Effect of Deformation and Heat Treatment on Grain Boundary Characteristic Distribution of 825 Alloy Tubes (Postprint)

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Abstract

Nickel-based 825 alloy tubes were cold drawn using an industrial cold drawing machine and subsequently annealed for grain boundary engineering (GBE) treatment. The effects of various cold drawing deformations and annealing temperatures on the grain boundary character distribution (GBCD) of 825 alloy were investigated using EBSD and orientation imaging microscopy (OIM). The results indicate that with 5% cold drawing deformation and annealing at 1050 ℃ for 10 min, the proportion of low-Σ coincidence site lattice (ΣCSL, coincidence site lattice, Σ$ $29) grain boundaries can be increased to over 75% (Palumbo-Aust criterion), while simultaneously forming a large-scale microstructure of clusters of grains with Σ3n orientation relationships (n=1, 2, 3,). As the cold drawing deformation prior to recrystallization annealing increases, the grain cluster size decreases, while the proportion of low-ΣCSL grain boundaries likewise decreases, and the proportion of low-ΣCSL grain boundaries also decreases with increasing grain size. For the alloy subjected to 5% cold drawing deformation, the grain boundary character distribution remains essentially unchanged upon annealing at 1050–1125 ℃ for 10 min, and the annealing temperature exerts a minor influence on the proportion of low-ΣCSL grain boundaries in the alloy; following 3%, 7%, and 10% cold drawing deformations, the proportion of low-ΣCSL grain boundaries in the alloy continuously decreases with increasing annealing temperature.

Full Text

Abstract

Alloy 825 is widely used for chemical and petrochemical applications due to its good combination of 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 field of grain boundary engineering (GBE) has developed a lot over the last two decades since the concept of grain boundary design was proposed. The aim of GBE is to enhance the grain-boundary-related properties of materials by increasing the frequency of low ΣCSL ($\Sigma \leq 29$) grain boundaries (GBs) and tailoring the grain boundary network. It was reported that in some fcc materials with low stacking fault energy, such as Ni-based alloys, lead alloys, austenitic stainless steels and copper alloys, the frequency of low ΣCSL GBs can be greatly increased by using proper thermomechanical processing (TMP), and as a result the grain boundary related properties were greatly enhanced.

In this work, GBE is applied to the manufacture of Ni-based alloy 825 tubes by cold drawing using a draw-bench on a factory production line and the subsequent annealing. The effect of thermomechanical processing on the grain boundary character distribution (GBCD) of alloy 825 was studied by means of the EBSD technique and orientation image microscopy (OIM). The results show that the proportion of low ΣCSL grain boundaries increase to more than 75% by the TMP after 5% cold drawing and subsequent annealing at 1050 ℃ for 10 min, and simultaneously the large-size highly-twinned grain-cluster microstructure is formed. The size of the grain-cluster and proportion of low ΣCSL grain boundaries decrease with the increase of pre-strain. The proportion of low ΣCSL grain boundaries decreases with the increase of the mean grain size. The annealing temperatures in the range of 1050~1125 ℃ have no obvious effect on the GBCD of the specimen with 5% cold drawing deformation; while the proportions of low ΣCSL GBs of the sample with 3%, 7% and 10% cold drawing deformation decrease with the increase of annealing temperature.

Introduction

Nickel-based Incoloy 825 alloy is a Ti-stabilized Ni-Fe-Cr-Mo-Cu solid solution strengthened alloy developed by Inconel Company in 1952. Due to its excellent resistance to stress corrosion cracking, crevice corrosion, and good performance in both oxidizing and non-oxidizing hot acidic environments, it has been widely adopted in industrial equipment for machinery, chemical processing, electronics, boilers, environmental protection, aviation, and instrumentation applications [1,2]. With rapid economic development, the demands on equipment and components in industrial production continue to increase, making further improvement of material properties a critical concern.

