

Article

Effects of Friction Stir Additive Deposition Process Under Different Cooling Conditions on the Microstructure and Mechanical Properties of 2195 Al-Li Alloy

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Abstract

This study investigates the effects of air cooling and water mist cooling on the microstructure and mechanical properties of friction stir additive manufactured Al-Cu-Li alloy. The results show that under air cooling, coarse grains (average 15.37 μm) form, accompanied by coarse θ phase (Al_2Cu), S phase (Al_2CuMg), and Fe-containing brittle intermetallic compounds. A strong texture is observed ($P_{\text{max}} = 6.45$), and the tensile fracture surface exhibits a mixed ductile–brittle fracture mode with the coexistence of cleavage facets and dimples. The tensile strength, yield strength, and elongation are 325 MPa, 165 MPa, and 21%, respectively. Water mist cooling significantly refines the grain size (average 5.15 μm), weakens the texture ($P_{\text{max}} = 5.41$), suppresses the formation of detrimental Fe-containing phases, and promotes the precipitation of fine, dispersed θ phase and T1 phase (Al_2CuLi) (~ 200 nm). The fracture surface transforms into a uniformly dimpled ductile fracture. Mechanical properties are simultaneously improved: tensile strength reaches 348 MPa (+7%), yield strength 182 MPa (+10%), and elongation 24.5% (+17%). Rapid cooling achieves a synergistic optimization of strength and ductility through grain refinement strengthening, precipitation strengthening, and elimination of harmful phases.

Keywords: Al-Li alloys; additive friction stir deposition; mechanical properties; microstructure

1. Introduction

With the rapid development of the aerospace field, the performance requirements for materials have become increasingly stringent. Materials are not only required to possess high specific strength and high specific stiffness, but also need to exhibit excellent fatigue resistance and corrosion resistance. Against this backdrop, the third-generation aluminum–lithium (Al-Li) alloys have emerged as outstanding candidates, with 2195 aluminum–lithium alloy (Al-Cu-Li) becoming a focal point of research for numerous scientists [1,2]. The addition of lithium significantly alters the properties of aluminum alloys. Compared to traditional 2XXX and 7XXX series alloys, aluminum–lithium alloys are stronger, lighter, and possess higher strength. Specifically, for every 1% (by weight) increase in lithium, the alloy's density decreases by approximately 3%, while its elastic modulus increases by 6% [3,4]. However, the high volatility of lithium and its narrow solidification temperature range pose challenges. When processing 2195 aluminum–lithium alloy using traditional fusion manufacturing methods or fusion additive manufacturing techniques, such as laser powder bed fusion (LPBF) and wire arc additive manufacturing (WAAM), the alloy is highly prone



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to solidification-related defects, including hot cracking, porosity, and micro-compositional segregation [5,6].

Additive Friction Stir Deposition (AFSD) is a groundbreaking solid-state additive manufacturing technology that operates at temperatures significantly lower than the material's melting point [7,8]. Unlike traditional fusion methods, AFSD relies on frictional heat generation and intense plastic deformation to achieve material joining, effectively avoiding common solidification defects [9,10]. According to research by Yu and colleagues [11], through the continuous dynamic recrystallization (CDRX) mechanism, AFSD can form dense, fine-grained equiaxed deposited layers. For 2195 alloy, the solid-state nature of AFSD helps maintain the stability of its chemical composition, making it a strong candidate for manufacturing large structural components. However, the thermal history during the AFSD deposition process has a profound impact on the alloy's final properties. Since AFSD is conducted in ambient air and involves substantial heat input along with a relatively slow cooling rate, this often leads to recrystallized grain coarsening and the dissolution or over-aging of strengthening precipitates (such as the T_1 phase in aluminum-copper-lithium alloys) [12]. Previous studies have shown that although AFSD can eliminate porosity, due to the inevitable thermal cycling, the strength of precipitation-hardened aluminum alloys in the "as-deposited" state is typically lower than that of the raw material in its peak-aged condition [13,14]. Therefore, controlling the thermal field distribution during deposition is crucial for suppressing grain growth and preserving the desired microstructure. In addition, Khan et al. proposed a solid-state additive manufacturing strategy based on AFSD and successfully fabricated AA2014/SiC_p composites, indicating the promising development prospects of this technology [15].

In the field of traditional friction stir welding (FSW), active cooling strategies have been extensively studied to enhance weld joint performance. For example, techniques such as underwater FSW and liquid nitrogen cooling have demonstrated significant effects in reducing the size of the heat-affected zone (HAZ) and refining the microstructure [16,17]. The review by Patel et al., entitled 'Critical Review on Advanced Cooling Strategies in Friction Stir Processing for Microstructural Control,' systematically summarizes and assesses the topic. The article comprehensively reviews the various mainstream methods for controlling sample cooling during and after the AFSD process, thoroughly analyzes the fundamental mechanisms, advantages, disadvantages, and limitations of each cooling strategy, and discusses their effects on microstructural evolution and mechanical properties of the materials [18]. However, the application of active cooling techniques in AFSD is more complex due to its continuous layer-by-layer deposition process and the dynamic nature of the material feeding system. Although some studies have explored the impact of substrate cooling, the effects of direct ambient cooling (such as water mist spraying on the deposition area) on 2195 aluminum–lithium alloy have not been thoroughly investigated. This study presents a comparative analysis of 2195 aluminum–lithium alloy processed by AFSD under two different ambient conditions: ambient air cooling and water mist spraying cooling. The objective is to comprehensively investigate the effects of accelerated cooling on the macroscopic appearance, grain development, and mechanical properties of the deposited layers. By integrating water mist cooling technology into the AFSD process, this study aims to mitigate the adverse effects of accumulated heat, promote grain refinement, and enhance the overall mechanical properties of solid-state additive-manufactured aluminum–lithium alloy.

2. Materials and Methods

2.1. Experimental Material

In this study, we selected 10 mm × 10 mm × 300 mm 2195-T34 aluminum–lithium alloy square bars as the core raw material and employed 7 mm-thick plates of the same material as the base substrate for the deposition process. The detailed chemical composition of the 2195-T34 aluminum–lithium alloy (produced by Zhengzhou, China) is shown in Table 1, and its mechanical properties are listed in Table 2. Figure 1a illustrates the schematic diagram of the AFSD technology. During the experiment, we utilized an AFSD3020-06T-C15 model friction stir additive manufacturing machine produced by Shanghai, China.

Table 1. Main chemical composition of alloy 2195 (wt.%).

Element	Cu	Li	Mg	Ag	Fe	Zr	Mn	Si	Ti	Al
Content	4.18	1.19	0.40	0.38	0.052	0.094	0.047	0.014	0.018	Bal.

