBSD mapping. The EBSD (Channel 5 of HKL) system was integrated with a CamScan Apollo 300 field emission scanning electron microscope (SEM). Many areas were mapped by using the EBSD system. The EBSD field of view was 800 μm × 500 μm or 1000 μm × 800 μm with a step size of 2 μm. Channel 5, the EBSD data analysis software, was used to process the EBSD data. The CSL boundaries were determined based on the Palumbo-Aust criterion ($\Delta\theta_{\max}=15^\circ\sum^{-5/6}$). The assembly of grains, between which are of $\sum 3^n$ ($n=1, 2, 3$) misorientations, was determined as a grain-cluster. TexTools was used to calculate the misorientations of grain-pairs.

3. Results

Large grain-clusters were observed in these EBSD maps, as shown in Fig. 1. The IPF-color highlighted region is a grain-cluster, in which all inner grain boundaries are $\sum 3^n$ ($n=1, 2, 3$) types. Grain-clusters in the map can be automatically determined by using Channel 5. This highlighted grain-cluster has 112 grains and 28 grain-orientations. Some grains have the same orientation in a

grain-cluster. The 5 largest-area orientations, i.e. the area fractions of grains with each of the 5 orientations in the grain-cluster are largest, were designated as $a\sim e$ in descending order. Some grains were marked using their orientation-labels in Fig. 1. Distributions of the area fraction and the number of grains per orientation in the grain-cluster are shown in Fig. 2. Neither of them is evenly distributed. Most orientations occupy small area and a few grains, whereas several orientations ($a\sim e$) take up most area and most number of grains in the grain-cluster.

The other 29 grain-clusters were investigated similarly, including cluster area, number of orientations, number of grains and the 5 largest-area orientations. These data were listed in Table 1 and Table 2. Total 30 grain-clusters (C1-C30) were included in this statistic. Typically, the highlighted grain-cluster in Fig. 1 is C17. It should be noted that some grain-clusters intersecting with the edges of scans were inevitably used in the statistics due to the large size of grain-clusters with respect to the scanned area. In addition, the 5 largest-area orientations in each of these grain-clusters were designated as $a\sim e$ in descending order by area. The area-fraction, the number of grains and the mutual misorientations of the 5 orientations $a\sim e$ were calculated, as shown in Table 1 and Table 2.

Distributions of the cluster area, the number of orientations and the number of grains for the 30 grain-clusters were shown in Fig. 3. Each grain-cluster has many orientations and more grains. For example, the grain-cluster C27 has 98 orientations and 232 grains. Generally, the number of grains is directly proportional to the number of orientations. The average number of orientations and the average number of grains per grain-cluster are 32.8 and 90.0 respectively for the 30 grain-clusters. Each orientation has 2.7 grains on average.

4. Discussion

4.1. Statistical analysis of grain-clusters

Grain-cluster is formed by multiple-twinning, i.e. the iterative process of twinning operations starting from a single nucleus during recrystallization as discussed in Ref. [10-12, 29, 30], so all the inner grains of a cluster are twins or twins of twins and so on of the nucleus (parent grain), and they could be reconstructed as a tree-shape twin-chain [11, 24]. On the other hand, the process of multiple-twinning can be demonstrated as a twin-chain of orientations [11, 24-26], as shown in Fig. 4a. It was reconstructed on the basis of the rule that one orientation has four twinning-orientations at most [11, 25]. In the k th generation there are $N_k=4\times 3^{k-1}$ orientations at most [23, 25].

A grain-cluster may have a great number of grains, and a number of orientations but less than the number of grains. The statistics of grains and orientations in grain-clusters would help to explore the general behavior of multiple-twinning. Mason et al. [31] carried out statistics of the number of grains in grain-clusters, and Cayron [24] calculated the numbers of grains and the lengths of the longest chain of grain-clusters to investigate the multiple-twinning quantitatively. However, only the number of grains in a grain-cluster is not enough to reflect the physical process of multiple-twinning. The number of orientations, the number of grains per orientation, and the area fraction of each orientation in a grain-cluster also should be taken into account.

