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ARTICLE

Microstructure Evolution and Mechanical Properties of Al-Zn-Mg-Cu-Zr Alloy of Heterogeneous Lamellar Structure Processed by High Pressure Torsion

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Abstract: The microstructures and mechanical properties of the Al-Zn-Mg-Cu-Zr alloy processed by high pressure torsion (HPT) at 400 °C were analyzed by transmission electron microscopy (TEM), electron back-scattered diffraction (EBSD), and Instron testing machine. The results show that the grain boundaries and the secondary phases in the grains of deformed specimens are significantly broken and refined, and the width of precipitation-free zones at the grain boundaries narrows, which greatly improves the strength and plasticity of the deformed specimens. The grain orientation of the initial specimen is randomly distributed. When the strain is small, the grain size, grain orientation, and the local orientation difference of the specimen all present heterogeneous lamellar distribution. The deformed specimens of 0.5 turn exhibit the optimal mechanical properties due to the back stress strengthening effect caused by the heterogeneous lamellar structure during deformation.

Key words: Al-Zn-Mg-Cu-Zr alloy; high pressure torsion; heterogeneous lamellar structure; secondary phase; mechanical properties

Due to the attractive properties, such as low density, high strength, ductility, toughness, and fatigue properties, 7XXX series Al-Zn-Mg-Cu alloys are widely used in aerospace and automobile industries[1]. The strength can be further improved by producing ultrafine grains through severe plastic deformation (SPD) techniques, such as high pressure torsion (HPT), equal channel angular pressing (ECAP), and cyclic extrusion compression (CEC) [2-6].

Precipitation strengthening, solid solution strengthening, fine grain (FG) strengthening, and deformation strengthening are the main strengthening mechanisms of Al-Zn-Mg-Cu alloy, among which the precipitation strengthening and fine grain strengthening play important roles[7-9]. However, when the grain size is reduced to a certain scale, it may lead to the decrease of plasticity and the weakening of work hardening effect[10,11]. Therefore, it is difficult to further improve the material properties only through grain refinement. In recent years, different multi-scale structure designs of metal materials have provided a new way to improve the material properties, including gradient structure, dual-phase nano-structure, core-shell structure, and heterogeneous lamellar structure (HLS)[8,12-16]. The heterogeneous lamellar microstructure is composed of fine and coarse lamellar layers, which simultaneously improves the strength and plasticity of materials. Therefore, it is considered to be one of the most effective methods to improve the strength and ductility[17,18]. Belyakov et al[19] indicated that the 316L stainless steel of HLS possesses enhanced strength and ductility. Wu et al[20] prepared Ti of HLS by asymmetric rolling, which realized the comprehensive improvement of mechanical properties. Ma et al[21,22] produced the nanostructured (NS) bronze and coarse grain (CG) copper of HLS by accumulative roll bonding, which improves the ductility greatly. In addition, the existing studies have also shown that HPT is an effective method to prepare heterogeneous lamellar structure, and has stronger ability to refine grains and break secondary phase[23,24].

In this research, the Al-Zn-Mg-Cu-Zr alloy of high Zn content (12wt%) was prepared by multi-pass HPT at 400 °C, and the microstructure evolution and mechanical properties of the deformed Al-Zn-Mg-Cu-Zr alloys were analyzed. The

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research provides a comprehensive investigation of Al-Zn-Mg-Cu-Zr alloy during HPT process and lends supports to the application of SPD techniques in material modification.

1 Experiment

The material used in this study was Al-Zn-Mg-Cu-Zr alloy, whose chemical composition is presented in Table 1. Disk-shaped blanks with dimension of *Φ*100 mm×8 mm were obtained from the alloy plate for subsequent HPT process. As shown in Fig. 1, the disk-shaped specimens were HPT processed by 0.5 and 2 turns at the temperature of 400 °C with a rotation speed of 1 turn per minute. Then, according to T6 temper procedure, the specimens after HPT were solution treated at 473 °C for 120 min, quenched in water at room temperature, and subsequently aged at 135 °C for 16 h. The specimens were cut at the place of 40 mm away from the center of deformed blanks for subsequent microstructure analysis and performance tests.

Optical microscope (OM) and transmission electron microscopy (TEM) were used to observe the microstructure of as-cast and HPT processed Al-Zn-Mg-Cu-Zr alloys.

TEM observations were performed on a FEI TECNAI G2 S-TWIN F20 instrument operated at 200 kV. The specimens for TEM observation were mechanically thinned to about 100 μm in thickness, and then the disk-shaped specimens of 3 mm in diameter were cut out from these specimens. Subsequently, the disk-shaped specimens were thinned by twin-jet electro-polishing until they reached a certain electron transparency. More than 10 TEM images were taken for each specimen to obtain the size of the grain boundary precipitate (GBP) and matrix precipitate (MPt). The width of precipitation-free zone (PFZ) was analyzed by ImagePro software.

