Rare Metal Materials and Engineering
Volume 53, Issue 3, March 2024
Available online at www.rmme.ac.cn

Science Press

Cite this article as: Liu Jiwei, Li Xuewen, Fang Wenbin. Abnormal Grain Growth Induced by Stress During Annealing of Industrial Pure Titanium TA1[J]. Rare Metal Materials and Engineering, 2024, 53(03): 625-631. DOI: 10.12442/j.issn.1002-185X.20230080.

ARTICLE

Abnormal Grain Growth Induced by Stress During Annealing of Industrial Pure Titanium TA1

Liu Jiwei[1,2], Li Xuewen[1], Fang Wenbin[2]

[1] Key Laboratory for Light-Weight Materials, Nanjing Tech University, Nanjing 210009, China; [2] School of Materials Science and Chemical Engineering, Harbin University of Science and Technology, Harbin 150080, China

Abstract: The influence of stress on the recrystallization process of pure titanium was investigated. Bending stress was applied during the annealing process of a pure titanium plate to observe the recrystallization process under tensile and compressive stresses on one section. Industrial pure titanium TA1 rolled plates with different deformation levels of 20%, 40%, and 60% were prepared. These samples were then subjected to a temperature of 600 °C and a stress of 30 MPa for 10 min. It is found that in the samples with 20% and 40% deformation levels, only a few recrystallized grains are observed. Additionally, the average grain size in the region under stress becomes larger than that in the region without stress. To further investigate the grain growth, the rolled sheet with 40% deformation level was kept at 600 °C and 30 MPa for 60 and 120 min. It is observed that the abnormal growth of grains in the tensile stress area continues until the critical size is reached, after which they stop growing. The increased grain growth during recrystallization can be attributed to the stress-promoting dislocation adjustment. The grains with favorable conditions tend to grow abnormally along the direction of the applied stress. However, the high density of residual dislocations within the titanium plate results in a reduction in the driving force for grain growth, leading to the existence of critical size. These findings provide an explanation for different recrystallization behavior observed in titanium under tensile and compressive stresses.

Key words: TA1; recrystallization; abnormal grain growth; stress

Industrial pure titanium has a wide range of applications in aerospace, shipping, nuclear energy and other high-tech fields[1–2] and has become a research hotspot in recent years. However, due to the high yield strength ratio of pure titanium, its ability to coordinate deformation during mechanical processing is relatively weak and it is difficult to deform. Therefore, recrystallization is needed to reduce the internal dislocation density of the crystal so as to soften the material before secondary processing. The current research on recrystallization is mainly focused on the microstructure evolution. For example, Miller et al[3] studied the relationship between recrystallization nucleation and grain orientation in Mg-Zn alloys, observed different deformation states of basal and non-basal grains by transmission electron microscopy and explained their different effects on recrystallization nucleation. Zheng et al[4] investigated the effects of different deformations and temperatures on the recrystallization of CoCrFeMnNi high-entropy alloys and established a static recrystallization model for different deformed high-entropy alloys. Sourabh et al[5] explored the coupling effect of stress and temperature on dynamic recrystallization through creep experiments. Zhang et al[6] found that high residual elastic strain still exists in recrystallized grains of single-phase pure iron and investigated the factors influencing the local residual strain. They concluded that this residual elastic strain is caused by the local defect density between the recrystallized grain and the matrix and the redistribution of long-range residual stresses.

In recent years, there is an increased interest in recrystallization and thermomechanical processing. Li et al[7] conducted physical simulations of the dynamic recrystallization behavior and hot workability of 300M steel and proposed a mechanism to study the coupled regulation of recrystallization and hot workability. A large number of kinetic models have been developed to quantify the volume fraction of recrystallization[8–10], while the effect of stress on recrystallization when the critical deformation conditions are

