



Research Article

Analysis of grain structure, precipitation and hardness heterogeneities, supported by a thermal model, for an aluminium alloy 7075 deposited by solid-state multi-layer friction surfacing

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ABSTRACT

Thermomechanical cycles during multi-layer friction surfacing (MLFS) cause microstructural and mechanical heterogeneities in the deposited high-strength Al alloy, 7075. The thermal profile and heat accumulation were investigated in this study using a multilayer numerical thermal model of the MLFS process; additionally, these variables were linked to experimentally observed microstructural heterogeneities. Compared with the feedstock, grain sizes decreased by 55–80 %. The mean grain size at the bottom and top areas of a given layer was finer than that in the middle of the layer because of the enhanced recrystallisation, which resulted from the friction and shear deformation experienced by the deposited material. The differences in the thermal cycle and plastic strain rate of the bottom and top areas along the layers resulted in a gradual increase in the grain size at the bottom of each layer and a reduction in the grain size at the top of each layer. The grain growth and continuous dynamic recrystallisation mechanisms are governed by the temperature and strain rate, those mechanisms determine the intra- and inter-layer grain sizes. The accumulated heat, owing to subsequent experimental deposition, resulted in excessive growth of the precipitates in the bottom layers. The strengthening of the solid-solution and Guinier-Preston zones significantly increased the microhardness of the top layer. Post-deposition T6 heat treatments confirmed the restoration of a uniform distribution of microhardness.

1. Introduction

The 7xxx series of aluminium alloys are precipitation-hardened alloys that are widely used in load-bearing components in aerospace applications, such as frames, spars, and stringers, owing to their high strength-to-weight ratio, good machinability, and high toughness [1]. For these applications, optimised structures facilitate a further reduction in aircraft weight and reduce energy consumption. Moreover, repetitive loading cycles can induce stress concentrations and lead to premature failure during the service of aircraft components or other aerospace components. Therefore, various additive manufacturing (AM) technologies, including fusion-based and solid-state ones, have been explored to additively repair or manufacture complex high-strength aluminium

components.

In fusion-based AM, such as laser powder bed fusion (LPBF) and wire arc additive manufacturing (WAAM), Martin *et al.* [2] applied a direct energy source to locally melt the alloy powders or wires. This process incorporates a high solidification rate between 0.1–5 m/s. Fusion-based AM continuously melts successive layers of powder or wire in a similar manner to that of welding. Therefore, this method is particularly suitable for “weldable” alloys. However, the use of “non-weldable” high performance 7xxx series aluminium alloys in fusion-based AM is challenging. Compared with other series of aluminium alloys, Zhou *et al.* [3] explained that the 7xxx series aluminium alloys have a wider solidification interval, greater tendency for hot cracking, higher alloying elemental content, and higher thermal conductivity. Therefore, the

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fusion-based AM of 7xxx series aluminium alloys increases the likelihood of elemental vaporisation (for example, 19 % of Mg and 33 % of Zn vaporisation during LPBF of AA7075) [4], and the production of microstructures that comprise columnar grains and are highly susceptible to hot cracking [5].

Thermomechanical processing-based solid-state AM technologies, such as cold spray, ultrasonic additive manufacturing, additive friction stir deposition (AFSD), lateral friction surfacing [6], and multilayer friction surfacing (MLFS) [7], have been recently developed. Melting of the alloy powders or wires does not occur during these solid-state AM processes; therefore, according to Karmakar *et al.* [7], the solidification defects can be avoided. Solid-state AM has been shown to produce parts with small nondendritic grains and fewer oxide residues than those produced using AM techniques that incorporate melting. Therefore, AM techniques that incorporate melting are generally subjected to reduced mechanical properties or residual tensile stresses, resulting in reduced fatigue life and susceptibility to corrosion. Among these processes, as stated by Karmakar *et al.* [7], AFSD and MLFS are known for their ability to utilise solid feedstock (such as rods or wires), metallic powder, or recycled machine chips [8] to build components additively, or repair complex-shaped components. They leverage the fundamental physics of friction stir welding (FSW) or friction stir processing (FSP) [9] to fabricate wrought-like microstructures that exhibit mechanical properties. A study by Lezaack *et al.* [10] showed that the stirred zone of a friction stirred AA7475 alloy exhibited a higher ductility (by 180 %) than that of the wrought 7475 alloy.

In AFSD and MLFS, layers are deposited by pressing a rotating consumable solid feedstock against a substrate. A layer is deposited when the consumable rod moves above the substrate. A strong metallurgical bond is formed through frictional heat and severe plastic deformation. During the AFSD process, the feed material is introduced through a hollow tool at the same height for each layer deposition, whereas the feed material is consumed using specific AM machines, such as the MELD® system developed and patented by Aeroprobe Corporation [11]. Compared with AFSD, MLFS directly uses a rotating consumable rod for material deposition without the use of a hollow tool; additionally, Prabhakar *et al.* [12] described that this method exhibits reduced deposition efficiency because of the presence of a flash around the consumable feedstock. MLFS is also prone to weak interfacial bonding and porosity between or inside the layers. This is particularly applicable to the sides parallel to the y-z plane of the deposition because the axial force is not fully transferred to the substrate owing to the absence of a tool shoulder, which, results in peripheral flashes on the rods [7]. However, a tool shoulder is utilised in the AFSD process. One of the key advantages of MLFS over AFSD is that it can be easily performed by any FSW machine or even using a computer numerical control (CNC) machining instrument. The process can then be combined with machining to improve the finish of the surface and dimensional precision, which can enhance the widespread use of MLFS in engineering applications.

Suhuddin *et al.* [13] concluded that the thermal cycles during MLFS result in microstructural changes and lead to grain size refinement owing to dynamic recrystallisation. Previous studies investigated the microstructure and mechanical properties of friction surfacing (FS) monolayers. In the experiment of Mukhopadhyay and Saha [14] with pure aluminium, the grain size reduced by 90 % of that of the feedstock material. While in the work of Ehrich *et al.* [15], a higher dislocation density led to a reduced grain size and higher fraction of high-angle grain boundaries in the FS AA6060 monolayers. As mentioned by Yu *et al.* [16], dynamic recrystallisation and grain refinement are primarily driven by continuous dynamic recrystallisation and discontinuous recrystallisation. Grain structure recrystallisation was considered a contributing factor to the enhancement of the material strength of non-heat-treated alloys. The shear strength of FS AA5083 increased with decreasing grain size, following the classical Hall-Petch relationship [17]. However, compared with FS monolayers, MLFS involves the

addition of multiple layers, which cause repetitive thermal cycles, potentially resulting in additional microstructural heterogeneities.

There have been several studies related to the multi-layer friction surfacing of aluminium alloys. Dilip and Ram [18] observed strengthening precipitates over the aging gradient in the MLFS of AA2014; however, the evolution of precipitates along the deposition height was not explained. The authors suggested that the alloying elements can be dissolved by solution heat treatment (SHT). However, the fine grains generally undergo abnormal grain growth during solution heat treatment at temperatures higher than 450 °C in aluminium alloys, which will influence the mechanical performances of the deposits [19]. Shen *et al.* [20] showed that all layers of MLFS AA5083 exhibited fine and equiaxed recrystallised grains, which were 4–5 µm in size. They concluded that multilayer deposition exhibits a homogenous microstructure over the height of the structure, which may occur because AA5083 is a none-heat-treatable alloy and thus less sensitive to the thermal history. However, the microstructure of only the middle of each layer was investigated in this study; therefore, the lower and upper portions of the same layer were not examined. Rath *et al.* [21] assessed the influence of successively deposited layers on the layer interface and anisotropy of MLFS AA5083. No significant gradients in the hardness and tensile strength along the length and height of the deposit were observed. However, the layered nature of MLFS and its complex thermal history are assumed to influence the microstructure and mechanical properties of precipitate-hardened aluminium alloys, for example, via the dissolution and coarsening of precipitates.