Grain boundaries are one of the most important structural features of polycrystalline materials and significantly influence material properties. Many material characteristics are closely related to grain boundary properties, including grain boundary diffusion [3], corrosion [4], intergranular fracture [5], slip [6], and segregation [7], all of which are affected by grain boundary structure. Low Σ-value coincidence site lattice (CSL) grain boundaries are considered to have highly ordered structures with lower energy and superior properties compared to random high-angle grain boundaries. To optimize and improve material performance, Watanabe [8] proposed the concept of "grain boundary design and control," which researchers later developed into the field of grain boundary engineering (GBE) [9~12]. GBE aims to enhance grain-boundary-related properties by increasing the proportion of special grain boundaries and controlling the grain boundary network distribution through appropriate deformation and heat treatment processes.

GBE technology can significantly improve grain-boundary-related properties without altering the chemical composition. Lehockey et al. [11] applied small-strain cold working combined with annealing at 975~1200 ℃ to alloys 625, V-57, and 738, achieving low ΣCSL grain boundary proportions three times higher than conventional alloys, thereby enhancing corrosion resistance, creep resistance, and fatigue performance. Thomson and Randle [13] deformed Ni200 alloy by 6%~7% and annealed at 750 ℃ for 12, 21.5, 24, and 26.5 h, finding that after 24 h of annealing, the proportion of low ΣCSL grain boundaries increased significantly with abundant Σ3 annealing twins formed, effectively preventing intergranular crack propagation. Fang et al. [14] cold-rolled 304 stainless steel by 6% and annealed at 900 ℃ for 1, 2, and 3 h, obtaining low ΣCSL grain boundary proportions of 47.5%, 57.1%, and 72.4%, respectively. Xia et al. [15] annealed cold-drawn (7%) alloy 690 tubes at 1100 ℃ for 5 min, increasing the low ΣCSL grain boundary proportion to 75% and substantially improving intergranular corrosion resistance. Kumar et al. [16] deformed alloy 600 by 20%, annealed at 1000 ℃ for 15 min, and repeated this thermomechanical process seven times, achieving a low ΣCSL grain boundary proportion of 70% with random grain boundary networks interrupted by special grain boundaries. Ding and Chen [17] annealed cold-rolled (5%) 316 stainless steel at 1050 ℃ for 30 min, resulting in a low ΣCSL grain boundary proportion exceeding 80%.

Although numerous studies have reported methods to increase the proportion of low ΣCSL grain boundaries, different materials require different deformation and heat treatment parameters, and their effects on grain boundary character distribution (GBCD) vary. Therefore, further understanding of how processing parameters affect GBCD during GBE treatment is needed. To apply GBE technology in actual production, it is essential to combine factory-scale manufacturing conditions and investigate processing methods that can increase the proportion of low ΣCSL grain boundaries, ensuring that research outcomes can be integrated with current industrial process parameters. This work utilizes production equipment to conduct cold drawing with various deformation levels and annealing treatments on nickel-based alloy 825 tubes, employing electron backscatter diffraction (EBSD) to investigate the effects of cold drawing deformation and heat treatment on the proportion of special grain boundaries and the characteristics of grain boundary network distribution in alloy 825.

1 Experimental Methods

The experimental material was nickel-based alloy 825 tube with a chemical composition (mass fraction, %) of: Cr 21.5, Fe 22.00, C 0.01, Al 0.20, Ti 0.90, Si 0.45, Mo 3.00, Mn 0.80, Cu 2.25, S 0.015, P 0.025, and Ni balance. Initially, solution-treated tube samples were subjected to cold drawing deformation of 3%, 5%, 7%, and 10% using a YLB-B-5/20-15 draw-bench in the factory. Subsequently, samples with different cold drawing deformations were annealed at 1050, 1075, 1100, and 1125 ℃ for 10 min, followed by rapid quenching after heating. This work employed small-strain cold working combined with high-temperature (equivalent to solution temperature) short-time annealing, which can be integrated with factory production process parameters.

Specimens were prepared by electrolytic polishing using an electrolyte composition of 20% $HClO_4$ + 80% CH_3COOH (volume fraction) at 30 V DC for approximately 90 s at room temperature. The polished samples were then etched using an etchant of HNO_3:HCl:H_2O = 1:1:1 (volume ratio) for about 5~10 s to reveal grain boundaries. A HKL-EBSD attachment in a CamScan Apollo300 thermal field emission gun scanning electron microscope (SEM) was used to scan the sample surface point by point and line by line with a step size of 3 m over an area of 1050 m × 660 m, collecting and indexing backscattered electron Kikuchi diffraction patterns to obtain crystallographic information. The test results were analyzed using HKL-Channel5 software, with the measurement system adopting the Palumbo and Aust standard [18] (Δ _{max} = 15°/Σ^(-5/6), where Δ _{max} refers to the maximum deviation angle between the experimentally measured CSL orientation relationship and the standard geometric CSL orientation relationship) to determine grain boundary types. A VHX-100 digital optical microscope (OM) was used to observe the etched sample surfaces.

