Enhanced Plastic Stability: Achieving High Performance in a Al6xxx Alloy Fabricated by Additive Manufacturing

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Abstract

Additive manufacturing (AM) facilitates the creation of materials with unique microstructural features and distinctive phenomena as compared to conventional manufacturing methods.

Among the various well-fabricated AM alloys, aluminum alloys garner substantial attention due to their extensive applications in the automotive and aerospace industries. In this work, an Al6xxx alloy is successfully fabricated with outstanding performance. A nucleation agent is introduced to diminish the susceptibility to cracking during the AM process, thereby inducing a heterogeneous microstructure in this alloy. However, the introduction of ultrafine grains induces plastic instability, evidenced by the presence of Lüders band. This work investigates the evolution of the Lüders band and the strategy to reduce their undesirable effect. The heterogeneity destabilizes the band propagation and thus deteriorates the ductility. Through a T6 heat treatment, the local Lüders strain decreases from 10.0% to 6.2%, leading to a substantial enhancement in plastic stability. With the increase in grain growth and the enlargement of coarse grain regions, the mismatch between the local and macroscopic Lüders strain disappears. Importantly, the strength and the thermal conductivity are concurrently increased. The findings demonstrate the significance of ensuring plastic stability to achieve improved strength-ductility trade-off in AM alloys with heterogeneous microstructures.

1. Introduction

Additive manufacturing (AM), commonly referred to as 3D printing, stands as a pivotal strategic technology driving innovation and ensuring sustainability.[1] AM technologies offer the remarkable capability of near net-shape component fabrication, which heralds a revolution in both design and manufacturing.[2] Laser powder bed fusion (LPBF), one of the most promising AM techniques, utilizes a laser to selectively melt a pre-laid metallic powder bed layer by layer.[3] LPBF provides the ability to finely adjust composition and microstructure at a very small scale.[4] Additionally, laser-induced ultrarapid solidification facilitates the creation of profoundly nonequilibrium microstructures.[5] Owing to the unique microstructure, the LPBF alloys exhibit exceptional performance across various aspects, including mechanical properties,[6, 7] oxidation resistance,[8] and other functional attributes.[9] These advanced achievements suggest the potential for further enhancing various materials through LPBF, which in turn attracts researchers due to the promising opportunities it offers.

Wrought aluminum alloys, including 2xxx, 6xxx, and 7xxx series, find extensive applications in the aerospace and automobile industries due to their high specific strength, corrosion resistance, and cost-effectiveness.[10, 11] As the demand for geometrically complex structures in these fields grows, LPBF of wrought aluminum alloys is gaining significant attention from both academia and industry. However, unlike the widely investigated near-

eutectic Al-Si alloys such as AlSi10Mg and Al12Si, which exhibit a minimal solidification range, LPBF of wrought aluminum alloys such as Al2024, Al6061, and Al7075 face challenges due to their susceptibility to cracking caused by a steep drop in temperature near the end of solidification.[12-16] For instance, through processing manipulation, the production of crack-free Al6061 using LPBF necessitates an exceedingly high preheating temperature of 500°C.[17] Luckily, significant advancements have been made through the introduction of nucleating agents like Al_3X (X= Zr, Sc, Nb, Ta, or Hf).[18-20] After functionalization with nucleating agents, crack-free samples with heterogeneous microstructures can be obtained.[21, 22] Crack free Al6xxx alloys have been achieved by incorporating various forms of Zr, including mixed with Yttrium Stabilized Zirconia,[15] ZrH_2,[19] and pre-alloying with Zr.[23] The 2xxx and 7xxx aluminum alloys with superior mechanical properties have been developed after the modification.[24, 25] The heterogeneous microstructure has also been considered as the reason for the improved strength-ductility trade-off.[26] However, with the introduction of ultrafine grains (UFGs), the well-known phenomenon of plastic instability, the Lüders band, has commonly been observed in LPBF-fabricated modified wrought aluminum alloys.[24, 27]

Lüders band is a plastic instability phenomenon characterized by localized strain regions that start from the yield point drop and correspond to the stress plateau stage.[28] This plastic instability phenomenon has been found in a wide range of materials, including steels,[29] aluminum alloys,[30] Cu,[31] shape memory alloys,[32] etc. For LPBF-fabricated wrought aluminum alloys, the presence of the Lüders band is commonly attributed to insufficient mobile dislocations.[24, 25, 33] Both the high volume fraction of grain boundaries in the UFG regions and the high concentration of solute atoms serve as obstacles to dislocation motion, hence limiting dislocation slip. Although the presence of the Lüders band in LPBF-fabricated aluminum alloys has been reported, its effect on the mechanical properties remains unknown.

The plasticity of metals is a fundamental and crucial property in structural engineering.[34] Achieving plastic stability is an ideal aspect for enhancing the material's performance. Generally, the Lüders band is considered an obstacle to the application due to the localized strain caused during deformation.[29] However, a stable propagation of the Lüders band can contribute to more than half of the total elongation, resulting in a favorable combination of strength and ductility.[29, 35-37] Hereby, the question arises: Is the presence of the Lüders band beneficial or detrimental to the performance of LPBF-fabricated high strength aluminum alloys? To address the question and gain insights into the non-uniform deformation, as well as to assess the structural integrity, it is essential to study the kinetics of the Lüders band in LPBF-fabricated aluminum alloys.

In this work, we aim to understand the importance of plastic stability on the mechanical properties of the LPBF-fabricated wrought aluminum alloys. Al6061, which is a 6xxx series alloy widely used in the heat exchanger industry due to its good balance between the mechanical properties and the thermal conductivity,[38] is chosen as the matrix material. Sc and Zr are added into the alloy to enhance the printability. The laser defocusing distance is changed to tailor the UFG fraction. Nevertheless, the plastic instability is observed in all the samples, irrespective of their ultrafine grain fraction. The occurrence of unstable band propagation is found, attributed to the heterogeneous microstructure generated during LPBF. We look for multiple heat treatment profiles to eliminate the Lüders band during the deformation to enhance the stability of the materials. By applying a T6 treatment, the plastic stability is enhanced.

