

Grain refinement and mechanical properties improvement of additively manufactured Al-Cu alloy through pre-deformation in thermo-mechanical treatment

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Abstract: Inferior mechanical properties induced by porosity and coarse grains of wire-arc additively manufactured Al-Cu alloy restrict its industrial application. The inter-layer cold rolling, conventional heat treatment (T6) and thermo-mechanical heat treatment (T8) was applied to Al-Cu alloy, and the effect of pre-stretched strain in T8 treatment was investigated. The pre-stretched strain induces equal increments of hardness and YS for both the as-deposited and cold-rolled conditions before aging treatment, while the increments for cold-rolled samples are larger after aging treatment. The cold-rolled sample shows better tensile properties than the as-deposited sample with the same pre-stretched strain in T8 treatment, proving that T8 treatment further increased the strengths compared to the T6 treatment. It shows the best tensile properties with the pre-stretched strain of 5%, exceeding the properties of 2219-T87 plate. With the increased pre-stretched strain, the density of the θ' phases significantly increases; while further increase to 10% pre-stretched strain induces severe coarsening of θ' phases. Therefore, an optimized pre-stretched strain leads to high-density and tiny dispersed θ' phases, thus resulting in the best tensile properties. Grain refinement induced by inter-layer cold rolling and uniformly precipitated phases resulted from proper pre-stretched strain both contribute to the strengths enhancement. The strengthening mechanism of hybrid additive manufacturing and inter-layer cold rolling followed by T8 treatment was discussed.

Key words: Wire-arc additive manufacture; inter-layer cold rolling; pre-stretched strain; grain refinement; tensile properties; strengthening mechanism

1 Introduction

With the development of aerospace industry, the carrier rockets with the characteristic of large load, lightweight and high reliability are in great need. Adopting high strength and lightweight materials is an effective method to achieve these goals. 2219 aluminum alloy is the key material for the critical components of the rockets due to good mechanical properties at low temperature, such as the tank and the transition ring [1-3]. 2219 aluminum alloy is strengthened through the precipitation of strengthening phases with heat treatment [4-5].

Additive manufacture is beneficial for preparation of components with complex geometry due to its flexibility [6]. Compared to powder bed fusion (PBF), wire - arc additive manufacture

(WAAM) is an arc-based additive manufacture with the melting of the metal wire [7]. Therefore, WAAM shows advantages of low cost, high efficiency and the capability of large-scale components without vacuum chamber compared to the PBF [8-9].

Previously published articles showed that WAAM was suitable for the manufacturing of aluminum components [10-11]. Nevertheless, the porosity defect of deposited aluminum component decreased its mechanical properties and prevented their wide application in the aerospace industry [12-13]. Gu [14] found that the distribution of the hydrogen micropores and solidification microvoids was homogeneous of the deposited wall. It was related to the different distribution of the microstructure and second-phase. Fang [15] and Cong [16] studied the effect of different cold metal transfer (CMT) modes on the microstructure and properties of aluminum alloy. CMT pulse advanced (CMT-PA) contributed to

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the porosity decrease and the tensile properties enhancement compared to other CMT modes. Wang [17] found that small pulse frequency and arc current contributed to the increased cooling rate of WAAM deposited Al alloy, thus decreasing the porosity and the grain size. Liu [18] studied the double-electrode gas metal fabricated 2219 aluminum alloy components, which can decrease the heat input. The bypass current obviously changed the morphology of the grains and it also showed a significant influence on tensile properties. The equiaxed and cellular grains both decreased or disappeared gradually with the increased bypass current ratio. Meanwhile, the UTS and ductility also increased.

Besides the control of deposition modes and the parameters of the WAAM deposition, the plastic deformation was integrated into the WAAM to refine the grains and enhance the properties. Interlayer pneumatic hammering was adopted to the WAAM deposited 2219 aluminum alloy [19]. The microstructure of samples with interlayer hammering exhibited highly refined grains, thus increasing the dislocation density and strengths of the material. Sun [20] combined laser shock peening with WAAM deposition of 2219 aluminum alloy, the grain size can be significantly decreased and the fraction of low angle boundaries was increased. Additionally, the tensile residual stress in WAAM deposition changes to compressive stress after laser shock peening. Gu [21-22] integrated inter-layer cold rolling into the WAAM deposition of Al-Cu alloy. Cold rolling was beneficial for decreasing the fraction and size of the micropores of the material. The micropores were obviously decreased with the force of 45 kN even after heat treatment as heat treatment will induce the growth of the micropores. However, the cold-rolled samples showed equal strengths compared to that in as-deposited samples after heat treatment.

It was known that the thermo-mechanical treatment (T8) was beneficial for enhancement of strengths of the metal alloy [23-24]. The thermo-mechanical heat treatment was achieved through the cold deformation before the solution, or

between solution and aging process [25]. Chen [26] found that the strengths and plasticity were increased simultaneously for 2E12 aluminum alloy by the T8 treatment compared to that with T3 treatment. Huo [27] studied the effect of T8 treatment on Al-Zn-Mg-Cu alloy. The deformation induced the precipitation of the particles and they contributed to the recrystallization and grain refinement, thus obviously increasing the strength and the elongation. Chen [28] studied the influence of novel thermo-mechanical treatment on the microstructure and mechanical behavior of Al-Zn-Mg-Cu alloy. The mechanical properties and corrosion resistance can be increased simultaneously. The co-clusters and nano-precipitates showed positive effect on increasing the strengths, while the small dislocations recovery maintained the ductility of the material. Wang [29] applied pre-deformation to 2219 aluminum alloy to improve the mechanical properties. The strain of 3% shows larger strengths and lower anisotropy compared with the non-deformed forgings. The pre-deformation increased the dislocations, thus contributed to the heterogeneous nucleation and dispersive precipitation of strengthening phases.

As discussed above, the cold rolling can enhance the properties of WAAM deposited aluminum alloy, and the T8 heat treatment can further enhance the strengths. Therefore, the combination of inter-layer cold rolling and T8 heat treatment was adopted to refine the grains and increase the mechanical properties of WAAM deposited Al-Cu alloy in this study. The effect of the pre-stretched strain between solution and aging treatment on the grain morphology, precipitation behavior and tensile properties of the material was analyzed. The strength contribution of as-deposited and cold-rolled material was analyzed, and the complex strengthening mechanism with inter-layer cold rolling and T8 heat treatment was discussed.

2 Materials and methods

The experiments of WAAM deposition and

inter-layer cold rolling were conducted on the integrated deposition and rolling system, as shown in Fig.1. The WAAM deposition can be achieved by the robot controlled equipment, consisting of a CMT power source, a wire feeder, a torch and a gas shielding device. The cold rolling was achieved by the hydraulic CNC system, which was integrated into the deposition equipment. The cold rolling force can be adjusted by the hydraulic cylinder.