Received date: March 16, 2023
Corresponding author: Li Xuewen, Ph. D., Professor, Key Laboratory for Light-Weight Materials, Nanjing Tech University, Nanjing 210009, P. R. China, Tel: 0086-25-83589160, E-mail: lixuewen2000@163.com

not reached is less studied. Myrjam et al[11] demonstrated that mechanical loading has a significant effect on the recrystallization behavior of aluminum alloys. This system was loaded with an applied tangential stress, the applied stresses were 0.035 and 10 MPa, the incubation time increased, and recrystallization at high stress was almost completely suppressed. Saintoyant et al[12] analyzed the recrystallization mechanism of zirconium alloys under stress conditions. It is found that the application of stress during recrystallization slows down the phase growth. The recrystallization nucleation rate is either accelerated or slowed down under different stress amplitudes and a simple model is proposed as an explanation. For titanium alloys, a lot of thermal deformations and heat treatments have been done to investigate the deformation mechanism, microstructure evolution and strengthening mechanism. Zheng et al[13] concluded that the recrystallization mechanism in near-α titanium matrix composites is related to a strong recovery process. Since the high stacking of dislocations in the α-phase in titanium alloys prevents the decomposition of dislocations and reduces the tendency of discontinuous recrystallization, the mechanism of refinement is considered to be a continuous recrystallization process with merging of subgrain boundaries. Yu et al[14] studied the dynamic recrystallization behavior of bimodal TC17 titanium alloys at high strain rates and concluded that a special recrystallization mechanism of particle stimulated nucleation (PSN) mechanism becomes dominant under high strain rate conditions. The local stress concentration due to the difference in the lattice structure of the two phases prompts a large number of dislocation walls to convert into subgrains, providing favorable conditions for DRX. The above studies on the recrystallization mechanism of titanium alloys show that stress and recrystallization are inextricably linked.

The aim of this study is to investigate the effect of applied stress on the static recrystallization process of industrial pure titanium TA1, which is of great significance for the precise control of the microstructure of titanium alloys. A pure bending experimental design was used to simultaneously observe the effect of compressive and tensile stresses on the recrystallization process in one cross section. EBSD was used to characterize the original microstructure and the recrystallized microstructure, and the experimental results were verified by calculating the geometrically necessary dislocation (GND) density using AZtec data analysis software.

1 Experiment

In this experiment, three pure titanium plates with different deformation amounts were prepared to explore the influence of stress on the recrystallization process. The blanks selected were TA1 commercial pure titanium sheets with original dimension of 100 mm×75 mm×4 mm, which were cut into three plates of 100 mm×25 mm×4 mm by wire-electrode cutting. The plates were then hot-rolled to thicknesses of 3.2, 2.4 and 1.6 mm with deformation levels of 20%, 40% and 60%, recorded as H20, H40 and H60, respectively. Ten equally spaced positions in the length direction were taken to measure the thickness of the rolled plate, the average value was taken as the thickness value and the shortest side length was taken as the length value. The dimensional data of the final three plates are shown in Table 1. The thickness of the rolled plate was uniform, and a long strip with 4 mm in width was cut off the plate for the stress recrystallization experiments.

Fig. 1 shows the experiment design where the slat is placed on the mold and the counterweight is placed in the middle of the slat to apply the load. This experimental design allows for the observation of recrystallization behavior in different stress states in a cross section. By applying bending stress to the specimen, the middle part of the slat is subjected to pure bending stress, while the inner side is subjected to compressive stress and the outer side is subjected to tensile stress. The maximum bending stress can be found from the load P and the spacing between the two sides.

$$\sigma_{\max} = 3Pa\frac{1}{bh^3} \tag{1}$$

where a is the spacing marked in Fig. 1, b is the width of the strip and h is the thickness of the strip. As seen from Eq. (1), stress control can be easily achieved by regulating the load and the spacing between the two sides. It is known from Ref. [15] that the flow stress of pure titanium at 800 °C is approximately 80 MPa. To reduce the effect of high temperature creep, the load set in this study was 30 MPa. The metal strips with 40% deformation were held at 600 °C for 10, 60 and 120 min, and the metal strips with 20% and 60% deformations were held at 600 °C for 10 min, which are recorded as H40-10, H40-60, H40-120, H20-10, and H60-10, respectively.

A small section was cut in the middle of the strip as a specimen for lateral cross-sectional observation by EBSD. To prepare the sample for EBSD observation, it was metallographically polished and then electrolytically polished. The composition of the electrolytic polishing fluid was 6% perchloric acid+34% n-butanol+60% methanol. A TESCAN S800 GMH field emission swept electron microscope was used to obtain the EBSD orientation map.