The present statistics of the 30 grain-clusters show that neither the area fraction nor the number of grains per orientation in a grain-cluster has even distribution. The grain-cluster C17, as shown in Fig. 2, is a typical example. Fig. 5 shows the distributions of the average area fraction and the average number (and number fraction) of grains per orientation for the 30 grain-clusters. The 5 largest-area orientations $a\sim e$ were calculated respectively, but other orientations (27.8 on

average) were sorted into one category, and it was divided by the orientation number 27.8 to get the average value $O_{27.8}$. Fig. 5a shows that the orientations in a grain-cluster have quite different area fractions. The 5 largest-area orientations take up 69.9% of area of a grain-cluster on average. Especially the largest-area orientation a covers 28.3% of area of the grain-cluster on average, but the other 27.8 orientations has a much smaller area fraction. Therefore, the grain-cluster has dominant orientations which occupy apparently higher fractions of the area of the grain-cluster. In addition, the 5 largest-area orientations take up 36.7% of the total number of grains in a grain-cluster, which are higher than any of the other 27.8 orientations. Hence the dominant orientations on area of a grain-cluster also dominate the number of grains of the grain-cluster.

Statistics of the mutual misorientations between the 5 dominant orientations (the 5 largest-area orientations) in the 30 grain-clusters was shown in Table 3. Most of them have low-order $\Sigma 3^n$ -relationships ($n=1, 2$). As an example, in the 30 grain-clusters, there are 23 grain-clusters in which the misorientation between orientation a and b ((a,b) -relationships) is $\Sigma 3$. Fig. 6 shows the distributions of $\Sigma 3^n$ ($n=1, 2, 3$) relationships between the 5 dominant orientations $a \sim e$. The $(a,b,c,d,e) \times (a,b,c,d,e)$ grid shows orientation-pairs. The vertical-axis shows the frequency of an orientation-pair having the given $\Sigma 3^n$ -relationship in the 30 grain-clusters. As an example, in Fig. 6b, the orientation-pair (b,c) points at 0.6, which means that 60% of the 30 grain-clusters have $\Sigma 9$ -misorientation between b and c . Fig. 6d illustrate the orientation-pairs that have the highest frequency in three boxes of Fig. 6(a, b, c), respectively. The orientation-pairs (a,b) , (b,c) and (d,e) have the highest frequency of $\Sigma 3$, $\Sigma 9$ and $\Sigma 27$, respectively. It can be derived that orientation-pairs that have higher area-fractions in a grain-cluster are prone to having lower order of $\Sigma 3^n$ -misorientation, and vice versa.

Based on the formation mechanism of grain-cluster, i.e. the multiple-twinning, the relationship of orientations in a grain-cluster can be reconstructed as a tree-shape twin-chain [11, 25, 26], as shown in Fig. 4a. Any two orientations in the twin-chain has $\sum 3^n$ -misorientation, where the exponent n indicates the number of steps between the two orientations. Therefore, the statistical result that most of the mutual misorientations between the 5 dominant orientations $a \sim e$ of a grain-cluster are low-order $\sum 3^n$ reflect that the 5 dominant orientations are not randomly located in the twin-chain, but they should locate close positions. The first-dominant orientation a in 54% of the 30 grain-clusters has $\sum 3$ -misorientation with the 4 sub-dominant orientations $b \sim e$, and the mutual misorientations between $b \sim e$ in 41% of the 30 grain-clusters are $\sum 9$. Therefore, it can be concluded that, in most cases, the 4 sub-dominant orientations $b \sim e$ are the first generation twinning-orientations of the first-dominant orientation a .

4.2. General behavior of multiple-twinning

Grain-cluster is formed by multiple-twinning [10-12, 29, 30], so the statistical characteristics of grain-clusters described in the above section can reflect the general behavior of multiple-twinning. Firstly, a twinning-operation may generate a new orientation (higher generation) or an orientation that already exists (lower generation) [23, 25]. For example, in Fig. 4a, a first-generation twinning-orientation '1' may generate a second-generation twinning-orientation (forward) or the parent-orientation (backward) when twinning-operation happens. This raises a question whether multiple-twinning goes straight from the parent-orientation to higher-generations or performs back-and-forth pattern. If multiple-twinning performs straight forward pattern, the formed grain-cluster should have similar numbers of orientations and grains. In fact, the number of grains is much larger than the number of orientations in a grain-cluster, as shown in Table. 2.

Therefore, multiple-twinning most likely perform back-and-forth pattern, which is indicated by the bidirectional arrow in Fig. 4b.