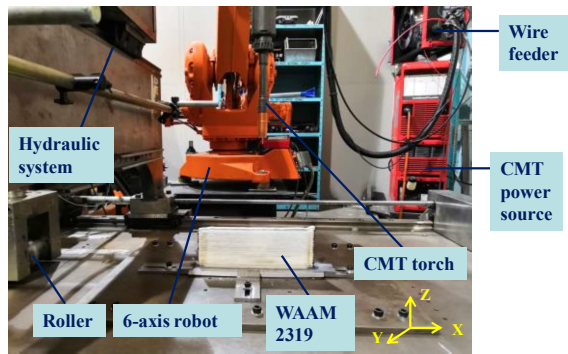


Fig.1 Set up of WAAM deposition and integrated cold rolling system

Compared to the conventional CMT mode, the heat input of CMT-PA mode was 42% lower under the same parameters in this study. CMT-PA mode was adopted for the WAAM deposition of ER2319 aluminum alloy as it was beneficial for the porosity decrease [30]. The reasons for the porosity decrease can be described as follows: the alternating polarities of the arc in CMT-PA mode produced a significant oxide cleaning effect on the end of the wire, thus impeding the hydrogen entering into the molten pool. Additionally, the stirring of molten pool with the polarity change in every cycle contributed to the gas pore escape. Finally, CMT-PA mode was observed to have low dilution and a fine equiaxed grain structure with low heat input, which showed efficient effect in reducing and even eliminating the porosity.

The material of the WAAM deposition was ER2319 aluminum alloy wire with a diameter of 1.2 mm. The wire feed speed and torch travel speed was 6 m/min and 0.6 m/min, respectively. Pure argon was adopted as the shielding gas and its flow rate was 25 L/min. To avoid collapse during the multi-layer

deposition, inter-layer cooling was adopted and the cooling time for each deposited layer was 2 mins. The 80 layers WAAM deposited wall was obtained with a total height of about 100 mm and a length of 200mm. In this study, rolling force of 45 kN was adopted as this rolling force is effective in decreasing the porosity [31] and will not cause collapse of the deposited wall of ER2319 aluminum alloy.

In this study, the microstructure and tensile properties of two walls were comparatively investigated: as-deposited and inter-layer cold-rolled. As shown in Fig.2. It can be seen that the inter-layer cold rolling obviously increase the width of each deposited layer, which increases from 6.5 mm in as-deposited condition to 11.7 mm in cold-rolled condition. Different heat treatments were applied to the walls. The procedures of conventional heat treatment (T6) were solution treated at 535 °C with 1.5 h in the furnace followed by water quenching; and then aging treated at 175°C with 3 h followed by furnace cooling. Thermo-mechanical heat treatment (T8) was investigated in this study by adding pre-stretched strain between the solution and aging process. Three different pre-stretched strains (3%, 5% and 10%) were applied using the electro-mechanical tensile machine and its effect on the microstructure and tensile properties in T8 heat treatment were investigated compared to that with T6 heat treatment.

As shown in Fig.2, the hardness and microstructure of the samples on YZ plane after heat treatments were analyzed. Meanwhile, the horizontal and vertical tensile coupons were prepared for the tensile test in different directions of the walls. The samples on YZ plane were ground, polished and etched with Keller's reagent (HF: HCl: HNO₃: H₂O=2:3:5:190) for grain observation using optical microscopy (OM). The precipitation distribution and the fracture surface of the tensile samples were observed using scanning electron microscopy (SEM) and energy dispersive spectrometry (EDS). Then the samples were ground to a thickness of 80 nm and a diameter of 3 mm with twin-jet electropolishing. The

electrolyte formula was $\text{HNO}_3:\text{CH}_3\text{OH}=1:3$ with a temperature of $-35\sim 25^\circ\text{C}$. The strengthening phases and dislocations were observed using transmission electron microscopy (TEM). The hardness was measured under a load of 200 g and a holding time of

15 s. The tensile test was conducted at room temperature. The tensile speed was 1 mm/min and laser extensometer was used to monitor the variation of the elongation.

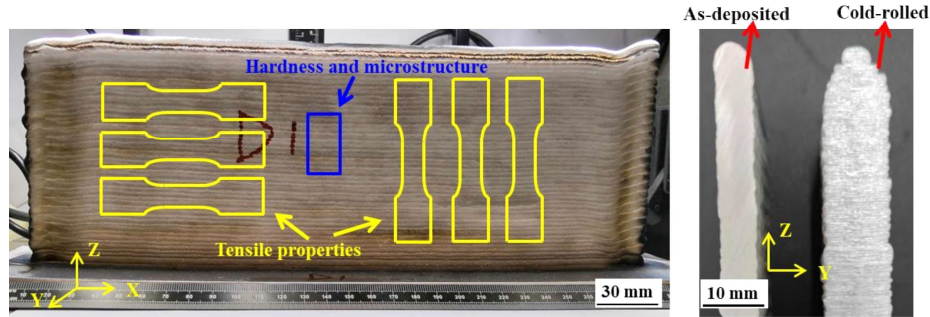


Fig.2 WAAM deposited walls and its cross section

3 Results and discussions

3.1 Hardness

As shown in Table 1, the inter-layer cold rolling increases the hardness with an increment of 26 HV compared to the as-deposited condition. The reason for the hardness increase is the strain induced strengthening resulting from cold rolling. The hardness of as-deposited sample increases obviously after solution treatment; while that of cold-rolled sample changes little. As a result, the hardness in two conditions almost keeps the same, indicating that solution strengthening shows different effect on the as-deposited and cold-rolled samples. The hardness and its increment both increase with the increased pre-stretched strain before aging process. It is interesting that the hardness increment induced by pre-stretched strain in as-deposited sample nearly equals to that in cold-rolled condition. The increments are 7 HV, 12 HV and 27 HV for as-deposited sample with the pre-stretched strain of 3%, 5% and 10%, respectively, which are 8 HV, 13 HV and 28 HV for cold-rolled sample. It can be seen that the pre-stretched strain between solution and aging process shows equal strengthening effect for two conditions before aging process.

The aging process significantly increases the hardness for both conditions compared to that in

solution state due to the precipitation strengthening. Nevertheless, different to the hardness variation with solution treatment, the hardness first increases and then decreases with the increased pre-stretched strain after aging process for both conditions. As a result, the hardness with pre-stretched strain of 10% locates between the value of 3% and 5%. It should be noted that the cold-rolled samples show larger hardness than the as-deposited samples after aging process; while it is almost the same after the solution and pre-stretched process. Therefore, the cold-rolled samples show different strengthening effect compared to that of as-deposited samples, which will be explained in the subsequent sections.

