Evolution of grain boundary character distributions in alloy 825 tubes during high temperature annealing: Is grain boundary engineering achieved through recrystallization or grain growth?

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Abstract: Grain boundary engineering (GBE) of nickel-based alloy 825 tubes was carried out with different cold drawing deformations by using a draw-bench on a factory production line and subsequent annealing at various temperatures. The microstructure evolution of alloy 825 during thermal-mechanical processing (TMP) was characterized by means of the electron backscatter diffraction (EBSD) technique to study the TMP effects on the grain boundary network and the evolution of grain boundary character distributions during high temperature annealing. The results showed that the proportion of $\Sigma 3^n$ coincidence site lattice (CSL) boundaries of alloy 825 tubes could be increased to more than 75% by the TMP of 5% cold drawing and subsequent annealing at 1050°C for 10 min. The microstructures of the partially recrystallized samples and the fully recrystallized samples suggested that the proportion of low Σ CSL grain boundaries depended on the annealing time. The frequency of low Σ CSL grain boundaries increases rapidly with increasing annealing time

associating with the formation of large-size highly-twinned grains-cluster microstructure during recrystallization. However, upon further increasing annealing time, the frequency of low Σ CSL grain boundaries decreased markedly during grain growth. So it is concluded that grain boundary engineering is achieved through recrystallization rather than grain growth.

Keywords: Grain boundary engineering; Electron backscatter diffraction; Grain boundary character distribution; Recrystallization; Grain growth; Nickel-based alloy 825

1. Introduction

Nickel-based alloy 825 is widely used for chemical and petrochemical industrial applications due to the combination of good mechanical properties and corrosion resistance. However, intergranular corrosion (IGC) is one of the serious problems for alloy 825 exposed to aggressive environments, which could result in unexpected failures and lead to huge losses. The grain boundary structure, which can partly be described by coincidence site lattice (CSL) model, can influence the grain boundary chemistry and the susceptibility to intergranular corrosion. The research field of ‘Grain Boundary Engineering’ (GBE) ^[1-3] has been developed a lot over the last three decades since the concept of “Grain Boundary Design” ^[4] was proposed by Watanabe. The aim of GBE is to enhance the grain-boundary-related properties of materials by increasing the frequency of low Σ CSL grain boundaries ($\Sigma \leq 29$) and tailoring the grain boundary network. It was reported that in some face centered cubic materials with low stacking fault energy, such as Ni-based alloys ^[5-8], lead alloys ^[9, 10], austenitic stainless steels ^[11-14] and copper alloys ^[15, 16], the frequency of low Σ CSL grain boundaries can be greatly increased by using proper thermo-mechanical processing (TMP), and as a result the grain boundary related properties were greatly enhanced.

GBE is usually applied through thermo-mechanical processing which includes different combinations of strain and annealing. Many researchers have studied the effects of thermo-mechanical processing on the low Σ CSL grain boundary population and grain boundary network. The high proportion of $\Sigma 3$ boundaries and twin-related higher-order $\Sigma 3^n$ ($n=2, 3, \dots$) boundaries are formed during GBE processing. Multiple twinning^[17, 18] is the basic process of $\Sigma 3^n$ boundaries formation. Regarding to the changes of grain boundary character distribution (GBCD) during GBE processing, different models were proposed to understand the grain boundary network evolution. Randle^[19, 20] proposed the “ $\Sigma 3$ regeneration model”. The two newly recrystallized grains with the same crystallographic texture and containing twins impinged to produce more mobile boundaries. These boundaries migrate to interact with the pre-existing grain boundaries. A large number of $\Sigma 3^n$ boundaries were formed. Kumar et al^[21] proposed the “boundary decomposition mechanism”. They thought that a small deformation during GBE processing was not sufficient to create the general conditions for conventional nucleation of recrystallization. They attributed the enhancement of the $\Sigma 3^n$ boundaries to the decomposition of high energy boundary during strain-induced boundary migration. Wang^[22] proposed that the nuclei of recrystallized grains had incoherent $\Sigma 3^n$ boundaries with the deformed microstructure. The migration and interactions of these grain boundaries played a significant role on the evolution of the grain boundary network. Our previous work^[23, 24] showed that the GBE was a recrystallization process which was featured by the formation of large grain-clusters with a large number of inner-connected $\Sigma 3^n$ type triple junctions. The grain-cluster was formed by multiple twinning starting from a single nucleus during the recrystallization process.