Secondly, grain-cluster has dominant orientations which take up most area and most number of grains of the grain cluster. (Note: the dominant orientations belong to the grain-cluster but not the specimen. The grain-clusters in a specimen have different dominant orientations.) This reflect that the multiple-twinning process should have preferential orientations. The preferential orientations are prone to forming during the multiple-twinning, and the twins with the preferential orientations are easy to grow, which result in that the area fractions of the dominant orientations in a grain-cluster are higher than their grain number fractions, as shown in Fig. 5. A grain has four possible twinning-orientations, but the probabilities to form the four orientations are unequal. The preferential orientations or orientations access to the preferential orientations are prone to formation during multiple-twinning.

Thirdly, the grain-cluster statistics show that the 5 dominant orientations of a grain-cluster are located in close positions in the twin-chain of orientation (Fig. 4a). The 4 sub-dominant orientations $b\sim e$ are the first generation twinning-orientations of the first-dominant orientation a in most cases. Therefore, we could assume that the first-dominant orientation is the parent-orientation to form the grain-cluster, and the 4 sub-dominant orientations are its first-generation twinning-orientations. In addition, it had been known that the multiple-twinning has propensity to form or access to the preferential orientations. It can be concluded that a twinning operation during the multiple-twinning has higher probability to generate lower-generation twinning-orientations (backward) than the probability to generate higher-generation twinning-orientations (forward), which was indicated by the rainbow color in

Fig. 4b. Red color means high probability to form, blue color means low probability.

5. Conclusions

Large grain-cluster formed by multiple-twinning is a prominent characteristic of the microstructure after GB-engineering. The grain-orientation statistics of 30 large-sized grain-clusters from GB-engineered alloy 690 specimens shown that grain-cluster has apparently dominant orientations. A grain-cluster may have a great number of grains and a number of orientations, but a few dominant orientations take up most area and grains of the grain-cluster. In addition, most mutual misorientations between the 5 dominant orientations are low-order of $\Sigma 3^n$ -type ($n=1, 2$), furthermore, in most cases, the 4 sub-dominant orientations are the twinning-variants of the first-dominant orientation. These statistical characteristics of grain-clusters reflect the general behavior of multiple-twinning. Firstly, the multiple-twinning process has a back-and-forth pattern. Secondly, the twinning operations during multiple-twinning are not random to form the four twinning-variants, but preferential orientations exist. The multiple-twinning has propensity to form or access to the preferential orientations, which result in the dominant orientations of the formed grain-cluster finally. Furthermore, if we assume the first-dominant orientation of a grain-cluster is the parent-orientation to form the grain-cluster, it can be deduced that the multiple-twinning has backward propensity rather than forward.

Acknowledgments

This work was supported by the Shanghai Science and Technology Commission (grant number 13520500500) and the fund from Jiuli Hi-Tech Metals Co., Ltd.. The authors gratefully thank Wei Guo who gave much help to improve the language.

References

- [1] H. Gleiter, The formation of annealing twins, *Acta Metallurgica*, 17 (1969) 1421-1428.
- [2] S. Mahajan, C.S. Pande, M.A. Imam, B.B. Rath, Formation of annealing twins in f.c.c. crystals, *Acta Materialia*, 45 (1997) 2633-2638.
- [3] P. Galizia, C. Baldisserri, C. Capiani, C. Galassi, Multiple parallel twinning overgrowth in nanostructured dense cobalt ferrite, *Materials & Design*, 109 (2016) 19-26.
- [4] G. Liu, R. Xin, F. Liu, Q. Liu, Twinning characteristic in tension of magnesium alloys and its effect on mechanical properties, *Materials & Design*, 107 (2016) 503-510.
- [5] B. Song, N. Guo, T. Liu, Q. Yang, Improvement of formability and mechanical properties of magnesium alloys via pre-twinning: A review, *Materials & Design* (1980-2015), 62 (2014) 352-360.
- [6] S.S. Katnagallu, S. Mandal, A.C. Nagaraja, B. De Boer, S.S. Vadlamani, Role of Carbide Precipitates and Process Parameters on Achieving Grain Boundary Engineered Microstructure in a Ni-Based Superalloy, *Metall Mater Trans A*, 46a (2015) 4740-4754.
- [7] S. Kobayashi, R. Kobayashi, T. Watanabe, Control of grain boundary connectivity based on fractal analysis for improvement of intergranular corrosion resistance in SUS316L austenitic stainless steel, *Acta Materialia*, 102 (2016) 397-405.
- [8] S.A. Xia, B.X. Zhou, W.J. Chen, Effect of single-step strain and annealing on grain boundary character distribution and intergranular corrosion in Alloy 690, *J Mater Sci*, 43 (2008) 2990-3000.
- [9] T. Watanabe, Approach to grain boundary design for strong and ductile polycrystals, *Res Mechanica: International Journal of Structural Mechanics and Materials Science*, 11 (1984) 47-84.
- [10] V. Randle, Twinning-related grain boundary engineering, *Acta Materialia*, 52 (2004) 4067-4081.
- [11] S. Xia, B.X. Zhou, W.J. Chen, Grain Cluster Microstructure and Grain Boundary Character Distribution in Alloy 690, *Metall Mater Trans A*, 40a (2009) 3016-3030.
- [12] T.G. Liu, S. Xia, H. Li, B.X. Zhou, Q. Bai, The highly twinned grain boundary network formation during grain boundary engineering, *Mater Lett*, 133 (2014) 97-100.
- [13] V. Randle, Mechanism of twinning-induced grain boundary engineering in low stacking-fault energy materials, *Acta Materialia*, 47 (1999) 4187-4196.