Table 2. Mechanical properties of 2195-T34 aluminum alloy.

Material	Yield Strength (YS/MPa)	Tensile Strength (UTS/MPa)	Elongation (EL/%)	Hardness (HV3)
2195	533	580	14	120

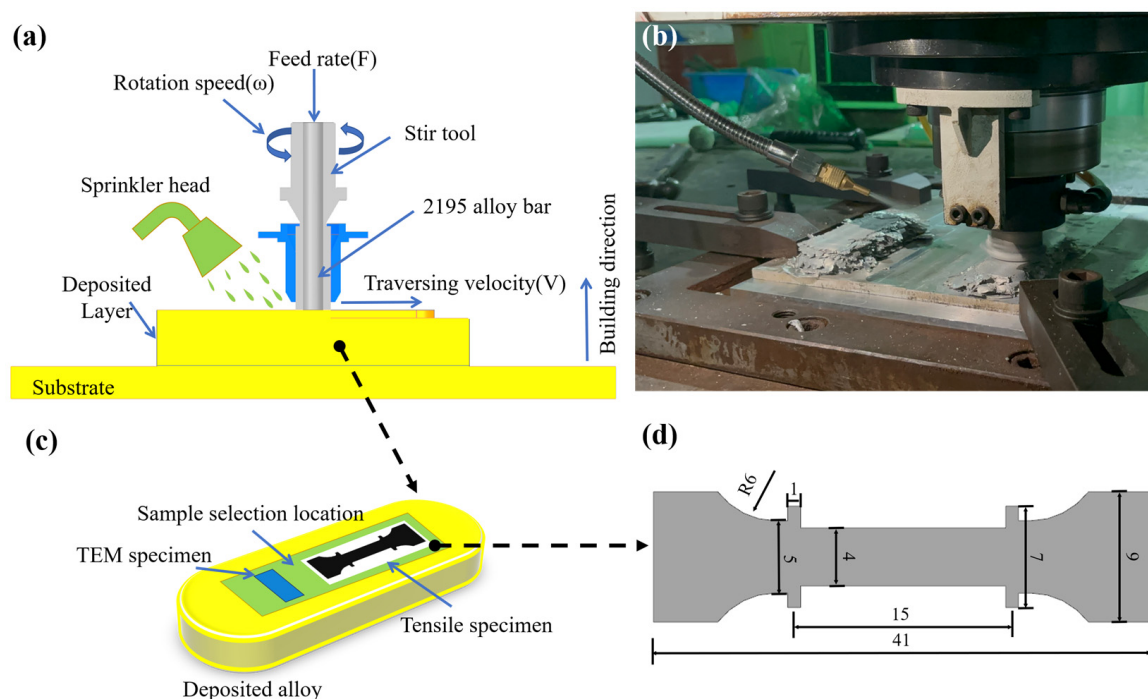


Figure 1. (a) Schematic representation of the AFSD process (under water mist spray conditions); (b) actual deposition manufacturing process; (c) sampling locations for EBSD, TEM characterization, and mechanical property testing; and (d) geometry and corresponding dimensions of the tensile specimens.

The key parameters set during the deposition process were as follows: a preheating time of 62 s, a tool rotation speed of 475 revolutions per minute, a rod feed rate of 110 mm/min, and an overall feed rate of 175 mm/min. The specific deposition manufacturing process is shown in Figure 1b. The alloy sample prepared through the AFSD process was constructed by stacking five deposited layers, with each layer being approximately 1.5 mm thick, 35 mm wide, and 100 mm long.

To ensure the quality of the prepared samples, we extracted a portion of material from the central region of the deposited sample (the specific extraction location is shown in Figure 1c). This extracted material will be used for subsequent tensile specimen fabrication, mechanical property testing, and detailed characterization analysis of the microstructure, with the specific dimensions of the tensile specimens shown in Figure 1d (GB/T 228.1 [19]).

2.2. Microstructure Characterization

Metallographic specimens were wet-ground sequentially using 400#, 800#, 1200#, 2000# and 5000# metallographic abrasive papers. Mechanical polishing was performed with 3 μm and 1 μm diamond suspensions, followed by final polishing using an aqueous detergent solution. After polishing, the specimens were rinsed with anhydrous ethanol and dried with cold air, then etched in Keller's reagent for 15 s prior to metallographic microstructure observation. Hardness measurements were carried out on a Huayin 200HVS-5 digital Vickers hardness tester (Laizhou Huayin Testing Instrument Co., Ltd., Laizhou, China), with a constant test load of 3 kg and a dwell time of 15 s. The grain characteristics of the material were observed using an MR2100 metallurgical microscope from Nanjing Jiangnan Yongxin Optics Co., Ltd., Nanjing, China. Microstructural characterization was conducted on two groups of specimens-with and without spray treatment-using a JEM-F200 transmission electron microscope (TEM) from JEOL Ltd., Akishima, Japan. The preparation process for TEM specimens was as follows: The alloy was mechanically ground to a thickness of 80–100 μm and then fabricated into circular discs with a diameter of 3 mm. Subsequently, further thinning was performed at $-25\text{ }^{\circ}\text{C}$ and 30 V using a twin-jet electrolytic polishing apparatus in a mixed electrolyte consisting of 30% nitric acid (HNO_3) and 70% methanol (CH_3OH).

2.3. Mechanical Measurements

Uniaxial tensile tests were conducted on the specimens at room temperature using a CMT4304 electronic universal testing machine from Shenzhen Sansi Experimental Equipment Co., Ltd., Shenzhen, China, at a tensile rate of 0.375 mm/min (GB/T 228.1-2021). Three parallel specimens were tested for each group, and the average values were taken as the final results.

3. Results

3.1. Mechanical Properties

As shown in Figure 2, there are significant differences in the morphology of deposited bodies formed through AFSD under air cooling and water mist cooling conditions. In an air environment, due to its relatively poor heat dissipation capacity, heat tends to accumulate continuously in the deposition zone during the additive manufacturing process, keeping the material at a high temperature and in a highly plasticized state for an extended period. This leads to excessive overflow of the plasticized metal to both sides under the pressure and rotational shear action of the stir tool, resulting in the formation of substantial flash and irregular accumulations. In contrast, under water mist cooling conditions, the overall contour of the deposited body is more concentrated, with reduced lateral spreading and a relatively regular forming area. This is primarily because water mist cooling significantly enhances the heat dissipation rate during the additive manufacturing process, lowering the peak temperature and heat accumulation in the deposition zone. Consequently, the plasticized material can cool and stabilize more rapidly after stir deposition, thereby suppressing excessive flow and lateral flash expansion. Overall, the cooling environment significantly influences the final forming morphology by altering the temperature field, material plasticization degree, and flow behavior during friction stir additive manufac-

turing. The differences in macroscopic morphology indicate that the microstructure and mechanical properties are inevitably different as well.