The development of multiple twinning can affect the microstructural evolution that results in different proportions of $\Sigma 3^n$ boundaries. However, it is not clear that the phenomenon of multiple twinning is occurred more dominantly during the process of recrystallization or grain growth. Chen et al. ^[25] have reported an investigation on the evolution of annealing twins in a cold rolled high-purity nickel as a function of annealing temperature. It is revealed that the fraction of $\Sigma 3$ boundaries increases rapidly with increasing annealing temperatures during recrystallization process; while during grain growth after full recrystallization, the fraction of $\Sigma 3$ decrease significantly with further increasing temperature. Jin's research work ^[26] on 304L austenitic stainless steel has also shown that the annealing twins are generated mostly during the recrystallization regime and determined by the interaction, the propagation or the disappearance of the existing twins in the grain growth regime. However, Horton et al. ^[27] have proposed that the percentage of twins increase in impure nickel during grain growth. The study of Gindraux et al. ^[28] on copper has revealed that twins are progressively annihilated during grain growth. Moreover the investigation of Randle et al ^[29] on nickel has presented that if a certain value of $\Sigma 3$ ratio is established, it is not changed by grain growth. Nevertheless during the annealing process after deformation, the recrystallization and grain growth are readily happen. It is not quite clear that the enhancement of the proportion of twin-induced low Σ CSL grain boundary is achieved during recrystallization or during grain growth.

Results from previous work on the effect of twins are rather contradictory. Besides, the relationship between the formation of the grain-cluster and the microstructural evolution during recrystallization needs to be further studied. So it is important to study the change process of low Σ CSL grain boundaries in alloy 825 during annealing. In the present work, the effects of TMP on the GBCD of alloy 825 tubes were studied. The evolution of GBCD in alloy 825 tubes during high

temperature annealing after deformation was focused on through the analysis of the partial recrystallization samples which were annealed at 1050°C or 1100°C for various time after 5% cold deformation. The result of this work was an attempt to answer the question: Is grain boundary engineering achieved through recrystallization or grain growth?

2. Experimental

The chemical composition of alloy 825 tube specimens used in this study is as follow (in *wt %*): 21.5 Cr , 22.00 Fe , 0.01 C, 0.20 Al, 0.90 Ti, 0.45 Si, 3.00 Mo, 0.80 Mn, 2.25 Cu, 0.015 S, 0.025 P, and the balance Ni. First, the solution-annealed alloy 825 tubes were deformed with cold drawing of 3%, 5%, 7%, and 10% respectively using a draw-bench on a factory production line. These cold-drawing strained tubes were then annealed at 1050, 1075, 1100 and 1125 °C for 10 min, respectively. Thereafter, the specimens were quickly quenched into water to obtain the different types of specimens, which have been designated as specimens A1~A4, B1~B4, C1~C4 and D1~D4 correspondingly, as shown in Table 1. In this work, all types of specimens were subjected to a small cold-drawing deformation and subsequent high temperature short-time annealing. To study the evolution of grain boundary network during recrystallization and grain growth, the partially recrystallized specimens were prepared. These specimens of 5% cold-drawing deformation were annealed at 1050 °C or at 1100°C for 60, 90, 120, 150, 180, 210, 240, 420 and 480s, respectively. The specimens are classified into A6p (5% cold drawing – 1050 °C / 60s), A9p, A12p, A15p, A18p, A21p, A24p, A42p and B6p (5% cold drawing – 1100 °C / 60s), B9p, B12p, B15p, B18p, B21p and so on. “A” stands for cold drawing deformation of 5% and annealing temperature of 1050 °C. “B” stands for cold drawing deformation of 5% and annealing temperature of 1100 °C.

“6p/9p/12p/15p/18p/21p/24p/42p” stands for annealing time of 60s/90s/120s/150s/180s/210s/240s/420s, respectively, as shown in Table 1.

The specimens for electron backscatter diffraction (EBSD) experiments were sequentially grinded using SiC papers to 400, 800, 1000 and 2000 grit, and subsequently electropolished in a solution of 20% HClO_4 + 80% CH_3COOH at room temperature with 30V direct current for 2 min. Metallographic specimens were etched for 5~10s in an etching solution containing HNO_3 : HCl : H_2O = 1: 1: 1 (volume fraction) and measured by VHX-100 optical microscopy. The surface crystallographic orientation data of specimens were characterized with HKL-Technology EBSD system attached to a CamScan Apollo 300 scanning electron microscope (SEM). The EBSD analysis was performed using a step size of 1 or 3 μm depending on grain size. An area of 500 μm $\times$ 400 μm or 1050 μm $\times$ 660 μm was scanned on each sample. HKL-Channel5 software was used for analyzing EBSD data. The CSL boundaries were defined according to the Palumbo–Aust criterion ($\Delta\theta_{\max} = 15^\circ \sum^{-5/6}$)^[30]. The grain boundaries with misorientation angle ranged from 2° to 15° were regarded as $\Sigma 1$ boundaries, grain boundaries with $3 \leq \Sigma \leq 29$ were defined as low- Σ CSL boundaries.