- [14] T. Watanabe, Grain boundary engineering: historical perspective and future prospects, *J Mater Sci*, 46 (2011) 4095-4115.
- [15] M. Masoumi, L.P.M. Santos, I.N. Bastos, S.S.M. Tavares, M.J.G. da Silva, H.F.G. de Abreu, Texture and grain boundary study in high strength Fe–18Ni–Co steel related to hydrogen embrittlement, *Materials & Design*, 91 (2016) 90-97.
- [16] V. Randle, The role of the coincidence site lattice in grain boundary engineering, Cambridge University Press, London, 1996.
- [17] T.G. Liu, S. Xia, H. Li, B.X. Zhou, Q. Bai, Effect of the pre-existing carbides on the grain boundary network during grain boundary engineering in a nickel based alloy, *Materials Characterization*, 91 (2014) 89-100.
- [18] T.G. Liu, S. Xia, H. Li, B.X. Zhou, Q. Bai, C. Su, Z.G. Cai, Effect of initial grain sizes on the grain boundary network during grain boundary engineering in Alloy 690, *J Mater Res*, 28 (2013) 1165-1176.
- [19] K. Kurihara, H. Kokawa, S. Sato, Y.S. Sato, H.T. Fujii, M. Kawai, Grain boundary engineering of titanium-stabilized 321 austenitic stainless steel, *J Mater Sci*, 46 (2011) 4270-4275.
- [20] Y. Cao, H.S. Di, Grain boundary character distribution during the post-deformation recrystallization of Incoloy 800H at elevated temperature, *Mater Lett*, 163 (2016) 24-27.
- [21] V.Y. Gertsman, C.H. Henager, Grain boundary junctions in microstructure generated by multiple twinning, *Interface Science*, 11 (2003) 403-415.
- [22] B.W. Reed, M. Kumar, Mathematical methods for analyzing highly-twinned grain boundary networks, *Scripta Materialia*, 54 (2006) 1029-1033.
- [23] G. Gottstein, Annealing texture development by multiple twinning in f.c.c. crystals, *Acta Metallurgica*, 32 (1984) 1117-1138.
- [24] C. Cayron, Quantification of multiple twinning in face centred cubic materials, *Acta Materialia*, 59 (2011) 252-262.
- [25] C. Cayron, Multiple twinning in cubic crystals: geometric/algebraic study and its application for the identification of the $[\Sigma]3n$ grain boundaries, *Acta Crystallographica Section A*, 63 (2007) 11-29.
- [26] J. Lind, S.F. Li, M. Kumar, Twin related domains in 3D microstructures of conventionally processed and grain boundary engineered materials, *Acta Materialia*, 114 (2016) 43-53.