Table 1 Chemical composition of as-cast Al-Zn-Mg-Cu-Zr alloy (wt%)

Zn	Mg	Cu	Zr	Ti	Fe	Si	Al
12	3	2.6	0.2	0.05	0.05	0.05	Bal.

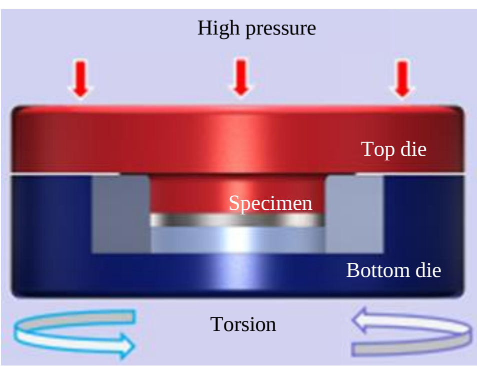

Fig.1 Schematic diagram of HPT process

The JSM-7001F electron back-scattered diffraction (EBSD) operated at a voltage of 20 kV was used to investigate the grain orientation and texture evolution of specimens before and after deformation. Specimens were prepared by cold mounting, grinding, and polishing. The cold mounting was conducted using conductive resin Demotec 70 at room temperature. After mounting, the specimens were ground by 600#, 800#, 1200#, and 4000# SiC papers. An initial polishing was conducted by the Metprep Alpha cloth, diamond polishing solution (1 μm), and water-based lubricant. Then a further polishing was performed by Metpr Alpha cloth and colloidal silica solution (0.04 μm). Finally, the specimens were ion polished for EBSD observation.

Tensile test was conducted at room temperature (~298 K) with an initial strain rate of 1.0×10^{-3} s^{-1} using the Instron testing machine (CMT5105) with optical sensors for strain measurement. Planar dog-bone-shaped tensile specimens were machined to the ones with dimension of 6 mm×2 mm×2.2 mm and polished with a finishing SiC paper of 2000# to remove the defects before testing. At least three specimens were tested under each condition to verify reproducibility.

2 Results and Discussion

2.1 Grain size

The OM images of Al-Zn-Mg-Cu-Zr alloy before and after HPT of 0.5 and 2 turns are shown in Fig.2. The *x*, *y*, and *z* axes represent the cutting, radial, and axial directions of specimens, respectively. The grain size distribution is shown in Fig.3. The microstructure of the as-cast alloy mainly consists of coarse equiaxed grains with grain size of 80~300 μm. After HPT of 0.5 turn, the grains are refined obviously but show inhomogeneous structure, and the grain size ranges from 5 μm to 60 μm. Besides, the microstructure consisting of alternating coarse and fine grained layers is defined as heterogeneous

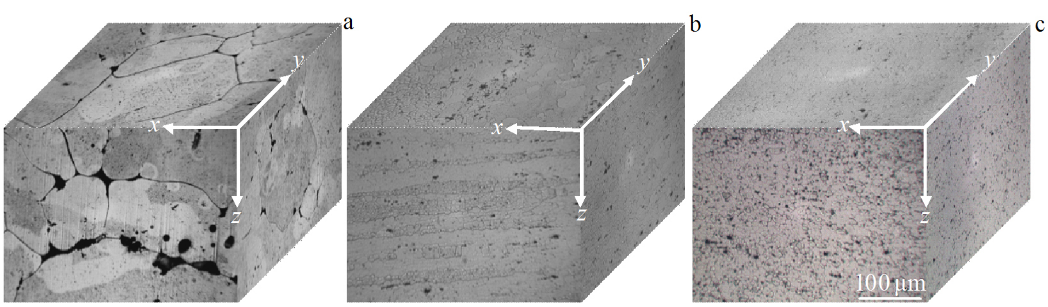

Fig.2 OM images of Al-Zn-Mg-Cu-Zr alloys before (a) and after HPT of 0.5 turn (b) and 2 turns (c)

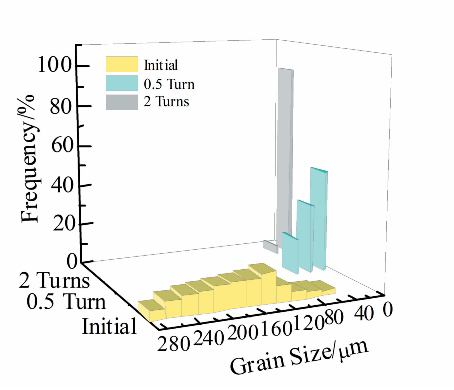

Fig.3 Grain size distribution of Al-Zn-Mg-Cu-Zr alloys before and after HPT process

lamellar structure. After HPT of 2 turns, the grains are continuously refined to 5 μm in size and the uniformity improves.