Table 1 Dimensional data of the final three plates

Plate	Thickness/mm	Variance	Deformation/%	Width/mm	Length/mm
H20	3.352	0.000 107	16.2	24.5	116.1
H40	2.593	0.000 268	35.17	25	148
H60	1.714	0.001 227	57.15	25	215

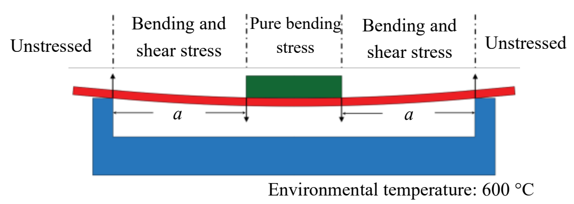

Fig.1 Experimental design diagram

2 Results and Discussion

2.1 Microstructure of as-rolled plates

Fig.2 displays the microstructures of the as-rolled plates. As shown in Fig. 2a – 2c, the larger the deformation, the more elongated the grains, and the more severe the lattice distortion. Additionally, twinning is more frequent in the 20% deformation rolled plate, but rare in the other two deformation rolled plates. The intense deformation of titanium alloys during rolling can lead to uneven deformation, especially in the thickness direction[16]. The kernel average misorientation (KAM) is calculated for each pixel to examine the structural variation in the thickness direction of the rolled plates, as shown in Fig.2d. The KAM of a given pixel is defined as the average misorientation angle between that pixel and its nearest three layers of pixels, where pixels belonging to another grains are excluded. The results show that the deformation degree of the rolled plate is roughly the same in the thickness direction, and the grain size distribution is uniform, indicating that the recrystallization energy storage inside and outside is the same. Fig.2e shows the polar diagram of 20% deformation rolled plate, and it can be seen that the (0001) polar diagram shows a typical cold rolled weave with the crystalline *c*-axis tilted by 30°–40° from the ND towards the TD. At the same time, a new texture component appears on the $(11\bar{2}0)$ and $(10\bar{1}0)$ pole diagrams, probably due to the higher temperature during hot rolling, more slip systems can be initiated, and slip replaces twinning as the main mode of deformation. As the deformation increases, the proportion of twins decreases and the influence of twins on the texture disappears, resulting in a prismatic conical surface texture.

2.2 Calibration of recrystallized grains and grain size statistics

The identification of recrystallized grains in the microstructure is based on the grain orientation spread (GOS) criterion, defined as the degree of deviation of the orientation for each pixel point within the grain from the average orientation of the grain. In different literatures, the cut off angle for GOS is chosen differently, but the common denominator is that grains with GOS less than 1° are regarded as recrystallized grains[17–18]. Fig. 3 shows the GOS maps of samples with different deformations. In this study, grains with GOS less than 1° are classified as fully recrystallized grains, which are labeled as blue, while grains with GOS greater than 1° and less than 5° are labeled as yellow as a reference for the amount of recovery, and those with GOS greater than 5° are labeled as red. The area shares of grains with GOS less than 1°, 1°–5 ° and greater than 5° are represented by G1, G2 and G3, respectively.

Fig.3 shows that recrystallization occurs in the H60 sample, while abnormal growth of grains appears in the H20 and H40 samples. The abnormal growth in the H20 sample is obviously influenced by the surface, so the H40 sample is selected for further annealing experiments. Fig.4 shows the GOS maps of the H40 sample annealed for 10, 60 and 120 min.

It can be seen from Fig. 3 that the larger the deformation, the smaller the grain size of the original structure and the greater the number of recrystallized grains. The distribution of recrystallized grains has a close relationship with stress. According to the experimental design of pure bending stress, the closer the distance to the sides, the higher the level of stress, and the more the recrystallized grains, indicating that stress has a facilitating effect on recrystallization, regardless of the tensile stress or compressive stress. This process is also influenced by the degree of deformation, so the recrystallized grains show a diffused distribution.