3. Results and discussion

3.1 Effects of TMP on GBCD

The optical micrograph and distribution of the types of grain boundaries of the solution annealed starting Ni-based alloy 825 tube sample are given in Figure 1(a) and Figure 1(b) respectively. The average grain size (circle equivalent grain diameter) of the starting sample is 10.64 μm . In this work, all types of boundaries are included as defining individual grains, and thus the annealing twins are regarded as grains. The GBCD of this sample is shown in Figure 1(c). The

proportion of low- Σ CSL grain boundaries is 43.4%. Most of them are $\Sigma 3$ twin boundaries, with a length fraction of 41.6%; the sum of $\Sigma 9 + \Sigma 27$ fraction is only 1.44%, which are regarded as higher-order twin boundaries produced by multiple twinning. Almost all of the $\Sigma 3$ boundaries appear in the shape of straight single line or parallel line pairs.

The measurement results of the fraction of low- Σ CSL boundaries in all the specimens which were cold drawn of 3-10% and then annealed at 1050-1125°C for 10 min are shown in Figure 2. The proportions of low- Σ CSL boundaries have been improved significantly after GBE processing compared to the solution annealed specimens. Except for samples with 10% pre-strain: D1, D2, D3 and D4, others are enhanced to more than 70%, which predominantly are $\Sigma 3$ boundaries, then are multiple twin boundaries ($\Sigma 9$, $\Sigma 27$). However, the proportions of low- Σ CSL boundaries in the D1, D2, D3 and D4 specimens still accounts for about 60%. It is proved that large amount of annealing twins ($\Sigma 3$) will be formed after GBE. The proportions of $\Sigma 3$ boundaries decrease with the increase of pre-strain. The $\Sigma 3$ boundary fraction in sample D4 is only 53.7%, which is significantly lower than other specimens with lower pre-strain and lower annealing temperature. When specimens were subjected to 3%, 5%, 7% or 10% cold drawing deformation, the fraction of low- Σ CSL boundaries decreased gradually with annealing temperature in the range of 1050-1125°C. However, for the specimen with 5% cold drawing deformation, the phenomenon of decline of the fraction of low- Σ CSL boundaries is not obvious. In contrast, it is obvious that the proportions of low- Σ CSL boundaries of the specimens with 5% cold drawing deformation are higher than that with 7% or 10% cold drawing deformation. Therefore, for the alloy 825 tubes in this work, the GBE thermal-mechanical-processing(TMP) of 5% cold drawing deformation followed by different temperature annealing for 10 min is suitable for industry application, because of the high fraction of

low- Σ CSL boundaries (75%) obtained and insensitivity to the annealing temperature. Of course, the adoption of reasonable TMP for GBE should also take into consideration of the original microstructures^[31, 32], such as the original grain size, carbide precipitation, texture, and etc.

The specimen A1 with 3% cold drawing and subsequent annealing at 1050°C for 10 min has the highest fraction of low- Σ CSL boundaries among all the investigated specimens. On one hand, small deformation leads to a lower stored energy and resulting in lower density of recrystallization nuclei during the subsequent annealing. Thus, recrystallization nuclei had larger potential space to grow by consuming the deformed area. This is beneficial to forming a high proportion of low- Σ CSL grain boundaries. On the other hand, the post-recrystallization high temperature annealing above 1050°C promoted the grain growth (as shown in figure 3), and in the meanwhile, the migration of general high angle grain boundaries swept away the existing low- Σ CSL grain boundaries (annihilated during grain growth). Thus the fraction of low- Σ CSL grain boundaries decreased.