- [27] C.V. Kopecky, A.V. Andreeva, G.D. Sukhomlin, Multiple twinning and specific properties of $\Sigma = 3n$ boundaries in f.c.c. crystals, *Acta Metallurgica et Materialia*, 39 (1991) 1603-1615.
- [28] F. Hanning, D.L. Engelberg, Metallographic screening of grain boundary engineered type 304 austenitic stainless steel, *Materials Characterization*, 94 (2014) 111-115.
- [29] M. Shimada, H. Kokawa, Z.J. Wang, Y.S. Sato, I. Karibe, Optimization of grain boundary character distribution for intergranular corrosion resistant 304 stainless steel by twin-induced grain boundary engineering, *Acta Materialia*, 50 (2002) 2331-2341.
- [30] M. Kumar, A.J. Schwartz, W.E. King, Microstructural evolution during grain boundary engineering of low to medium stacking fault energy fcc materials, *Acta Materialia*, 50 (2002) 2599-2612.
- [31] J.K. Mason, O.K. Johnson, B.W. Reed, S.F. Li, J.S. Stolken, M. Kumar, Statistics of twin-related domains and the grain boundary network, *Acta Materialia*, 61 (2013) 6524-6532.

Figure Captions

Fig. 1. EBSD map of a GB-engineered alloy 690, in which a grain-cluster was highlighted using the IPF color code. The 5 largest-area orientations in the grain-cluster were designated as $a\sim e$ in descending order.

Fig. 2. Distributions of the area fractions and the numbers of grains per orientation in the grain-cluster highlighted in Fig. 1.

Fig. 3. Distributions of the grain-cluster area, the number of orientations and the number of grains for the thirty grain-clusters according to Table 2.

Fig. 4. (a) Twin-chain of orientation schematically showing the process of multiple-twinning according to the rule that each orientation has four twinning-variants. (b) Modified twin-chain of orientation in which the general behaviors of multiple-twinning were drew: the bidirectional arrows indicate the back-and-forth pattern; the rainbow color indicates the twinning probability.

Fig. 5. Distributions of average area fraction and average number (number fraction) of grains per orientation per grain-cluster in the 30 grain-clusters, where orientations $a\sim e$ are the 5 largest-area orientations in each of the 30 grain-clusters; ‘ $O_{27.8}$ ’ indicates the average value of the other 27.8 orientations except orientations $a\sim e$ because the average orientation number per grain-cluster is 32.8 for the 30 grain-clusters.

Fig. 6. (a, b, c) Distributions of the number fractions that the mutual misorientations between the 5 dominant orientations $a\sim e$ have $\Sigma 3$, $\Sigma 9$ and $\Sigma 27$ respectively in the 30 grain-clusters. The data are in accordance with Table 3. (d) Showing the orientation-pairs that have the highest frequencies in figure a, b and c.

Table 1

The area fractions of the 5 largest-area orientations ($a\sim e$) in each grain-cluster and their mutual misorientations. 30 grain-clusters C1-C30 were analyzed in total.