The contents of G1, G2 and G3 in Fig. 3 were calculated and plotted in Fig. 5. The proportion of G1 in H20 and H40 samples is very small, and no obvious recrystallization occurs, while the proportion of G1 on both sides of H60 samples is

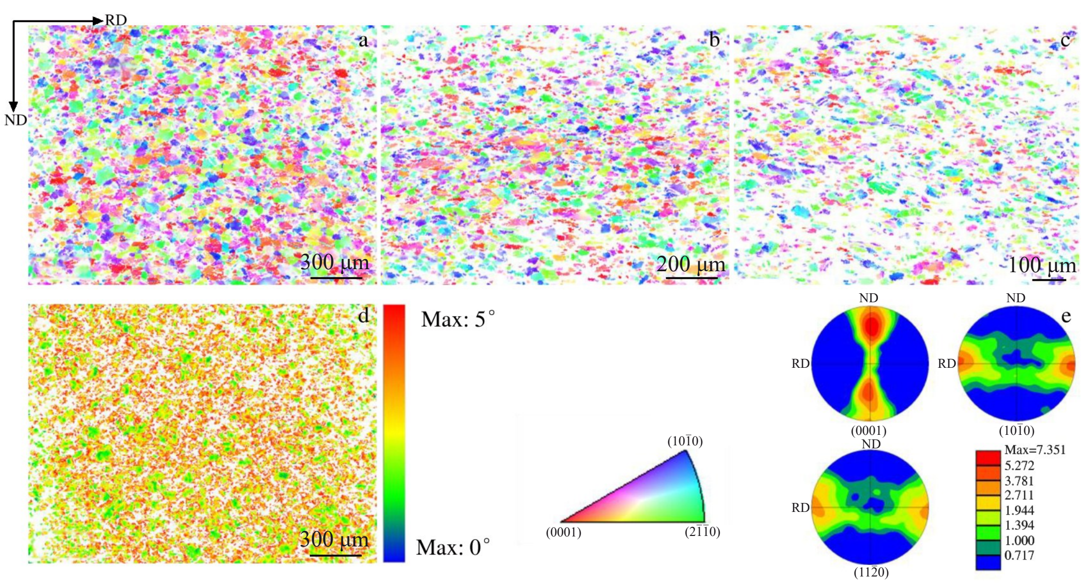

Fig.2 EBSD maps for 20% (a), 40% (b) and 60% (c) deformation rolled plates; KAM map (d); polar plots for 20% deformation rolled plates (e)

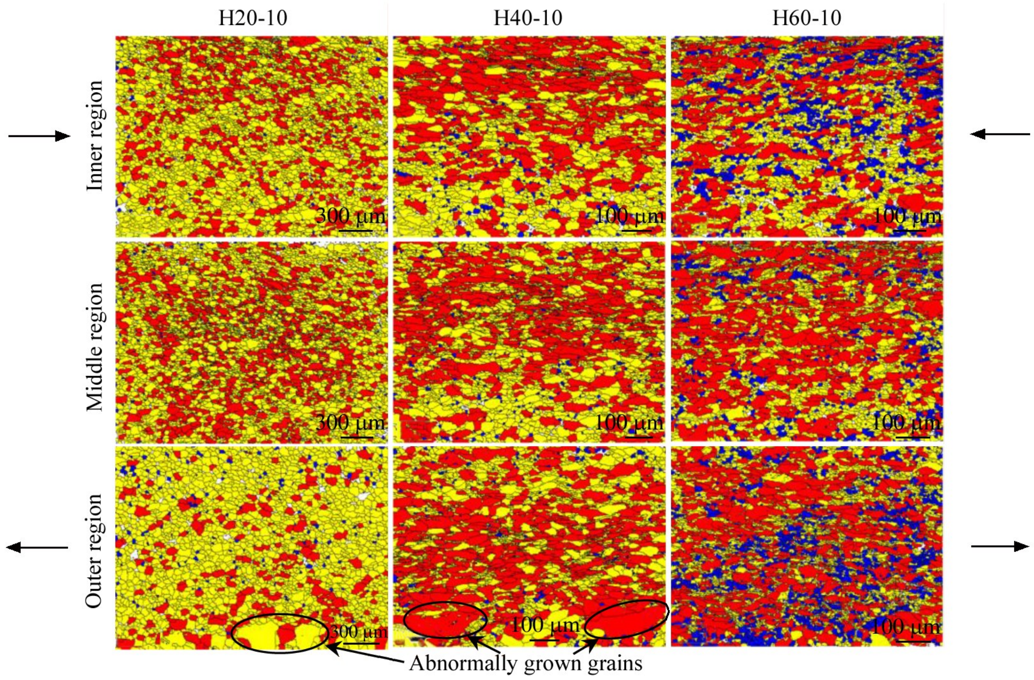

Fig.3 GOS maps of H20, H40 and H60 samples annealed for 10 min in different regions (blue represents G1, yellow represents G2 and red represents G3)