The aim of this study is to expand the understanding of a heat-treatable aluminium alloy of the 7xxx series produced by MLFS. Studies on 7xxx series Al alloys for FS deposition [22] and MLFS [23] are limited compared with other aluminium alloy series. Heat-treatable alloys are significantly affected by multiple thermal cycles. This study addresses the following question: How are the microstructure and resulting hardness of AA7075 affected by the MLFS process? The local heterogeneity of the microstructure is the focus of this study.

As stated by Seidi *et al.* [9], a detailed understanding of the temperature evolution and distribution during MLFS is useful. However, owing to the layered nature of MLFS, the in situ experimental monitoring of temperature evolution is complex. Predictive numerical models facilitate the development of insights into the expected thermal cycles by comparing various positions within a single layer and across layers. Modelling thermal cycles can link the heat input to the microstructure (such as the grains and precipitates) and expose the effect of temperature on the deposited material, within a single layer or sequentially across layers.

Studies on the simulation of friction surfacing are limited. Vitanov and Javaid [23] developed an FE thermal model to predict the temperature at the interface with a substrate. The heat distribution at the frictional interface and variation in the material properties with temperature significantly affected the thermal cycles. Bararpour *et al.* [17] showed that the strain rate and temperature of the top layer of the deposit was higher than the bottom of the deposit. This analysis is crucial to the present study because the increased strain rate and temperature contribute to microstructural heterogeneity. The first technique to investigate the influence of FS configurations, such as process parameters, substrate thickness, and backing plate material, on the heating and cooling cycles and temperature distribution in the substrate was developed by Kallien and Klusemann [24]. Based on a thermo-mechanical model, Rahmati *et al.* [25] showed that the maximum temperature and heat generation at the deposition interface increased with increasing axial force (F), and rotational speed (ω), or decreasing advancing speed (v). However, these models were restricted to monolayer deposition. This study extends the model proposed by Rahmati *et al.* [25] for multilayer deposition.

The multilayer AA7075 structures were processed using MLFS. This study aims to assess the link between the grain properties (grain boundary, misorientation, and grain size), precipitate heterogeneities,

and hardness evolution. This link will be investigated for each layer and along the height of the deposition to determine the impact of MLFS on the AA7075 alloy. This will enable future users to become proficient in the MLFS process and adjust it to applications based on their knowledge of heterogeneities. As MLFS is associated to temperature gradients, is it possible to predict temperature distribution of MLFS? This work proposes a multilayer thermal model, experimentally verified. Then, can this model be related to microstructural features? The present study establishes a qualitative link between the (simulated) thermal cycle and microstructural heterogeneities, which represents a novel contribution to the know-how of multilayer AA7075 manufactured by MLFS. Finally, the influence of T6 post-treatment on the MLFS specimens was analysed, aiming to answer the following questions: can T6 treatment help to recover and homogenize the mechanical properties of the feedstock? What are the changes in the microstructure?

2. Experimental methods

2.1. Materials and processing

The deposits were produced using a dedicated FSW machine (FSW-LM-AM-16-2D, Tra-C Industries) that incorporated in-process torque measurements. Fig. 1a shows a 3D schematic of the layer-by-layer deposition process consisting of a substrate and consumable rod. AA7075 in the T6 state (20 mm diameter and 120 mm length) was used as the consumable rod (initial feedstock). AA6060 (400 mm in length, 70 mm in width, and 10 mm in thickness) in the T6 state was used as the substrate in this study. Oxide was removed from the surface of the substrate plate using sandpaper; thereafter, the surface of the substrate plate was cleaned with ethanol and acetone, and dried. The deposition was cooled in air to ambient temperature before the addition of the next

layer.

During the FS process, the main process parameters, F , ω and ν , must be optimised to obtain high-quality deposits [26]. Based on a parametric study for single layers by Li *et al.* [27], deposition specimens exhibited favourable interfacial mixing between the deposition and the substrate when processed with $F = 5$ kN, $\omega = 1000$ rev/min and $\nu = 180$ mm/min (Fig. 2a). These parameters were used to construct multiple layers to further investigate the thermal cycles and resulting changes in the microstructure and mechanical properties of the deposition during MLFS. The length of the multi-layer specimen, average thickness per layer, and average width were 270 mm, 1.65 ± 0.20 mm, and 22.74 ± 1.01 mm, respectively. These measurements were taken between the outermost points on the advancing and retreating sides of each cross section. The variation in the thickness and width of the layers was marginally influenced by the deposition temperature. Indeed, the thickness is slightly decreasing and the width is slightly increasing from bottom to top layer owing to the increase of the temperature. A 270 mm-long layer was synthesised via the consumption of 52 mm of feedstock; however, flashes remained on the feedstock. This implies that the feed rate of the feedstock in the z-direction was 34.7 mm/min. The cross-section at the centre length (Fig. 2c) of the deposition of the multilayer is shown in Fig. 2b.

A new specimen containing thermocouples was built to validate the thermal model. The walls used for the temperature measurements were constructed using the same materials (AA7075 for the wall and AA6061 for the substrate) and the same process parameters. The travel length, mean width, and mean thickness of the layers of the sample used for numerical validation were 100, 24, and 1.72 mm, respectively. The thickness of this sample was within the standard deviation of all the samples processed using the selected parameters. The wall contained six layers and was thermally scanned using thermocouples that were directly in contact with the material (without drilled holes) during the deposition process. These thermocouples were located at five positions at half the travel length of the deposition. The thermocouple positions

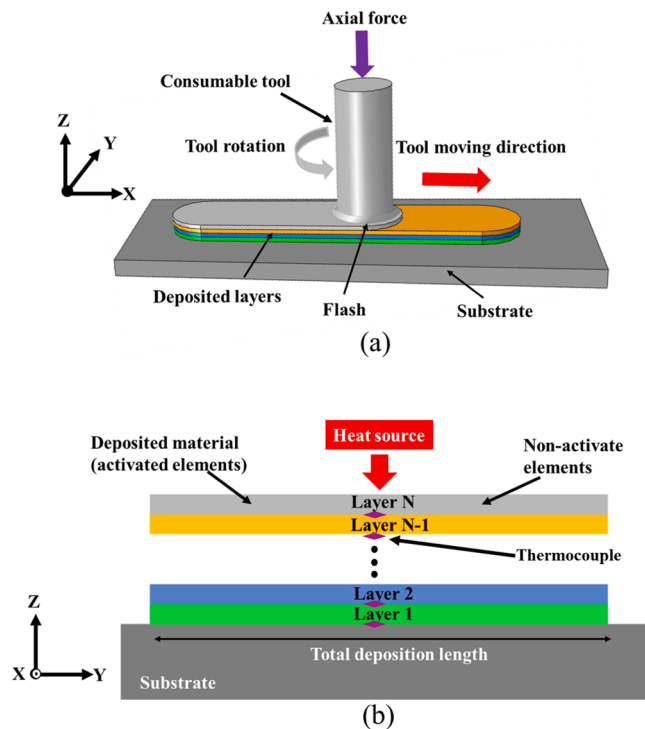


Fig. 1. Schematic of deposition via multi-layer friction surfacing. (a) 3D schematic of the multi-layer friction surfacing. The three main parameters are highlighted: axial force, rotation speed, and traverse speed. Due to the deposition process, the feedstock generates flashes. (b) Schematic of the multi-layer heat transfer in the multi-layer friction surfacing with the position of the experimental thermocouple. For each deposited layer, heat is generated and transferred to the lower layers.