We demonstrate the presence, negative effect, and potential solution of the plastic instability in an LPBF-fabricated heterogeneous microstructural Al6xxx alloy. These findings may be also applied to other high-strength aluminum alloys fabricated by LPBF, which possess the heterogeneous microstructures. Considering the substantial reduction in elongation caused by plastic instability, it is crucial to weaken or even eliminate the Lüders band phenomenon, especially in situations where ductile materials are required.

2. Results

2.1 Presence of Lüders band

The suppression of cracking in a LPBF-fabricated modified wrought aluminum alloy relies on the introduction of the UFGs. All the samples presented and discussed in this work are crack-free. **Figure 1**a shows the grain morphology of the samples printed with different laser defocusing distances. All the samples display a heterogeneous microstructure with the UFGs at the melt pool edge and the coarse grains (CGs) at the melt pool center. The UFGs are generated through heterogeneous nucleation, a process dependent on the nucleating agents - mostly Al_3Sc, as there is no obvious Zr segregation shown in the energy dispersive spectroscopy (EDS) maps (**Figure 1**b). The fast Fourier transform (FFT) pattern (**Figure 1**c) shows the superlattice reflections from $L1_2$-structured phase. The interfacial coherence between the Al matrix and the $L1_2$-structure Al_3Sc is proven by the high-resolution transmission electron microscopy (HRTEM, **Figure 1**c), which demonstrates the effectiveness of the nucleating agent. Within a conductive melt pool, the cooling rate increases from the edge to the center, resulting in the formation of the UFGs at the melt pool edge and the CGs at the

melt pool center. At the melt pool edge, the average size of the UFGs is ≈600 nm, but it increases as it gets closer to the center. As the defocusing distance increases, the fraction of the UFGs increases. The fraction increases from 52% to 81% when the defocusing distance (Δz) increases from + 0.5 to + 6.5 mm. The laser defocusing diverges the laser beam, which leads to a flat laser intensity distribution, resulting in a reduction in the melt pool ratio.[39] By increasing the defocusing distance, the cooling rate and the thermal gradient significantly decrease.[40] As the cooling rate decreases, the fraction of locations where Al_3Sc nucleates as the nucleation site increases.[41] Hence, the fraction of the UFGs increases along with the defocusing distances. The average grain size decreases with the increase in the UFG grain regions, as well as with the defocusing distance.

As one of the consequences of the introduction of the UFGs, all the samples show the evidence of the Lüders band regardless of the UFG fraction (**Figure 1**d). During tension, the stress drops after the sample yields, which is due to the activation of the Lüders band. There is a stress plateau followed, which corresponds to the propagation of the Lüders band.[42] The macroscopic Lüders strain (ε_L) is characterized by the macroscopic strain at the termination of the yield stress plateau. As the defocusing distance increases from +0.5 to +6.5 mm, the macroscopic Lüders strain decreases (**Figure 1**e). After the yield stress plateau, the samples show a significantly weak work hardening ability before fracture. The macroscopic Lüders strain was generally reported to be negatively correlated with the grain size.[31, 43, 44] However, this trend has not been observed in this study. To gain insight into the underlying mechanism of this abnormal phenomenon, we investigate the Lüders band kinetics.

2.2 Lüders band kinetics

Since the Lüders band kinetics are affected by the strain rate,[45] and the effect varies among different microstructures or alloys,[46, 47] the strain rate is chosen as the variable during the in-situ tensile tests to examine the stability of the Lüders band propagation. From the respective stress-time curves of the tensile test (Figure S1a-c), all the curves demonstrate a load drop after the onset of the plastic deformation. A yield stress plateau follows the stress drop, suggesting the presence of the Lüders band. The results are similar with the phenomenon described in the last section regardless of the strain rate. However, the yield point elongation (YPE) increases from ≈4% to ≈6% as the strain rate increases from 1.5×10^{-4} s^{-1} to 2.9×10^{-3} s^{-1}. The total elongation shows an obvious change while the strain rate increases, and the trend

is not monotonous (**Figure 2**i). The total elongations are 16.7 ± 1.9 %, 21.2 ± 1.3%, and 18.3 ± 1.7 %, respectively.

Batch I observations (Figure S1a-c, **Figure 2**a-c) capture the initiation and the propagation of the Lüders band. In Batch II observations, five equidistant points (P1-P5, **Figure 3**a-c) were selected to monitor the local strain evolution. If the necking region was not captured in the five points, an additional point (PM) was added. Combining both the observations, five deformation stages can be identified.

The first stage corresponds to the period before yielding. The overall local strain stays in a low level. The second stage corresponds to the initiation of the Lüders band. The Lüders band initiates when the stress drop appears, which is proven by the bright lines appeared on the specimens (second insert photos in Figure S1a-c). The Lüders band initiates at the location close to one shoulder and mainly propagates to another shoulder in the tensile specimens. The local strain increases accordingly (**Figure 3**a-c).