Cluster	Area fraction					Mutual misorientations of the 5 largest-area orientations: Σ -value/rotation angle-axis									
ID	a	b	c	d	e	ab	ac	ad	ae	bc	bd	be	cd	ce	de
C1	0.23	0.19	0.13	0.10	0.05	3/59.8°, [1 1 1]	3/60.0°, [-1-11]	9/38.9°, [011]	81c/38.3°, [-531]	9/38.8°, [0-1-1]	27b/35.8°, [02-1]	243g/31.7°, [-411]	3/59.8°, [-11-1]	27b/35.4°, [-10-2]	9/39.1°, [0-1-1]
C2	0.39	0.15	0.10	0.07	0.06	3/59.9°, [1-1-1]	3/60.0°, [1-11]	9/39.1°, [011]	81c/38.3°, [53-1]	9/39.1°, [10-1]	27b/35.5°, [02-1]	243g/31.6°, [41-1]	27b/35.2°, [-10-2]	243e/49.6°, [-11-1]	9/39.2°, [01-1]
C3	0.23	0.11	0.10	0.10	0.08	3/59.8°, [-1-1-1]	3/59.5°, [-111]	3/59.9°, [1-1-1]	3/59.8°, [-111]	9/38.6°, [-101]	9/38.9°, [-101]	9/38.7°, [0-1-1]	9/38.5°, [-10-1]	9/39.2°, [0-1-1]	9/39.7°, [0-11]
C4	0.36	0.15	0.15	0.08	0.06	3/59.7°, [-1-11]	3/60.0°, [1-1-1]	9/38.6°, [10-1]	27b/35.5°, [20-1]	9/38.8°, [-110]	3/59.8°, [-11-1]	9/39.1°, [0-11]	27b/35.3°, [012]	81a/38.9°, [-114]	3/59.9°, [-1-11]
C5	0.26	0.24	0.18	0.10	0.04	3/59.7°, [-1-1-1]	3/59.8°, [-1-1-1]	3/59.8°, [1-11]	3/60.0°, [111]	9/39.0°, [10-1]	9/38.8°, [-110]	9/39.5°, [-101]	9/38.9°, [0-1-1]	9/39.1°, [101]	9/38.4°, [01-1]
C6	0.40	0.20	0.09	0.06	0.05	3/59.6°, [-11-1]	3/59.9°, [-1-1-1]	3/59.7°, [1-11]	9/38.9°, [10-1]	9/39.1°, [011]	9/39.4°, [-101]	27b/35.1°, [-102]	9/38.7°, [10-1]	27a/31.6°, [10-1]	3/60.0°, [-1-1-1]
C7	0.28	0.14	0.08	0.06	0.06	3/59.9°, [1-11]	9/38.9°, [-10-1]	3/60.0°, [1-11]	27b/35.3°, [0-2-1]	3/59.8°, [1-1-1]	9/39.0°, [0-1-1]	9/38.9°, [01-1]	27b/35.6°, [0-12]	3/59.8°, [-1-1-1]	81a/39.3°, [-1-4-1]
C8	0.25	0.21	0.13	0.08	0.06	3/59.6°, [1-1-1]	9/38.6°, [10-1]	27b/35.2°, [01-2]	81b/58.1°, [-32-2]	3/59.9°, [111]	9/39.5°, [0-11]	27b/35.6°, [419]	3/60.0°, [1-1-1]	35.8°, [01-1]	3/59.0°, [1-11]
C9	0.37	0.18	0.14	0.06	0.05	9/38.9°, [0-1-1]	3/59.8°, [-11-1]	3/59.9°, [-1-11]	27b/35.2°, [0-21]	3/59.5°, [11-1]	27b/35.7°, [-201]	3/59.9°, [-1-11]	9/38.5°, [-101]	9/39.1°, [011]	81a/39.3°, [141]
C10	0.30	0.15	0.15	0.13	0.05	9/39.1°, [0-11]	3/59.6°, [-1-11]	3/59.8°, [111]	9/39.5°, [-110]	27b/35.6°, [012]	3/60.0°, [-11-1]	9/38.8°, [0-11]	9/39.6°, [-10-1]	27a/31.0°, [-1-10]	3/59.7°, [11-1]
C11	0.49	0.16	0.09	0.08	0.03	3/59.8°, [-1-1-1]	3/59.4°, [111]	81a/38.9°, [11-4]	3/59.8°, [1-11]	9/39.1°, [10-1]	243a/43.4°, [-2-1-1]	9/39.6°, [10-1]	27b/35.3°, [0-21]	9/39.1°, [011]	42.7°, [-104]
C12	0.19	0.17	0.13	0.10	0.09	3/59.8°, [1-11]	3/59.9°, [-11-1]	9/39.0°, [-101]	9/39.1°, [10-1]	9/38.8°, [10-1]	27a/31.6°, [-101]	3/59.8°, [1-11]	3/59.7°, [111]	27b/35.1°, [02-1]	81c/38.6°, [3-5-1]
C13	0.08	0.08	0.08	0.07	0.07	3/59.1°, [11-1]	9/37.7°, [-101]	27b/36.3°, [012]	27a/32.6°, [0-1-1]	3/59.8°, [-111]	9/38.9°, [10-1]	9/38.8°, [-10-1]	3/59.7°, [111]	3/59.9°, [-11-1]	9/39.0°, [10-1]
C14	0.18	0.14	0.09	0.06	0.06	3/60.0°, [-111]	27b/35.4°, [021]	9/39.6°, [011]	3/59.9°, [-111]	9/38.8°, [0-1-1]	3/59.8°, [-11-1]	9/39.3°, [01-1]	3/59.6°, [1-1-1]	81c/38.6°, [3-1-5]	27a/30.6°, [01-1]
C15	0.24	0.16	0.13	0.12	0.09	3/59.9°, [11-1]	3/59.9°, [-11-1]	9/39.8°, [0-1-1]	3/59.7°, [1-1-1]	9/39.3°, [011]	3/59.1°, [11-1]	9/38.7°, [011]	27a/30.8°, [1-10]	9/38.9°, [10-1]	27b/34.0°, [10-2]