Table 1 Hardness variation under different treatments

Hardness	As-deposited	Cold-rolled
Before heat treatment	74 ± 2.4	100 ± 1.6
S	100.7 ± 3.5	102.2 ± 3.8
S+3%	107 ± 2.9	110.3 ± 2.2
S+5%	112.8 ± 6.1	115.4 ± 5.3
S+10%	127.6 ± 6.2	130.3 ± 2.8
T6	139.2 ± 4.4	143.7 ± 4.4
T8-3%	145.3 ± 3.3	159.1 ± 3.8

T8-5%	152.4±6.7	163.9±4.0
T8-10%	153.1±5.6	161.6±4.3

S: solution treatment; S+3%: solution + pre-stretched strain of 3%; T8-3%: solution+ pre-stretched strain of 3% +aging

3.2 Microstructure

The as-deposited sample shows equiaxed dendrites and grains with porosity, which is quite different from the typical columnar dendrites of WAAM deposited metal alloy. The microstructure formation is related to the heat input and its flow direction in WAAM deposition. In this study, CMT-PA mode is adopted and its heat input is much lower than other CMT modes, thus resulting in equiaxed grains instead of the columnar dendrites. The average grain size of as-deposited sample is $65.4\pm 9.3\ \mu\text{m}$, and the size of the porosity is about $37.2\pm 6.7\ \mu\text{m}$ with the density value of 98.92%, as shown in Fig.3. CMT-PA mode can decrease the number of the porosity, but it cannot completely eliminate the porosity. The solubility of hydrogen in liquid aluminum is much larger than that in solid aluminum, and it can be as large as twenty times. The dissolved hydrogen will be precipitated during the cooling process, thus resulting in the formation of the porosity. Cold rolling transforms the equiaxed grains into elongated grains; meanwhile, the porosity is significantly decreased, as it cannot be seen with the OM, as shown in Fig.3.

Fig. 4 shows the morphology of the secondary phases of the Al-Cu alloy before heat treatment using SEM. The white network-like secondary phases are generated at the grain boundaries and the grain interiors of the as-deposited material. Nevertheless, the secondary phases mainly appear at the grain boundaries for the cast Al-Cu alloy. The distribution of secondary phases is quite different for the WAAM deposited and cast Al-Cu alloy, which can be explained as follows: first, the cooling rate in WAAM deposition is much higher than that in cast process due to its small weld pool. Large cooling rate is beneficial for the formation of equiaxed grains with small size and the decreased microscopic segregation.

Additionally, the layer-by-layer deposition provides the transient aging effect to the previously deposited material, which may result in the precipitation of secondary phases in the grain interiors. The network-like secondary phases are fragmented into small pieces with cold rolling process. The fraction of the secondary phases is about 6.8% for both conditions with the Image software. Therefore, the cold rolling process contributes to the formation of finer secondary phases and its dispersive distribution.

Fig.5 shows the effect of pre-stretched strain on microstructure after heat treatment. There is a slight increase for the grain size, porosity size and the number of porosity for the as-deposited sample with T6 heat treatment. The average grain size increases to $72.8\pm 7.5\ \mu\text{m}$; meanwhile, the porosity size increases to $40.7\pm 6.2\ \mu\text{m}$ with the density value of 98.7%. With the increased pre-stretched strain, the average grain size of as-deposited sample first decreases to $65.7\pm 6.4\ \mu\text{m}$ and $33.2\pm 4.5\ \mu\text{m}$ with the strain of 3% and 5%, respectively, and then increases to $55.4\pm 5.2\ \mu\text{m}$ with the strain of 10%. Meanwhile, the size and the number of porosity show similar variations and they are obviously decreased with the strain of 3% and 5%. However, the size and the number of porosity both increase with the strain of 10%. The porosity size reaches $39.6\pm 4.3\ \mu\text{m}$ with the density value of 99.07%, which is comparable to that with T6 heat treatment.

The heat treatment transforms the elongated grains into equiaxed grains for cold-rolled samples. Similar to that of as-deposited samples, the average grain size first decreases and then increases with the increased pre-stretched strain. As a result, the sample with the strain of 5% shows the smallest grain size of $25.1\pm 3.1\ \mu\text{m}$. The porosity is significantly decreased for the cold-rolled samples after heat treatment; however, the size and number of porosity obviously increases with the strain of 10%, which is comparable to that of as-deposited sample. The cold rolling process increases the dislocation density of the material, which will increase the solubility of hydrogen, as there was linear relationship between the hydrogen solubility and the dislocation density [32]. It

can be inferred that excessively large pre-stretched strain may contribute to the formation and coalescence of the porosity.

Different to the network-like distributed secondary phase of as-deposited samples, heat treatment decreases its size and induces discrete distribution of the secondary phase, as shown in Fig.6. It can be seen that the cold-rolled samples show finer secondary phases with more dispersive distribution compared to that of as-deposited samples. The white secondary phases prove to be Al-Cu eutectics with EDS analysis. The solubility of the Cu element increases with the increased temperature. In this study, the solution temperature is 535 °C and it is much higher than the aging temperature of 175 °C. The precipitated Cu element in the WAAM deposition will be dissolved into the matrix in solution treatment, and

it will be precipitated again as the strengthening phases during the subsequent aging process.

The variation of pre-stretched strain does not obviously change the morphology of the secondary phases. To qualitatively investigate the effect of pre-stretched strain on the size of secondary phases, the average size and its distribution are calculated using Image software, as shown in Fig.7. The average size of secondary phases decreases with the increased pre-stretched strain for as-deposited samples, while it first decreases and then increases for cold-rolled samples. Meanwhile, it can be seen that the size of secondary phases for cold-rolled samples is smaller than that for as-deposited samples. The size of secondary phases play vital role in the strengths of the material, which will be discussed in the followed section.

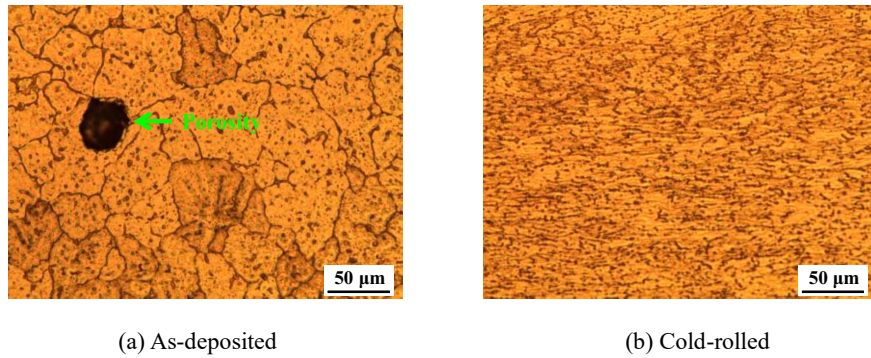


Fig.3 Microstructure of as-deposited and cold-rolled 2219 alloy before heat treatment

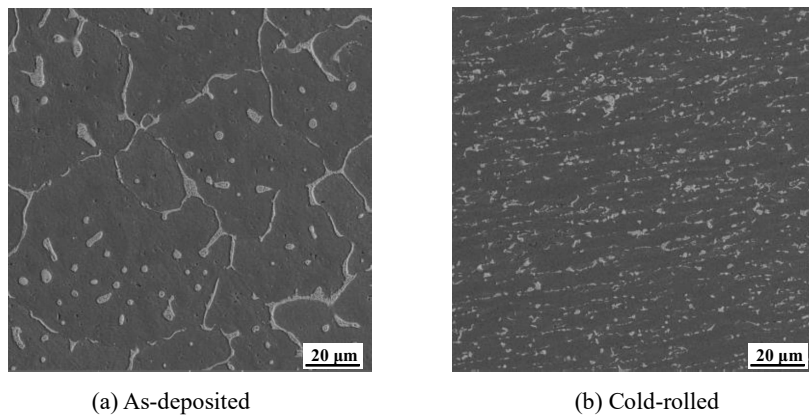


Fig.4 Secondary phase of as-deposited and cold-rolled 2219 alloy before heat treatment