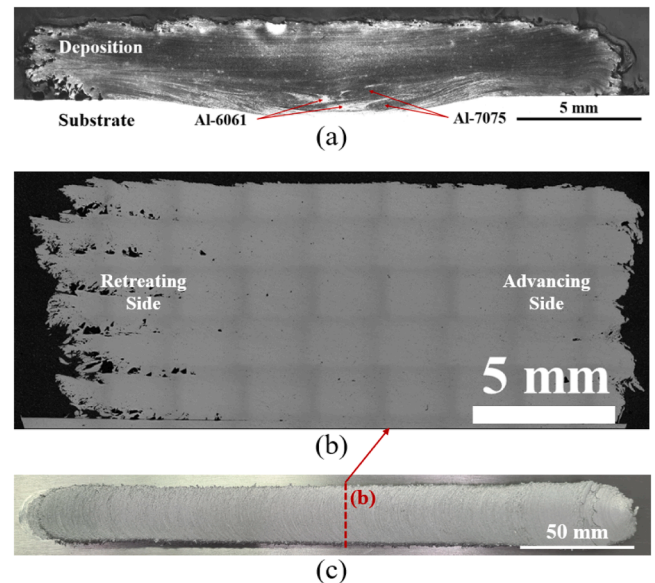


Fig. 2. Images of single and multi-layer depositions. (a) Interfacial mixing between the AA7075 (deposition) and AA6061 (substrate) during monolayer friction surfacing deposition. This is an indicator of high interface quality. (b) Optical microscopy image of the multi-layer specimen constructed with an axial force $F = 5$ kN, a rotation speed $\omega = 1000$ rev/min and a traverse speed $\nu = 180$ mm/min. The different profiles of retreating and advancing side are visible, the retreating side exhibits more defects. (c) Top view of the multi-layer specimen constructed via the multi-layer friction surfacing process. It indicates the cross-section location of (b).

for $N = 6$ —below the first layer (L1-below), above the first layer (L1-above), below the fourth layer (L4-below), above the fourth layer (L4-above), and below the sixth layer (L6-below)—are shown in Fig. 1b (3-D schematic in Supplementary Fig. S1). These locations were chosen for the inclusion of the bottom, middle, and top layers of the deposition in the analysis. Because of these locations, the variation along the depositional height can be highlighted, which integrates the three most important zones for analysis. L1 represents the region in proximity of the substrate, L4 is the behaviour of the medium layer, and L6 highlights the microstructure of the layer with no additional thermal cycles.

SHTs were performed on the MLFS deposits in a Nabertherm 1100 furnace at $470 \pm 5^\circ\text{C}$ for 30 min, as proposed by Lezaack et al. [19] for the friction-stir-processed 7xxx alloy. High-temperature SHT aimed to dissolve hardening precipitates in deposits. Water quenching was applied immediately after SHT. Subsequently, artificial aging was performed at 120°C for 24 h to regenerate the fine T6 temper precipitation and attain the hardest T6 state. Moreover, other heat treatments were performed on the specimens analysed using transmission electron microscopy (TEM). The solution heat treatment (SHT) and as-quenched temper (W heat treatment) aimed to dissolve the hardening precipitates of the deposits with SHT at $470 \pm 5^\circ\text{C}$ for 30 mins followed by quenching; aging was not included for the W state. The T4 heat treatment involved the same steps as the W heat treatment; however, an additional week of natural aging at ambient temperature was included. This aimed to regenerate the elements in the solid solution and Guinier-Preston (GP) zones in AA7075.

2.2. Microstructure and hardness characterization

The deposits were dissected using a cutting machine to prepare specimens for microstructural characterisation. The specimens cut from the deposits were polished using standard metallurgical procedures—successive abrasive SiC papers, a diamond suspension, and a final oxide polishing suspension.

The polished samples were examined using a field-emission gun scanning electron microscope (SEM) Ultra 55. SEM was used to analyse the precipitates from the top to the bottom of the deposition section. Images were captured at the mid-height of each deposition layer along the z-direction (Fig. 1a,b for axis definitions). The size of the precipitates was then quantified using ImageJ software.

Electron backscatter diffraction (EBSD) characterisation was performed with a field emission gun-SEM (Zeiss SigmaTM) equipped with an EBSD detector (Oxford Symmetry ® S3). An accelerating voltage of 10 kV, tilt of 70° , working distance of 20 mm, and steps of 100 and 70 nm based on the grain size, were used for EBSD measurements. EBSD observations were performed at different positions of the investigated multilayer deposits along the deposition direction. Further details regarding the selected positions are provided in Section 4.1. EBSD maps were post-processed using AZtecCrystal software. The criterion for the transition from low-angle grain boundaries (LAGBs) to high-angle grain boundaries (HAGBs) was set as usual at 15° .

TEM was performed using a CM20 Philips TEM at an accelerating voltage of 200 kV. A 3 mm diameter cylinder was extracted from the deposition by electro-erosion along the z-axis. Chemical etching was performed on the extracted cylinder to identify the different deposited layers. Subsequently, a lamella was extracted from layer 1, 4, and 6 (area A2; see Section 4), mechanically polished to 100 μm and electropolished using a twin-jet Struers Tenupol 3. TEM was used to characterise the fine precipitation via selected area electron diffraction (SAED) pattern identification and to characterise crystal defects, such as dislocations, on the different layers of deposition.

An EMCO-TEST DuraScan G5 tester was used to measure the microhardness of the deposits and initial feedstock. The 0.1 kg loading and 0.5 mm indent spacing were selected for this study. The initial feedstock exhibited a hardness of 171 ± 4 HV0.1.

2.3. Thermal model

A multilayer thermal model was developed to predict thermal cycles during the MLFS process. The Abaqus 2020 finite element package was used. Fig. 3 shows a schematic of the modelled domain and mesh, which are the geometries used in the experiments. The mesh was optimised for computational efficiency and numerical accuracy. The “element birth and death” method [28] was applied to simulate the layer-by-layer deposition. The dimensions of each layer corresponded to a specified experiment that was performed to validate the thermal model. The thermal properties of the deposited materials were assigned to the active elements.

The model was meshed with linear hexahedral elements (eight nodes, EC3D8 in Abaqus) in a rectangular cuboid shape. The sizes of the horizontal faces (in the x–y plane) of the mesh were selected such that they typically corresponded to 3 mm. The perpendicular faces (in the z-direction) of the mesh (Fig. 3b) were separated into several finer sections to improve the simulation accuracy in the direction of the deposition section and for data processing purposes. Ambient temperature (25°C) was initially assigned to the entire mesh geometry. All free surfaces were assigned convective conditions. The convection coefficient was set to $1000 \text{ W} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ at the bottom of the substrate (imitating the backing plate) and to $20 \text{ W} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ for all other surfaces [17]. The material input data for the thermal properties are listed in Table 1. Temperature-dependent material properties were considered in this study.

The simulation replicated the additive manufacturing of the MLFS process by adding elements to the mesh at the same rate as that of the experimental process (Fig. 3a and Video S1) along with the movement of the heat source (Fig. 3b and Video S1). The total durations of layer deposition for the simulation and experiment over travel lengths of 100 mm (validation of the numerical model) and 270 mm (deposition used for the analysis) were 33 s and 90 s, respectively.