2.2 Microstructure

Typical PFZ width and size of GBP and MPt of Al-Zn-Mg-Cu-Zr alloys before and after HPT of 0.5 and 2 turns are shown in Fig. 4 and Fig. 5. The microstructure characteristics of all specimens are summarized in Table 2. It can be seen from Fig. 4a and 4d that coarse secondary phases are distributed incessantly along the grain boundaries in as-cast alloy, accompanied by PFZ of 171 nm in width. After HPT of 0.5 turn, the coarse GBP is violently broken, which restrains the formation of PFZ, as shown in Fig. 4b and 4e[25]. With further increasing the torsion extent, the coarse GBP is continuously refined and the uniformity is improved. Besides, MPt becomes denser, GBP is distributed more incessantly, and PFZ widens to near 90 nm, because HPT can promote the precipitation (Fig.4c and 4f).

The size of MPt was obtained from bright-field TEM images. The microstructure characteristics of all specimens are summarized in Table 3.

2.3 Grain orientation

Fig.6 shows the grain orientation of Al-Zn-Mg-Cu-Zr alloys before and after HPT and the misorientation angle distribution by correlated and uncorrelated methods. Different colors represent different corresponding grain orientations: red, blue, and green represent the <001>, <111>, and <101> directions, respectively. The difference of adjacent orientation indicates the angle of grain boundary. However, when the difference of orientation between adjacent and non-adjacent grain boundaries is small, the grain boundaries of different angles are randomly distributed. When the difference is large, they are unevenly distributed. It can be seen from Fig. 6 that the orientation of different grains in the initial alloy is quite different. The orientation difference between adjacent and non-adjacent grain boundaries in the deformed alloys is small, and their microstructure is layered distribution, which is consistent with the results of OM observation. The grains of different sizes are randomly distributed in alloy, and the difference between adjacent and non-adjacent orientations is small. In addition, the orientation difference of grains in the same layer

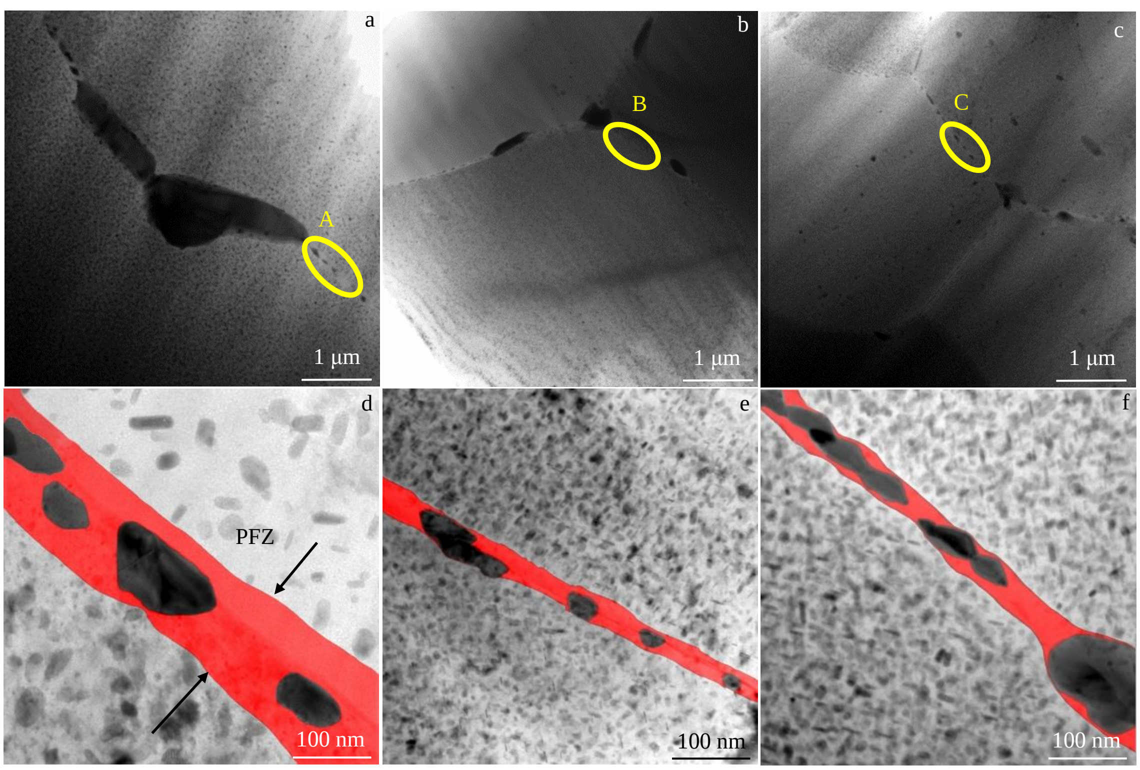

Fig.4 TEM images of Al-Zn-Mg-Cu-Zr alloys before (a) and after HPT of 0.5 turn (b) and 2 turns (c); enlarged images of area A (d), area B (e), and area C (f) in Fig.4a~4c, respectively