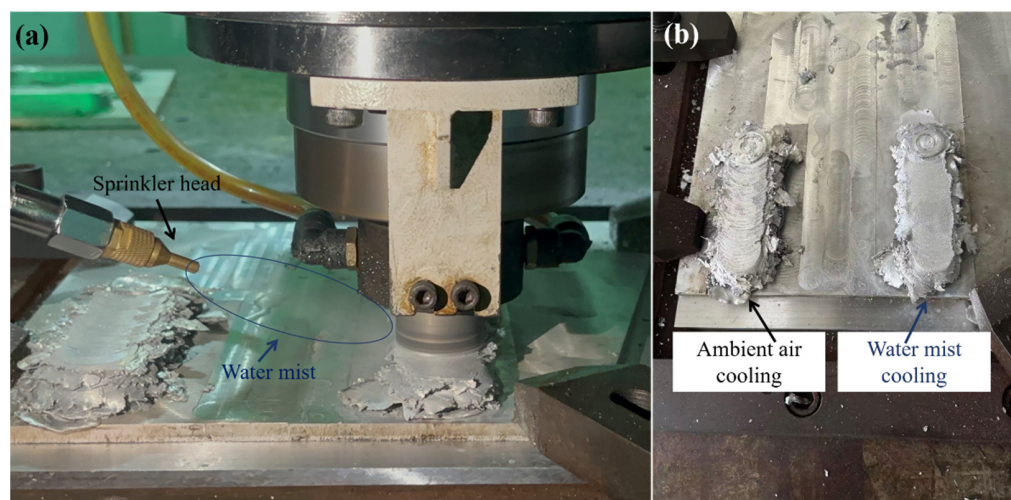


Figure 2. (a) Deposition manufacturing site under water mist spray conditions; (b) morphological features of the deposited alloys under ambient air cooling and water mist spray conditions.

Figure 3a presents the hardness distribution under the two cooling conditions. It can be observed that the AFSD Al-Li alloy exhibits an identical symmetrical distribution centered in the middle region, where the hardness remains stable, while the hardness at both sides is relatively lower. The mechanical properties in the middle region are superior, so all specimens in this study were sampled from this middle region. Additionally, it is found that the hardness of the AFSD Al-Li alloy under water mist cooling is consistently higher than that under ambient air cooling, indicating that water mist cooling exerts a positive effect on the microstructure and implies an improvement in mechanical properties. Figure 3b presents the engineering stress–strain curves of AFSD Al-Li alloy under two cooling conditions: ambient air cooling and water mist cooling. From the curve morphology, both samples exhibit a continuous yielding behavior without a distinct yield plateau, indicating that the material undergoes uniform plastic deformation and sustained work hardening during tensile loading. However, significant differences exist in the position and elongation of the two curves: the entire stress–strain curve of the water mist cooling sample lies above that of the air cooling sample, with a longer uniform plastic deformation region and a larger fracture strain.

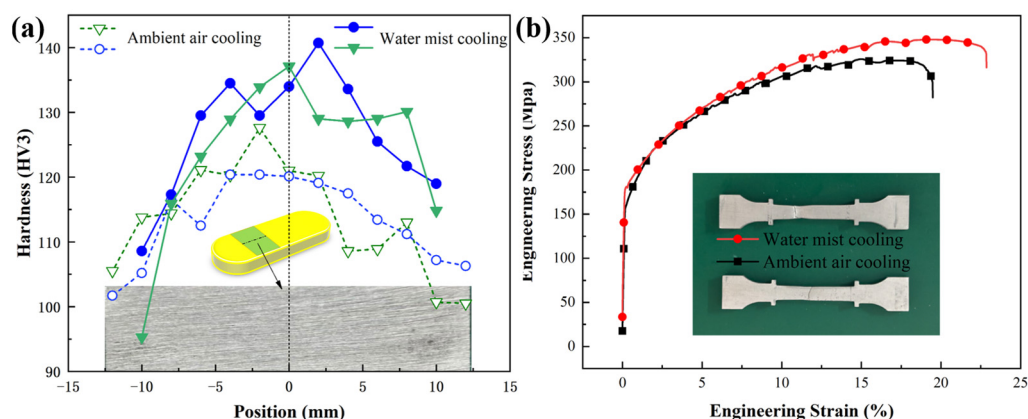


Figure 3. Hardness distribution (a) and engineering stress–strain curves of AFSD specimens under different cooling conditions (b).

Table 3 summarizes the mechanical properties obtained from the tensile tests. The air-cooled sample shows an ultimate tensile strength of 325 MPa, a yield strength of 165 MPa, and an elongation of 21%. The water mist cooling sample exhibits improved UTS of 348 MPa, YS of 182 MPa, and EL of 24.5%. Compared with the water mist cooling condition, the air-cooled sample shows a reduction of approximately 7% in UTS, 10% in YS, and 17% in EL. Notably, the relative improvement in elongation (17%) exceeds that in strength (7–10%), suggesting that water mist cooling has a more pronounced effect on enhancing ductility.

Table 3. Mechanical properties of AFSD specimens.

Cooling Conditions	Yield Strength (MPa)	Ultimate Tensile Strength (MPa)	Elongation (%)
Ambient air cooling	165 ± 4	325 ± 3.5	21 ± 1.2
Water mist cooling	182 ± 5	348 ± 3	24.5 ± 1.5

From the perspective of work hardening behavior, both samples show a gradually rising trend after yielding up to peak stress, followed by a slight drop before fracture. The water mist cooling sample maintains a higher flow stress throughout the entire plastic deformation stage, and the strain value corresponding to the peak stress (uniform elongation) is significantly larger than that of the air-cooled sample. This indicates that water mist cooling not only elevates the strength level of the material but also delays the onset of necking, thereby endowing the material with superior plastic formability.