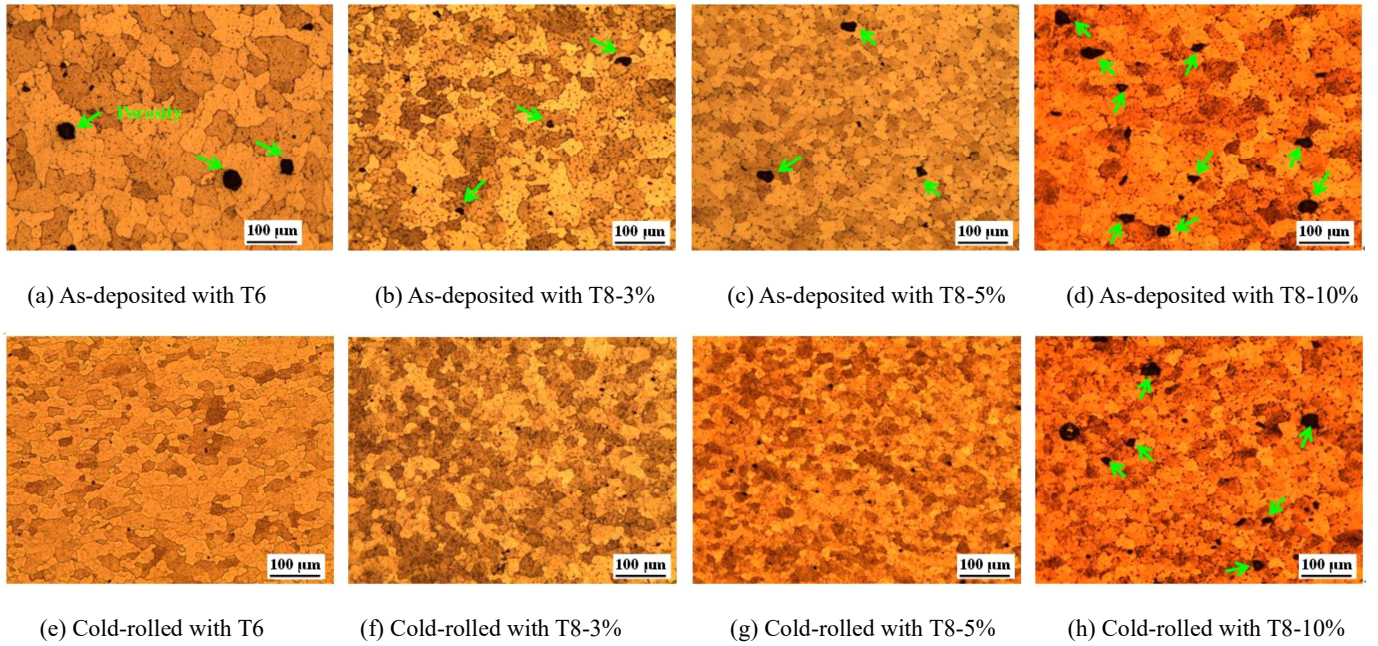


Fig.5 Effect of pre-stretched strain on microstructure after heat treatment

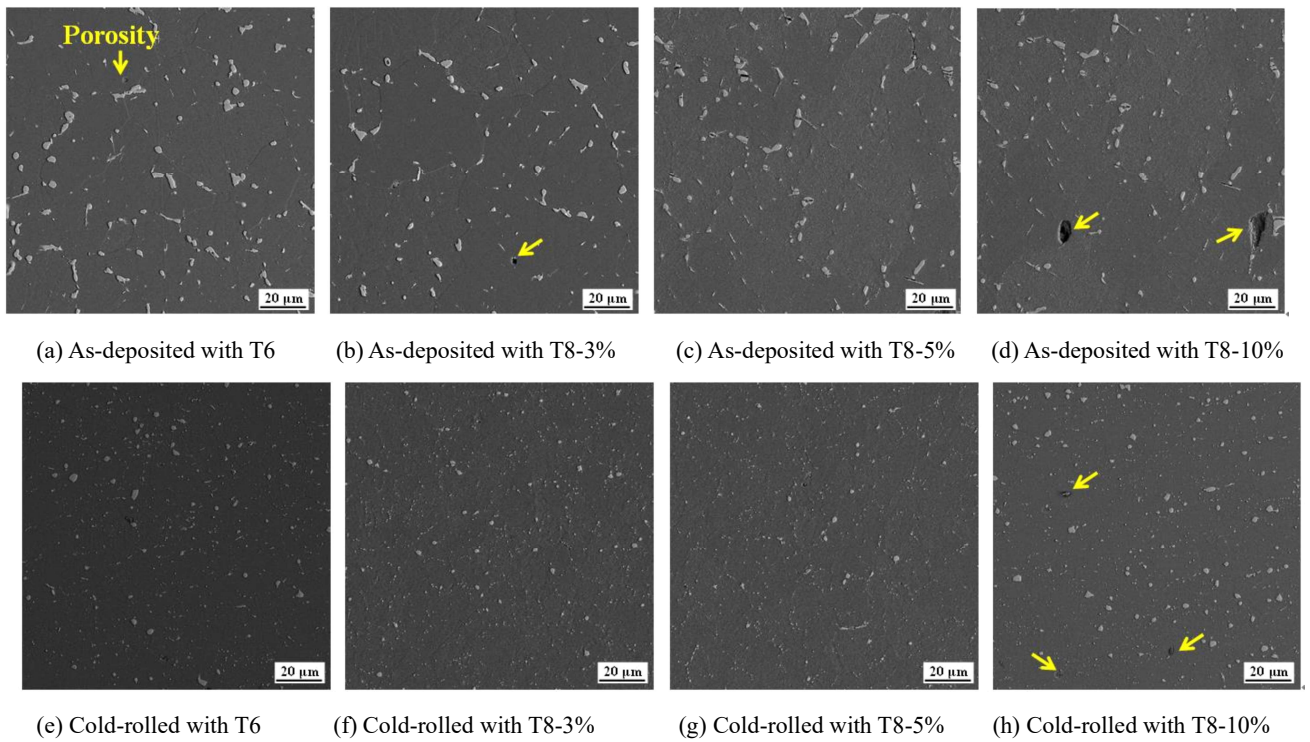
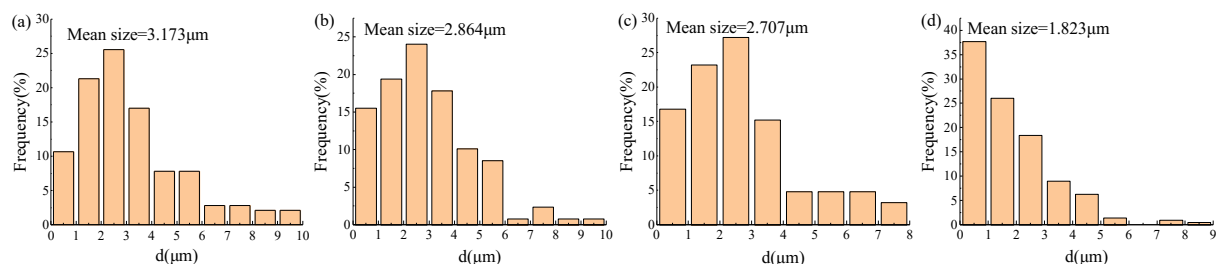


