Effect of Pre-deformation on Age Hardening and Microstructure of Al-Mg-Si-Cu Alloy: Postprint

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Abstract

The effects of different deformation amounts on the mechanical properties and microstructure of Al-Mg-Si-Cu alloys prepared by combining deformation and aging were investigated using microhardness testing, tensile testing, EBSD, and TEM. The results indicate that with increasing deformation amount, the hardness of the as-rolled alloy gradually increases; during subsequent aging, the deformed alloys can be further strengthened, but the age-hardening capacity gradually decreases. Grains are gradually elongated into a lamellar structure along the rolling direction, forming numerous subgrain boundaries. At small deformation amounts, the dislocation density within the alloy increases with increasing deformation; at larger deformation amounts, dislocations become tangled and form subgrains. The changes in dislocation configuration induced by deformation significantly affect the precipitation behavior of the alloy, with precipitates gradually evolving from a discrete distribution to a continuous distribution. The continuously distributed precipitates result from the interaction between solute atom precipitation and defect recovery; adjusting the deformation amount and aging process can help prepare aluminum alloys with a good combination of strength and ductility.

Full Text

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Abstract

The 6××× series aluminum alloys Al-Mg-Si-Cu are widely used in the transportation and building industries due to their comprehensive mechanical properties, adequate formability, high corrosion resistance and good weldability. For decades, ultrafine grain structure (UFG) produced by severe plastic deformation (SPD) has been proved to be a promising way in strengthening Al alloy materials. Although this method can guarantee a great improvement in strength, the obtained ductility is always disappointing. Besides, this method has a limitation to fabricate products suitable for practical use. Recently, combining deformation and aging has been proposed to produce high-strength Al alloys. This strategy is very effective in achieving Al alloys with strength-ductility synergy even through conventional producing process, for example, rolling and aging. The strain ratio of deformation is critical in tuning the mechanical properties which could be acquired by the above method. The effect of deformation strain ratio on the age-hardening behaviors and microstructure in Al-Mg-Si-Cu alloy produced by combining cold-rolling and aging are investigated using hardness test, tensile test, EBSD and TEM in this work. The results show that the as-rolled hardness increases gradually with deformation strain ratio. The age-hardening potential declines with the increase of strain ratio, though post-aging could further strengthen the as-rolled alloys. The grains elongate along the rolling direction during deformation and finally have a lamellar structure. Fragmentation and extensive defects like sub-grain boundaries occurs inside the grains. The dislocations become denser inside the alloy with the increase of the deformation ratio. When the deformation ratio is large (above 60%), formation of dislocation tangling and sub-grains are observed. Deformation-induced change of the dislocation configuration affects the precipitation significantly. Due to the interaction between solutes precipitation and defects annihilation, the distribution of precipitates undergoes a change from being isolated to a continuous manner.

KEY WORDS aluminum alloy, deformation, aging, dislocation, precipitation

Introduction

Compared with other commonly used aluminum alloys, 6××× series Al-Mg-Si-Cu alloys are widely used in the automotive and aerospace industries due to their light weight, good mechanical properties, formability, and weldability

[1,2]. However, the strength of these alloys is relatively low, and improving their strength has been a common concern for researchers. Traditional methods for obtaining high strength mainly rely on precipitation strengthening and solid solution strengthening. Over the past decade, several techniques collectively known as severe plastic deformation (SPD) have emerged [3~8], such as equal-channel angular pressing (ECAP), high-pressure torsion (HPT), accumulative roll-bonding (ARB), cryogenic rolling, dynamic plastic deformation (DPD), and high-ratio differential speed rolling (HRSDR). These methods can improve the strength of metallic materials by producing ultrafine-grained (UFG) structures and have become promising approaches for fabricating high-performance aluminum alloys [9~12]. However, these methods significantly reduce the ductility of materials while improving their strength [13~16]. Additionally, SPD methods have limitations on sample size and involve complex processing, which has prevented their widespread application in industrial production of aluminum alloys.

The combination of deformation and aging is currently one of the most promising methods for improving the strength of aluminum alloys [17~21]. This approach not only significantly increases material strength but also maintains good elongation and is suitable for industrial production. Researchers [22] have performed pre-aging treatment before cold rolling, followed by aging treatment after rolling, producing Al-Mg-Si-Cu alloys with significantly improved strength compared to traditional T6 treatment while maintaining the same elongation. For alloys without deformation, subsequent aging is equivalent to traditional T6 treatment, where the precipitation process is mainly controlled by bulk diffusion of solute atoms. Aluminum alloys prepared by combining deformation and aging utilize both crystal defects and precipitates to strengthen the material, and the precipitation process during subsequent aging is affected by deformation. Therefore, understanding the effect of deformation amount on precipitation during subsequent aging is crucial for elucidating the mechanism of excellent strength-ductility combination in aluminum alloys prepared by deformation and aging. This work investigates the evolution of microstructure and internal defects in Al-Mg-Si-Cu alloys and their influence on precipitation during subsequent aging by controlling the deformation amount during rolling, which helps to understand the reason for the good combination of strength and ductility in Al-Mg-Si-Cu alloys prepared by deformation and aging and provides guidance for process optimization.