2 Results and Discussion

[Figure 1: see original paper] shows the OM image, different types of grain boundary distribution maps, and grain orientation distribution maps of the solution-annealed nickel-based alloy 825 tube. Using the equivalent circle diameter method in HKL-Channel5 software, the average grain size was measured to be 10.5 m (in this work, twin boundaries were included in grain size calculations). The proportion of low ΣCSL grain boundaries in this solution-treated tube was 43.4% (length fraction, same below), with the vast majority being Σ3 twin boundaries at 41.6%, while multiple twin boundaries Σ9+Σ27 accounted for only 1.4%.

EBSD measurements were performed on alloy 825 after different small-strain cold drawing deformations and short-time annealing treatments at various temperatures. The variation in the proportion of different low ΣCSL grain boundaries with cold drawing deformation and annealing temperature is shown in [Figure 2: see original paper]. It can be seen that after GBE processing, the proportion of low ΣCSL grain boundaries in the samples increased significantly compared to the solution-treated sample. Except for the samples with 10% cold drawing deformation annealed at 1050~1125 ℃, all other samples achieved low ΣCSL grain boundary proportions above 70.0%, with Σ3 grain boundary proportions exceeding 60.0%. Even the samples with 10% cold drawing deformation annealed at 1050~1125 ℃ exhibited low ΣCSL grain boundary proportions around 60.0%, with Σ3 grain boundary proportions over 50.0%. Whether in the sample with higher low ΣCSL grain boundary proportion (3% cold drawing + 1050 ℃ annealing) or the sample with lower proportion (10% cold drawing + 1125 ℃ annealing), Σ3 grain boundaries dominated the low ΣCSL grain boundaries, accounting for approximately 87.0% of the total, followed by multiple twin boundaries Σ9 and Σ27, with other low ΣCSL grain boundaries present in minimal amounts. This indicates that after small-strain cold drawing and high-temperature short-time recrystallization annealing, the samples developed a high proportion of annealing twins, but the Σ3 grain boundary proportion continuously decreased with increasing pre-deformation. The Σ3 grain boundary proportion in the sample with 10% cold drawing deformation and 1125 ℃ annealing was only 53.7%, significantly lower than in other deformation-annealing samples. Furthermore, for samples with 3% cold drawing deformation, the low ΣCSL grain boundary proportion after annealing at 1050 ℃ was notably higher than after annealing at higher temperatures. For samples with 10% cold drawing and 1125 ℃ annealing, the low ΣCSL grain boundary proportion was significantly lower than in samples with other deformation levels. However, samples with 5% cold drawing deformation and different annealing temperatures showed essentially identical low ΣCSL grain boundary proportions, which were significantly higher than those in samples with 7% or 10% cold drawing deformation. Therefore, for alloy 825 tubes in this work, 5% cold drawing deformation followed by annealing at various temperatures for 10 min represents an ideal GBE processing method, capable of increasing the low ΣCSL grain boundary proportion to above 75.0% with minimal influence from annealing temperature. Of course, appropriate GBE processing parameters also depend on the initial microstructure [19,20], such as original grain size, carbide precipitation state, and texture. Additionally, [Figure 2: see original paper]c shows that alloy 825 cold-drawn by 3% and annealed at 1050 ℃ for 10 min achieves the highest low ΣCSL grain boundary proportion, which decreases with increasing annealing temperature. This occurs because, during GBE processing, samples with smaller deformation have lower recrystallization nucleation density, providing larger potential space for grain growth. As the recrystallization front migrates through the deformed matrix, it continuously generates annealing twins, facilitating the formation of high proportions of low ΣCSL grain boundaries. On the other hand, processing above 1050 ℃ promotes grain growth ([Figure 2: see original paper]d). After recrystallization is complete, during subsequent grain growth, general high-angle grain boundaries migrate and eliminate existing low ΣCSL grain boundaries, causing the proportion of low ΣCSL grain boundaries to decrease.