C16	0.29	0.20	0.09	0.09	0.08	3/60.0°, [11-1]	9/38.7°, [1-10]	27b/35.7°, [0-21]	3/60.0°, [-1-11]	27b/35.6°, [02-1]	81b/54.4°, [-322]	9/38.7°, [0-1-1]	3/59.8°, [1-11]	3/59.7°, [-1-1-1]	9/39.0°, [-101]
C17	0.22	0.19	0.10	0.09	0.08	3/59.5°, [11-1]	3/59.7°, [-1-11]	9/37.3°, [011]	9/38.5°, [-10-1]	9/39.3°, [-101]	27a/33.2°, [0-11]	3/59.7°, [-11-1]	3/59.9°, [-1-11]	27a/31.8°, [0-1-1]	3/60.2°, [1-1-1]
C18	0.10	0.08	0.06	0.06	0.05	27a/32.0°, [-101]	81c/37.2°, [7-41]	9/38.6°, [0-11]	9/38.8°, [011]	3/58.1°, [1-11]	3/59.8°, [-1-11]	42.5°, [0-3-1]	9/40.2°, [101]	729h/45.6°, [42-3]	81a/39.0°, [-141]
C19	0.16	0.13	0.13	0.12	0.07	9/38.9°, [101]	3/59.9°, [11-1]	27b/35.8°, [201]	3/59.5°, [-1-11]	3/60.0°, [111]	3/59.8°, [1-1-1]	27b/35.4°, [201]	9/39.1°, [101]	9/39.6°, [101]	81b/53.7°, [23-2]
C20	0.25	0.23	0.13	0.07	0.05	3/59.9°, [111]	3/59.8°, [-1-11]	9/39.5°, [101]	27b/35.5°, [0-1-2]	9/39.1°, [101]	27b/35.2°, [021]	9/38.9°, [10-1]	27a/31.0°, [0-1-1]	81a/38.8°, [41-1]	243b/49.5°, [-1-7-6]
C21	0.37	0.10	0.09	0.08	0.06	81c/37.8°, [315]	9/38.8°, [-101]	3/59.8°, [-1-11]	3/59.8°, [111]	729q/50.4°, [-110]	3/59.6°, [0-1-1]	43.0°, [-10-4]	3/59.9°, [-111]	27a/31.8°, [0-11]	9/38.9°, [101]
C22	0.40	0.30	0.11	0.09	0.03	3/59.9°, [1-11]	3/59.8°, [-111]	3/59.9°, [111]	9/38.2°, [0-1-1]	9/39.0°, [0-1-1]	9/39.3°, [10-1]	3/59.9°, [-1-11]	9/39.0°, [01-1]	27b/35.5°, [-102]	27b/35.6°, [2-10]
C23	0.13	0.11	0.10	0.10	0.05	3/59.8°, [111]	27b/35.4°, [021]	243c/12.2°, [113]	243e/50.2°, [-1-1-1]	9/38.9°, [0-11]	81b/55.2°, [3-2-2]	81c/38.4°, [5-1-3]	81a/39.2°, [-411]	9/38.9°, [-101]	729a/39.1°, [-11-1]
C24	0.18	0.11	0.10	0.10	0.08	3/59.6°, [1-11]	81b/54.2°, [2-3-2]	3/59.8°, [11-1]	9/38.5°, [10-1]	27b/35.5°, [0-2-1]	9/38.3°, [110]	3/59.8°, [-111]	243d/35.6°, [231]	81c/38.6°, [-135]	27b/35.8°, [20-1]
C25	0.61	0.15	0.14	0.06	0.02	3/59.9°, [11-1]	3/60.0°, [111]	3/59.9°, [-1-11]	3/59.7°, [1-11]	9/39.2°, [-101]	9/39.1°, [-1-10]	9/38.6°, [-10-1]	9/38.7°, [-10-1]	9/39.3°, [-110]	9/38.7°, [-101]
C26	0.31	0.24	0.10	0.07	0.04	3/59.8°, [111]	3/59.3°, [1-1-1]	3/59.8°, [111]	9/39.3°, [-110]	9/39.0°, [0-1-1]	9/38.8°, [-101]	27a/31.3°, [011]	9/39.2°, [01-1]	3/60.0°, [11-1]	27b/34.6°, [10-2]
C27	0.11	0.09	0.08	0.05	0.05	81b/54.4°, [2-23]	3/59.9°, [-111]	9/38.6°, [011]	27b/35.6°, [20-1]	27b/35.5°, [-120]	81c/38.5°, [14-6]	3/60.0°, [-111]	3/59.7°, [-11-1]	9/38.6°, [-101]	27a/31.9°, [10-1]
C28	0.37	0.11	0.09	0.07	0.07	3/59.9°, [1-11]	3/59.8°, [-11-1]	9/38.8°, [-10-1]	3/59.9°, [-1-1-1]	9/38.8°, [0-1-1]	3/59.8°, [-1-11]	9/38.7°, [10-1]	27b/35.5°, [201]	9/38.8°, [-101]	27b/35.6°, [120]
C29	0.20	0.19	0.12	0.11	0.07	9/39.5°, [10-1]	3/59.8°, [-1-1-1]	27a/31.5°, [10-1]	9/38.5°, [0-11]	3/59.7°, [11-1]	3/59.4°, [111]	9/38.4°, [0-11]	9/39.1°, [-101]	3/59.8°, [-1-11]	27b/34.6°, [-201]
C30	0.53	0.39	0.04	0.03	0.00	3/59.8°, [11-1]	9/38.1°, [011]	9/39.0°, [011]	3/59.8°, [-1-1-1]	9/38.3°, [01-1]	3/59.6°, [1-11]	9/38.3°, [01-1]	9/39.4°, [10-1]	27a/32.7°, [0-11]	27b/35.4°, [-10-2]