Experimental

The original material used in this study was a hot-rolled Al-Mg-Si-Cu alloy plate with a chemical composition (mass fraction, %) of: Mg 0.75, Si 0.75, Cu 0.8, and Al balance. The contents of Fe, Cr, and Mn were controlled below 0.12% to exclude the influence of excessive second-phase particles. The alloy was first solution-treated at 560 °C for 30 min, water-quenched, and then naturally aged for 1 day as a pre-treatment. Subsequently, cold rolling was performed at room temperature, with the plate thickness reduced from 5 mm to 4.75, 4.5, 4, 3, 2, and 1 mm, corresponding to deformation amounts (thickness reduction) of 5%, 10%, 20%, 40%, 60%, and 80%, respectively. For samples with deformation amounts greater than 10%, the reduction per pass was 10%, while for others it was 5%. Finally, the cold-rolled samples were subjected to post-aging treatment at 120, 150, and 180 °C in an oil bath furnace. For comparison, a traditional process was also applied to the rolled plates, where the plate was directly rolled from 5 mm to 1 mm with 10% reduction per pass, then solution-treated at 560 °C for 30 min, water-quenched, and subjected to T6 treatment at 180 °C (the temperature at which Al-Mg-Si-Cu alloy exhibits maximum peak-aging strength).

Vickers microhardness testing was performed on the rolling plane of the alloys using an HXD-1000T hardness tester with a load of 4.9 N and loading time of 10 s. At least 7 points were measured for each sample to obtain the average hardness value. Tensile testing was conducted using an Instron 3369 testing machine. Tensile specimens were machined according to the ASTM E517-00 standard, with the gauge length cut parallel to the rolling direction. Room-temperature tensile tests were performed at a constant speed of 2 mm/min until fracture. For microstructural observation, alloy samples were mechanically ground with sandpaper and then electropolished in an electrolyte with a volume ratio of 1:3 nitric acid to methanol. Electron backscatter diffraction (EBSD) observations were carried out using a Quanta 200 scanning electron microscope (SEM), and transmission electron microscopy (TEM) was performed using a Tecnai F20 microscope to examine the microstructure. High-resolution TEM (HRTEM) was used to observe the crystal structure of precipitates along the <001> direction of the Al matrix. Thin foils for TEM observation were first mechanically polished and punched, then electropolished in an electrolyte at -30 °C with a voltage of 15 V.

Results

2.1 Age-Hardening Characteristics

The age-hardening curves of Al-Mg-Si-Cu alloys with different deformation amounts at various temperatures are shown in Figure 1 [Figure 1: see original paper]. As the deformation amount increases, the hardness of the as-rolled samples gradually increases. During subsequent aging, the hardness of all deformed alloys can be further improved, and the aging hardening kinetics accelerate with increasing aging temperature.

At 120 °C, the hardness of the alloys increases with aging time, but the increase is relatively slow, and the hardness still shows a tendency to increase even after 480 h. Compared with 120 °C, the time to reach the peak hardness is significantly shortened at 150 °C. As the deformation amount increases, the time to reach the peak becomes shorter, while the difference between the peak value and initial value decreases. For samples with deformation amounts less than 60%, the hardness changes little with further aging after reaching the peak, forming a hardness plateau. However, for samples with deformation amounts greater than 60%, the hardness decreases with prolonged aging after reaching the peak.

At 180 °C, dislocation recovery occurs, and the hardness of all deformed alloys decreases slightly within the first few minutes, then increases rapidly to reach a peak. Larger deformation amounts result in shorter times to reach the peak. Additionally, Figure 1c shows that the peak hardness of all deformed alloys after aging at 180 °C is higher than that of the traditional T6-treated sample at the same temperature, indicating that even small deformations have a positive effect on hardness.