A notable feature of grain boundary networks in medium-to-low stacking fault energy fcc metals after GBE processing is the formation of numerous annealing twins. These twins have specific orientation relationships, and when twins on different {111} planes meet, multiple twin boundaries (Σ9 and Σ27, etc.) are derived [21,22]. Therefore, the sum of Σ9 and Σ27 grain boundary proportions should correlate well with the Σ3 twin proportion—when Σ3 proportion is high, the sum of Σ9 and Σ27 proportions is also relatively high. The curves in [Figure 2: see original paper] showing low ΣCSL grain boundary proportion versus annealing temperature reveal that for alloy 825 cold-drawn by 3%, 5%, 7%, and 10% and annealed at different temperatures for 10 min, the variation trend of Σ9 and Σ27 grain boundary proportion sum with annealing temperature is similar to that of Σ3 grain boundary proportion. For samples with 3% cold drawing deformation, both Σ3 proportion and Σ9+Σ27 proportion sum decrease with increasing annealing temperature.

[Figure 2: see original paper]d presents the variation of average grain size in alloy 825 with cold drawing deformation and annealing temperature. Compared to the solution-treated sample, all samples subjected to different cold drawing deformations and annealing treatments exhibited slight grain growth, with grain size decreasing as pre-deformation increased and increasing with annealing temperature. Chen et al. [23] studied the evolution of annealing twins in 95% cold-rolled high-purity Ni during annealing at different temperatures and found that the variation of Σ3 twin boundary proportion during annealing occurs in two stages: during recrystallization, Σ3 grain boundary proportion increases with grain size; however, after recrystallization is complete, during subsequent grain growth, the formation of new small-angle grain boundaries causes Σ3 grain boundary proportion to decrease significantly with further grain size increase. In this experiment, [Figure 2: see original paper]c and d show that for samples with 3%, 7%, and 10% cold drawing deformation annealed at 1050~1125 ℃, the proportion of low ΣCSL grain boundaries decreases with increasing grain size. However, for samples with 5% cold drawing deformation and annealing treatment, the proportion of low ΣCSL grain boundaries did not change significantly despite noticeable grain size increase.

The full development of multiple twins is key to controlling grain boundary network distribution through GBE processing. The higher the (Σ9+Σ27)/Σ3 ratio, the more fully multiple twins develop. [Figure 3: see original paper] shows the (Σ9+Σ27)/Σ3 ratio in alloy 825 after different cold drawing deformations and annealing temperatures. For samples with 3% or 10% cold drawing deformation, the (Σ9+Σ27)/Σ3 ratio gradually decreases with increasing annealing temperature. In contrast, samples with 5% or 7% cold drawing deformation and different temperature annealing treatments exhibit higher (Σ9+Σ27)/Σ3 ratios than those with 3% or 10% deformation. When annealed at 1050~1125 ℃, the (Σ9+Σ27)/Σ3 ratio remains high, indicating that multiple twins develop more fully in alloy 825 tubes when pre-deformation is 5% or 7%.

[Figure 4: see original paper] presents different character grain boundary maps of alloy 825 after various cold drawing deformations and annealing temperatures. The GBE-processed samples contain numerous annealing twins and derived multiple twin boundaries that interconnect to form abundant $\Sigma3^n$-type triple junctions (n = 1, 2, 3, …), creating a microstructure characterized by large-size "grain clusters with $\Sigma3^n$ orientation relationships" [24,25], hereafter referred to as grain clusters, as shown by C1, C2, C3, C4 in [Figure 4: see original paper]a. Within these grain clusters, all grains maintain $\Sigma3^n$ misorientation relationships regardless of whether they are adjacent, forming numerous interconnected $\Sigma3^n$-type triple junctions, primarily Σ3-Σ3-Σ9 and Σ3-Σ9-Σ27. The boundaries between different grain clusters are mainly composed of random grain boundaries.