Fig.6 Effect of pre-stretched strain on secondary phase after heat treatment



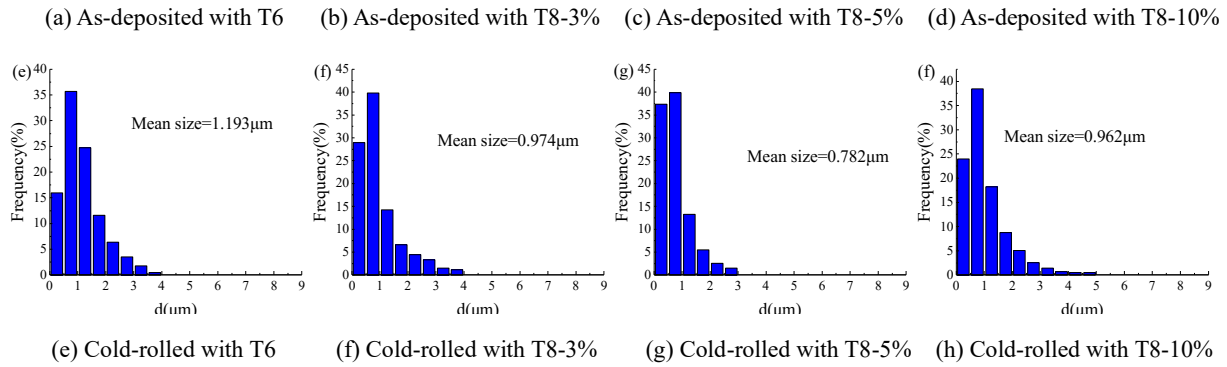


Fig.7 Particle size distribution under different pre-stretched strain after heat treatment

3.3 Tensile properties

The as-deposited samples show low UTS and YS but high elongation before heat treatment. The YS of the cold-rolled samples is over twice compared to that of the as-deposited samples, as shown in Table 2. The UTS increases with the cold rolling process accompanied by obvious decreased elongation. It can be seen that the work hardening effect induced by the inter-layer cold rolling process contributes to the enhancement of strengths and decrease of elongation before heat treatment.

Solution treatment increases the UTS and elongation for both the as-deposited and cold-rolled conditions. Similar to the variation of hardness, the solution treatment increases the YS of as-deposited samples while decreases the YS of cold-rolled samples. Based on the same YS for the as-deposited and cold-rolled samples after solution treatment, it can be concluded that the solution treatment shows different strengthening effect on the as-deposited and cold-rolled samples.

It can be seen that the pre-stretched strain increases the strengths and reduces the elongation for both conditions after solution treatment. The increase in strengths and reduction in elongation both increase with the increased pre-stretched strain. The YS increments for the as-deposited samples are 100 MPa, 143 MPa and 152 MPa with pre-stretched strain of 3%, 5% and 10%, respectively. They are 115 MPa, 156 MPa and 168 MPa for the cold-rolled samples before heat treatment. It can be concluded that the pre-stretched strain shows equal strengthening effect on both conditions as the YS increments are nearly the same considering the error bar of the YS. Compared to the

variation of YS, the pre-stretched strain slightly increases the UTS. The strengthening mechanism for the pre-stretched strain is the strain induced strengthening, which usually increases the strengths accompanied by the decreased elongation.

The aging treatment significantly increases the strengths and decreases the elongation for both conditions compared to the solution treated samples. The UTS and YS of both conditions almost reach 450 MPa and 300 MPa, which exceeds the 2219 aluminum alloy plate with T62 treatment. The strengths are further enhanced with T8 heat treatment accompanied by slight decrease of elongation for as-deposited samples. The increase in strengths and reduction in elongation for as-deposited samples both increase with the increased pre-stretched strain. It should be noted that the tensile properties of as-deposited samples with T8 heat treatment are inferior to the 2219 aluminum alloy plate with T87 treatment. The cold-rolled samples show different variation with T8 heat treatment. The strengths first increase and then decrease with the pre-stretched strain, as a result, the sample with 5% pre-stretched strain shows the highest tensile strengths. Although the elongation of cold-rolled samples decreases with the pre-stretched strain, it is still larger than 10%. It can be seen that the cold-rolled sample with pre-stretched strain of 5% and 10% both exceeds the tensile properties of the plate with T87 treatment.

It can be seen that the YS increments of cold-rolled samples are larger than that of as-deposited samples with T8 treatment. The variation of YS increments with T8 treatment is different from that before heat treatment, indicating that the pre-stretched strain contributes to the

strengthening effect. In summary, the pre-stretched strain plays different role in the enhancement of strengths for the as-deposited and cold-rolled samples. Therefore, an optimal

pre-stretched strain should be adopted to obtain the higher mechanical properties.

Table 2 Tensile test results of 2219 alloy

AD	UTS/MPa	YS/MPa	Elongation (%)	CR	UTS/MPa	YS/MPa	Elongation (%)
Before HT	267.3±2.9	127±3.8	19.2±1.2	Before HT	334.6±5.2	272±2.3	14.8±1.8
S	373.7±1.4	174.2±5.3	22.3±0.8	S	403.8±1.6	183.7±4.9	30.5±1.2
S+3%	388.5±10.3	274.8±13.2	20.9±0.6	S+3%	405.6±10.5	298.7±10.8	24.8±1.5
S+5%	397.3±0.4	317.7±2.5	18±0.7	S+5%	413.5±5.5	339.4±8.3	18.8±1.8
S+10%	428.9±1.4	326±1.5	16±0.9	S+10%	440.8±2.5	351.2±6.3	17.8±1.2
T6	450.5±11.9	297.4±3.3	13.9±0.9	T6	458±1.3	292.4±12.8	18.8±4.9
T8-3%	464.5±1.5	351.8±5.7	13.1±2.2	T8-3%	495.5±1.2	387.8±1.2	15.6±1.5
T8-5%	478.9±8.8	384.2±7.2	10.4±0.5	T8-5%	513.4±2.8	412.9±0.1	14.1±1.62
T8-10%	481.6±20.2	398±5.7	4.5±1.4	T8-10%	482±2.9	398±6.4	10.4±0.5
2219-T62	415	290	10	2219-T87	475	395	10

HT: heat treatment; S: solution treatment; S+3%: solution + pre-stretched strain of 3%; T8-3%: solution+ pre-stretched strain of 3% +aging

3.4 Fracture morphology

Fig. 7 shows the fracture morphology of the samples with T6 and T8 heat treatments. Coarse and deep primary dimples accompanied by plenty of finer dimples are generated for as-deposited samples with T6 heat treatment, indicating ductile fracture mode and high tensile elongation. The size of the primary dimples decreases while the number of finer dimples increases after cold rolling process, as shown in Fig. 7(b). Meanwhile, deep primary dimples with small size also exist for the cold-rolled samples; therefore, the cold-rolled samples with T6 heat treatment also show good ductility. The finer and denser dimples indicate higher tensile strength as it is beneficial for the formation of a large number of micro voids during the tensile deformation process [33]. As a result, higher strength needs to be applied for the fracture of the samples.