The third stage corresponds to the propagation of the Lüders band. The yield point elongation (YPE) does not always correspond to the full propagation of the Lüders band (Figure S1d-f). The propagation of the Lüders band may undergo two substages. For the first substage, the stress plateau corresponds to the partial propagation of the Lüders band. In this stage, all the specimens exhibit a sharp decrease in propagation speed after the initiation of the Lüders band, and then remain constant (**Figure 2**d-f). The average constant propagation speeds are 10, 86, and 248 μm/s, respectively. By comparing with the crosshead speed, the similarity of the propagation can be identified (**Figure 2**g). The initial average relative speed (=propagation speed/crosshead speed) is ≈20. The average constant propagation speed is positively correlated to the strain rate, which is ≈10 times the crosshead speed in all specimens. The local strain increases when the Lüders band is passing (**Figure 3**d-f). At a specific strain rate, all points exhibit the similar maximum local strain rates during the Lüders band propagation (Figure S2a-c). By dividing the strain rate, the relative maximum location strain rate is found irrelevant to the strain rate, which is ≈400% (Figure S2d-f). The local strain stops rapid increasing after Lüders band passed. Hence, the local strain rates decrease. For the most points, it will decrease to 0. For locations where the Lüders band has passed, the local strain is similar. Except the points at the shoulder (P1&P5), the local strain for the location where the Lüders band passed remains constant (10-12%) regardless of the strain rate. This constant value is called as local Lüders strain (ε_{LL}). For the second substage, the Lüders band still propagates after the stress plateau stage, which is abnormal comparing with what is typically described in the literature.[28, 29, 42] In this stage, the stable propagation cannot be maintained, manifesting as

a decreasing propagation speed. When the speed decreases to 0, the propagation of the Lüders band comes to an end. However, not all specimens experience a completed propagation of the Lüders band throughout the gauge length area. The local strain evolution keeps the same as the previous stage until necking occurs.

The fourth stage corresponds to the necking. Sharp brightness is observed near the initiation of the bands, which corresponds to the necking of the specimens. The brighter points are firstly observed over the bands, and then they become wider and coalesce. A sharp increase in the local strain rate can be found in the points in necking region (red line in Figure S2a-c), which corresponding to stress concentration. After necking, the local strain in other regions stops increasing, even though the Lüders band propagation is yet to complete. The final stage is fracture, which takes place when the local strain reaches ≈80%.

The local Lüders strain is larger than the macroscopic Lüders strain, which is abnormal. If we assume that the Lüders band fully propagated within the stress plateau range, the Lüders strain would be equal to the local Lüders strain (or slightly lower when considered the lower local strain value at the shoulder). The discrepancy between the two strains aligns with the observation that the Lüders band propagation has not been completed within the stress plateau range. In **Figure 3**d and f, some points show a lower value or even close to 0 in maximum local strain, which is attributed to the fact that the Lüders band is passing or has not passed the monitoring points. The strain rate influences the total elongation by affecting the termination of Lüders band propagation. **Figure 2**h suggests that the total elongation is proportional to the Lüders band propagation ratio. Therefore, it can be concluded that the Lüders band contributes to the plasticity of the LPBF-fabricated modified Al6061. However, this contribution is unstable due to the potential suspension of the Lüders band propagation. The suspension of the Lüders band propagation depends on the loading conditions, and the partial band propagation results in significant loss in the ductility. Therefore, the presence of the Lüders band is undesirable in this material during applications, especially in scenarios where ductile behavior is required.

2.3 Weakening of the Lüders band

As the Lüders band appears after the introduction of the UFGs, the most straightforward method to weaken or eliminate it is to increase the grain size in the samples, thereby weakening the effect of the UFGs. As Al6061 is a heat-treatable alloys, we investigate the effect of six typical heat treatment profiles on the plastic deformation behavior.

The direct aging (DA) treatment is specifically designed for the aluminum alloys which is modified with Sc/Zr. After DA, the changes in the grain size are negligible. No decrease in the fraction of UFG regions is observed (**Figure 4**a). The UFGs are retained with a mean size of 500-600 nm, which is similar to that of the as-built samples. At a large scale, Zr segregation becomes apparent, presenting a notable difference from the as-built samples (Figure S3). At the HRTEM scale, the results indicate the nucleation of the nano-scaled secondary Al_3(Sc, Zr) particles (**Figure 4**b), which is the main strengthening phase in the DA samples.

The grain growth of the solution treated samples (ST) depends on the temperature. With the increasing temperature, the grain growth becomes more obvious. The solution + artificial aging (STA) treated sample exhibits same trend (**Figure 4**a). For the samples solution treated under 480 ºC, there is no significant grain growth, indicating the temperature is not high enough or the duration is not long enough to result in the obvious grain growth. For the samples solution treated under 520 ºC, the holding temperature for the solution treatment is high enough to trigger the UFG growth. However, the grain growth is not homogenous. While partial grains grow, there are still quite a few UFGs, which is attributed to the low solution temperature and short duration. For the samples solution treated under 550 ºC, the resulting significant grain growth is more homogenous. The Al_3(Sc, Zr) maintains the $L1_2$ structure (**Figure 4**c, d). The Thermo-Calc calculation results indicate that the Mg_2Si will be fully dissolved at ≈519 ºC (Figure S4a). From the XRD results (Figure S4b), the Mg_2Si (111) and (422) peaks disappear when solution-treated at 520 ºC and 550 ºC while they remain when solution-treated at 480 ºC. The intensity of the (220) peak decreases after the solution treated at 520 ºC and 550 ºC. The ratios of the total peak intensity of Mg_2Si to the total peak intensity of α-Al are 0.81, 0.98, 0.23, 0.22 in AB, ST-L, ST-M, ST-H samples. The drops in the ratio indicate the dissolve in Mg_2Si. As the ratios are comparable in the ST-M and ST-H samples, we believe that the solution treatment is complete after solution treated at 520 ºC for 0.5 h. After artificial aging treatment, needle-shaped precipitates are observed. Based on the fast Fourier transformation (FFT) results, streaks of β" are present in both STA-1 and STA-2 samples, representing the nucleation of the β" phase during aging (**Figure 4**c, d). The EDS results (Figure S3) also shows the Zr segregation in STA-2 sample. However, the Mg segregation is not visible in the map, which is due to the small thickness of the β" precipitates.