3.2. Tensile Fracture Behavior

Figure 4 shows the fracture surface morphologies of friction stir additively deposited alloy specimens under different cooling conditions. In the air-cooled sample (Figure 4a), numerous flat cleavage steps and localized quasi-cleavage features are observed, with a relatively small number of dimples unevenly distributed, indicating a mixed ductile–brittle fracture mode with a certain tendency toward brittle fracture. In contrast, the water mist-cooled sample (Figure 4b) exhibits a large number of fine dimples and tearing ridges on the fracture surface, with obvious void coalescence features, suggesting that its fracture mechanism is dominated by typical ductile fracture. This difference indicates that water mist cooling can effectively suppress heat accumulation during deposition, improve microstructural homogeneity, and enhance plastic deformation compatibility, thereby improving the plasticity and fracture toughness of the material. The fracture surface analysis results are consistent with the tensile property test results (Figure 3): the water mist-cooled sample shows more dimple features, corresponding to its higher plasticity and superior comprehensive mechanical properties, whereas the air-cooled sample exhibits more pronounced quasi-cleavage features, corresponding to its relatively lower post-fracture plasticity [20].

3.3. Microstructure

Figure 5 shows the metallographic microstructures and grain size distributions of AFSD Al–Li alloy under different cooling conditions. Under both cooling conditions, the deposited alloys exhibit predominantly equiaxed grain structures, indicating that the material underwent severe plastic deformation and dynamic recrystallization during the AFSD process [21]. Under ambient air cooling (Figure 5a), the alloy grains are generally coarser with an uneven size distribution, and obvious abnormal grain growth is observed. The corresponding grain size statistics (Figure 5c) show an average grain size of 15.37 μm with a wide distribution range exhibiting a typical multi-peak feature, reflecting the inhomogeneity of grain growth. In contrast, under water mist cooling (Figure 5b), the alloy

grains are significantly refined with uniform overall size and no obvious coarse grains. The grain size statistics (Figure 5d) reveal an average size of only 5.15 μm , with a narrowed distribution curve showing a single-peak profile, indicating a significant improvement in grain size uniformity.

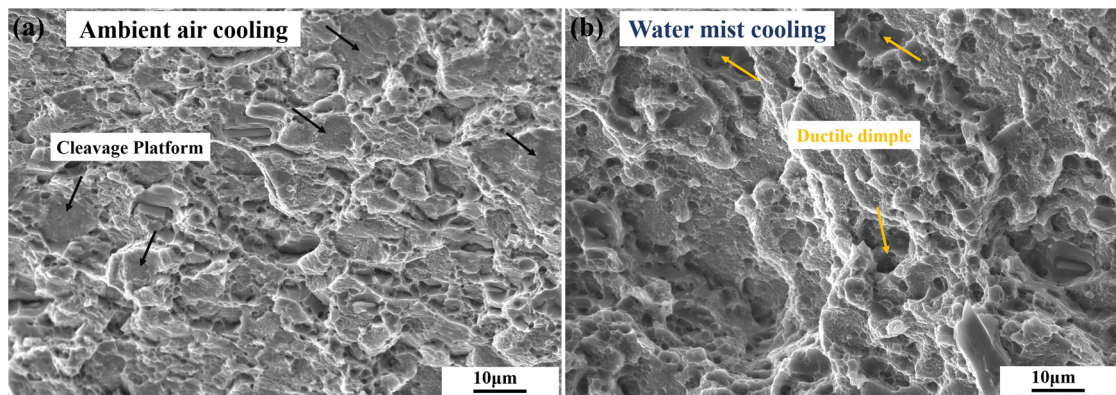


Figure 4. Fracture surface morphologies of AFSD specimens under different cooling conditions: Ambient air cooling (a) and Water mist cooling (b).

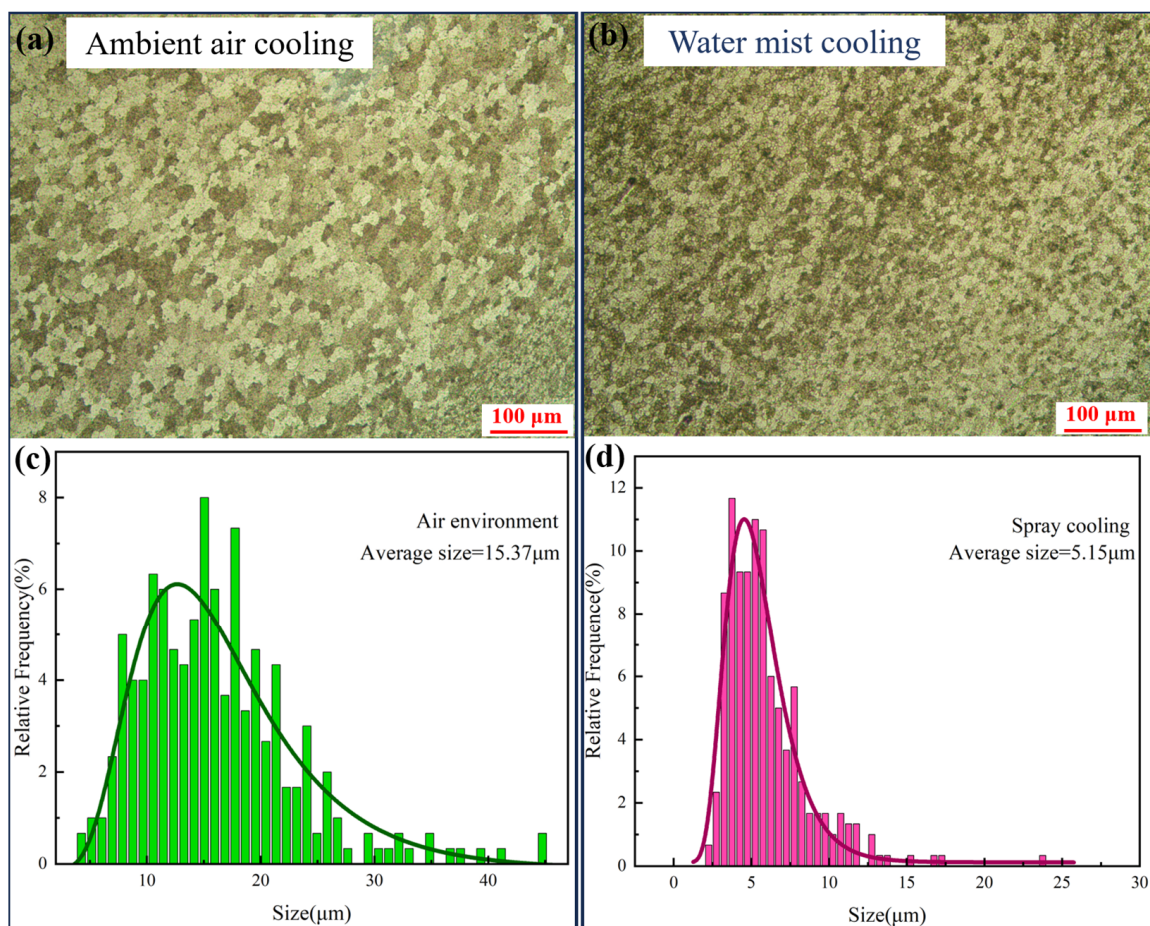


Figure 5. Grain characteristics of AFSD specimens under different cooling conditions (a,b), along with the corresponding size distribution of grains (c,d).