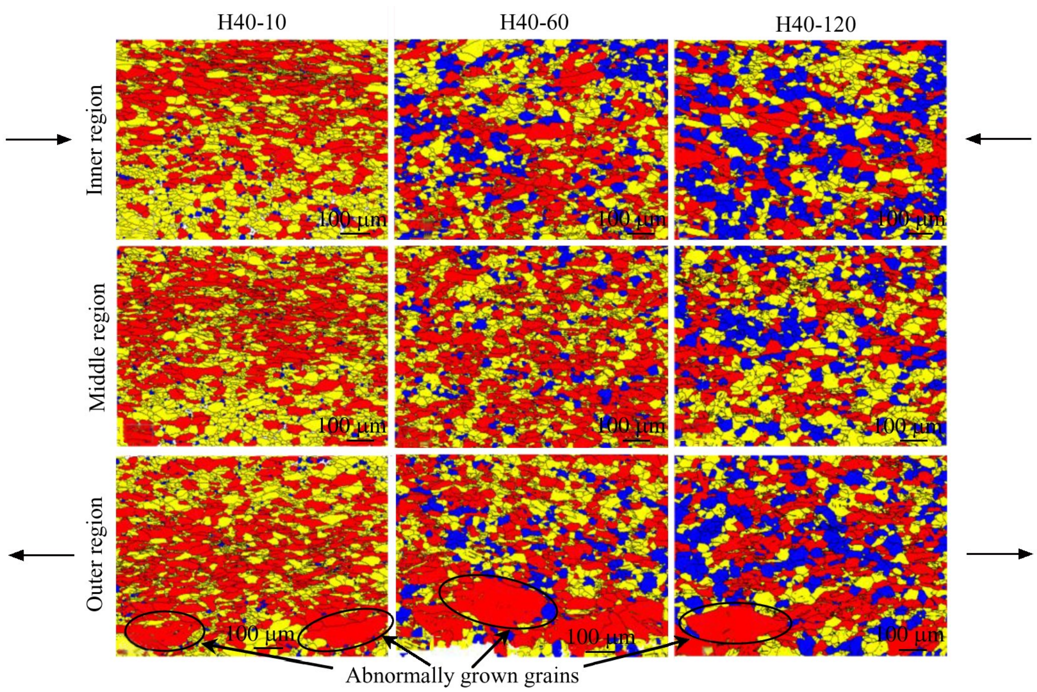

Fig.4 GOS maps of H40 sample annealed for 10, 60 and 120 min

significantly higher than that in the middle region, indicating that the stress promotes recrystallization. G2 represents the area percentage of grains with GOS values at 1° – 5°, indicating that the dislocation density and storage energy inside these grains are higher than those of recrystallized grains, but lower than those represented by G3. As shown in Fig. 5, the proportion of G2 decreases with increasing the deformation. In H20 sample, the grains with GOS in the range of 1°–5° are the main type. This part of the grains releases stored energy by optimizing the dislocation structure through dislocation motion at high temperature.

To quantify the growth of grains in different regions during the annealing process and to exclude the influence of recrystallized fine grains and abnormally grown grains, grains