After the heat-treatment, all the samples show the improved strength. However, most samples still show the Lüders band characteristics (**Figure 4**e). The DA samples only show work-softening after yielding. The in-situ tensile test (Figure S5) shows that the Lüders band forms but fails to propagate further. The yield strength (YS) and ultimate tensile strength (UTS)

are 361 ± 15 MPa and 379 ± 18 MPa, respectively. The improved strength is due to the modulus hardening from the secondary Al_3(Sc, Zr) nucleated during the aging process.[27] The total elongation is only 10.4 ± 1.3 %. The DA samples fabricated under Δz= +6.5 mm demonstrate the similar working softening phenomenon, with the YS of 349 ± 6 MPa and UTS of 375 ± 7 MPa. The statistical analysis indicates that these two samples yield similar values for YS and UTS, with *p*-values of 0.46 and 0.54, respectively. With a higher fraction of UFGs, Sc and Zr trapped in the matrix are less. The nucleation of the secondary Al_3(Sc, Zr) relied on the trapped Sc and Zr. Therefore, there is an offset effect between the grain boundary strengthening and the precipitation strengthening, which is the reason for the comparable strength. For the ST samples, only ST-L and ST-M samples show the Lüders band. The macroscopic Lüders strain decreases to 5-6%. The degree of strain reduction depends on the holding temperature, with higher holding temperatures resulting in lower strains. The in-situ tensile test demonstrates that the local Lüders strain is similar with the macroscopic Lüders strain (Figure S5), which means the mismatch-induced instability is eliminated. The decreased local Lüders strain also suggests that the Lüders band becomes weaker. Additionally, the work-hardening ability increases. After the completion of the Lüders band propagation, the local strain increases uniformly before necking. With the highest temperature (550 ºC) for the solution treatment, the Lüders band is eliminated. The ST-H samples show the best work-hardening ability but the lowest YS (192 ± 4 MPa). The UTS (328 ± 3 MPa) and total elongation (19.9 ± 0.9 %) are maintained when compared to the as-built samples. For the STA-1 samples, after artificial aging, the macroscopic Lüders strain slightly increases (≈6%). The in-situ tensile test shows that the local Lüders strain is similar to the macroscopic Lüders strain (Figure S5), which indicates a weakened Lüders band phenomenon when compared to the as-built samples. The STA-1 samples show the highest UTS (386 ± 6 MPa) and YS (373 ± 12 MPa), while the total elongation is 16.1 ± 0.9 %. For STA-2 samples, there is no Lüders band observed. After artificial aging, the YS and the UTS improve to 294 ± 5 MPa and 339 ± 5 MPa, respectively, while the elongation drops to 15.3 ± 1.3 %. β" precipitates provide significant strengthening contributions from modulus strengthening, chemical strengthening, and coherency strengthening,[48] which are the reasons for the improved strength.

As thermal conductivity is important when the material is used as the heat exchanger, the associated effect of the heat treatment is clarified. The as-built sample demonstrates a thermal conductivity of 133 ± 1 W m^{-1} K^{-1}, which is the lowest among all the samples. The ST sample shows a thermal conductivity of 150 ± 3 W m^{-1} K^{-1}, which is not affected by the solution temperature. The DA sample shows a good thermal conductivity of 159 ± 6 W m^{-1} K^{-1}, while

the STA-1 sample shows a comparable thermal conductivity of 160 ± 5 W m^{-1} K^{-1}. The solute redistribution caused by the DA treatment contributes to the improvement in thermal conductivity.[38] For the ST samples, during the solution treatment, the dissolution of Mg and Si can cause lattice distortion, negatively impacting thermal conductivity. However, the precipitation of Sc and Zr during this process leads to an improvement in thermal conductivity. Therefore, the ST samples exhibit higher thermal conductivity than the as-built samples, but lower thermal conductivity compared to the DA samples. By further artificial aging treatment, the solute redistributions of Mg and Si take place, which results in an improved thermal conductivity for the STA samples. The results indicate the modification for the Lüders band via heat treatment can also contribute to the thermal conductivity improvement.

3. Discussion

The observed abnormal relationship between the laser defocusing distance and the macroscopic Lüders strain can be attributed to the presence of a heterogeneous microstructure. The crystal plasticity simulation results (**Figure 5**a) show the differences in the stress concentration between the UFG regions and CG regions. The strength of the slip system varies depending on the grain size, with greater strength observed in the UFG regions. Conversely, the von-Mises stress is lower in the CG regions, indicating that a significant portion of the plasticity is likely to be localized in these regions (or along the interface). With the increase in the strain, the distinction in von-Mises stress between the UFG regions and the CG regions increases, which indicates the hardening in the UFG regions. The UFG regions are interconnected as the melt pools are well-bonded, and thus the CG regions are isolated. The deformation of the CG regions is constrained by the UFG regions. With the same local strain (after band swept), dislocations generate easily in the CG regions (soft). Thus, dislocations are accumulated in the CG regions, and a few stacking faults are observed to from associated with the large lattice dilatation due to the massive accumulation of plastic strain (**Figure 5**b). Meanwhile, the UFG regions have a poor ability to accumulate the dislocation due to the interaction between the dislocations and the grain boundaries. The hardening in the UFG regions is attributed to the necessary extra stress to activate new dislocation source to accommodate strain.[49] After band swept, the dislocation increment can be observed in both CG and UFG regions (Figure S6). A <111>//TD (tensile direction) texture can be observed, indicating lattice rotation in both UFG regions and CG regions (**Figure 5**c). By comparing the pole density between the global area and the UFG regions, it is evident that lattice rotation

occurs more readily in the CG regions, which is consistent with the simulation results. In addition, no twin boundaries are generated during the deformation. Further deformation will increase the <111>//TD texture intensity. The maximum pole density of 18.04 was found near the fracture surface area (Figure S7).