2.2 Tensile Properties

Since the hardness of alloys aged at 120 °C still shows an upward trend even after 480 h, tensile tests were only performed on peak-aged alloy samples at 150 and 180 °C, with the results shown in Figure 2 [Figure 2: see original paper]. For alloys aged at different temperatures, both yield strength and tensile strength gradually increase with deformation amount. The uniform elongation is between 6% and 8% when the deformation amount exceeds 10%, and slightly lower when the deformation amount is less than 10%. At 180 °C, when the deformation amount is 20%, the tensile strength (355 MPa) is comparable to that of the traditional T6 process (361 MPa). When the deformation amount reaches 80%, the tensile strength is 450 MPa, representing a 24.7% increase compared to the T6 condition, while the uniform elongation is 7.1%. Although this is slightly lower than that of the T6 treatment (9%), the ductility is well maintained. The same phenomenon occurs at 150 °C. As the deformation amount increases, deformation defects gradually accumulate, and the recrystallized structure becomes increasingly refined after heat treatment, leading to enhanced grain refinement strengthening. At larger deformation amounts, the subgrain structure becomes more abundant and can strengthen the material, resulting in increased alloy strength with deformation amount.

2.3 Grain Structure

Figure 3 [Figure 3: see original paper] shows EBSD images of the Al-Mg-Si-Cu alloy in the as-rolled state with different deformation amounts, sampled from the normal direction-rolling direction (ND-RD) plane. At small deformation amounts (5%), the alloy exhibits a near-equiaxed grain structure with an average grain size of about 41 m. As deformation increases, grains become elongated along the rolling direction under rolling stress, the grain width decreases, and grain fragmentation occurs. The sample texture gradually transforms into a typical rolling texture. Texture analysis reveals that the texture components are consistent with the rolling plane, mainly consisting of B, C, and S textures [23], which increases the Taylor factor of the alloy and requires greater stress for yielding under tensile stress, representing one reason for the gradually increasing yield strength with deformation amount. Meanwhile, the orientation inside the grains changes. At large deformation amounts (80%), micron-sized subgrain structures exist within grains, resulting from dislocation interaction and rearrangement during recovery. The black areas in the images represent bad points in the collected data, mainly caused by crystal defects induced by deformation. As the deformation amount increases, the black regions in the alloy increase significantly, indicating that the density and distribution area of crystal defects gradually increase. Since the subsequent aging temperature is far below the recrystallization temperature of the alloy, the grain structure after aging is similar to that in the as-rolled state [21~23], and the difference cannot be distinguished in EBSD images.

2.4 Subgrain Structure and Precipitates

The strengthening effect of Al-Mg-Si-Cu alloys is closely related to dislocation structure and the size, distribution, and type of precipitates. Figure 4 [Figure 4: see original paper] shows TEM images of Al-Mg-Si-Cu alloys with different deformation amounts after aging at 150 °C. When the deformation amount is less than 20%, the alloy contains high-density randomly distributed dislocations. As the deformation amount increases, the dislocation density gradually increases. When the deformation amount reaches 20%, the dislocation configuration begins to change: randomly distributed dislocations start to tangle together, forming dislocation tangles or cell structures, and even subgrains. Due to the increase in dislocation density and the formation of dislocation cells and subgrains, the dislocation strengthening effect becomes increasingly significant, which is the main reason for the increasing alloy strength with deformation amount.

Figure 5 [Figure 5: see original paper] shows dark-field TEM images of precipitates in Al-Mg-Si-Cu alloys with different deformation amounts after peak-aging at 150 °C. The precipitate morphology changes significantly with deformation amount. At 5% deformation, the precipitates consist mainly of discrete precipitates distributed in the matrix and a small number of continuous precipitate particles on dislocations. When the deformation amount increases to 10%, the continuous precipitate particles on dislocations increase, and the size of precipitates in the matrix begins to decrease. At 20% and 40% deformation, the precipitates in the matrix are significantly refined compared to small deformations, and the length of continuous precipitate particles on dislocations decreases, with fine continuous precipitates beginning to appear. At 60% deformation, discrete precipitates continue to decrease while fine continuous precipitates gradually increase. At 80% deformation, a large number of nanoscale continuous precipitates form in the sample. The continuous precipitates likely form preferentially at dislocation cell boundaries or subgrain boundaries. Due to the combined effect of precipitation strengthening and crystal defect strengthening, the sample strength gradually increases with deformation amount.

Discussion

The Al-Mg-Si-Cu alloy undergoes short-term natural aging before deformation. During cold rolling, solute atoms or extremely fine clusters can hinder dislocation motion to some extent, resulting in work hardening. At small deformation amounts, uniformly distributed slip dislocations exist in the Al matrix. As deformation increases, dislocation density gradually increases and dislocations begin to interact and tangle. With further deformation increase, dislocation tangles transform into three-dimensional cellular structures. If dislocations accumulated at cell boundaries partially annihilate, they can transform into subgrain boundaries during continued deformation. Under severe plastic deformation, nanocrystalline grains can even form through dynamic recrystallization in aluminum alloys. This study employed conventional cold rolling deformation. Based on EBSD and TEM observations, the original coarse grains generate numerous defects and undergo fragmentation during elongation. The general microstructural evolution process is as follows: randomly distributed dislocations evolve into tangled configurations, then into cellular structures, and finally into subgrain boundaries.