Figure 6 shows the {111} pole figures of AFSD alloy under different cooling conditions. Both samples exhibit multiple discrete pole density peaks rather than a completely uniform distribution, indicating that the deposited alloy developed a certain degree of

preferred orientation under the combined effects of intense shear plastic deformation and dynamic recrystallization during the AFSD process, but no extremely strong single texture concentration is observed overall. Under ambient air cooling, the high pole density regions in each pole figure are more concentrated with more prominent local peaks, reaching a maximum pole density of 6.45, indicating a relatively higher texture intensity. In contrast, under water mist cooling, the pole figure peaks are more dispersed with weakened local peaks, and the maximum pole density decreases to 5.41, approximately 16.1% lower than that of air cooling, suggesting that water mist cooling significantly weakens the texture of the deposited alloy and promotes a more uniform grain orientation distribution.

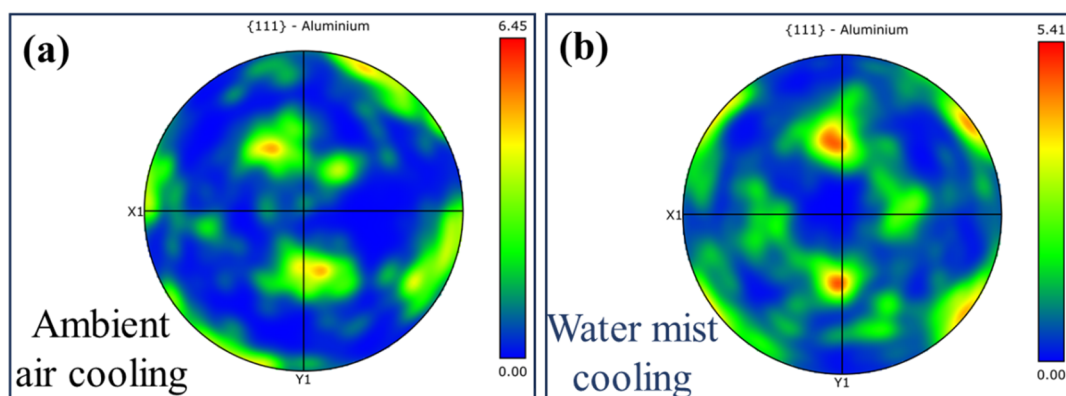


Figure 6. Pole figures under two cooling conditions: ambient air cooling (a); water mist cooling (b).

To further quantitatively reveal the local strain state and microstructure evolution law of AFSD Al-Li alloy under different cooling conditions, systematic statistical analysis was conducted on the Kernel Average Misorientation (KAM), grain boundary misorientation distribution, and recrystallized grain fraction of the two groups of specimens. The average KAM value of the deposited alloy under ambient air cooling is 3.3, while that of the alloy under water mist cooling rises to 4.6, indicating that the rapid cooling process of water mist cooling effectively suppresses the cross-slip, climb, and annihilation of dislocations at the high-temperature stage. This enables the deposited material to retain a higher local dislocation density and residual strain after hot deformation, thus providing sufficient driving force for dynamic recrystallization nucleation. According to the statistical results of grain boundary characteristic distribution, the fraction of low-angle subgrain boundaries with a misorientation of 2° – 10° in the air-cooled specimen is about 23%, and the fraction of grain boundaries with a misorientation greater than 10° is about 77%; while the fraction of low-angle grain boundaries in the water mist-cooled specimen drops to about 15%, and the fraction of grain boundaries with a misorientation greater than 10° increases to about 85%. The statistical results of the recrystallized grain fraction further verify the regulatory effect of cooling rate on the recrystallization process: the fraction of fully recrystallized grains under ambient air cooling is 49.5%, and this fraction increases to 63.5% under water mist cooling. The above results demonstrate that the high thermal gradient and short high-temperature residence time introduced by water mist cooling not only avoid excessive coarsening and merging of subgrains during dynamic recovery but also promote the transformation of low-angle subgrain boundaries into high-angle grain boundaries via continuous dislocation absorption, driving the dynamic recrystallization process to proceed more sufficiently. This microstructure evolution law forms a complete evidence chain with the previously observed results of significant grain refinement, reduced texture intensity, and improved overall hardness, clarifying the mechanism by which water mist cooling optimizes the microstructure of the deposited alloy through regulating recrystallization behavior.

Figure 7 shows the TEM morphologies of precipitates in the deposited Al-Li alloy under different cooling conditions. As seen in Figure 7a, the air-cooled sample contains a relatively large number of rod-shaped, plate-shaped, and ellipsoidal precipitates with relatively large sizes and an uneven distribution. Among these precipitates, the θ phase (Al_2Cu) is dominant. The θ phase is a common Cu-enriched precipitate in aluminum alloys that can impede dislocation motion to a certain extent and provide strengthening. However, when the θ phase is large in size and unevenly distributed, its strengthening efficiency is limited, and it may serve as local stress concentration sites during deformation [22].

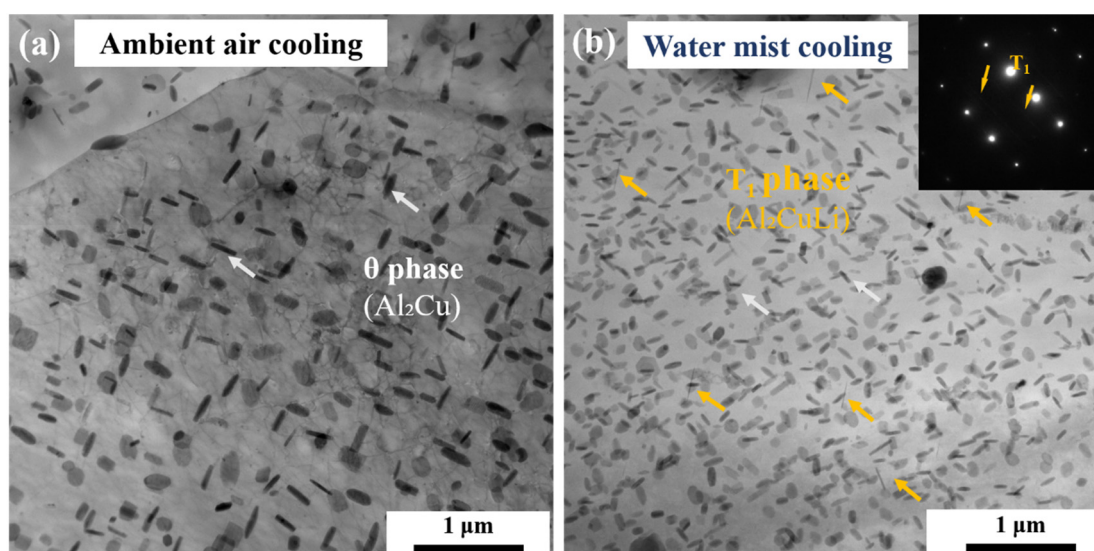


Figure 7. TEM images of precipitates in AFSD Al-Li alloy under different cooling conditions: (a) ambient air cooling; (b) water mist cooling.