Table 2

Statistics of the grain-cluster area, the number of orientations and the number of grains for the 30 grain-clusters; and the numbers of grains with the 5 largest-area orientations $a\sim e$.

Cluster ID	Area μm^2	Orientation number	Grain number	Number of grains with given orientation				
				a	b	c	d	e
C1	345628	50	180	19	7	12	18	5
C13	126704	97	283	5	13	16	17	10
C18	105784	77	205	5	11	6	7	3
C3	76256	47	151	25	16	7	11	8
C14	71652	56	153	8	12	9	2	5
C17	69184	28	112	20	18	7	10	8
C6	60392	29	83	9	6	11	9	3
C27	59300	98	232	7	8	10	8	12
C12	51524	37	109	9	8	11	7	8
C4	51112	27	99	20	14	11	13	2
C2	40824	15	33	4	3	3	5	2
C26	40712	37	151	22	24	12	16	3
C9	38832	21	41	4	3	3	3	1
C5	35132	18	58	12	4	5	6	6
C24	33788	28	62	4	8	2	4	3
C23	31388	44	80	2	4	6	1	3
C10	30060	24	62	8	4	5	10	3
C8	26872	25	77	8	13	11	4	1
C15	26512	20	57	6	7	5	6	4
C19	26140	25	50	2	3	6	1	2
C22	24204	13	41	7	4	4	6	4
C7	22328	27	54	4	4	4	5	1
C20	20908	26	74	7	11	4	2	4
C16	20632	23	48	6	2	3	5	1
C30	18452	5	13	3	3	1	4	2
C21	18252	27	61	7	2	1	10	3
C29	18064	19	37	3	3	5	2	2
C11	17804	14	28	2	7	3	1	4
C28	15960	20	41	3	2	5	3	6
C25	10668	6	24	4	5	9	2	3

Table 3

Statistics of the number of grain-clusters, in which the misorientation of the given orientation-pair has the certain value of $\sum 3^n$. The data were calculated according to Table 1.

Number		Relationship										
		<i>ab</i>	<i>ac</i>	<i>ad</i>	<i>ae</i>	<i>bc</i>	<i>bd</i>	<i>be</i>	<i>cd</i>	<i>ce</i>	<i>de</i>	Sum
Σ -value	3	23	20	11	11	7	10	6	9	5	5	107
	9	4	6	12	9	18	10	15	12	11	8	105
	27	1	2	5	6	4	6	4	7	8	9	52
	81	2	2	1	3	0	3	1	1	4	5	22
	243	0	0	1	1	0	1	4	1	1	2	11
	729	0	0	0	0	1	0	0	0	1	1	3

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6