The change in dislocation configuration inevitably affects microstructural evolution during subsequent aging, and precipitate morphology also changes with deformation amount. Figure 6 [Figure 6: see original paper] shows HRTEM images of precipitates in Al-Mg-Si-Cu alloys under different deformation amounts after peak-aging at 150 °C. Figures 6a and b display HRTEM images of two types of discrete precipitates. In Figure 6a, a parallelogram outlines the unit cell of a needle-shaped precipitate with circular cross-section, revealing a monoclinic structure. Fast Fourier transformation (inset in Figure 6a) further confirms the monoclinic structure with lattice parameters of a = 1.49 nm, b = 0.66 nm, and $\gamma = 105°$, which matches the unit cell parameters of the b phase [24]. Figure 6b shows a precipitate with rectangular cross-section. Although its crystal structure cannot be identified, its shape suggests it is an L or Q' phase [25]. Figure 6c shows continuously distributed granular precipitates on dislocations that are not interconnected. Figure 6d shows an HRTEM image of a typical continuous precipitate, where arrows indicate a misalignment angle between crystals on both sides of the precipitate. Such precipitates likely form through heterogeneous nucleation at dislocation cell boundaries or subgrain boundaries. At low dislocation densities (<40%), the alloy contains mainly needle-shaped phases, L phases, and granular continuous precipitates, all relatively coarse. As deformation amount increases, the size of L phases and needle-shaped phases dispersed in the matrix gradually decreases, and continuous precipitate particles on dislocations also refine. At large deformation amounts (>40%), continuous precipitates increase and are no longer composed of continuous granular precipitates. Similarly, as deformation amount increases, continuous precipitates become increasingly refined. At 80% deformation, this continuous precipitation phenomenon is not visible at lower magnifications, but higher magnifications reveal numerous fine precipitates growing along interfacial defects.

The mechanical properties of the Al-Mg-Si-Cu alloy prepared in this work result from the synergistic effect of deformation and aging. Solute atoms can affect the multiplication and evolution of defects during deformation, while defect annihilation during aging can influence the precipitation process of solute atoms. Generally, larger deformation amounts result in more stored dislocations and higher strength, which is confirmed by our experimental results. However, the presence of defects also reduces material ductility. This work found that Al-Mg-Si-Cu alloys still exhibit considerable uniform elongation even under large rolling deformations. At small deformation amounts, precipitation is mainly dominated by bulk diffusion, with accelerated precipitation kinetics but little difference in strength compared to T6-treated alloys. Meanwhile, residual dislocations cause some reduction in elongation. At large deformation amounts, the interaction between precipitation and crystal defects is significantly enhanced, and the nucleation and growth of precipitates are strongly influenced by defect configuration, leading to significant changes in precipitate characteristics. Therefore, there likely exists a critical deformation amount above which the interaction between precipitation and defect reorganization can maintain defect strengthening while reducing its impact on ductility.

Conclusions

(1) With increasing deformation amount, the hardness of Al-Mg-Si-Cu alloy gradually increases. Although all deformed alloys can be further strengthened during subsequent aging, the age-hardening capacity gradually decreases. At small deformation amounts, the achievable peak hardness is slightly higher than that of traditional T6 treatment, while at large deformation amounts, the hardness is significantly higher than that of peak-aged T6 samples.

(2) With increasing deformation amount, the yield strength and tensile strength of the alloy gradually increase, while the elongation first decreases and then increases. Even at large deformation amounts, the Al-Mg-Si-Cu alloy with significantly improved strength still exhibits good elongation.

(3) Deformation amount has a significant effect on the microstructure of the alloy. Grains are gradually elongated into a lamellar structure along the rolling direction, with fragmentation occurring inside grains and numerous subgrain boundaries forming. At small deformation amounts, the dislocation density in the alloy increases with deformation; at large deformation amounts, dislocations tangle and form subgrains. Deformation-induced changes in dislocation configuration significantly affect precipitation characteristics, with precipitates evolving from discrete distribution to continuous distribution. The continuously distributed precipitates result from the interaction between solute atom precipitation and defect annihilation. By adjusting deformation parameters and aging processes, aluminum alloys with a good combination of strength and ductility can be prepared.

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