With the increased pre-stretched strain in T8 heat treatment, the size of the dimple and its number are both further decreased. It can be seen that shallow dimples with smaller size appear with the pre-stretched strain of 3%. Brittle fracture characteristic with cleavage planes starts to

appear with the pre-stretched strain of 5%. The mixed distribution of cleavage planes and finer dimples with the pre-stretched strain of 5% implies decreased ductility. The number of cleavage planes is significantly increased accompanied by hardly any dimples for the as-deposited samples with the pre-stretched strain of 10%. Meanwhile, several cracks are mainly generated on the grain boundaries, showing brittle fracture mode. Al-Cu eutectics are the main secondary particles for 2219 aluminum alloy, which are the positions for the initial formation of the micro voids. Primary crack can be triggered at these positions during the tensile test through the coalescence and propagation of micro voids. Then the crack propagation occurs and the trans-crystalline fracture is induced. However, there are still many finer and shallow dimples for the cold-rolled samples with pre-stretched strain of 10% in spite of the increased cleavage planes. Additionally, there is no obvious crack in the cold-rolled samples with the pre-stretched strain of 10%, indicating that the inter-layer cold rolling is beneficial for the closure of the crack. It can be seen that the number of dimples in cold-rolled samples is much higher than that of

as-deposited samples on the same conditions. As a result, the cold-rolled samples show good elongation and ductile

fracture mode even with the pre-stretched strain of 10%.

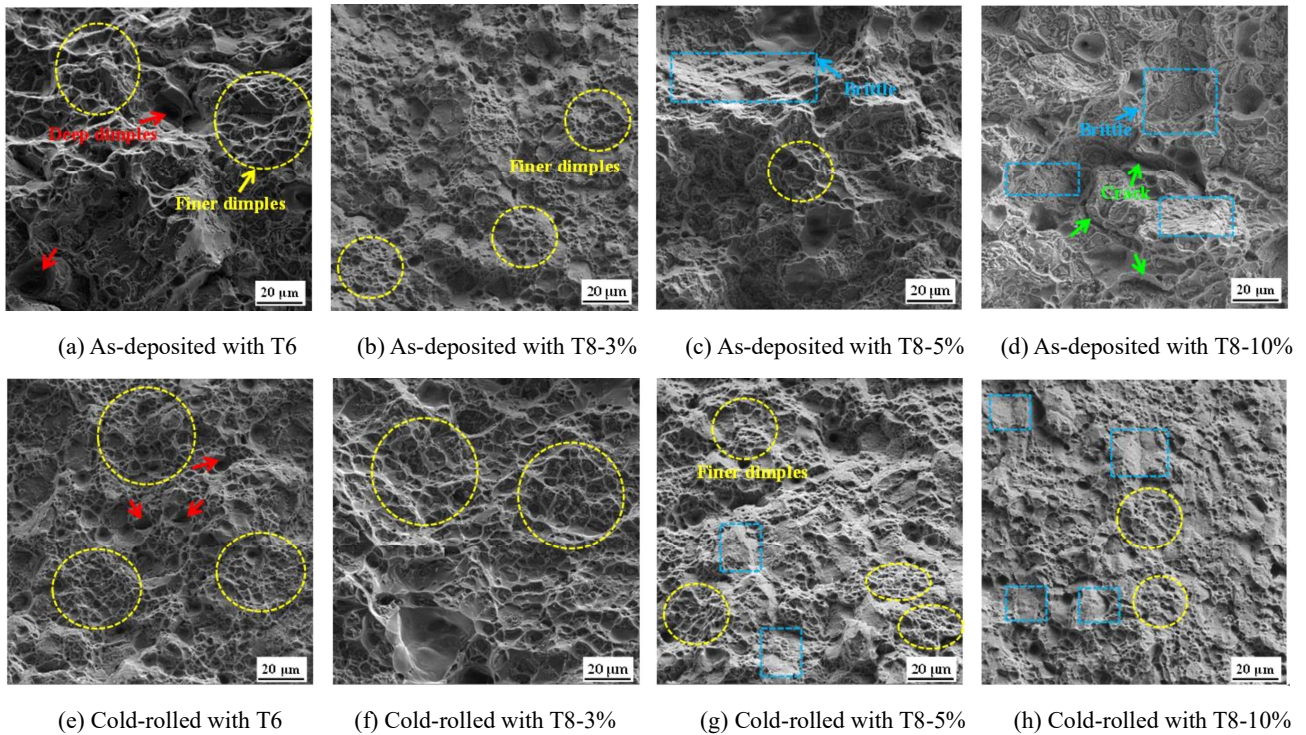


Fig.8 Effect of pre-stretched strain on fracture morphology after heat treatment

3.5 Strength contribution in T8 treatment

There are numerous stages for the hybrid WAAM deposition and inter-layer cold rolling process followed by T8 heat treatment, including inter-layer cold rolling, solution treatment, pre-stretching process and aging treatment. The strength enhancement is quite complex during these processes, consisting of strain induced strengthening (inter-layer cold rolling and pre-stretching deformation), solution strengthening, grain boundary strengthening and precipitation strengthening. The hardness increases from 74 HV to 100 HV, and the YS ranges from 127 MPa to 272 MPa after inter-layer cold rolling before heat treatment. It can be seen that the inter-layer cold rolling obviously enhances the strength of the as-deposited material, which is the strain induced strengthening.

Solution treatment obviously contributes to the hardness increment (74 HV to 101 HV) and YS increment (127 MPa to 174 MPa) of as-deposited samples. Nevertheless, the YS of cold-rolled sample decreases from 272 MPa to 184 MPa after solution treatment. It can be seen that the solution strengthening shows different effect on two conditions.

The alloying elements will be dissolved into the matrix in the solution treatment, thus increasing the dislocation density of the matrix and causing severe lattice distortion. There will be interaction between the stress induced by the lattice distortion and the elastic stress around the dislocations [34], which plays a pinning effect on the movement of the dislocations, thus increasing the strength of the material. There is plenty of alloying elements in Al-Cu alloy, such as Cu, Fe, Si and Mn. The difference of atomic size between alloying elements and the matrix is essential for the solution strengthening effect. The solution treatment increases the intragranular solid-solution capacity of the solute and effectively increases the intragranular solid-solution amount of solute elements in the $\alpha(\text{Al})$ matrix. The increased degree of supersaturated solid-solution provides the driving force of precipitates in the aging process [35]. Therefore, the YS of the as-deposited sample increases obviously during the solution treatment. As for the cold-rolled samples, the dislocation density increases with the cold rolling process, however, it will be decreased at high solution temperature through the dislocation climb and cross slip. The accumulated dislocation density in the cold

rolling process will be recovered in the solution treatment, thus resulting in the decreased YS compared to that before solution process.