Compared with the air-cooled sample, the precipitate morphology in the water mist cooling sample changes significantly. As shown in Figure 7b, the precipitates in the water mist cooling sample are notably refined with an increased number density and a more uniformly dispersed distribution. A large number of fine plate-shaped and short rod-shaped precipitates can be observed. Combined with the selected area electron diffraction results, the presence of the T1 phase (Al_2CuLi) is confirmed. The T1 phase is one of the most important strengthening phases in Al-Cu-Li aluminum–lithium alloys, typically appearing as thin plates and preferentially precipitating along the $\{111\}$ planes of the aluminum matrix [23]. Its high density and fine, dispersed distribution can significantly increase the resistance to dislocation motion, which is a major reason for the strength improvement in the water mist cooling sample.

It can also be seen from Figure 7 that the number of coarse precipitates under water mist cooling is significantly reduced, indicating that rapid cooling effectively suppresses the growth and coarsening of precipitates. In contrast, air cooling, due to its lower cooling rate, allows the deposited material to remain at higher temperatures for a longer duration, giving solute atoms such as Cu, Mg, and Li more sufficient diffusion time, thus making it easier to form larger precipitates. To further clarify the elemental composition of the precipitates, elemental analysis was performed on the precipitates under both cooling conditions.

Figure 8 shows the STEM-EDS elemental mapping results of precipitates in the air-cooled sample. It can be seen that the matrix is primarily composed of Al, while noticeable Cu enrichment is observed in some rod-shaped, plate-shaped, and particle-shaped precipitate regions, indicating that these precipitates are mainly Cu-enriched phases. Previous studies have shown that the temperature during AFSD can reach above 300 °C, at which

the T1 phase dissolves [24]. Combined with the TEM morphology, the main precipitate phases in the air-cooled sample are identified as the θ -Al₂Cu and S-Al₂CuMg phases.

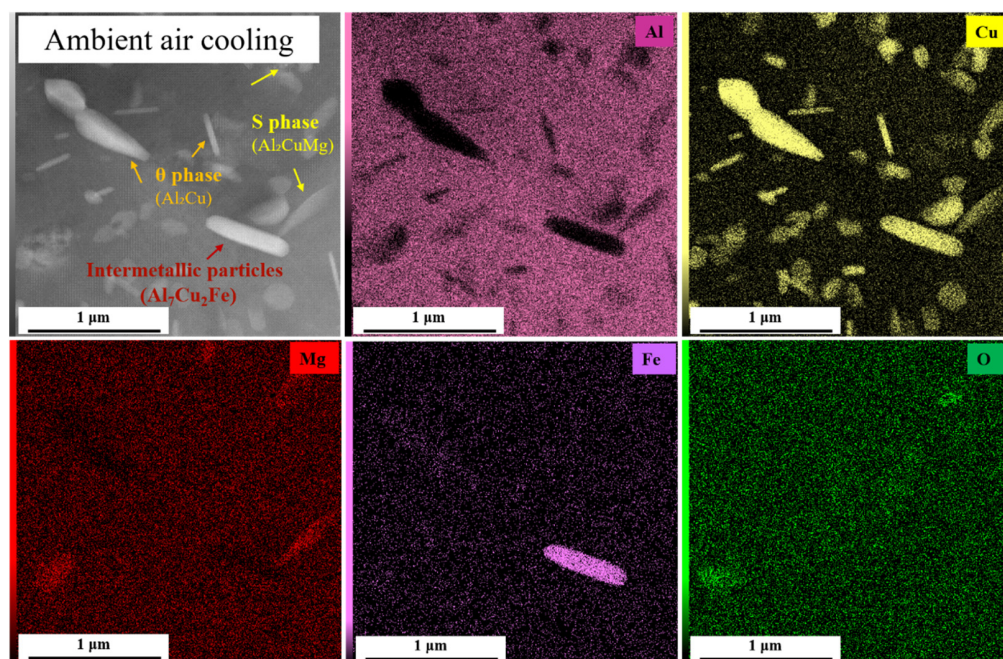


Figure 8. STEM-EDS elemental maps of precipitates in AFSD Al-Li alloy under ambient air cooling.

The θ -Al₂Cu phase regions exhibit distinct Cu enrichment, while the S-Al₂CuMg phase is enriched in both Cu and Mg. The Mg enrichment in some elongated precipitate regions further confirms the presence of the S phase. In addition, certain large irregular particles show a significantly enhanced Fe signal accompanied by Cu enrichment, indicating that these particles are Al-Cu-Fe iron-rich intermetallic compounds, such as Al₇Cu₂Fe. Such Fe-rich phases typically possess high hardness and low ductility and tend to cause local stress concentration during tensile deformation [25].

The presence of θ phase, S phase, and Fe-rich intermetallic compounds in the air-cooled sample indicates that the relatively slow cooling process provides sufficient conditions for solute atom diffusion, segregation, and precipitate growth. Due to their large size and uneven distribution, these precipitates offer a relatively limited resistance to dislocation motion. Meanwhile, the interface incompatibility between coarse secondary phases and the aluminum matrix may promote interfacial debonding, void nucleation, or crack propagation under external loading, thereby limiting the ductility of the material [26].

Figure 9 shows the STEM-EDS elemental mapping results of precipitates in the water mist cooling sample. Compared with the air-cooled sample, the precipitates in the water mist cooling sample are significantly smaller, mainly appearing as fine plates or short rods, with a more uniform distribution. Cu enrichment is clearly observed in the precipitate regions. Combined with the TEM morphology and diffraction results in Figure 7b, the main precipitates in the water mist cooling sample are identified as θ -Al₂Cu phase along with a small amount of T1-Al₂CuLi phase.