During the propagation of the Lüders band, the band passes through the region where sufficient mobile dislocations have accumulated. Given the presence of a limited fraction of UFGs, it requires only a minor localized Lüders strain, as CG regions readily accumulate a significant number of dislocations. As the fraction of the UFGs increases, the local Lüders strain increases for the sufficient dislocation accumulation. If the band fully propagates within the yield stress plateau range, the Lüders strain increases along with the UFG fraction. However, as observed before, the full propagation of the Lüders band does not always occur. In the LPBF-fabricated heterogeneous microstructural samples, the UFG regions are interconnected. As the UFG regions increase and occupy most of the samples, the microstructure can be considered as a lamellar structure with a heterogeneous microstructural layer (CG + UFG) and a near-full UFG layer. Consequently, the misfit between these two layers becomes significant and cannot be ignored. The near-full UFG layer needs more stress to activate new dislocation source to accommodate strain. When the Lüders band is passing through a certain area, the required flow stress for the near-full UFG layer is expected to be higher than that of the heterogeneous microstructural layer. The potential increase in flow stress at the interface of the layers results in the instability of band propagation, and global work hardening may occur before the full propagation of the Lüders band. The likelihood of this occurrence increases with an increase in the near-full UFG layer width. Hence, the Lüders strain decreases as the UFG fractions increases. A larger strain rate corresponds to a faster band propagation rate, the difference in the band interaction with the microstructure between the different layers can be more easily compensated. Conversely, the instability becomes stronger as there is more interacting time for the misfit-induced early global work hardening, especially at lower strain rates. Consequently, the Lüders strain increases along with the strain rate. The cessation of Lüders band propagation occurs between the initiation of work hardening and the completion of the Lüders band propagation. The cessation is associated with the necking, which corresponds to the stress concentration in the deformed area. The increased flow stress may lead to additional dislocation generation in the deformed area, which is one of the reasons for the necking phenomenon. The samples subjected to the lowest strain rate exhibit the earliest initiation of the work hardening stage, making them more susceptible to necking. On the other hand, the dislocation generation is directly proportional to the strain rate. From Kernel average

misorientation (KAM) map (Figure S6), the global geometrically necessary dislocation density increases from 1×10^{14} m^{-2} to 1.41×10^{14} m^{-2} (1.5×10^{-4} s^{-1}) or 1.52×10^{14} m^{-2} (2.9×10^{-3} s^{-1}). High dislocation density is another factor which induces the necking. Therefore, both the samples under the smallest strain rate and the highest strain rate were observed to fail to complete the Lüders band propagation.

The changes in the Lüders band phenomenon caused by the heat treatments are attributed to the grain growth and the precipitate nucleation. For the DA treatment, as the holding temperature is not sufficiently high, the UFGs do not undergo an obvious growth. Due to the dislocation sink caused by the UFGs, the Lüders band initiates. As a result of the maintained UFGs, the local Lüders strain will not drop, or even increase due to the nucleation of the secondary Al_3(Sc, Zr). However, the high stress leads to the activation of the plastic instability, and thus the Lüders band is not able to propagate. A worse plastic stability is obtained. Therefore, in the DA samples, only work softening can be observed, resulting in the lowest elongation.

A larger grain size corresponds to a lower grain boundary density, which leads to reduced dislocation sinks during deformation. Therefore, the increase in grain size contributes to the weakening or elimination of the Lüders band. As the grain size increases along with the holding temperature, the moderate solution temperature (520 ºC) results in a weaker Lüders band phenomenon compared with the low solution temperature (480 ºC), manifesting a lower local Lüders strain. Further increasing the holding temperature to 550 ºC promotes extensive grain growth, which eventually lead to the elimination of the Lüders band. Owing to the nucleation of the β", the mobile dislocation density decreases, which enhances the Lüders band phenomenon. Therefore, a slight increase in the local Lüders strain is observed in the STA-1 samples compared to the ST-M samples. Nevertheless, the Lüders band in the STA-1 samples is still weaker than that in the as-built samples, which is due to the increased CGs. The STA-2 samples demonstrate no Lüders band due to the significant grain growth during the solution treatment.

An overall appreciation for the LPBF-fabricated Sc/Zr modified Al6061 is attained from the radar chart shown in **Figure 6**. We evaluate the material from the strength, hardness, plasticity, plastic stability, and thermal conductivity. The plastic stability is defined as $(1-\varepsilon_{LL}/\varepsilon_T)$. While DA treatment is a common choice for LPBF-fabricated Sc or Zr modified aluminum alloys,[20, 27, 41] it is not recommended for the current material as it demonstrates work softening and the worst plasticity, making it unsuitable for applications that require high ductility. For STA-1 samples, except the plasticity, all the remaining properties show

improvement compared to the as-built samples. The total elongation of 16.1% is sufficient for most applications where Al6061 is required. If the local Lüders strain is excluded from the total elongation, considering that the Lüders band may not fully propagate, it becomes evident that the strength-ductility trade-off improves after the STA-1 treatment. The strength increases while the remaining elongation remains the same. Therefore, STA-1 treatment is the most recommended heat treatment for the material among all the heat treatments investigated. Questions might arise when considering the extensive utilization of the DA treatment for nucleation of Al_3X in LPBF heat-treatable aluminum alloys: Why do we choose this base material, and what about the other elements originally present in the alloy? Our results demonstrate that STA treatment can be the preferred choice for LPBF-fabricated modified aluminum alloys. If the base material is a heat-treatable aluminum alloy, STA treatment can be considered as the potential optimal heat treatment profile. By comparing STA-1 and STA-2 samples, one of the key factors in achieving excellent mechanical properties through the STA heat treatment is the control of the grain growth during the solution treatment. Both temperature and duration need to be evaluated while choosing the solution treatment profile. The comparison between our results and those of other Al6061-based AM materials is available in **Figure 4**f.[17, 23, 38, 50, 51] After the STA-1 treatment, the alloy exhibits excellent mechanical properties in the AM Al6061 community.