It is known that the strength of the material is in inverse to its average grain size based on Hall-Petch equation. The equiaxed grains in WAAM deposition change to banded structure after cold rolling, which cannot be described with the average grain size. The banded structure changes to equiaxed grains after T6 heat treatment. As shown in Fig.5, the average grain size of cold-rolled sample with T6 heat treatment is 58.4 μm compared to that of 72.8 μm for as-deposited sample. However, there is no difference of strengths between as-deposited and cold-rolled samples with T6 heat treatment, indicating that the grain refinement does not contribute to the strengths enhancement.

As shown in Table 1, the hardness increments are 7 HV, 12 HV and 27 HV with the pre-stretched strain of 3%, 5% and 10% of the as-deposited sample compared to the solution treated condition, respectively. Meanwhile, these hardness increments are 8 HV, 13 HV and 28 HV for the cold-rolled sample. It can be seen that the pre-stretched strain shows equal hardness increment for both conditions before aging treatment. It is known that the YS of the material is positively related to the hardness. As shown in Table 2, the pre-stretched strain shows equal strengthening effect on both conditions before aging treatment as the increments of YS are nearly the same considering the error bar. The pre-stretched strain increases the dislocation density inside the material and then enhances the strengths, thus is the strain induced strengthening. The high-density dislocations were shown to effectively break the coarse second phase particles, the degree of supersaturation of Cu atoms in Al matrix was greatly improved, which caused a greater driving force for precipitation of the θ' phase during the subsequent aging process [36-37].

Nevertheless, the effect of pre-stretched strain on the strengthening effect is quite complex after aging treatment. First, the increments of hardness and YS for cold-rolled samples are higher than that in as-deposited condition after aging treatment, which is different to that before aging treatment. Additionally, the hardness and strengths of as-deposited sample both increase with the increased pre-stretched strain after aging process, while they first

increase and then decrease in the cold-rolled condition. As a result, the maximum values locate with the pre-stretched strain of 5% for cold-rolled sample. It implies that the combination of cold rolling, pre-stretched strain and the subsequent aging will result in complex strengthening mechanism.

As discussed above, there may be interactions between the inter-layer cold rolling, pre-stretching process and aging treatment, thus resulting in complex strengthening mechanism. In order to investigate this, the morphology and size of the strengthening phases are examined by TEM. As shown in Fig.9, the dislocation density significantly increases with the increased pre-stretched strain. Dislocation lines and a small portion of tangled dislocations appear with no pre-stretched strain, while large amount of tangled dislocations and dislocation cells start to appear with the increased pre-stretched strain. Dislocation cells and even dislocation walls play a dominant role in the sample with the pre-stretched strain of 10%. It can be seen that the pre-stretched strain contributes to the clustering and migration of vacancies to dislocations, inducing the formation of dislocation cells and dislocation walls, which results in the obvious increase of the dislocation density.

A few needle-like θ' phases distribute sporadically along the $\{001\}$ Al planes with plenty of small θ'' phases scattering in the matrix for the samples without pre-stretched strain, as shown in Fig.10 (a) and (b). The needle-like θ' phases orthogonally precipitate along a certain direction, which is induced by the stress orientation effect, as reported by Xu [38] and Yang [39]. It can be seen that the density of the θ' phase and θ'' phase is roughly the same for the as-deposited and cold-rolled samples, accounting for the equivalent strengths of two conditions after T6 treatment. The density of the θ' phase remarkably increases with the pre-stretched strain ranging from 0% to 5%, while the size of the θ' phase changes little with the length of 70 ~166 nm and the average thickness of 5 nm. The increased density of the θ' phase indicates that a large part of the θ'' phases have transformed into the θ' phases with the increased pre-stretched strain compared to that in T6 treatment. It is obvious that the density of the θ' phase in cold-rolled condition is larger than that of as-deposited sample with the same pre-stretched strain. Coarse θ' phases

are generated for both the as-deposited and cold-rolled samples with the pre-stretched strain of 10%. The acicular θ' phases change to rod-like precipitates with the average thickness increasing from 5 ~ 8 nm to 15.5 nm for the as-deposited and cold-rolled conditions, respectively.

The precipitated θ' phase and θ'' phase contribute to the strengths enhancement as the strengths for both conditions increase significantly after aging process. These strengthening phases show pinning effect on the dislocations inside the material and larger stress needs to be applied for the dislocation slip; as a result, the material is strengthened. As shown in Fig.11 (a), the precipitates preferentially form around the dislocation pile-ups or the positions with high dislocation density, indicating that the dislocation provides advantageous nucleation sites for the θ' phase and θ'' phase. For the samples without pre-stretched strain in T6 treatment, there are only a small number of defects generated in the quenching stage, such as the vacancy and dislocation. The precipitated phases preferentially form at these positions with high free energy during the subsequent aging stage. Increased pre-stretched strain significantly increases the dislocation density, which provides more nucleation sites for the precipitates; as a result, the high-density uniformly distributed θ' phases are generated. The precipitation of Guinier-Preston zone (G.P. zones) and the transition to θ'' shifts to a lower temperature

when pre-stretched is performed [40]. The high density of dislocations developed during the stretching contributes to the acceleration in precipitation. Additionally, the uniformly distributed θ''/θ' -Al₂Cu particles effectively reduce the stress concentration in the plastic deformation process [41], and the elastic interaction between the particles and the dislocations is greatly reduced, which hinders the crack initiation and improves the strengths.

With further increase of the pre-stretched strain (to 10%), the accumulated dislocations provide driving force for the growth of the θ' phases, thus resulting in the obvious coarsening of θ' phases. It is interesting that there is little small θ'' phase scattering around the coarsened θ' phase as shown in Fig.11. The growth of the θ' phases is at the expense of small θ'' phases, as reported by Werz [42]. Based on Orowan strengthening mechanism [43], high density and tiny dispersed strengthening phases show positive effect on the strengths enhancement. As shown in Fig.10, the density of the θ' phases obviously increases with the pre-stretched strain ranging from 0% to 5% despite the unchanged length, thus significantly enhancing the strengths. Further increase of the pre-stretched strain induced severe coarsening of θ' phases, which in turn decreases the strengths. Therefore, the cold-rolled samples with the pre-stretched strain of 5% show the largest strengths.