To simulate the heat source in MLFS, a heat flux (q_{heat}) was applied to

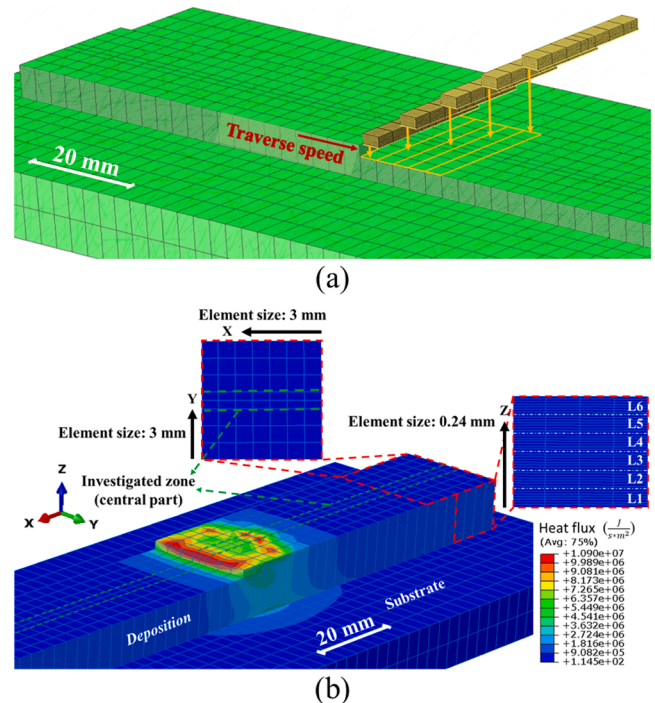


Fig. 3. Representations of the numerical model. (a) Elements addition method used in the simulation. The traverse speed direction is given and follows the direction of the added elements. (b) Finite element mesh of the simulation with the top and lateral overview. The figure includes also the heat source colour map induced by friction stir contact (see Video S1).

Table 1

Density, specific heat, and thermal conductivity of AA7075 and AA6060. Temperature-dependent material properties assigned in the thermal model.

Material	Density (kg/m ³)	Specific heat (J/kg K)		Thermal conductivity (W/m K)	
		T (K)	value	T (K)	value
AA7075 [29,30]	298	293	835.4	293	166.7
	400	2810	293	293	
	400	2805	373	897	373
	500	2782	423	916.3	423
	600	2755	473	974	473
	800	2700	573	1012.5	573
	900	2505	673	1128	673
AA6060 [31,32]	980	2480	773	1205	773
	298	298	900	298	237
	400	2700	298	298	
	400	2681	350	931	350
	500	2662	400	956	400
	600	2640	500	995	500
	800	2560	673	1060	673
	931	2379	873	1160	873

the deposited material (Fig. 3b). The rate of travel of the heat source was set to an experimental rate of traverse of 180 mm/min. The heat source was composed of a surface heat source that imitated the frictional heat source, and a volume heat source that represented the plastic deformation effect on heat dissipation, as suggested by Simar *et al.* [33] for AA6005A FSW. The total power input was extracted from the torque measurement during the tool-travelling phase (steady-state regime). The torque generated by the MLFS tended to exhibit a horizontal asymptote when more layers were added (Fig. 4a). This was attributed to the difference in the thermal and dissipation conditions of these layers on the substrate material. The total power input P [W] is given by

$$P = \eta M_z \omega \quad (1)$$

where M_z is the torque [Nm], ω is the rotational speed [rad/s], and η is the deposition efficiency of MLFS, which was lower than 100 % considering the heat loss in the tool and estimated at 95 % by Li *et al.* [26] for FSW process. Volumetric efficiency can be estimated by assuming that the shape of each layer is a rectangular parallelepiped rounded at the corners (see Supplementary Fig. S2). Karmakar *et al.* [34] defined the deposition efficiency as the ratio of the deposition volume to the consumed volume. This variable was quantified as 82 %. The η value is assumed to be equivalent to the deposition efficiency justified by the material ratio. The assumed surface (Eq.2) and volume heat sources (Eq.3) were 80 % and 20 % of the total heat source as proposed by Simar *et al.* [35] for AA6005A FSW. The values of the surface and volume heat

sources used in the validation of the simulation (Fig. 4b) are given by the following equations:

$$Q_s = \frac{0.8}{\pi r^2} P \quad (2)$$

$$Q_v = \frac{0.2}{\pi r^2 t} P \quad (3)$$

where, P is the input power, r is the feedstock radius, and t is the thickness of the layer.

A numerical model was established based on the initial conditions (shape and process parameters) and heat source values. The rectangular section shape of each layer was modelled to ensure an equivalent surface area and average thickness to those of the experiments. This guarantees that the deposited quantity of material was equivalent to that of the experiment.

3. Results

3.1. Thermal history during MLFS

Temperature profiles are for locations above L1 and below L4 and shown in Fig. 5a. These profiles were predicted with an 82 % efficiency. Fig. 5b shows the maximum temperatures of the experiments and simulation. Fig. S3 depicts the errors for each location. The greatest and average errors were lower than 8.2 % and 3.6 %, respectively (Fig. S3). Therefore, the numerical model was considered reliable. Owing to the symmetry of the numerical model, only the centre (in the width direction) of the cross-section was investigated in this study (Fig. 3b). The difference between the advancing and retreating sides was not considered in the simulation. Furthermore, a steady-state zone was considered for the deposited layers. The initiation and termination of the deposition exhibited particular thermal conditions, which were mainly linked to varying deformation conditions and heat sinks. Conversely, the central zone of the deposition was considered to be under pseudo steady-state conditions for the heat input and deformation fields (Video S1).

The addition of multiple layers leads to complex thermomechanical phenomena (heating, cooling, reheating, and plastic deformation) during the MLFS process. Performing a thermomechanical simulation is beyond the scope of this study; however, the thermal model described in Section 3 can be used to discuss the influence of thermal cycles on microstructural evolution. The temperature of the three layers (bottom layer (L1), middle layer (L4), and top layer (L6)) of the analysed multilayer specimen (length of 270 mm) was predicted, as shown in Fig. 6a. For each layer, three thermal cycles were extracted from the model, marked by the red (A1), green (A2), and blue (A3) areas in

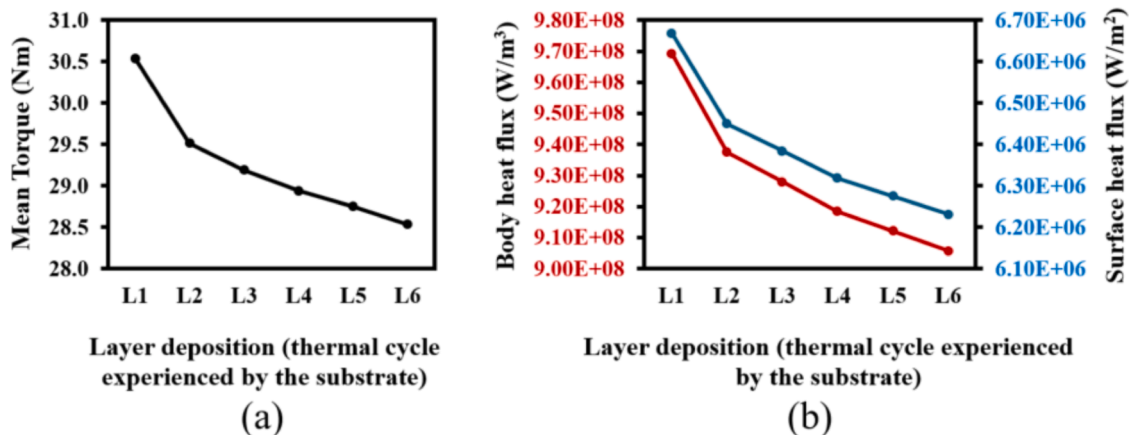


Fig. 4. Input data for the numerical model. (a) Torque measured by the tool during multi-layer friction surfacing along the different layers and (b) the heat flux assigned in the thermal model along the different layers. The heat flux is decomposed into body heat flux and surface heat flux. All these indicators decrease as the layer number increases.