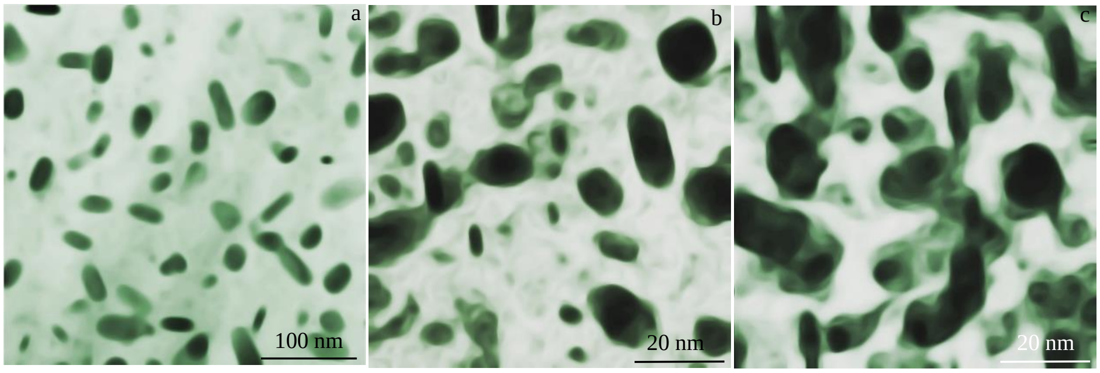

Fig.5 Bright-field TEM images of MPt of Al-Zn-Mg-Cu-Zr alloys before (a) and after HPT of 0.5 turn (b) and 2 turns (c)

Table 2 Grain boundary characteristics of Al-Zn-Mg-Cu-Zr alloys before and after HPT of 0.5 and 2 turns (nm)

Turn	Width of PFZ	Average GBP size
0	171.3	279.7
0.5	53.9	95.1
2	89.8	76.8

Table 3 Size and quantity of MPt of Al-Zn-Mg-Cu-Zr alloys before and after HPT of 0.5 and 2 turns

Turn	Average MPt size/nm	Number of η phase in 1 μm^2	Number of η' phase in 1 μm^2
0	30	153	126
0.5	12	2437	837
2	15	2172	861

is similar, while that between adjacent layers is quite different. The lamellar distribution of grain orientation further leads to a significant increase in the difference between adjacent and non-adjacent orientations, i.e., the grain boundary distribution of different angles is uneven. With the increase of torsion extent, the lamellar structures with different orientations restrict the subsequent deformation, resulting in the fact that the orientation of the whole grain tends to be the same. The phenomenon of layered distribution disappears. The orientation difference among only a small part of the surrounding grains is larger, and this part of the grains are distributed evenly in the microstructure. Meanwhile, the difference between adjacent and non-adjacent orientations in the alloy after HPT of 2 turns decreases, i.e., the uniformity of grain boundary distribution of different angles increases.

Effect of torsion extent of HPT deformation on the textures was investigated by orientation distribution function (ODF). It can be seen from Fig. 7 that the texture of the deformed specimen of 0.5 turn is obvious, but relatively dispersed. With the increase of torsion extent, the texture becomes more concentrated. For orthorhombic systems, the analytical relationship between orientation (φ_1, φ, φ_2) and texture type (hkl)<uvw> obeys Eq.(1) and Eq.(2), as follows:

$$h:k:l=-\sin\varphi\cos\varphi_2:\sin\varphi\sin\varphi_2:\cos\varphi \quad (1)$$

$$u:v:w=(\cos\varphi\cos\varphi_1\cos\varphi_2-\sin\varphi_1\sin\varphi_2):(-\cos\varphi\cos\varphi_1\sin\varphi_2-\sin\varphi_1\cos\varphi_2):\sin\varphi\cos\varphi_1 \quad (2)$$

The calibration results show that the texture types of the specimens after 0.5 turn of deformation are (221)<114>, (001)<010>, and (523)<141>. After 2 turns of deformation, the grain orientation becomes more concentrated, and the main texture type is (111)<121> and part texture type is (111)<112>. In the process of HPT deformation, the grains are rotated by specific and regular deformation forces, and the overall grain orientation is along the direction of force or has a specific angle with the direction of force, thereby revealing the texture characteristics. Due to the random grain orientations in the initial specimen, the transition process of grain orientations to stable state in the subsequent deformation process is also different. After the small deformation of 0.5 turn, the orientation of some grains is closer to the torsion direction, which makes them easier to deform and refine, eventually leading to the formation of heterogeneous lamellar structure. With the increase of deformation, the grains tend to be uniformly refined, and the orientation of grains tends to be similar.