4. Conclusion

In summary, our work discovers the importance of the enhanced plastic stability in a LPBF-fabricated Sc/Zr modified aluminum alloy. The as-built samples with the dominated UFGs result in the plastic instability. The plastic instability is observed to lead to a loss of up to 1/3 in elongation. The loss can further increase if the band propagation ratio decreases further. Our study reveals that the local Lüders strain can be larger than the macroscopic Lüders strain, indicating the continuous propagation of the Lüders band even after the initiation of work hardening. The increase in flow stress leads to an unstable Lüders band propagation, which may result in the failure to fully propagation of the Lüders band. Consequently, a sacrifice in the ductility can be expected. The instability of the Lüders band propagation is attributed to the lamellar structure comprising heterogeneous microstructure and near-full UFGs as the layers. For situations where the loading conditions are uncertain or ductility is required, weakening or eliminating the Lüders band in the LPBF-fabricated heterogeneous microstructural modified aluminum alloys is strongly recommended. By implementing a T6

heat treatment, grain growth is induced, and thus the plastic stability is enhanced. Remarkable improvements in strength, strength-ductility trade-off, and the thermal conductivity are achieved.

5. Experimental Section

Sample Preparation

The gas atomized pre-alloyed Sc/Zr modified Al6061 aluminum powder with the D50 of 39.4 μm was used as the starting materials in this work. The chemical composition of the powder, determined using inductively coupled plasma atomic emission spectroscopy (ICP-AES), was found to be Al-0.96Mg-0.64Si-0.52Sc-0.28Cu-0.26Zr-0.24Cr-0.08Fe (wt.%). The LPBF process was conducted using the ProX 300 System under the argon atmosphere. The cuboid samples were built with various laser defocusing distances. The definition and the detailed setting are shown in Figure S8. The measurements from Francis indicate that the default plane for the ProX 300 System is defocused with ≈1.5 mm and the Rayleigh length is 3-4 mm, depending on which kind of beam profile is used for fitting.[52] This result was used to deduce the changes in the laser defocusing distance. The selected defocusing planes consistently fall within the positive defocusing range, which correspond to the defocusing distances from +0.5 to +6.5 mm (+0.5, +2.5, +4.5, +6.5 mm). The latter two are larger than the Rayleigh length. By applying the fitted Gaussian beam profile,[52] the estimated laser power densities for the four conditions, $I=P/A$ (P is the laser power and A is the area of the laser beam), were 5.4, 3.4, 1.9, and 1.1 MW/cm^2, respectively. The laser power, scan speed (v), laser thickness (t), and hatching space (h) were kept constant during the process. The adopted laser volumetric energy density, $E=P/hvt$, was 441 J/mm^3. The XY scanning strategy was applied, representing that the scanning directions between adjacent layers were perpendicular to each other. There was no significant element loss after the LPBF process. The chemical composition of the as-built samples was Al-0.95Mg-0.59Si-0.53Sc-0.29Cu-0.27Zr-0.25Cr-0.08Fe (wt.%).

Three types of heat treatments were selected to study the weakening of the Lüders band: i) Direct aging (DA), ii) Solution treatment + natural aging (ST), and iii) the Solution treatment + artificial aging (STA, T6). The samples fabricated under Δz= +4.5 mm was chosen as the main set for heat treatments. For the DA treatment, samples were held at 400 ºC in air for 1 h and then cooled in the air. For the ST treatment, samples were held at 480 ºC (ST-L), 520 ºC (ST-M) or 550 ºC (ST-H) for 0.5 h in air and then quenched with the water at room temperature.

The samples were held at room temperature for 96 h before the tensile test. For the STA treatment, samples were held at 520 ºC (STA-1) or 550 ºC (STA-2) for 0.5 h in air and then quenched with the water at room temperature. Samples were subsequently aged at 175 ºC for 8 h. The samples fabricated under Δz= +6.5 mm was chosen as a comparison set. The same DA treatment was conducted on the comparison set. Thermo-Calc software 2023a with TCAl8 database was utilized to simulate the dissolution of the Mg_2Si phase.

Microstructural Characterization

The X-ray diffractometer (XRD, Panalytical Empyrean) was used to identify the phase dissolution of the ST treated samples. For microstructure analysis, a field emission scanning electron microscopy (FESEM, JEOL Model 7800) equipped with the Electron Backscatter Diffraction (EBSD) detector (symmetry, Oxford instrument) was used. All the data were analyzed via Channel 5 software. The EDS mapping was obtained by Transmission Kikuchi Diffraction-Electron Backscatter Diffraction (TKD-EBSD) on electron transparent lamellae. For precipitate and dislocation analysis, a transmission electron microscopy (TEM, JEOL JEM-ARM200F) was used. The geometric phase analysis (GPA) via the software Strain++ was used to evaluate the lattice dilatation.[53] The TEM lamellae for the deformed areas were prepared with a Focused Ion Beam (FIB, ZEISS SEM 550 Model). Other TEM lamellae and the TKD-EBSD lamellae were prepared using a Precision Ion Polishing System (PIPS).

Property Characterization

The tensile specimens were machined from blocks by electrical discharge machining, and then ground with abrasive SiC papers before the tensile tests. A universal Instron-5982 testing system equipped with a video extensometer to conduct tensile test was used. All the samples were tested under the strain rate of 1×10^{-3} s^{-1} at room temperature. Three samples were evaluated for each condition. A Laser Flash Apparatus (LFA, NETZSCH LFA 457 MicroFlash) was used to evaluate the thermal conductivity. Each test was replicated for three times. The density was measured via Archimedes' method. The specific heat capacity was calculated via the Thermo-Calc software 2023a with TCAl8 database. The Vickers' microhardness was performed on the mounted samples, and the load applied was 100 g with the holding time of 15 s. The result for the microhardness is shown in Table S1. Each value represented the average of 11 data points.