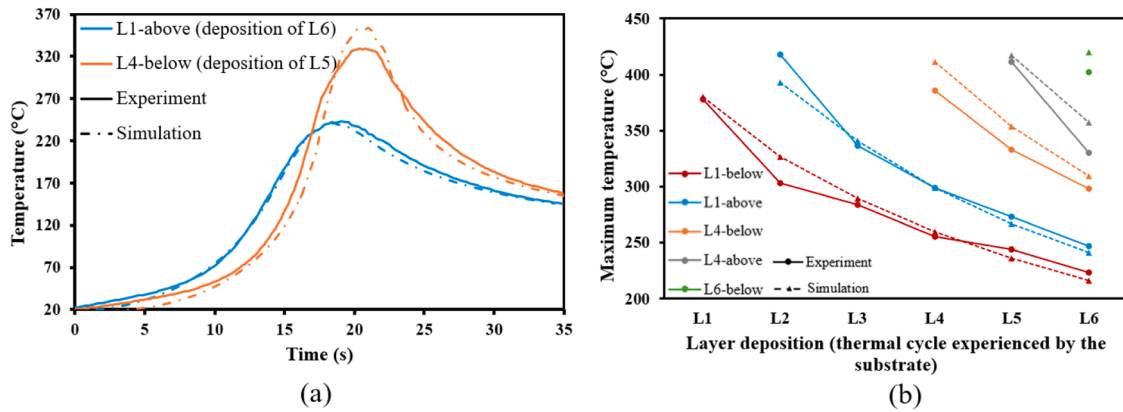


Fig. 5. Temperature profile of the numerical model and experiment. (a) Temperature profiles of two locations based on the experimental deposition and simulation. The first location corresponds to the part above layer 1 while layer 6 is being deposited, the second location is below layer 4 while layer 5 is being deposited. (b) Temperature maxima, of all the recorded locations, based on experimental deposition and numerical predictions. Each (plain or dotted) line corresponds to the same thermocouple / temperature simulated location while the layers are deposited. The model predicts accurately the trends in the temperature profile.

Fig. 6b. These locations aim to identify the variations occurring within a given layer. They were found to have different microstructures within a given layer and between the layers (see Section 4.2).

Fig. 6c shows the predicted spatial and temporal variations in temperature in A1 of L1 (L1_A1) of the deposit. In this study, the temperature cycles extracted from the simulation were used to qualitatively compare the temperature evolution at various locations of the deposition layers to explain the experimentally observed grain evolution (see Section 4.2). The predicted maximum temperatures agreed with those reported in the study of Kallien and Klusemann [24]. L1_A1 attained a maximum temperature of 404 °C when depositing layer L1 (**Fig. 6c**). Subsequent thermal cycles (deposition of layers 2, 3, 4, 5, and 6) resulted in the reheating of area L1_A1 with gradually decreasing temperatures, owing to the increasing distance between area L1_A1 and the layer being deposited. Two features of multiple thermal cycles were observed in **Fig. 6d**.

- The temperature of the top area (A3) of the layer remained higher than that of the lower area (A1).
- The maximum temperature of an area (A1, A2, or A3) increased when probing a higher layer (red dashed line in **Fig. 6d**).

The cooling rates followed a trend similar to that of the maximum temperature (**Fig. 6e**). **Fig. 6f** shows the calculated heat accumulation (integration of thermal cycles) in areas A1, A2, and A3 in the three investigated layers (L1, L4, and L6) of the deposit. The heat accumulation, which significantly decreased when probing higher layers, was calculated as the energy per kilogram of the material. The maximum heat accumulation of a layer was always observed near the top of the deposited layers (area A3).

3.2. Heterogeneities in grain structure

3.2.1. Grain structure evolution

To investigate the effects of the thermal profile on microstructural evolution, the layers selected for analysis were identical to the probed layers of the thermal model (**Fig. 6a,b**). **Fig. 7** shows the EBSD inverse pole figure maps of areas A1, A2, and A3 of selected layers in the deposit as well as the average grain size (AGS). Refined equiaxed grains were observed in the EBSD images of all areas. These microstructures are observed by Kalinenko *et al.* [36] for friction stir processed aluminium alloys owing to dynamic recrystallisation (DRX). Moreover, the grain structure exhibited alternating coarse and fine grains along the deposition height (Z direction in **Fig. 6a,b**); therefore, A2 exhibited coarser grains than that of A1 and A3, as discussed in Section 4.2.2.

3.2.2. Grain size distribution

The feedstock exhibited an average grain size of $3.3 \mu\text{m} \pm 4.6 \mu\text{m}$ in the plane perpendicular to the extrusion direction with a bimodal size distribution, which contained the smallest ($0.4 \mu\text{m}$) and largest ($40.4 \mu\text{m}$) grain sizes. **Fig. 8** shows the cumulative grain-size distributions for A1, A2, and A3 of selected layers of the deposit. **Fig. 9** shows the average grain size in a single graph adding also the standard deviations as functions of the same positions. The following conclusions were drawn.

- Compared with that of the feedstock, the grain size was reduced by 66 %.
- For all layers, the grains of the A2 zone were the largest among the three observed areas because the top (A3) and bottom (A1) areas experienced relatively higher plastic deformations owing to the friction and shearing of the deposited material [25]. This enhanced severe plastic deformation associated to high cooling rates (see **Fig. 6e**) favoured recrystallisation at the detriment of grain growth, which reduces the grain sizes of A3 and A1 compared with that of the A2 area. Indeed, the finer grains (areas A1 and A3) are thermally unstable and turn to secondary recrystallized material (possibly by abnormal grain growth mechanism) after SHT while coarser and more stable grains (area A2) do not face that reaction. This is proved by supplementary Figs. S4, S5, where abnormal grain growth is observed in areas A1 and A3 while the grains in area A2 remain unchanged.
- The grain size of the A1 (bottom) area of L1 was lower than that of the other L1 areas. During MLFS, the temperatures attained in the A1 area of L1 were lower than those of the A2 and A3 areas of the same layer because of the heat absorption of the backing plate (see **Fig. 6d**), which reduced grain growth in A1 [16].
- In the subsequent layers (L4 and L6), the grains in A3 were smaller than those in A1 and A2. Higher temperatures and plastic strain were exhibited in the A3 area of L4 and L6 than those of the A1 area of those layers (**Fig. 6d**). Rahmati *et al.* [25] predicted a lower maximum plastic strain in the A1 area (~ 1.62) than that of the A3 area (~ 7.29) in AA2024. This indicates that recrystallisation in the A3 area of L4 and L6 was likely greater than that of A1 in the same layers; therefore, the grains in the A3 area were finer than those of the A1 area.
- The stability of the grain size in A2 was higher than that of the A1 and A3 areas; additionally, this difference in stability was independent of the layer level.
- The grain size in A1 increased along the deposition height (Z direction), whereas the grain size in A3 decreased along the deposition height.