The abundant formation of T1 phase under water mist cooling is closely related to the rapid cooling rate. Rapid cooling shortens the dwell time of the deposited layer in the high-temperature range, suppresses the long-range diffusion of solute atoms such as Cu, Mg, and Li, and thereby reduces the formation and growth of coarse θ and S phases. Meanwhile, the severe plastic deformation during the AFSD process introduces a large number of dislocations, subgrain boundaries [27], and other crystal defects into the matrix, which can serve as heterogeneous nucleation sites for the T1 phase. Water mist cooling

helps retain these deformation-induced defects and supersaturated solid solution, thus promoting the formation of fine and dispersed T1 phase [28].

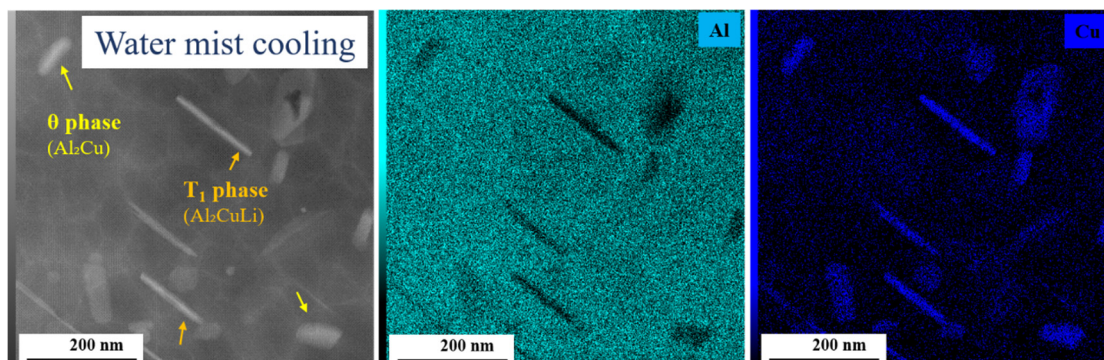


Figure 9. STEM-EDS elemental maps of precipitates in AFSD Al-Li alloy under water mist cooling.

The T1-Al₂CuLi phase has a significant strengthening effect on aluminum–lithium alloys. On one hand, the fine plate-shaped T1 phase can effectively impede dislocation slip, increasing the yield strength and flow stress. On the other hand, its dispersed distribution reduces the risk of local stress concentration, helping to maintain good ductility. Therefore, the appearance of fine and dispersed T1 phase in the water mist cooling sample is an important microstructural reason for its higher strength and elongation compared to the air-cooled sample.

4. Discussion

4.1. The Effect Mechanism of Cooling Rate on Recrystallization Behavior and Grain Size

AFSD is a thermo-mechanically coupled solid-state forming technique. During the deposition process, the rotation and traversal of the stir tool impose severe plastic deformation on the material, causing a sharp increase in dislocation density within the grains [29]. Continuous dynamic recrystallization (CDRX) or geometric dynamic recrystallization (GDRX) occurs, resulting in fine, equiaxed grains. However, the final retained grain size depends not only on the recrystallization nucleation stage but is more strongly governed by post-recrystallization grain growth, which is controlled by the cooling rate after deposition [30].

Grain growth is a thermally activated process, driven by the reduction of grain boundary energy. The grain growth rate exhibits an exponential relationship with temperature (following the Arrhenius equation) [31]:

$$\frac{dD}{dt} = k_0 \exp\left(-\frac{Q_g}{RT}\right) \quad (1)$$

where D is the grain size, t is time, Q_g is the apparent activation energy for grain boundary migration, R is the gas constant, and T is the absolute temperature. Under air cooling conditions, the deposited layer takes a relatively long time (possibly exceeding 10 min) to cool from high temperatures (approximately 400–500 °C) to room temperature, allowing sufficient time for grain growth and resulting in a final average grain size of 15.37 μm. In contrast, under water mist cooling, the cooling rate can reach as high as 20–50 °C/s (estimated), with an extremely short residence time at elevated temperatures (possibly only a few seconds). The time window for grain growth is severely compressed, and thus the grains are “frozen” at a smaller size (5.15 μm).

In addition, rapid cooling may also lead to a higher recrystallization nucleation rate $\dot{N}$. According to classical recrystallization nucleation theory [32], the nucleation rate ΔG^* with increasing undercooling ΔT :

$$\dot{N} \propto \exp\left(-\frac{\Delta G^*}{kT}\right) \quad (2)$$

where ΔG^* is the critical nucleation work. A larger undercooling reduces the critical nucleation work, thereby promoting the formation of more recrystallization nuclei. These nuclei compete for growth, ultimately resulting in finer grains with a more uniform size distribution.

4.2. Thermodynamic and Kinetic Analysis of Precipitation Behavior

Al-Cu-Li alloy is a typical age-hardenable aluminum alloy with a complex precipitation sequence, generally including supersaturated solid solution $\rightarrow$ GP zones $\rightarrow \delta'$ (Al_3Li)/ θ'' (Al_2Cu) $\rightarrow \theta'$ (Al_2Cu)/ T_1 (Al_2CuLi) $\rightarrow \theta$ (Al_2Cu)/ T (AlCuLi) equilibrium phases [33]. The cooling rate plays a decisive role in the nucleation, growth, and final type of precipitates [34].

Air cooling provides a relatively slow cooling rate. When the alloy cools from the high additive manufacturing temperature, the degree of supersaturation is low, resulting in a low nucleation rate. However, atomic diffusion is sufficient, favoring the formation of coarse equilibrium phases. Specifically, Cu atoms have adequate time to migrate, forming large θ phase (Al_2Cu). Mg combines with Cu and Al to form S phase (Al_2CuMg), also in a coarse morphology. Fe, as an impurity element in the alloy, diffuses during slow cooling and combines with other elements to form Fe-bearing intermetallic compounds (Al, Cu, Fe). Once formed, these compounds are highly stable and difficult to eliminate in subsequent heat treatment. They are brittle, incoherent with the matrix, serving as typical crack initiation sites [35].

Water mist cooling provides a very high cooling rate, equivalent to a quenching effect. Rapid cooling generates high supersaturation, forming a highly supersaturated solid solution, which provides the driving force for subsequent aging precipitation (whether natural aging or subsequent artificial aging). In this study, direct air cooling or spray cooling after additive manufacturing means that precipitation actually occurs during the cooling process. Due to the large undercooling, the critical nucleation radius is small, and a large number of fine precipitate nuclei undergo explosive nucleation during cooling. At low temperatures, the atomic diffusion rate is extremely low, and precipitates cannot grow after nucleation, resulting in a very fine final size (approximately 200 nm), with a strengthening effect superior to that of coarse phases. Furthermore, in terms of the overall uniformity of elemental area distribution, the distribution of Cu, Mg, Fe, and other elements in the water spray-cooled samples is more uniform, without the obvious local enrichment zones observed in the air-cooled samples. This further confirms that rapid cooling can suppress micro-segregation, yielding a more uniform solid solution matrix, which is beneficial for improving ductility.