As $\Sigma 9$ and $\Sigma 27$ grain boundaries are formed by multiple twinning, the fraction of ($\Sigma 9 + \Sigma 27$) boundaries should depend the fraction of $\Sigma 3$ boundaries. If obtained a high proportion of $\Sigma 3$ boundaries, the proportion of ($\Sigma 9 + \Sigma 27$) boundaries will be enhanced correspondingly. As shown in Fig. 2, the fraction of ($\Sigma 9 + \Sigma 27$) has a similar variation trend with that of the $\Sigma 3$ boundary as the annealing temperature varying. For the specimens after 3% cold drawing deformation, the proportions of $\Sigma 3$ boundaries decrease with the increase of annealing temperature. Similarly, the fraction of ($\Sigma 9 + \Sigma 27$) boundaries is also decreased gradually with temperature.

Fig. 3 shows the dependence of grain size on the pre-strain and the annealing temperature. The grain sizes after GBE processing are larger than the initial grain size of the solution annealed specimen. The deformation amount and annealing temperature have important influences on the

grain size during GBE processing. For a given pre-strained series specimens, the grain size increased with the annealing temperature. While for a specific annealing temperature, the grain size decreased with the pre-strain. Chen et al ^[25] reported that the fraction of $\Sigma 3$ boundaries increased at first with increasing of grain size, and then decreased significantly as the grain size further increased. In the current experiment, as can be seen from Fig. 3, the proportions of low- Σ CSL grain boundaries decrease with the increasing of grain size for the specimens with 3, 7 or 10% pre-strains and subsequent annealing in the temperature range of 1050-1125°C. However, there is no obvious change accordingly for low- Σ CSL grain boundaries fraction with the obvious increase of grain size in the specimens after 5% cold drawing deformation and annealing processing.

Fig. 4 shows the grain boundary maps of alloy 825 after GBE-processing. Most boundaries are of the $\Sigma 3^n$ type, and they connect with each other, forming many triple junctions of $\Sigma 3^n$ -type boundaries. They are encircled by random high-angle grain boundaries, inside of which can be recognized as a grains-cluster (gray background in Fig. 4). All of the grains within the grain-cluster can have $\Sigma 3^n$ mutual misorientations, no matter if they are adjacent to one another (or not). However, there are obvious differences in grain-cluster size for the specimens with difference pre-strain by cold drawing deformation, as shown in Fig. 4. The sample A1 with 3% pre-strain has the largest grain-cluster and highest proportion of low- Σ CSL grain boundary as well; whereas the sample with larger pre-strain often have smaller grain-cluster and the associated lower proportion of low- Σ CSL grain boundaries. As shown in Figure 5, the proportion of low- Σ CSL grain boundaries has a positive correlation with the value of $(D/d)^2$ in the specimens with similar grain size (B1, C4, D4), where the ‘ D ’ is the grain-cluster size and the ‘ d ’ is the grain size. The value of $(D/d)^2$ can be approximated as the number of grains within a grains-cluster. The larger the size of grains-cluster, the higher the ratio

of the grains-cluster size over grain size, which will result in a higher proportion of low- Σ CSL grain boundaries.

3.2 Grain boundary network evolution during recrystallization and grain growth

For the purpose of analyzing the evolution of grain boundary network during recrystallization and grain growth, two partially recrystallized specimens were selected. Fig 6 and Fig 7 shows the grain boundary networks with different methods of Channel5 in specimens with 1050°C or 1100°C annealing for different short time after 5% cold- drawing. As the figure shows, when annealing time increased from 60s to 420s, a clear recrystallization process could be seen. In Fig. 6a₁-a₄, no recrystallization nucleuses were observed in the deformation microstructure. With further extension of the annealing time at 1050°C to 90s, a small amount of recrystallization grains were formed, where some $\Sigma 3$ and $\Sigma 9$ boundaries were generated inside the newly formed grains. Recrystallized and non-recrystallized regions were separated by random grain boundaries. As the increase of annealing time, the recrystallized grains grow larger at the expense of consuming the deformed matrix. When the time was up to 210 s, recrystallized grains completely occupied the microstructure of the specimen, indicating that the sample was almost fully recrystallized. In this case, grain growth occurred upon further annealing. Nevertheless, when annealed at 1100°C, a few recrystallization nucleus were observed in the simple with 60 s annealing, as shown in Fig. 7a₁-a₄; upon annealing for 150 s, the sample were fully recrystallized. While for the samples annealed at 1050°C, the recrystallization kinetic was slower.