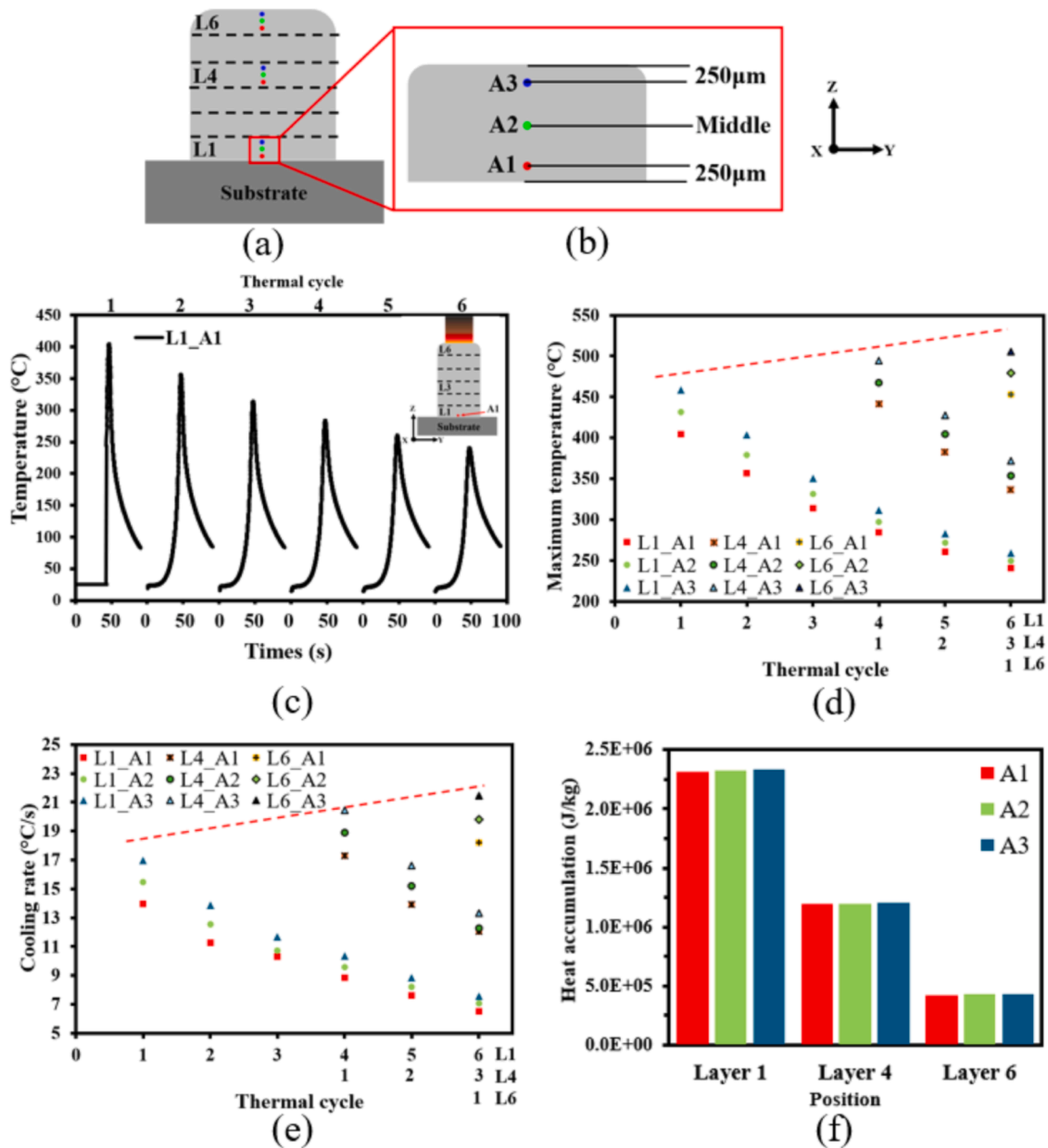


Fig. 6. Simulation results. (a) Schematic of selected layers and (b) areas (A1, A2 and A3) in the deposition. (c) Predicted temperature along time corresponding to layer 1 at A1 (L1_A1), while the layers (1–6) are deposited, determined using the numerical model. (d–f) Predicted maximum temperature, cooling rates and accumulated heat at the selected positions of the deposition with the addition of layers. The local maximum temperature decreases for higher layers and the overall temperature increases for higher layers (see red trend line).

3.2.3. Low- and high-angle grain boundaries

Fig. 10 shows the fraction of LAGBs ($2^\circ < \theta < 15^\circ$) and HAGBs ($>15^\circ$) in all selected areas (see Fig. 6a). For all layers in the deposit, the areas in proximity to the layer interfaces (A1 and A3) exhibited a higher fraction of HAGBs than A2 (centre). The high fraction of HAGBs indicated that DRX was the restoring mechanism during the friction surfacing process. In FS aluminium alloys, continuous dynamic recrystallisation (CDRX) is generally assumed to dominate the DRX mechanism because of their high stacking fault energy and low shear modulus [15]. During the initial stage of deformation, dislocation cells were formed within the larger grains and converted into LAGBs via the progressive accumulation of dislocations. The formation of well-developed subgrain structures, which contained limited dislocations, during deformation, and the misorientation of the subgrain boundaries progressively increased. This resulted in the gradual conversion of LAGBs into HAGBs [37].

3.3. Heterogeneities in coarse precipitates

AA7075 alloys are composed of Al_2CuMg , Mg_2Si , Fe-rich precipitates, such as $\text{Al}_6(\text{Fe}, \text{Mn})$, Al_3Fe , and $\text{Al}_7\text{Cu}_2\text{Fe}$, and Al and Cr-rich precipitates, such as $\text{Al}_{18}\text{Mg}_3\text{Cr}_2$, as well as $\eta(\text{MgZn}_2)$ precipitates [38]. As stated by Davis [39], only η precipitates are expected to dissolve into aluminium during SHT. The distribution of precipitates was correlated with the shape and distribution of the grains. The formation of η precipitates during aging in 7xxx series aluminium alloys occur via preferential precipitation at grain boundaries [10]. Fig. 11a shows the SEM images of precipitates in the deposit. These precipitates were quantified in the A2 area (see Fig. 6b). The average size of these precipitates was determined using ImageJ software. The average size of the precipitate decreased from the bottom to the top layer (Fig. 11c). The size of the precipitates varied between 175–205 nm. The surface fraction of coarse precipitates also decreased from the bottom to the top layer

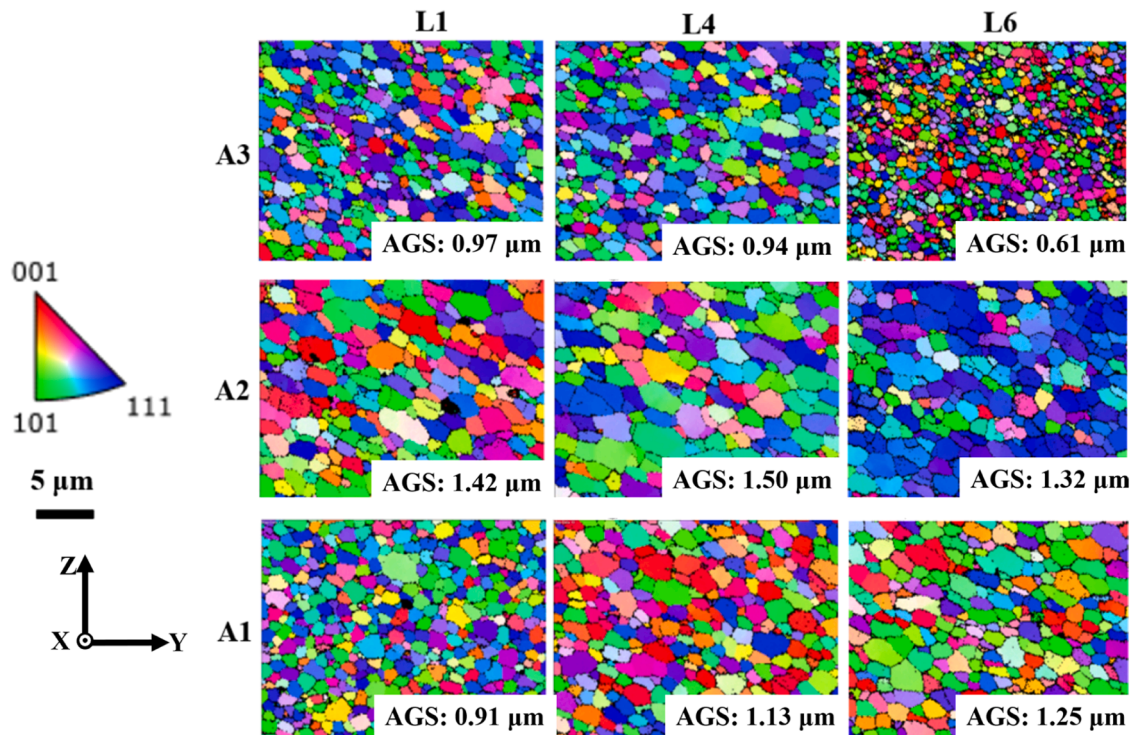


Fig. 7. Grains at different positions. Inverse pole figure maps made by electron backscatter diffraction at varying positions, corresponding to the ones shown in Fig. 6a,b, along the deposit. Fig. 9 provides the grain size tendencies that correspond to the present maps. AGS = Average grain size.