4.3. Comparative Analysis of Mechanical Properties of the Alloy Under Two Cooling Conditions

Based on the tensile test results (Table 3), the water spray-cooled alloy exhibits comprehensively superior mechanical properties compared to the air-cooled alloy: the ultimate tensile strength increases from 325 MPa to 348 MPa (+7.1%), the yield strength from 165 MPa to 182 MPa (+10.3%), and the elongation after fracture from 21.0% to 24.5% (+16.7%). Two points deserve particular attention: first, the relative increase in yield strength (10.3%) exceeds that of the ultimate tensile strength (7.1%), indicating that rapid cooling more significantly enhances the resistance to early plastic deformation; second, the relative increase

in elongation (16.7%) is the largest, demonstrating that water mist cooling is especially effective in improving ductility.

(1) Strengthening Mechanism

Yield strength is primarily governed by the initial resistance to dislocation motion. The average grain size decreases from 15.37 μm to 5.15 μm . According to the Hall–Petch relationship [36], the grain boundary area increases substantially, significantly enhancing the obstruction to dislocation motion. The coarse θ , S, and Fe-bearing phases in the air-cooled condition not only provide poor strengthening but also serve as stress concentration sources. In contrast, water mist cooling produces a large number of fine, dispersed θ and T_1 phases (approximately 200 nm in size). These particles effectively pin dislocations via the Orowan bypassing or shearing mechanisms, providing more pronounced precipitation strengthening [37]. Solid solution strengthening and texture weakening, and rapid cooling retain more solute atoms in the matrix, and the reduced texture intensity (P_{max} decreases from 6.45 to 5.41) facilitates easier activation of slip systems [38], although their direct contribution to yield strength is relatively small. Overall, the yield strength of the water spray-cooled sample increases by approximately 17 MPa, with grain refinement strengthening and precipitation strengthening playing the dominant roles.

The ultimate tensile strength reflects the maximum stress the material can sustain during uniform plastic deformation. The water spray-cooled sample shows a 23 MPa increase in UTS. In addition to the aforementioned strengthening mechanisms, this is also related to the enhanced work hardening capacity. Existing literature confirms that AFSD alloys under rapid cooling exhibit higher dislocation densities. Meanwhile, fine grains and dispersed precipitates promote more uniform dislocation storage, delay dynamic recovery, and allow the flow stress to rise continuously to a higher peak. In contrast, coarse secondary phases and Fe-bearing particles in the air-cooled sample initiate local microcracks at early stages of deformation, limiting the ultimate tensile strength.

(2) Improvement in Elongation (Ductility)

The elongation increases from 21.0% to 24.5%, a relative increase of 16.7%, representing the most significant performance improvement in this study. The reasons can be analyzed from the fracture process:

The coarse Fe-bearing phases and θ phases in the air-cooled sample undergo interfacial debonding or intrinsic cracking at low strain, forming early microvoids. Water mist cooling eliminates these coarse harmful phases; the fine precipitates bond well with the matrix, and microvoid nucleation requires higher local strain. Once microvoids nucleate in the air-cooled sample, the large inter-particle spacing allows rapid crack propagation, forming cleavage facets with brittle characteristics. In the water spray-cooled sample, the large number of fine, densely distributed precipitates enables uniform microvoid nucleation and slow growth. The crack propagation path becomes tortuous, consuming more energy, and ultimately producing a typical dimple fracture surface. Additionally, the weaker texture ($P_{\text{max}} = 5.41$) enhances the coordinated deformation capability among grains of different orientations, resulting in a more uniform strain distribution and delaying the onset of necking [39].

Conventionally, strength improvement is often accompanied by a loss of ductility. However, water mist cooling simultaneously enhances both strength and ductility, breaking this “trade-off relationship.” This is because fine grains benefit both strength and ductility; fine, dispersed precipitates provide strengthening while causing far less damage to ductility than coarse phases; and the elimination of harmful phases significantly improves ductility without compromising strength. Therefore, rapid cooling is an ideal strategy for synergistic optimization of strength and ductility.

A review of recent reports on friction stir additively manufactured Al-Cu-Li alloys [40] reveals that the as-deposited (without subsequent heat treatment) ultimate tensile strength

typically ranges from 300 to 350 MPa, with elongation between 15% and 25%. The air-cooled sample in this study (UTS = 325 MPa, EL = 21%) falls within the intermediate range, whereas the water spray-cooled sample (UTS = 348 MPa, EL = 24.5%) achieves a relatively excellent level for this process, particularly in terms of elongation improvement.

Compared with conventional cast Al-Cu-Li alloys (e.g., 2099, 2195) [41,42], the as-deposited alloy from friction stir additive manufacturing delivers a favorable strength-ductility combination without requiring subsequent T6 heat treatment, demonstrating the potential of this process. The rapid cooling introduced by water mist cooling further narrows the performance gap with age-hardened alloys.

From an engineering application perspective, water mist cooling offers the advantages of simple operation, low cost, and easy integration into additive manufacturing equipment. For the additive manufacturing of large thin-walled components or heat-sensitive structures, water mist cooling can effectively control heat accumulation, prevent grain coarsening and the formation of harmful phases, thereby improving the overall mechanical properties and service reliability of the components.

5. Conclusions

Through systematic comparison of the microstructure and mechanical properties of AFSD Al-Li alloy under ambient air cooling and water mist cooling conditions, the following main conclusions are drawn:

- (1) Water mist cooling reduces the average grain size from 15.37 μm to 5.15 μm , with a more uniform grain size distribution, weakened texture, and improved ductility.
- (2) The air-cooled sample contains coarse θ phase, S phase, and detrimental Fe-bearing intermetallic compounds, whereas water mist cooling produces fine, dispersed θ phase and T1 phase, eliminating the Fe-bearing phases.
- (3) Water mist cooling increases the ultimate tensile strength from 325 MPa to 348 MPa (+7%), yield strength from 165 MPa to 182 MPa (+10%), and elongation from 21% to 24.5% (+17%), achieving simultaneous enhancement of both strength and ductility.

In summary, water mist cooling is an effective thermal control strategy for the additive process. Rapid cooling enables the formation of fine grains, a weakened texture, and beneficial precipitates while suppressing harmful phases, thereby significantly improving the comprehensive mechanical properties of additively friction stir deposited Al-Cu-Li alloy.

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