The results of Fig. 6 show that sample “A9p” and sample “A15p”, which were cold drawn 5% and annealed at 1050°C for 90s and 150s respectively, were partially recrystallized. As shown in Figs. 6b₃ and 6d₃, there was a grain-cluster in the recrystallized region for each figure, where all the

grain boundaries inside are of $\Sigma 3$, $\Sigma 9$, $\Sigma 27$ or even higher-order twin boundaries types. Fig. 8 shows the growing grain-clusters in samples A9p and A15p. In order to analyze the mutual misorientations between any two grains inside of the cluster, eight grains were randomly selected, and the results are listed in Table II and Table III. The detailed analyzing of all the mutual misorientations between any two grains in the grain-cluster was reported elsewhere^[23, 33]. It can be demonstrated that all grains in cluster have $\Sigma 3^n$ misorientations regardless of whether they are adjacent. A large number of experimental results^[34-36] show that the grain-cluster began to grow from a single recrystallized nucleus during recrystallization, and then were formed by generating a series of $\Sigma 3$ annealing twin boundaries, and in the meanwhile, $\Sigma 3^n$ grain boundaries can be produced when different generation twins meet. It is easy to form annealing twins for fcc metal materials with low to medium stacking fault energy during the period of newly formed grain growing. Thus, new annealing twins are iteratively generated in the wake of the moving grain boundary during recrystallization, forming the interconnecting $\Sigma 3^n$ type grain boundaries. So any two grains in a grain-cluster remains $\Sigma 3^n$ misorientations relationship regardless of whether they are adjacent. Some black lines denoted random grain boundaries inside the grain-cluster are actually high order twin boundaries, such as $\Sigma 3^4$ ($\Sigma 81$), $\Sigma 3^5$ ($\Sigma 243$)..., which are arbitrarily defined as random ones by the channel 5 system due to their Σ values are larger than 29. The detailed result of misorientation examination between any grain pairs inside the cluster was reported elsewhere^[23].

In Fig 7, recrystallized regions with annealing twins could be clearly seen inside sample “B6p,” which was cold drawn 5% and annealed at 1100°C for 60s. The recrystallized region also consisted of a growing grain-cluster which is shown in Figure 9. All of the grains belonging to this grain-cluster retained specific $\Sigma 3^n$ mutual misorientations among them, as shown in Table IV.

The grain size, recrystallization fraction and proportion of low- Σ CSL grain boundaries as a function of annealing time for the specimen with 5% strain and subsequent annealing at 1050°C are shown in Fig. 10, 11. It is clearly seen that the average grain size increased with the extension of annealing time, meanwhile, the fraction of recrystallization also increased. When the annealing time was up to 210s, the recrystallized fraction increased up to 80%. Y.Jin et al.^[26] suggested that the samples approached the grain growth regime if the fraction of recrystallization reached more than 80%. In this case, as shown in Fig. 10, when annealed at 1050°C for 210s, the sample was fully recrystallized. When the annealing time increased over 210s, the sample was in the grain growth regime (post-recrystallization). The changes of the fractions of different character grain boundaries during recrystallization and grain growth are shown in Fig. 11. With the annealing time in the range of 60~210s, the proportion of Σ 1 grain boundaries gradually reduced, however, the proportion of Σ 3 grain boundaries increased significantly with the increase of annealing time, while the tendency of total low Σ CSL grain boundaries change was consistent with the variation trend of Σ 3. The fraction of low Σ CSL and Σ 1 grain boundaries reached its highest and lowest respectively at the time of 210s. In another word, the sample was fully recrystallized at the time of 210 s. The proportion of low- Σ CSL grain boundaries was maximally improved. As the annealing time was extended further, the proportions of Σ 3 and low- Σ CSL grain boundaries began to decrease gradually. The same phenomenon was also found in other samples, which were subjected to 5% strain deformation and subsequent annealing at 1100 °C for various times, as shown in Figs. 12 and 13. But it is clearly seen that the recrystallization fraction of the sample annealed at 1100 °C for 180s had been increased to more than 80%. With the annealing time in the range of 60~180s, the proportion of Σ 1 grain boundaries gradually reduced. When annealing time was up to 180s, the fraction of Σ 1

achieved minimum value. This demonstrates that when treated at 1100°C for 180s, samples were also recrystallized completely. Before the completion of the recrystallization, the proportions of $\Sigma 3$ and total low- Σ CSL grain boundaries increased with annealing time. On the contrary, after the completion of the recrystallization, the proportions of $\Sigma 3$ and total low- Σ CSL grain boundaries reduced with annealing time.