2.4 Local misorientation

Fig. 8 shows the distribution of local misorientation of Al-Zn-Mg-Cu-Zr alloys before and after HPT. The local misorientation represents the change of internal orientation of grains, and reflects the substructure distribution in the grains. It can be seen from Fig.8 that the orientation difference of the initial alloy is small, which is basically below 0.5°. After HPT of 0.5 turn, the local misorientation inside the grain increases obviously, and the maximum misorientation is 3°. Besides, the distribution of local misorientation is not uniform, showing a lamellar appearance. In addition, the local misorientation mainly occurs in the FG layer, which further explains the strain in some layers is much larger than that in other layers, i.e., the FG layers have experienced a much more violent deformation. With the increase of torsion extent, the overall local misorientation does not change obviously, compared with the results of alloy after HPT of 0.5 turn, but the lamellar structure of local misorientation disappears, resulting in a more uniform distribution.

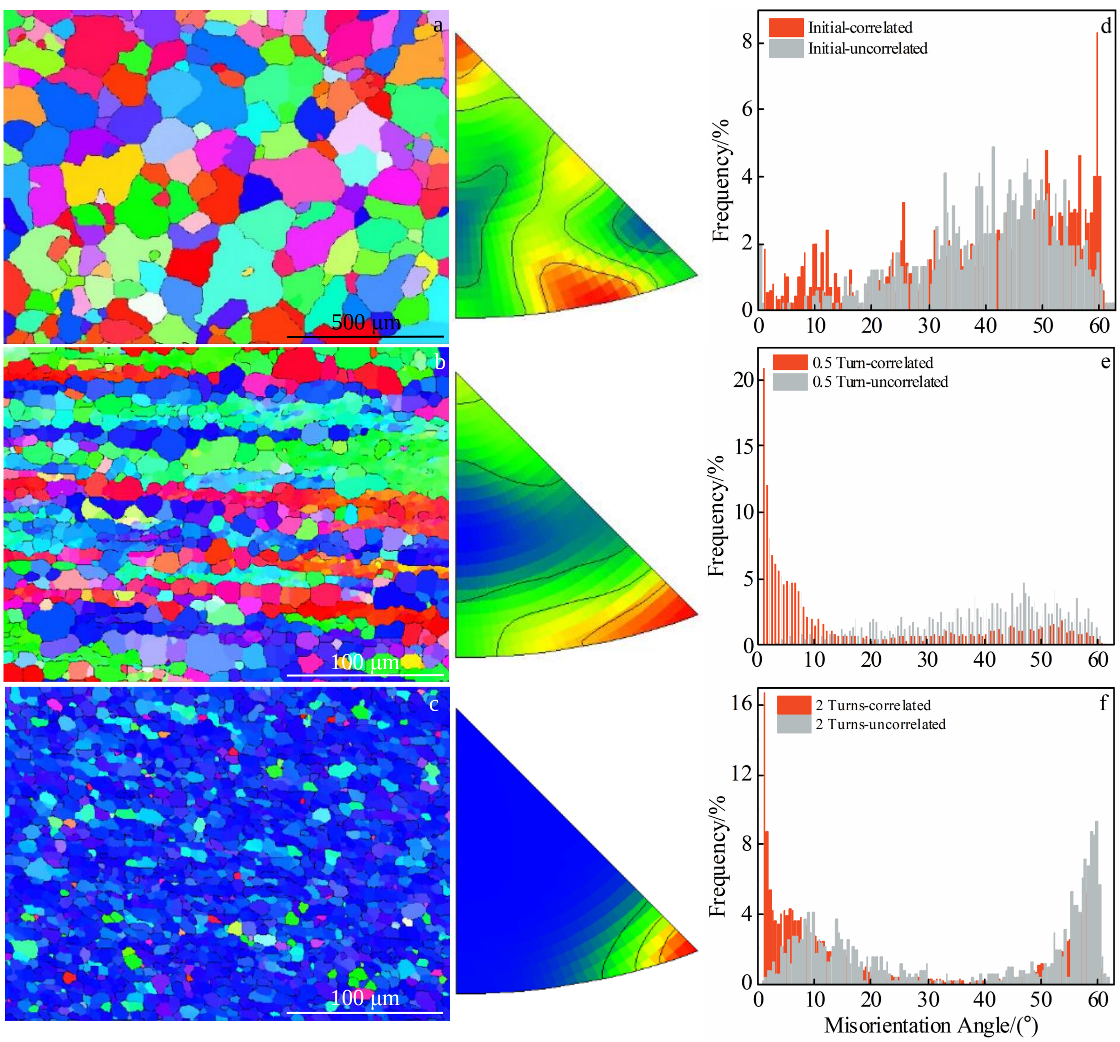

Fig.6 Grain orientations (a~c) and misorientation angle distributions (d~f) of Al-Zn-Mg-Cu-Zr alloys before (a, d) and after HPT of 0.5 turn (b, e) and 2 turns (c, f)

2.5 Mechanical properties

Fig. 9 shows the tensile curves and strain-strain hardening rate curves of Al-Zn-Mg-Cu-Zr alloys before and after HPT process. It can be seen from Fig.9 that the tensile strength and elongation of the initial specimen are 426 MPa and 3.6%, respectively. After HPT of 0.5 turn, the tensile strength of alloy increases obviously to 781 MPa, and elongation increases to 8.5%. After HPT of 2 turns, the tensile strength of alloy is 701 MPa and the elongation is 7.8%. The strength and elongation of the deformed specimen are obviously higher than those