For the in-situ tensile test, the samples fabricated under Δz= +4.5 mm was chosen. The printed samples were machined into in-situ tensile specimens with the dimension shown in

Figure S9. After the standard polishing procedure to a 3 μm diamond surface finish, the in-situ tensile specimens were divided into two batches. Batch I was used for direct in-situ observation. To study the kinetic details of the Lüders band propagation, 10 photos of each specimen were chosen to calculate the average propagation speed. The interval of the photos was decided to be 3, 10, and 66s as the strain rate decreases. The propagation speed was measured at a single site, specifically the left side in **Figure 2**a-c. The Lüders band front was determined by identifying the point at which the pixel turned brighter. Batch II was used for digital image correlation (DIC) observation. The random speckle pattern was prepared by spraying white and black paints on the surface. The in-situ tensile test equipment was put in a dark room. An annular incandescent lamp was switched on during the in-situ tensile tests. The crosshead speeds of 1.375 mm/min (2.9×10^{-3} s^{-1}), 0.48 mm/min (1×10^{-3} s^{-1}), and 0.072 mm/min (1.5×10^{-4} s^{-1}) were used during the tests. An industrial camera was used to capture the images during the tests. The interval for the capturing is 1 s. The controls for the in-situ tensile tests and the photo capture were under the same computer but different software. Three samples were tested for each crosshead speed. The images captured from DIC batch were analyzed via the ZEISS GOM correlate software.

Synthetic Representative Volume Elements (Monte Carlo method)

Due to the unavailability of the complete 3D data from the experiments, it is not possible to fully digitalize the representative volume elements (RVEs) for numerical simulations. Instead, we opted to generate synthetic RVEs that encompassed the fundamental characteristics of the bimodal grain size distribution within the melt-pool. Utilizing the experimental EBSD maps in both the XY and XZ planes as a reference (Figure S10), we created a synthetic RVE that represented the bimodal grain size distribution. The generation of this RVE was accomplished through the implementation of the Potts Monte Carlo method.[54] The implementation is briefly described below.

Each element of the 64x64x64 RVE is initially assigned a random state (orientation). At each simulation time step, a randomly selected element is assigned a new state (orientation) and that state is accepted with a certain probability following the probability transition function:

$$P = \frac{p_0}{2}\left[1 - \tanh\left(\frac{\Delta E}{2kT}\right)\right] \quad (1)$$

where kT defines a thermal energy of the simulation and ΔE is the energy difference in the system between the previous and the current state. The Potts Hamiltonian is of the following form:

$$H = \sum_{i=1}^{N} \left\{ \sum_{j=1}^{n} \frac{\gamma(s_i, s_j)}{2} \right\} \quad (2)$$

where γ is the grain boundary energy between the two lattice sites s_i, s_j. Volumetric energy is neglected as no preferential grain growth is modelled.

To achieve bimodality, our approach involves two separate nucleation events occurring at different times. The second nucleation event is introduced at a specific time to ensure that the overall ratio of grain sizes resembles what is observed in the experimental EBSD data. It is important to note that the method here is employed purely as an RVE design tool. However, since the grain growth is driven by curvature, the resulting grain shapes appear more realistic compared to other methods. To complete the process, random Euler angles are assigned to the individual states which survived the growth process. The XY scanning strategy between the adjacent layers was also considered. The result is presented in Figure S10.

Crystal Plasticity Formulation

In this work, the finite strain crystal plasticity model was employed for FCC metals.[55] The details were presented below.

The kinematics of crystal plasticity is defined by the multiplicative decomposition of the deformation gradient into elastic and plastic part:

$$\boldsymbol{F} = \boldsymbol{F}^e \boldsymbol{F}^p \quad (3)$$

where the deformation gradient $\boldsymbol{F}=\boldsymbol{\nabla_0 x}$ and *det* $\boldsymbol{F} > 0$ with $\boldsymbol{x}$ being the spatial coordinate and $\boldsymbol{\nabla_0}$ being the gradient with respect to the material coordinate. The Green-Lagrange strain tensor as $\boldsymbol{E}=(\boldsymbol{F}^T\boldsymbol{F}-\boldsymbol{I})/2$ was further defined. The evolution equation for the plastic deformation gradient is defined by:

$$\dot{\boldsymbol{F}}^p \boldsymbol{F}^{p-1} = \sum_{\alpha} \dot{\gamma}^{\alpha} \vec{m}_0^{\alpha} \otimes \vec{n}_0^{\alpha} \quad (4)$$

where $\dot{\gamma}^{\alpha}$ are the shear rates on slip systems α, and $\vec{m}_0^{\alpha}$, $\vec{n}_0^{\alpha}$ are the slip direction and slip plane normal vectors in their initial configuration. For an FCC crystal lattice, these vectors are listed in Table S2. The kinetics of dislocation slip $\dot{\gamma}^{\alpha}$ is define by a power-law function:

$$\dot{\gamma}^{\alpha} = \dot{\gamma}_0^{\alpha} \left(\frac{|\tau^{\alpha}|}{s^{\alpha} + s_{HP}} \right)^{\frac{1}{m}} \text{sign}\,(\tau^{\alpha}) \quad (5)$$

where τ^{α} is the resolved shear stress, s^{α} is the slip resistance due to the intragranular dislocation interactions and s_{HP} is the slip resistance due to the Hall-Petch effect taking into

consideration the grain size. Here, $\dot{\gamma}_0^{\alpha}$ is the reference shearing rate and *m* is the rate sensitivity parameter. The resolve shear stress τ^{α} at each slip system can be calculated by:

$$\tau^{\alpha} \approx \boldsymbol{S}^{2PK} : \vec{m}_0^{\alpha} \otimes \vec{n}_0^{\alpha} \tag{6}$$