According to the analyzing above, it is concluded that the proportion of low- Σ CSL grain boundaries of the sample after GBE processing increased firstly and then decreased with the increase of annealing time. In the early stages of annealing, the recrystallization occurred. During the recrystallization process, the $\Sigma 3$ grain boundaries are developed (see Fig. 6, 7) through multiple-twinning associated with the recrystallization nuclei growth. Therefore, the proportion of low- Σ CSL grain boundaries was enhanced. When annealed for 210 s or 180 s, very few $\Sigma 1$ grain boundaries were left, and the samples were in the fully recrystallized state, so the proportion of low- Σ CSL grain boundaries reached the peak value. If further annealing after 210s or 180s, the samples were in the grain growth stage after recrystallization. In contrast to the recrystallization process, the frequency of low- Σ CSL grain boundaries decreased continually during the grain growth. Li ^[37] and Bozzolo ^[38] had proposed a higher rate of grain boundary migration was more favorable to promote the generation of annealing twins. Compared with recrystallization, the lower driving forces in grain growth resulted in much slower grain boundary migration. Moreover, Jin ^[39] reported that the decrease in annealing twin density could be attributed to the disappearance of small grains containing several twin boundaries, which grew without producing new twins. During the grain growth process, small grains were consumed by large grains, due to the existing low- Σ CSL grain

boundaries being swept away by the migration of general high angle grain boundaries, so the proportion of low- Σ CSL grain boundaries decreased with the extension of the annealing time.

4. Conclusion

Grain boundary engineering was carried out in tubes of alloy 825. The GBE processing which was 5% cold drawing and subsequent annealing at 1050°C for 10 min, can increase the fraction of low- Σ CSL grain boundaries to more than 75% which mainly are of $\Sigma 3^n$ types. And meanwhile, the large-size highly-twinned grain-cluster microstructure was formed. Study on the evolution of grain boundary network during recrystallization and grain growth, showed that during the recrystallization stage, the proportion of low- Σ CSL grain boundaries increase significantly with increasing annealing time. However, during the grain growth after full recrystallization, the proportion of low- Σ CSL grain boundaries decrease with further extending of the annealing time due to being swept away of the existing low- Σ CSL grain boundaries. So it is concluded that grain boundary engineering is achieved through recrystallization rather than grain growth.

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Figure captions

Fig.1 OM map (a), distributions of the types of grain boundaries (b) and histogram of grain boundary character distribution (c) of the solution annealed starting material.

Fig.2 Effect of deformation and annealing temperature on the length fraction of $\Sigma 3$, $\Sigma 9 + \Sigma 27$, and other low- Σ CSL of alloy 825.

Fig.3 Effect of deformation and temperature on length fraction of low- Σ CSL and the mean grain size of alloy 825. Colored bars correspond to length fraction of low- Σ CSL grain boundaries and points/lines curves correspond to grain size.

Fig.4 Different character grain boundaries of 825 alloy after deformed at 3% (a, e, i, m), 5% (b, f, j, n), 7% (c, g, k, o), 10% (d, h, l, p) and heat treated at 1050 °C (a ~ d), 1075 °C (e ~ h), 1100 °C (i ~ l) and 1125 °C (m ~ p).

Fig.5 The proportions of low- Σ CSL grain boundaries, the values of $(D/d)^2$ and the mean grain size for specimen B1, C4, D4: the value of $(D/d)^2$ can be approximated as the number of grains encircled by a grain-cluster.

Fig.6 EBSD analysis of microstructural changes during high temperature annealing for specimen A6p (a₁-a₄), A9p (b₁-b₄), A12p (c₁-c₄), A15p (d₁-d₄), A18p (e₁-e₄), A21p (f₁-f₄), A24p (g₁-g₄), A42p (h₁-h₄) with map of different types of boundaries (a₁-h₁), grain orientation map contrasted by color of inverse pole figure (a₂-h₂), map contrasted by color of grain-mean misorientation (a₃-h₃), and component map of recrystallized fraction (a₄-h₄).

Fig.7 EBSD analysis of microstructural changes during high temperature annealing for specimen B6p (a₁-a₄), B9p (b₁-b₄), B12p (c₁-c₄), B15p (d₁-d₄), B18p (e₁-e₄), B21p (f₁-f₄), B24p (g₁-g₄), B42p (h₁-h₄) with map of different types of boundaries (a₁-h₁), grain orientation map contrasted by color of

inverse pole figure (a_2-h_2), map contrasted by color of grain-mean misorientation (a_3-h_3), and component map of recrystallized fraction (a_4-h_4).