of initial specimen for the following reasons. (1) Grain refinement: after HPT, the grains of alloy are obviously refined, while grains with different orientations hinder the slip, and the resistance of dislocation movement at the grain boundary is greater than that within the grain boundary. Therefore, the alloy strength improves. (2) Substructure: the alloy produces many substructures during deformation (Fig. 8), and the substructure hinders the movement of dislocations and plays an important role in strengthening the alloy. (3) Breaking and refinement of the secondary phases: the coarse secondary phase in the initial specimen agglomerates at the grain boundary, which reduces the bonding between the adjacent grains, resulting in the rapid propagation of cracks along the grain boundary during the tensile test[26,27]. Consequently, the obviously broken and refined GBP leads to the increase of mechanical properties. (4) The improvement of dispersion strengthening: the decrease in size and the increase of quantity of MPt bring a better dispersion strengthening effect and thus improve the alloy strength. In addition, due to the lack of the secondary phase strengthening effect in PFZ, the strength of this region is low, thereby promoting the crack propagation. Therefore, the narrowing of PFZ is beneficial to the improvement of mechanical properties. Finally, with all the influence mentioned above, the strength and plasticity of the alloy after deformation are significantly enhanced.

The strength and plasticity of the alloy are significantly improved by HPT, but those of alloy after torsion of 0.5 turn are higher than those of alloy after torsion of 2 turns. It can be seen from Fig.9b that the strain hardening rate of the specimen

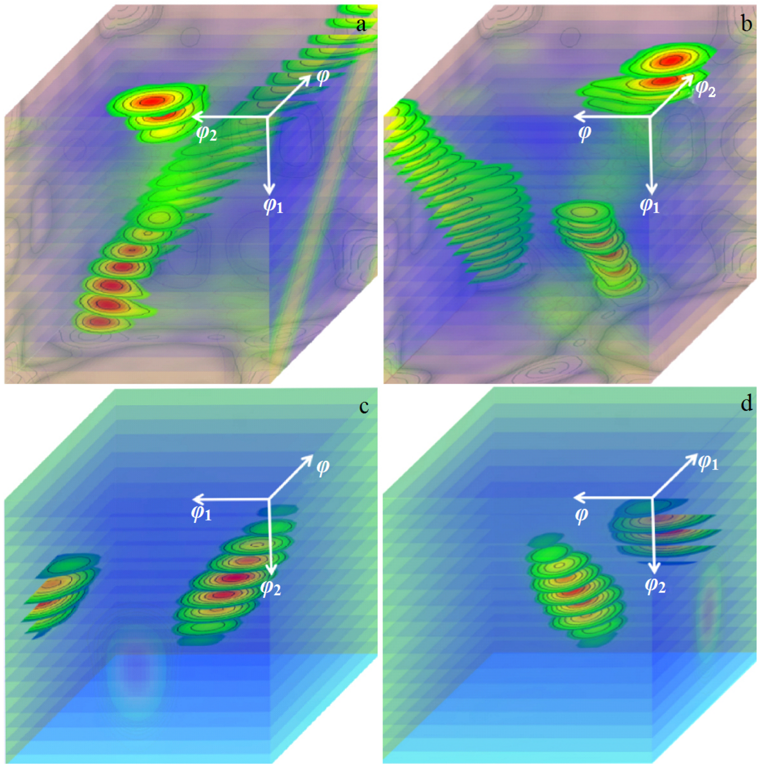

Fig.7 ODFs of Al-Zn-Mg-Cu-Zr alloys after HPT of 0.5 turn (a, b) and 2 turns (c, d)