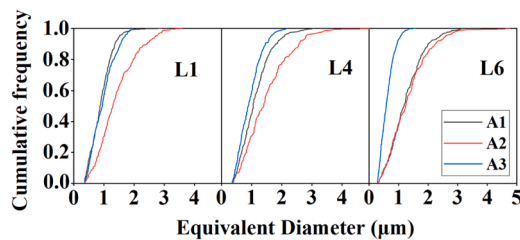


Fig. 8. Grain size at different positions. Cumulative grain size distribution of the various layers of the deposit.

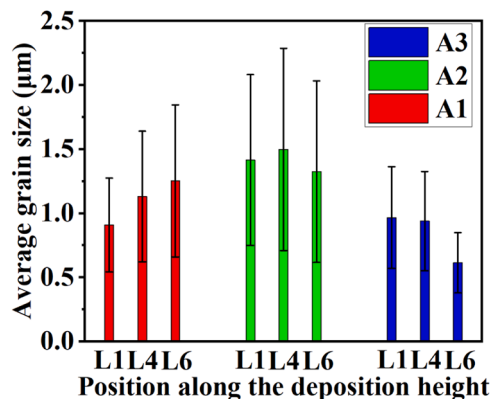


Fig. 9. Grain size distribution. Electron backscatter diffraction of the average grain size at various positions along the deposition height. Error bars represent the 1 standard deviation.

(Fig. 11c). This higher precipitate volume fraction and size in L1 occurred because of its exposure to additional thermal cycles. In opposition to the conclusions of the grain size analysis, here the accumulated

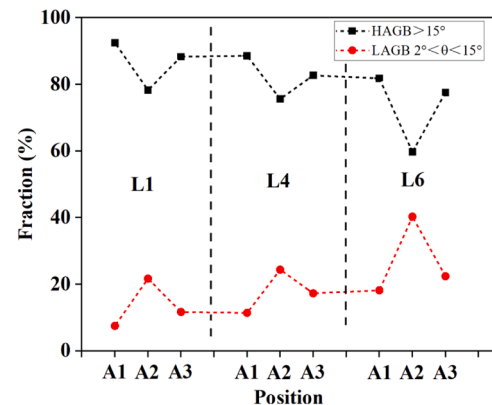


Fig. 10. Spatial distribution of the low and high angle grain boundaries (LAGBs and HAGBs). Fraction of LAGBs and HAGBs in the A1, A2 and A3 areas of the selected layers.

heat (Fig. 6 f) directly explains the observed trends.

TEM analysis of selected samples was performed to complement the SEM analysis. The TEM images (Fig. 11b) were obtained from the centre (A2 in Fig. 6) of positions L1, L4, and L6. The same qualitative reduction in the volume fraction of the precipitates from L1 to L6 was observed (Fig. 11c). These images were captured in proximity to the sample hole to ensure that the sample width of each image was similar. Three types of precipitates were identified in L1 (Fig. 12a).

- A coarse and hexagonal shaped precipitate (highlighted by a white arrow) was identified as an η precipitate using SAED (Fig. 12b) [40].
- A coarse rod-like precipitate (highlighted by a black arrow) was identified as $\text{Al}_{18}\text{Mg}_3\text{Cr}_2$ using SAED (Fig. 12c) [41].
- Thin, several nanometre-long, elongated precipitates (highlighted by a red arrow) were identified as a series of η' precipitates (Fig. 12d)

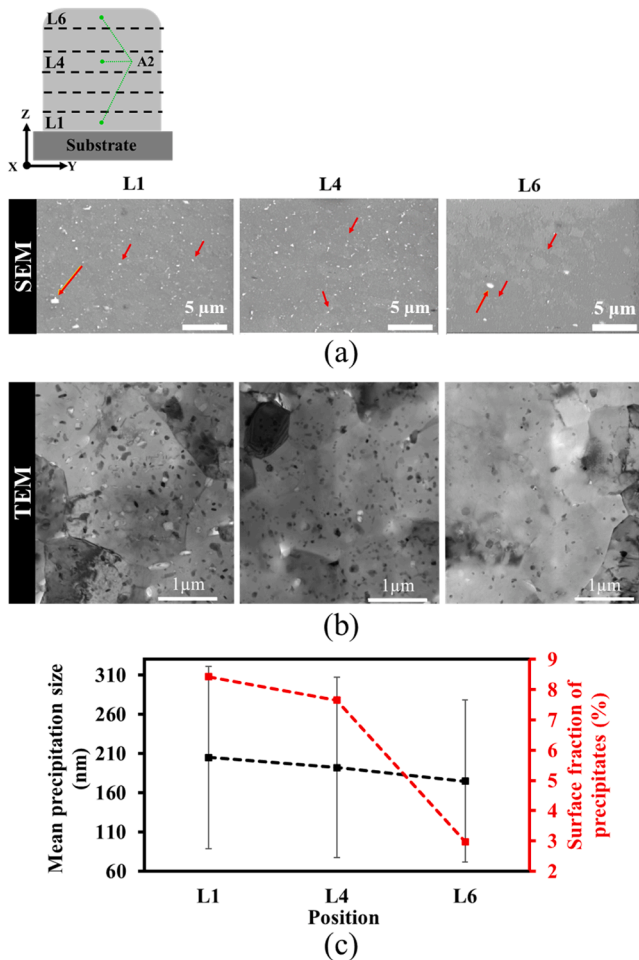


Fig. 11. Analysis of the coarse precipitates. (a) Schematic positions of the investigated zones by scanning electron microscopy. Precipitates and phases distributions are observed and quantified in area A2 of the deposition layers. The red arrows highlight the precipitates. (b) Transmission electron microscopy bright-field images of the precipitation evolution in area A2 of the respective layers, as schematically exposed in (a). (c) The mean size and surface fraction of the coarse precipitates (obtained by scanning electron microscopy image analysis). Error bars represent the 1 standard deviation. The mean precipitates size and surface fraction decrease for higher layers.

because of their alignment in specific crystallographic directions, which is expected from semi-coherent precipitates [42].

Precipitation was not the same in each layer (Fig. 11b). In addition to the overall decrease in the volume fraction of the precipitate from layers L1 to L6, a difference in the precipitate type was observed. Layer L1 contained all three types of precipitates. However, layer L6 exhibited no thin η/η' precipitation; only coarse precipitation was observed.

No evident difference in the dislocation density between the samples extracted from different layers was observed. However, L6 exhibited atypical wavy dislocations (Fig. 12d), which were attributed to the presence of multiple pinning points or helix-shaped dislocations. Dislocations can form helices via a climbing mechanism as explained by Narayan and Narayan [43]. When an obstacle blocks the gliding of a dislocation, it can climb and continue gliding. Although Al can exhibit a high stacking fault energy, Caillard and Martin [44] found that climbing can be caused by the high density of vacancies when high temperatures are attained. Obstacles, such as thin precipitation or other dislocations (the formation of a dislocation forest), were not identified. Thus, the obstacles causing these dislocation helices were assumed to be solid solutions and/or GP zones.