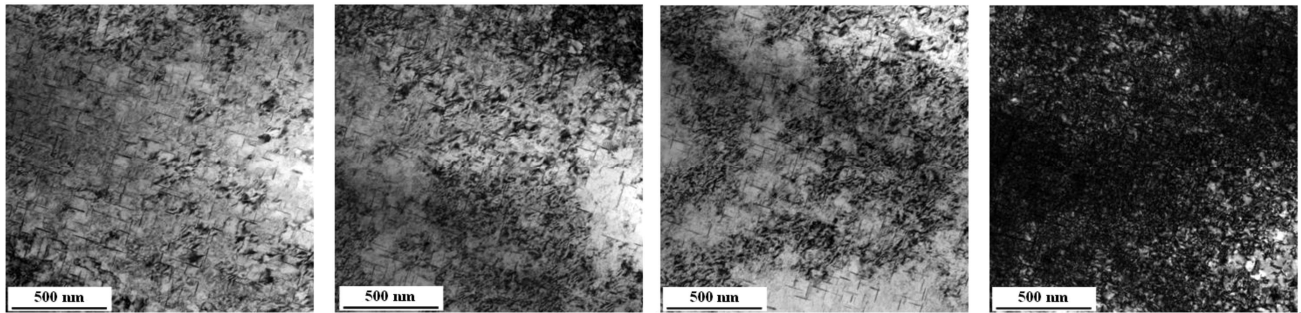
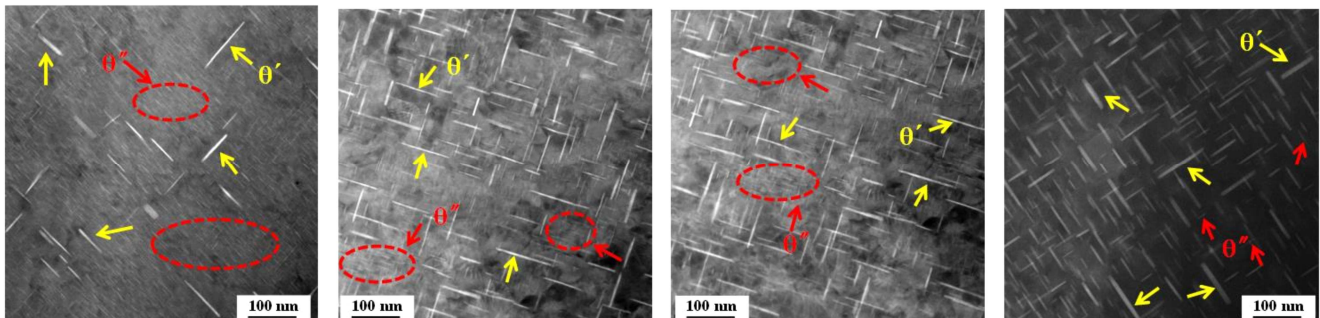


Fig.9 Effect of pre-stretched strain: (a) 0%; (b) 3%; (c) 5% and (d) 10% on dislocation after heat treatment



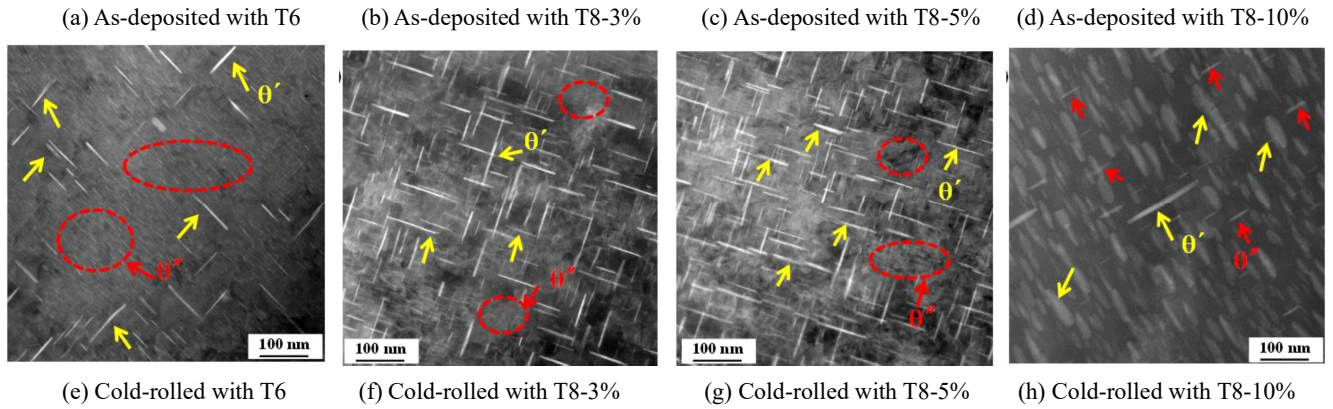


Fig.10 Effect of pre-stretched strain on precipitates distribution after heat treatment

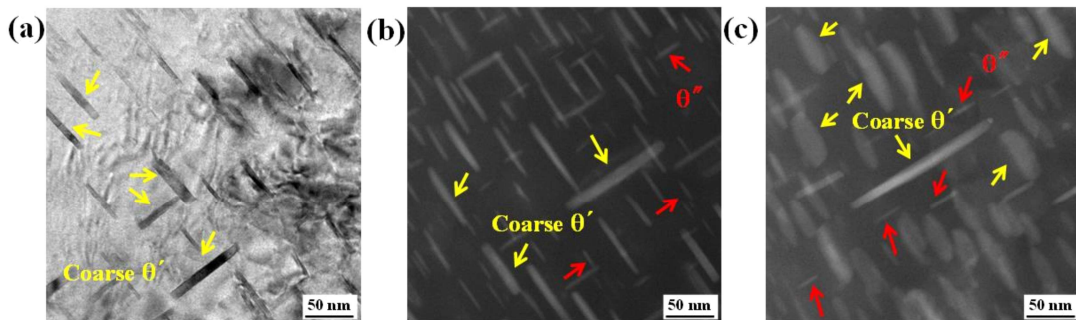


Fig.11 High magnification of precipitates distribution with 10% pre-stretched strain after T8 heat treatment: (a) TEM; (b) As-deposited sample; (c) Cold-rolled sample

4 Conclusions

Inter-layer cold rolling and thermo-mechanical heat treatment are adopted to refine the grains and enhance the strengths of WAAM deposited material. The effect of pre-stretched strain on the microstructure, hardness and tensile properties is analyzed.

1. The inter-layer cold rolling increases the hardness and strengths of the material. The pre-stretched strain induces equal increments of hardness and YS for both the as-deposited and cold-rolled conditions before aging treatment, while these increments of cold-rolled samples are larger than that in as-deposited condition after aging treatment.
2. Both T6 and T8 heat treatments increase the strengths and reduce the elongation. The increased pre-stretched strain increases the strengths and reduces the elongation after heat treatment for as-deposited samples. While the cold-rolled samples show best tensile properties with the pre-stretched strain of 5% after heat treatment, which exceeds the

properties of commercial aluminum plate with T87 treatment. The cold-rolled samples show better tensile properties than that of as-deposited samples with the same pre-stretched strain in T8 treatment.

3. The inter-layer cold rolling is beneficial for grain refinement and dispersive distribution of strengthening phases compared to that in as-deposited conditions. The samples with the pre-stretched strain of 5% show the smallest grain size. An optimized pre-stretched strain can further refine the grains and decrease the size of the strengthening phases.

4. Increased pre-stretched strain ranging from 0% to 5% is beneficial for the high-density and uniformly dispersed θ' phases, thus significantly enhancing the strengths. Further increase of the pre-stretched strain to 10% induces severe coarsening of θ' phases, which in turn decreases the strengths.

5. The strengthening mechanisms of integrated cold rolling and T8 heat treatment are discussed and its contribution to the strengths is revealed, including the strain induced strengthening, solution

strengthening, grain boundary strengthening and precipitation strengthening.

CRedit authorship contribution statement

Tao Zhang: conceptualization, experimentation, analysis and writing. Junwen Chen: methodology and analysis. Hai Gong and Yunxin Wu: conceptualization, resources and reviews. Xin Chen: experimentation assistance of rolling.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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