Grains 1, 2, and 3 in [Figure 5: see original paper] are three arbitrarily selected non-adjacent grains from cluster C1 in [Figure 4: see original paper]a. HKL-Channel5 software can calculate the average Euler angles of each grain in the scanned area, enabling the determination of orientation relationships between any two grains. The orientation relationship is expressed in angle-axis pair form [HKL] and CSL Σ value with deviation angle Σ/Δ , where represents the rotation angle, [HKL] the rotation axis, Σ the reciprocal of coincidence site density, and Δ the maximum deviation angle between the experimentally measured CSL orientation relationship and the standard geometric CSL orientation relationship. Statistical analysis shows that the orientation relationship between grains 1 and 2 is 38.9°[411], Σ81b/0.84°; between grains 1 and 3 is 38.4°[011], Σ9/0.89°; and between grains 2 and 3 is 54.5°[322], Σ81c/0.26° (b and c indicate CSL grain boundaries with the same Σ value obtained by rotating crystal axes through different angles), demonstrating that they all maintain $\Sigma3^n$ misorientation relationships.

Comparing [Figure 4: see original paper]a~d reveals that after small-strain cold drawing and recrystallization annealing at 1050 ℃, the low ΣCSL grain boundary proportion increased to varying degrees, forming large-size "grain clusters with $\Sigma3^n$ orientation relationships." However, grain cluster size varied significantly with different cold drawing deformation levels. [Figure 6: see original paper] shows the grain cluster sizes in samples annealed at 1050 ℃ after 3%, 5%, 7%, and 10% cold drawing deformation. The sample with 3% cold drawing deformation and 1050 ℃ annealing for 10 min exhibited the highest low ΣCSL grain boundary proportion and the largest corresponding grain cluster size, while the sample with 10% cold drawing deformation and 1050 ℃ annealing showed lower low ΣCSL grain boundary proportion and smaller grain cluster size. With increasing pre-deformation, grain cluster size continuously decreased and low ΣCSL grain boundary proportion also decreased, though some $\Sigma3^n$-type grain boundaries remained. Numerous experimental results [21,22,26,27] indicate that grain clusters grow from a single recrystallization nucleus during recrystallization, forming a series of annealing twins and multiple twins during growth. When twins of different generations meet, $\Sigma3^n$ grain boundaries form. After material deformation, the work done by external forces is stored as distortion energy within the material. Upon reheating and annealing, recovery, recrystallization, and grain growth occur. Recrystallization is a process where new strain-free grains replace the entire deformed structure, driven by the stored energy not released during recovery. During GBE processing, samples with smaller deformation have lower stored energy, resulting in lower recrystallization nucleation density and larger potential space for grain growth. For low stacking fault energy fcc metallic materials, annealing twins easily form during growth of new grains. As recrystallized grains grow, they continuously generate annealing twins, forming first-generation, second-generation, and higher-generation twins to constitute long twin chains [21,22,26], thereby creating numerous interconnected $\Sigma 3^n$ grain boundaries. The new grain clusters continuously consume the deformed matrix and grow. When the deformed matrix is completely consumed, recrystallization is complete, finally forming grain clusters of certain size. Consequently, all grains within a cluster maintain $\Sigma 3^n$ orientation relationships regardless of adjacency. However, if deformation is excessive, recrystallization nucleation density becomes high and space for grain growth becomes limited, making it difficult to form large-size grain cluster microstructures and resulting in lower low ΣCSL grain boundary proportions. Therefore, small deformation and high-temperature annealing can produce large-size grain clusters and achieve high proportions of low ΣCSL grain boundaries.

Conclusions

(1) After 5% cold drawing deformation and recrystallization annealing at 1050 ℃ for 10 min, the proportion of low ΣCSL grain boundaries in alloy 825 increased significantly, while simultaneously forming large-size "grain clusters with $\Sigma 3^n$ orientation relationships." The grain size within these clusters was relatively small.

(2) With increasing cold drawing deformation before recrystallization annealing, the grain cluster size decreased, the proportion of annealing twins declined, and the proportion of low ΣCSL grain boundaries also decreased.

(3) For alloy 825 subjected to 5% cold drawing deformation, annealing temperature had minimal effect on grain boundary character distribution. However, for samples with 3%, 7%, and 10% cold drawing deformation, the proportion of low ΣCSL grain boundaries decreased with increasing annealing temperature.

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Note: Figure translations are in progress. See original paper for figures.