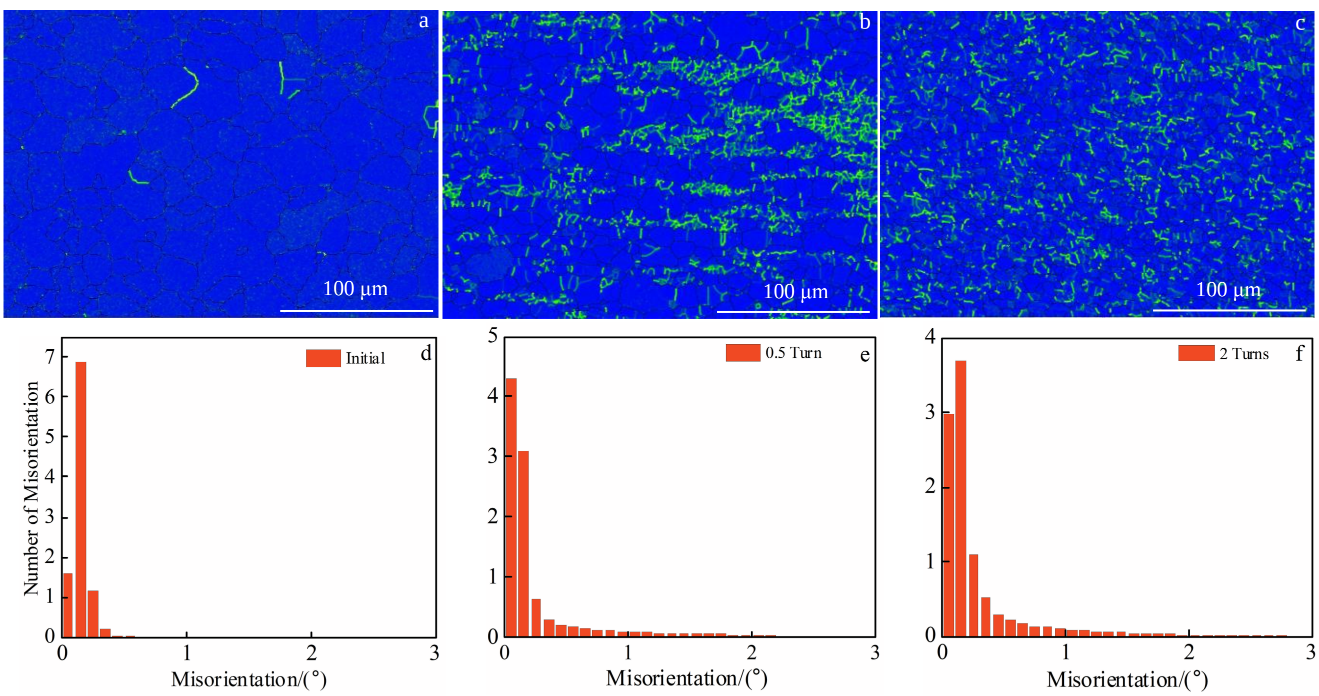

Fig.8 Local orientation difference distribution (a~c) and misorientations (d~f) of Al-Zn-Mg-Cu-Zr alloys before (a, d) and after HPT of 0.5 turn (b, e) and 2 turns (c, f)

after HPT deformation is obviously higher than that of the initial specimen. After the yield stage, the strain hardening rate of specimen after torsion of 0.5 turn is obviously higher than that of specimen after torsion of 2 turns, indicating that the former one has higher strength. The main reason is that the strain strengthening rate of the alloy is increased by the

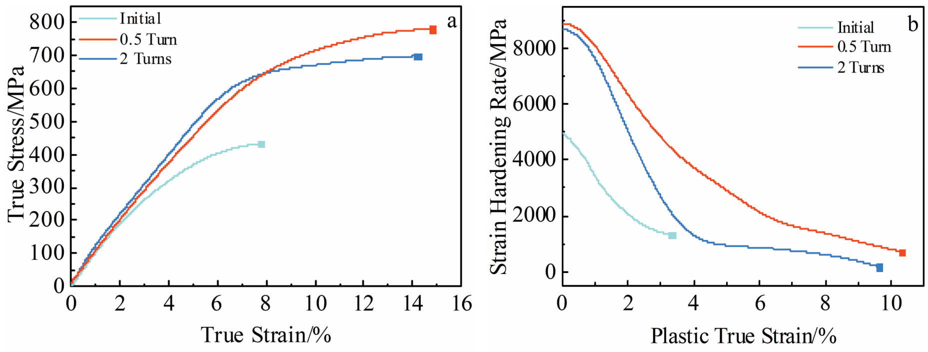

Fig.9 Tensile stress-strain curves (a) and strain hardening rate curves (b) of A1-Zn-Mg-Cu-Zr alloys