where the second Piola-Kirchhoff stress $\boldsymbol{S}^{2PK}$ is assumed to be linearly proportional to the elastic Green-Lagrange strain tensor $\mathbf{E^e}$:

$$\boldsymbol{S}^{2PK} = C : \boldsymbol{E}^{e} \tag{7}$$

The cubic elasticity was assumed, and therefore the elastic tensor is defined by three material parameters C_{11}, C_{12}, C_{44}. The Hall-Petch strength is defined by:

$$s_{HP} = \frac{k_{HP}}{\sqrt{d}} \tag{8}$$

$$d = \left(\frac{6V}{\pi}\right)^{\frac{1}{3}} \tag{9}$$

where k_{HP} is the Hall-Petch coefficient and *d* is the characteristic grain size calculated as Eq. (9), where *V* is the volume of the grain. The texture evolution can be conveniently captured by the elastic deformation gradient:

$$\vec{m}^{\alpha} = \boldsymbol{F}^{e} \vec{m}_0^{\alpha}, \qquad \vec{n}^{\alpha} = \boldsymbol{F}^{e^{-T}} \vec{n}_0^{\alpha} \tag{10}$$

The mechanical equilibrium equations $\boldsymbol{\nabla} \cdot \boldsymbol{\sigma} = \mathbf{0}$ are solved using the updated Lagrangian formulation and the crystal plasticity material model is integrated using a fully implicit scheme eliminating the requirement for a small-time step.

Statistical Analysis

Source data were obtained from the respective characterization device described in Section 5.3. All data obtained were used for statistical analysis without manipulation, as all data were considered valid (fractured within the gauge length area). Data were displayed as mean values ± SD. The sample size (*n*) was described in each figure caption. Statistical significance analysis was performed using one-way ANOVA. In all case, significance was defined as $p \leq 0.05$. Statistical analysis was carried out using Microsoft Excel.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data availability statement

The data that support the findings of this study are available from the corresponding author on reasonable request.

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Figure 1 a) Inverse pole figure (IPF) maps of the samples fabricated by different laser defocusing distances (Δz) showing the heterogeneous microstructure. b) Energy dispersive spectroscopy (EDS) maps of the samples fabricated under Δz=+4.5 mm showing the elemental segregation along the ultrafine grain (UFG) boundaries. c) High-resolution transmission electron microscopy (HRTEM) image and fast Fourier transform (FFT) pattern of the samples fabricated under Δz=+4.5 mm showing the coherent relationship between the matrix and the Al_3Sc. d) Representative tensile curves. e) Mechanical properties of the samples fabricated by different laser defocusing distances. Other than the macroscopic Lüders strain, the mechanical properties minorly change along with the defocusing distance. The yield strength (YS) of the samples is 302 ± 10 MPa (n=12, p=0.39), and the ultimate tensile strength (UTS) is 324 ± 12 MPa (n=12, p=0.21), while the total elongation (ε_T) is 20.4 ± 1.4 % (n=12, p=0.27). Statistical analysis was performed a one-way ANOVA. All data were displayed as mean values ± standard deviation (SD).

Figure 2 a-c) Captured images showing the Lüders band propagation in the in-situ tensile test at 1.5×10^{-4} s^{-1} a), 1×10^{-3} s^{-1} b), 2.9×10^{-3} s^{-1} c). d-f) Propagation speed during the band propagation in the in-situ tensile test at 1.5×10^{-4} s^{-1} d), 1×10^{-3} s^{-1} e), 2.9×10^{-3} s^{-1} f). g) Relative speed of the band propagation. h) Relationship between the band propagation ratio and the total elongation of the in-situ tensile tested samples. i) Relationship between the total elongation and the strain rate.

Figure 3 Contouring maps indicating the local strain distribution a-c) and local strain evolution for the selected points d-f) obtained by digital image correlation (DIC) analysis via in-situ tensile test at different strain rates: a, d) 1.5×10^{-4} s^{-1}, b, e) 1×10^{-3} s^{-1}, c, f) 2.9×10^{-3} s^{-1}.

Figure 4 a) IPF map for samples under different heat treatments. b) HRTEM result showing the coherent relationship between the matrix and the secondary Al_3(Sc, Zr) in DA sample. c) HRTEM result showing the β" and Al_3(Sc, Zr), and the coherent relationship between the matrix and the Al_3(Sc, Zr) in STA-1 sample. d) HRTEM results showing the β" and Al_3(Sc, Zr), and the coherent relationship between the matrix and the Al_3(Sc, Zr) in STA-2 sample. e) Representative stress-strain curves for the samples with different heat treatments (YS: n=21, p<0.0001; UTS: n=21, p<0.0001; EL: n=21, p<0.0001). Statistical analysis was performed a one-way ANOVA. All data were displayed as mean values ± SD. f) Yield strength (YS) and as

a function of total elongation for our samples plotted versus the literature data: Al6061,[17] ASM-Al6061-T6,[50] Al6061+Zr,[23, 38] Al6061+Ceramics.[51]

Figure 5 a) Slip system strength (I, II) and Von-Mises stress (III, IV) for the heterogeneous microstructural sample under tension with the strain of 1% (I, III) and 20% (II, IV). b) TEM results indicating the dislocation and stacking fault generation: (I) Bright field (BF) image showing the dislocations after band swept, (II) BF image showing the stacking fault after band swept, (III) HRTEM image showing the stacking fault, (IV) Geometric phase analysis (GPA) map conducted on Figure III using {111} frequencies. c) IPF//TD (tensile direction) maps and {111} pole figure before and after band swept: (I, III) Before tensile, (II, IV) after band swept, i.e., ≈10% local strain; (I, II) Overall regions, (III, IV) UFG regions.

Figure 6 Radar chart showing the feature comparison for the LPBF fabricated Sc/Zr modified Al6061. Reference used for deriving the chart: ASM-T6.[50]