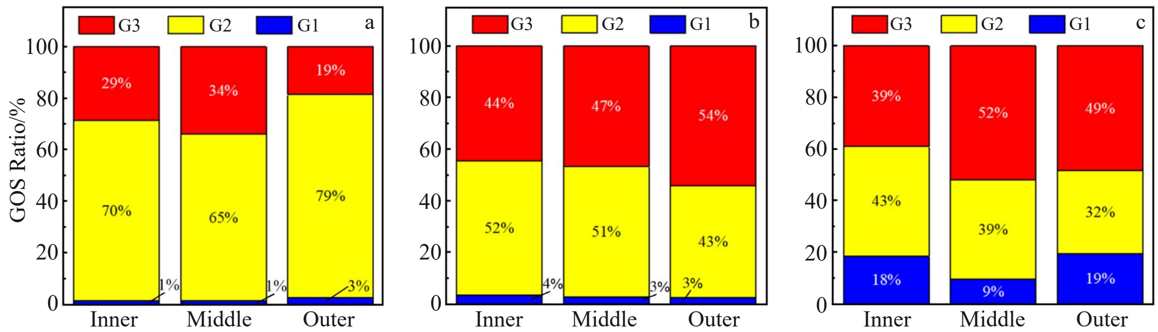

Fig.5 Contents of G1, G2 and G3 in H20-10 (a), H40-10 (b) and H60-10 (c) samples

with sizes of 10–100 μm were extracted from each sample, and then the average grain sizes of three regions were calculated, as shown in Table 2. In general, the grain size increases after annealing. The greater the degree of deformation, the higher the stored energy, and the stronger the DRX behavior. It is noted that the grain size in the stressed region is always larger than that in the intermediate region (Fig. 4 and Fig. 5) of the three samples, regardless of the microstructures after hot rolling. This explicitly indicates that the stress can cause and facilitate the growth of the grain before recrystallization.

In Fig. 4, the H40 sample continues to anneal for 1 h. The diameter of individual grains in the tensile stress region significantly exceeds that of the surrounding grains, and the GOS values remain high, indicating that high storage energy and dislocations still exist within the abnormally grown grains. After annealing for 120 min, such grains remains the same in size, but starts to decrease in number. Again, the contents of G1, G2, G3 in Fig. 4 are counted and plotted in Fig.6. The G1 content in the stress region of the H40 sample is consistently higher than that in the middle region, which confirms the previous findings. The G1 in the tensile stress region is smaller than that in the compressive stress region due to the effect of abnormal grains. The content of G2 in each region also decreases with prolonging the annealing time and it is subsumed by recrystallized grains during the annealing process, so the G2 content on both sides is lower than that in the middle. The tensile stress region may also be subsumed by abnormally grown grains, so the G2 content is even lower.

2.3 Geometrically necessary dislocation distribution

To reveal the grain growth mechanism and the abnormal growth phenomenon in the tensile stress region, the geometrically necessary dislocation (GND) density on the outer side of the H40 sample is calculated using AZtec software and plotted in Fig. 7. A line is also drawn through the abnormally grown grains in the sample to derive the distribution of GND density on the line. The GND density in the as-rolled sample is approximately $10^{16}/m^2$; after annealing, the GND density in the sample gradually decreases. In recrystallized grains, the GND density is close to $10^{13}/m^2$, such as that of the right side of points B and I and the FG segment in Fig. 7. The AB, CD, EF, and HI segments are abnormally grown grains, where internal GND density is the highest near the grain boundary and decreases with increasing the distance from the grain boundary. The lowest GND density can be

Table 2 Average grain size in three regions of H20, H40, H60 samples and the original microstructure (μm)

Location	H20-10	H40-10	H60-10	H20 original microstructure	H40 original microstructure	H60 original microstructure
Inside	34.47	29.48	23.65			
Middle	32.5	28.55	20.07	27.925	20.2697	13.47
Outside	39.33	30.82	23.067			

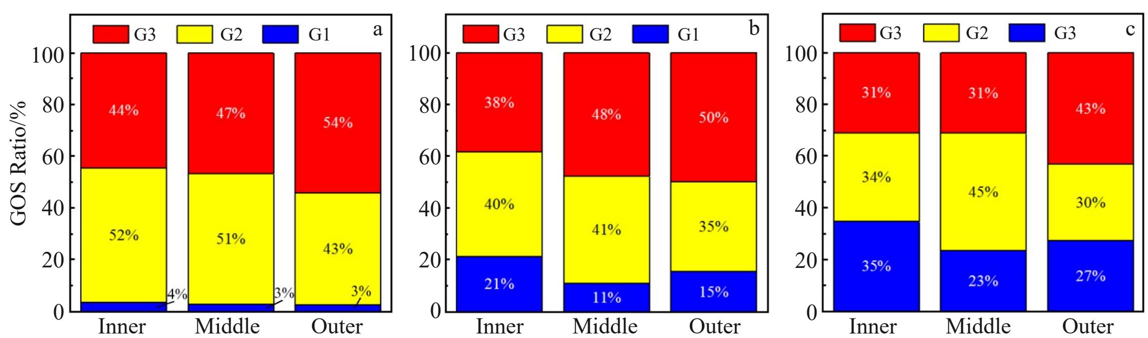

Fig.6 Contents of G1, G2 and G3 in H40-10 (a), H40-60 (b) and H40-120 (c) samples