Fig.8 A growing grain-cluster in the partially recrystallized sample A9p (a) and A15p (b) as shown in Fig.6.

Fig.9 A growing grain-cluster in the partially recrystallized sample B6p as shown in Fig.7.

Fig.10 Evolution of microstructural parameters for the specimens annealed at 1050°C: (a) average grain size and (b) recrystallized fraction as a function of annealing time.

Fig.11 Special boundary proportions as a function of annealing time for the specimen with 5% strain and subsequent annealing at 1050 °C.

Fig.12 Evolution of microstructural parameters for the specimens annealed at 1100°C: (a) average grain size and (b) recrystallized fraction as a function of annealing time.

Fig.13 Special boundary proportions as a function of annealing time for the specimen with 5% strain and subsequent annealing at 1100 °C.

Table 1 Thermomechanical treatments of the alloy 825 specimens

GBE				Partially recrystallized processing		
The initial	Cold-drawing /%	Annealing	Sample ID	Cold-drawing /%	Annealing	Sample ID
Solution annealed	3	1050°C×10	A1	5	1050°C×60 s	A6p
		1075°C×10	A2		1050°C×90 s	A9p
		1100°C×10	A3		1050°C×120	A12p
		1125°C×10	A4		1050°C×150	A15p
	5	1050°C×10	B1		1050°C×180	A18p
		1075°C×10	B2		1050°C×210	A21p
		1100°C×10	B3		1050°C×240	A24p
		1125°C×10	B4		1050°C×420	A42p
	7	1050°C×10	C1		1100°C×60 s	B6p
		1075°C×10	C2		1100°C×90 s	B9p
		1100°C×10	C3		1100°C×120	B12p
		1125°C×10	C4		1100°C×150	B15p
	10	1050°C×10	D1		1100°C×180	B18p
		1075°C×10	D2		1100°C×210	B21p
		1100°C×10	D3		1100°C×240	B24p
		1125°C×10	D4		1100°C×420	B42p

Table II. Mutual misorientations of the eight grains randomly selected from grain-cluster in sample

A9p from Fig.8 (a)

Grain	Misorientation							
	a	b	c	d	e	f	g	h
a		$\Sigma 3/0.3^\circ$	$\Sigma 9/0.5^\circ$	$\Sigma 81d/0.3^\circ$	$\Sigma 27a/0.2^\circ$	$\Sigma 9/0.6^\circ$	$\Sigma 27b/0.3^\circ$	$\Sigma 81b/0.3^\circ$
b	59.9, [1-11]		$\Sigma 3/0.2^\circ$	$\Sigma 27a/0.6^\circ$	$\Sigma 9/0.1^\circ$	$\Sigma 3/0.5^\circ$	$\Sigma 9/0.3^\circ$	$\Sigma 27b/0.8^\circ$
c	38.9, [-10-1]	59.5, [11-1]		$\Sigma 81b$ 39.9, [141]	$\Sigma 27b/0.2^\circ$	$\Sigma 9/0.1^\circ$	$\Sigma 27b/0.3^\circ$	$\Sigma 81c/0.3^\circ$
d	59.5, [4-3-4]	32, [-1-10]	38.4, [21-4]		$\Sigma 3/0.2^\circ$	$\Sigma 9/0.3^\circ$	$\Sigma 27b/0.1^\circ$	$\Sigma 81b/0.2^\circ$
e	32.5, [01-1]	39.5, [0-11]	35.8, [210]	59.2, [-111]		$\Sigma 3/0.1^\circ$	$\Sigma 9/0.3^\circ$	$\Sigma 27b/0.5^\circ$
f	38.8, [-101]	59.5, [1-11]	38.6, [0-1-1]	38.9, [10-1]	59.1, [111]		$\Sigma 3/0.2^\circ$	$\Sigma 9/0.3^\circ$
g	37.3, [02-1]	38.7, [-101]	35.3, [1-20]	34.9, [0-12]	38.8, [110]	58.9, [1-11]		$\Sigma 3/0.4^\circ$
h	38.7, [41-1]	35.9, [0-21]	54.5, [322]	40.0, [-1-14]	34.7, [120]	38.5, [-10-1]	59.7, [11-1]	

Table III. Mutual misorientations of the eight grains randomly selected from grain-cluster in sample

A15p from Fig.8 (b)