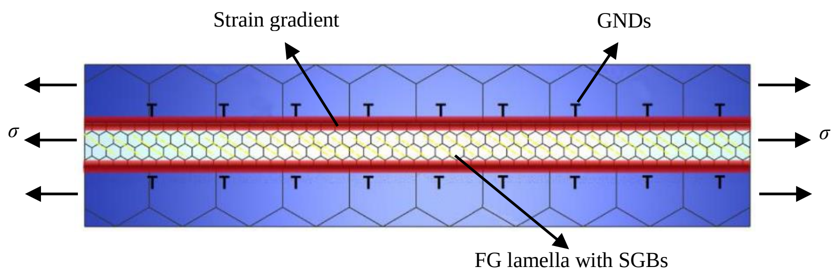

Fig.10 Schematic diagram of strain distribution of heterogeneous lamellar structure during tensile process

heterogeneous lamellar structure.

The existing studies show that the strength and plasticity of heterogeneous lamellar structure are not a simple superposition of the strength and plasticity of CG layer and FG layer. In addition, the common heterogeneous lamellar structure is mainly reflected in the grain size difference between the adjacent lamellae, i.e., the CG layers and the FG layers are arranged alternately. While the heterogeneous lamellar structure obtained by HPT is reflected in not only the difference of grain size, but also the distribution of grain orientation and the substructure.

Based on these analyses, it can be seen that due to different grain sizes and substructure distribution, the CG layer of low strength starts with plastic deformation firstly in the tensile process. However, the CG layer is restricted by the surrounding FG layers for the following reasons: (1) FG lamellae are stronger than the coarse lamellae, as they contain more grain boundaries and sub-grain boundaries (SGBs); (2) the misorientation angles between FG and CG lamellae near the domain boundaries have a significant difference, which makes it more difficult for the dislocations in CG lamellae to migrate to FG lamellae. These two reasons result in plastic strain gradient and the accumulation of geometrically necessary dislocations (GNDs) at the interface which cause long-range back stress and lead to the increase of strength as the back stress counterbalances the applied stress. The strain distribution of heterogeneous lamellar structure during the tensile process is shown in Fig.10. With the increase of strain, both FG and CG lamellae are deformed plastically, and the strain gradient becomes larger with the increase of strain, because the CG lamellae sustain much higher strain than FG lamellae do. The strain gradient improves the backing stress hardening which prevents necking phenomenon during tensile test, thereby improving the ductility. Besides, the geometrically necessary dislocations at the interface of lamellae, the interaction between geometrically necessary dislocations, and the statistically stored dislocations further promote the accumulation of dislocations, thus increasing the strain hardening rate.

3 Conclusions

1) The heterogeneous lamellar structure plays a critical role in improving the mechanical properties of Al-Zn-Mg-Cu-Zr alloy. After high pressure torsion (HPT) process, the grains of alloy are refined and the secondary phases are uniformly dispersed, which greatly improves the properties of the alloy compared with those of as-cast alloy.

2) The heterogeneous lamellar structure formed during the HPT process is mainly reflected in the heterogeneous lamellar distribution of grain size, grain orientation, and substructure. With increasing the torsion extent, the uniformity of microstructure is greatly improved.

3) Due to the influence of heterogeneous lamellar structure, the mechanical properties of alloy after torsion of 0.5 turn greatly improve.

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高压扭转法制备非均匀层状结构Al-Zn-Mg-Cu-Zr合金的组织演变与力学性能

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摘　要：采用透射电子显微镜（TEM）、电子背散射衍射（EBSD）和Instron试验机对试验温度400 ℃下高压扭转变形加工的Al-Zn-Mg-Cu-Zr合金进行组织和力学性能的表征与测试。结果表明，变形试样的晶界和晶粒中的第二相明显被破碎和细化，晶界无沉淀析出带宽度变窄，大大提高了变形试样的强度和塑性。初始样品的晶粒取向是随机分布的。当应变较小时，试样的晶粒尺寸、晶粒取向和局部取向差异均呈现非均匀的片层状分布。由于非均匀层状组织在变形过程中产生的背应力强化效应，0.5圈变形试样的力学性能最好。

关键词：Al-Zn-Mg-Cu-Zr合金；高压扭转；不均匀片层状组织；第二相；力学性能

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