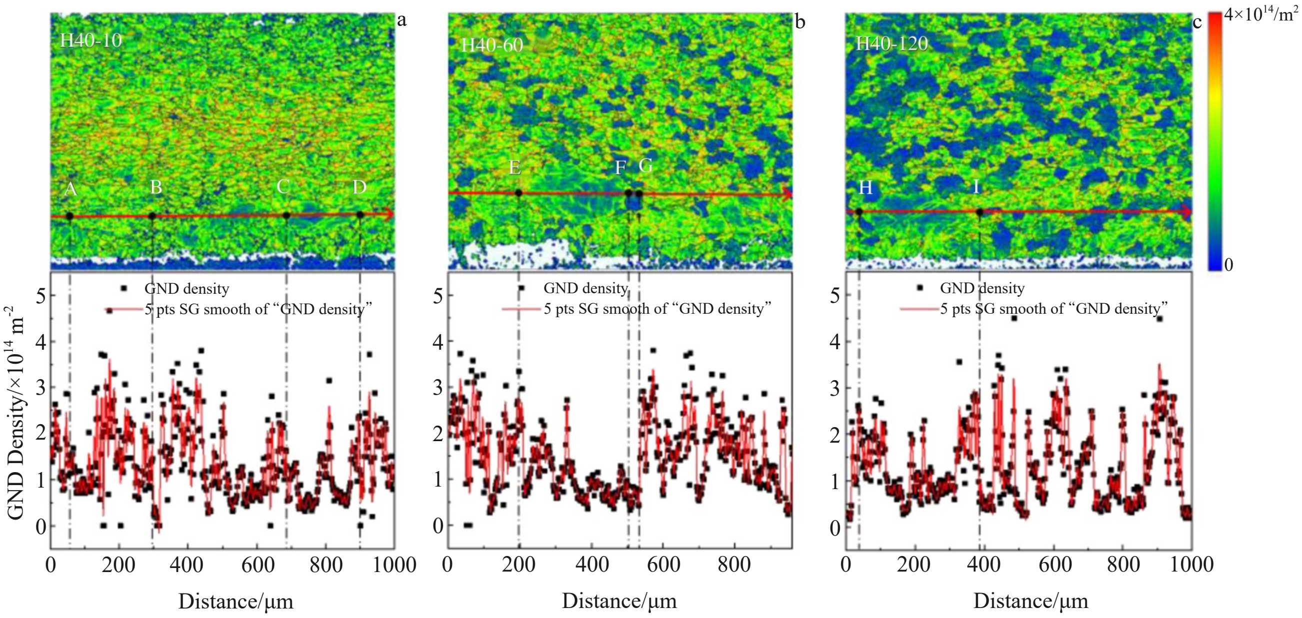

Fig.7 GND distribution of the outer region of the H40 samples with different annealing time: (a) 10 min, (b) 60 min, and (c) 120 min

observed at regions close to the recrystallized grains, and there is GND concentration at the center. The driving force of grain boundary migration is the difference in storage energy on both sides of the grain boundary, which can be explained by the difference in dislocation density. Under high temperature conditions, the stress stimulates the source of vacancies at the grain boundary to produce vacancies, and dislocations will attract vacancies and then climb. The stress also promotes the movement of dislocations and the adjustment of the dislocation structure, resulting in a reduction in the dislocation density inside the grain. Due to the differences in shape, size and orientation between grains, the tendency of dislocations to fall is different for different grains, resulting in differences in dislocation density on both sides of the grain boundary, providing a driving force for grain boundary migration. The thermal movement of dislocations takes time, while as the grain grows, the distance to be swept by the dislocation movement increases, the grain grows slowly and causes dislocations to accumulate at grain boundaries and to stop moving when they become in contact with recrystallized grains.

3 Conclusions

1) The stress applied during the annealing process of pure titanium can cause the grains to grow before recrystallization while promoting the occurrence of recrystallization. The grain size in the stress region is larger than that in the imiddle region in all three samples, and the amount of recrystallization has the same trend.

2) Tensile stress can cause abnormal grain growth. Abnormally grown grains appear on the outside of sample with 40% deformation, and as the annealing time increases, the abnormally grown grains stop growing.