Grain	Misorientation							
	a	b	c	d	e	f	g	h
a		$\Sigma 9/0.3^\circ$	$\Sigma 3/0.2^\circ$	$\Sigma 3/0.3^\circ$	$\Sigma 3/0.2^\circ$	$\Sigma 81b$ 38.9, [141]	$\Sigma 27a/0.3^\circ$	$\Sigma 243d$ 35.4, [52-4]
b	38.8, [101]		$\Sigma 27a/0.5^\circ$	$\Sigma 27b/0.3^\circ$	$\Sigma 3/0.3^\circ$	$\Sigma 729d$ 28.6, [05-8]	$\Sigma 243a$ 43.1, [0-11]	$\Sigma 243j$ 60, [-11,1,-11]
c	59.7, [1-11]	31.7, [011]		$\Sigma 9/0.2^\circ$	$\Sigma 9/0.4^\circ$	$\Sigma 243i$ 49.7, [565]	$\Sigma 81b$ 38.9, [141]	$\Sigma 81a$ 38.4, [-1-3-5]
d	59.9, [-111]	35.8, [-120]	39.2, [01-1]		$\Sigma 9/0.1^\circ$	$\Sigma 27b/0.3^\circ$	$\Sigma 9/0.3^\circ$	$\Sigma 81c/0.3^\circ$
e	59.9, [11-1]	59.6, [-11-1]	38.7, [011]	38.8, [-101]		$\Sigma 243a$ 43.1, [0-11]	$\Sigma 81d/0.4^\circ$	$\Sigma 729f$ 28.6, [7-62]
f	38.6, [-241]	28.7, [-30-2]	49.9, [-3-3-4]	35.3, [2-10]	43.1, [304]		$\Sigma 3/0.3^\circ$	$\Sigma 3/0.3^\circ$
g	31.6, [0-1-1]	43.1, [-304]	38.6, [-421]	38.9, [011]	60.2, [4-34]	59.9, [111]		$\Sigma 9/0.2^\circ$
h	35.2, [32-4]	57, [4-2-3]	38.8, [-314]	54.5, [-3-2-2]	28.3, [-3-41]	59.7, [-11-1]	38.6, [110]	

Table IV. Mutual misorientations of the eight grains randomly selected from Fig.9

Grain	Misorientation							
	a	b	c	d	e	f	g	h
a		$\Sigma 3/0.3^\circ$	$\Sigma 9/0.1^\circ$	$\Sigma 81b/0.2^\circ$	$\Sigma 81c/0.3^\circ$	$\Sigma 243b/0.6^\circ$	$\Sigma 243g$ 43.1, [59-5]	$\Sigma 27b/0.1^\circ$
b	59.5, [1-11]		$\Sigma 3/0.2^\circ$	$\Sigma 27b/0.3^\circ$	$\Sigma 27b/0.4^\circ$	$\Sigma 81c/0.3^\circ$	$\Sigma 81c$ 54.5, [2-3-2]	$\Sigma 9/0.2^\circ$
c	38.6, [101]	59.8, [1-1-1]		$\Sigma 9/0.2^\circ$	$\Sigma 9/0.3^\circ$	$\Sigma 27b/0.6^\circ$	$\Sigma 243b/0.3^\circ$	$\Sigma 3/0.4^\circ$
d	39.2, [1-41]	34.8, [-102]	38.9, [110]		$\Sigma 9/0.3^\circ$	$\Sigma 27b/0.1^\circ$	$\Sigma 243i$ 49.7, [5-6-5]	$\Sigma 3/0.1^\circ$
e	54.9, [32-2]	35.2, [012]	38.6, [-1-10]	38.8, [01-1]		$\Sigma 3/0.3^\circ$	$\Sigma 27b/0.4^\circ$	$\Sigma 3/0.1^\circ$
f	12.2, [-13-1]	54.1, [2-2-3]	35.3, [-2-10]	35.5, [20-1]	59.9, [-1-11]		$\Sigma 9/0.3^\circ$	$\Sigma 9/0.2^\circ$
g	45.7, [302]	54.9, [-233]	11.9, [31-1]	49.6, [41-4]	35.1, [-120]	38.9, [-1-10]		$\Sigma 81c/0.2^\circ$
h	35.3, [0-1-2]	39.4, [-110]	59.1, [-1-1-1]	59.6, [1-1-1]	59.7, [1-11]	38.7, [0-1-1]	54.6, [223]	

Fig 1

Fig 2

Fig 3

Fig 4

Fig 5

Fig 6

Fig 7

Fig 8

Fig 9

Fig 10

Fig 11

Fig 12

Fig 13