3) The reason for the abnormal grain growth induced by tensile stress is that the stress can improve the dislocation motility while promoting the dislocation structure adjustment and reducing the dislocation density, thus creating dislocation density differences on both sides of the grain boundary and driving the migration of the grain boundary. However, as the grain grows, the difficulty of dislocation movement increases, and the grain growth is slow and eventually stops.

References

1 Schutz R W, Watkins H B. *Materials Science & Engineering A*[J], 1998, 243(1): 305

2 Mohsen Ostad Shabani, Ali Mazahery. *Composites Part B*[J], 2013, 45: 185

3 Miller V M, Nie J F, Pollock T M. *Journal of Magnesium and Alloys*[J], 2022,10(11): 3041

4 Zheng Chaowei, Wang Ying, Jin Junsong *et al*. *Journal of Materials Research and Technology*[J], 2020, 20: 2277

5 Sourabh K, Singh J B. *Materials Characterization*[J], 2022, 194: 112429

6 Zhang Yubin, Yu Tianbo, Xu Ruqing *et al*. *Materials Characterization*[J], 2022, 191: 112113

7 Li Changmin, Huang Liang, Zhao Mingjie *et al*. *Materials Science & Engineering A*[J], 2020, 797: 139925

8 Sellars C M. *Materials Science and Technology*[J], 1990, 6(11): 1071

9 Kim Sung-Il, Lee Youngseog, Lee Duk-Lak *et al*. *Materials Science & Engineering A*[J], 2003, 355: 384

10 Liu Caiyi, Barella Silvia, Peng Yan *et al*. *International Journal of Mechanical Sciences* [J], 2023, 238(15): 107838

11 Myrjam Winning, Carmen Schäfer. *Materials Science & Engineering A*[J], 2006, 419: 18

12 Saintoyant L, Legras L, Bréchet Y. *Scripta Materialia*[J], 2011, 64(5): 418

13 Zheng Zhuangzhuang, Kong Fantao, Wang Xiaopeng *et al. Intermetallics*[J], 2022, 147: 107597

14 Yu Yangbo, Yan Hongge, Zhu Huaming *et al. Materialstoday Communications*[J], 2023, 34: 105255

15 Su Juanhua, Zhou Tiezhu, Ren Fengzhang *et al*. *The Chinese Journal of Nonferrous Metals*[J], 2015, 25(6): 1471 (in Chinese)

16 Zhu Xinjie, Fan Qunbo, Liu Xin *et al*. *Progress in Natural Science: Materials International*[J], 2021, 31: 105

17 Wright S I, Nowell M M, Field D P. *Microsc Microanal*[J], 2011, 17: 316

18 Athreya C N, Suwas S, Subramanya Sarma V. *Materials Science & Engineering A*[J], 2019, 763: 138153

工业纯钛TA1退火过程中的应力诱导晶粒长大行为

刘继伟[1,2]，李学问[1]，房文斌[2]

(1. 南京工业大学 先进轻质高性能材料研究中心，江苏 南京 210009)

(2. 哈尔滨理工大学 材料科学与化工学院，黑龙江 哈尔滨 150080)

摘　要：探究了应力对纯钛再结晶行为的影响。在纯钛板的退火过程中施加弯曲应力，并观察一个截面在拉伸和压缩应力下的再结晶过程。制备了变形量分别为20%、40%和60%的工业纯钛TA1轧制板，然后将这些样品置于600 ℃和30 MPa下保温10 min。结果表明，在变形20%和40%的样品中，只观察到少数再结晶晶粒。此外，受应力区域的平均晶粒尺寸大于无应力区域。进一步将变形40%的轧制薄板在600 ℃和30 MPa下保温60和120 min，拉伸应力区域中晶粒的异常生长一直持续到达到临界尺寸，之后晶粒停止生长。再结晶过程中晶粒生长的持续增加可归因于应力促进的位错调节。具有有利条件的晶粒倾向于沿着所施加的应力方向异常生长。然而，钛板内的高密度残余位错导致晶粒生长的驱动力降低，导致临界尺寸的存在。这些发现解释了钛在拉伸和压缩应力下观察到的不同再结晶行为。

关键词：TA1；再结晶；异常长大；应力

作者简介：刘继伟，男，1998年生，硕士，哈尔滨理工大学材料科学与化工学院，黑龙江 哈尔滨 150080，E-mail：douglasmarker@163.com