3.4. Heterogeneities in microhardness

Fig. 13 shows the microhardness maps measured along the thickness of the multilayer-deposited specimen before and after T6 heat treatment. The deposit was softer (95–130 HV0.1) than the initial feedstock (171 ± 4 HV0.1), as shown in Fig. 13a. Moreover, the deposition exhibited a hardness gradient along the Z direction that gradually increased from the bottom to the top layers. This hardness gradient along the deposition height was not reported in previous studies on MLFS AA5083 [20]. However, this difference between studies was attributed to the difference between the AA5083 and AA7075 alloys, wherein the former is not heat-treatable, whereas the hardening precipitation of the latter is heat-sensitive, as shown for other heat-treatable Al alloys by Seidi *et al.* [9].

Fig. 13b shows the hardness evolution of the deposition after T6 heat treatment. SHT at 470 °C for 30 mins and artificial aging at 120 °C for 24 h increased the hardness of the deposit to 181 ± 4 HV0.1, which was marginally higher than that of the initial feedstock in the T6 state (171 ± 4 HV0.1). The homogeneity of the hardness was adequate throughout the entire deposition section. However, localized secondary recrystallisation (or possibly abnormal grain growth) was observed in this T6 treated sample (Figs. S4 and S5 in the Supplementary). Ludtka *et al.* [45] showed that the columnar microstructure of high-solute 7xxx aluminium alloys was particularly favourable for crack propagation, owing to intergranular damage nucleation. This phenomenon is known to be a consequence of SHT; however, it may be prevented by using appropriate heat control, as proposed by Lezaack *et al.* [19] for the friction-stir-processed 7xxx alloy. Applying these strategies to MLFS is not a straightforward process. However, this finding creates opportunities for future research. Changing the temperature of the solution heat treatment (460–480 °C) did not prevent the occurrence of excessive grain growth.

4. Discussion

4.1. Analysis of the thermomechanical cycles

Repetitive thermomechanical cycles, owing to the addition of multiple layers during MLFS, significantly affected the microstructure. The significant grain size reduction (Fig. 7) indicated that all selected areas in all layers were subjected to dynamic recrystallisation owing to heating and plastic deformation during MLFS.

The heat sinks—substrate and backing plate—significantly affected the temperatures of the FS process [19]. As the substrate and backing plate absorbed heat, a thermal gradient was generated during MLFS (see Fig. 6d). Furthermore, the thermal, strain-rate, and cooling-rate gradients affected the microstructure at different levels in the deposited layer. Torsion/compression between the consumable rod and substrate/previously deposited layers resulted in severe plastic deformation.

Based on the thermal model, A1 attained lower temperatures than those of A3 in all deposited layers. The predicted thermal gradient corroborated the thermal gradients reported by Ehrich *et al.* [15], which were identified using an infrared camera during the FS of the 6060 Al alloy. According to the thermal model in this study, this thermal gradient was observed because the top area of the deposited layer was in contact with the heat source.

The predicted maximum temperature of L6 (~ 505 °C) was higher than that of L1 by approximately 47 °C (Fig. 6d). These differences in temperature induce small variations in the dimension of the layer, such as thinner depositions at higher temperatures. The influence of higher temperatures (higher layers) on microstructural features are highlighted in Figs. 7, 8, 9, 10, and 11. The higher temperature of the top layers was a consequence of heat absorption by the backing plate, which decreased with increasing deposition height. Thus, the following conclusions from the thermomechanical analysis of the process are used to explain

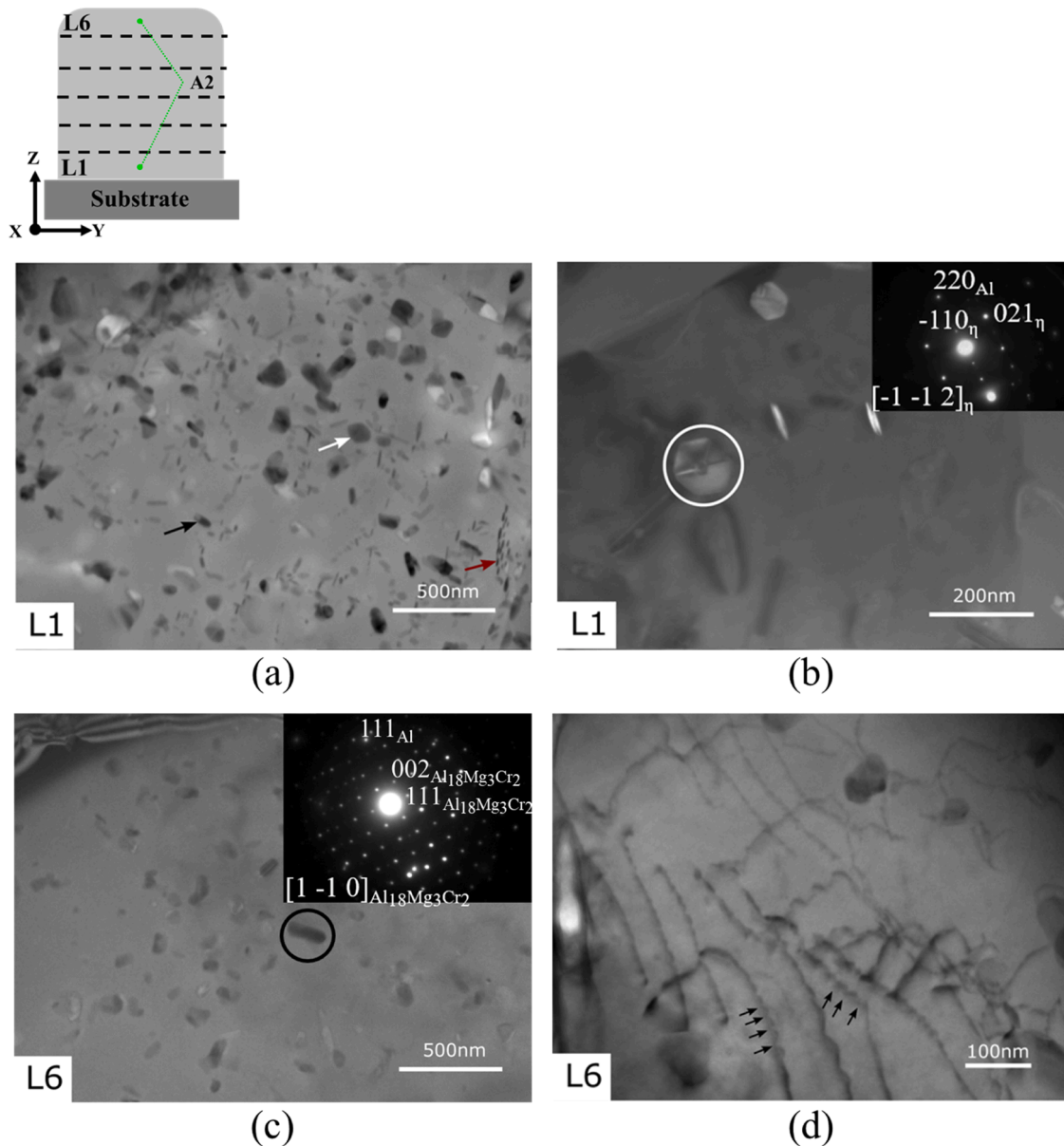


Fig. 12. Identification of precipitates and dislocations using transmission electron microscopy (TEM). TEM images of layers L1 and L6 in areas A2. (a) Brightfield image of three types of precipitates are highlighted using an arrow: the η precipitate (white), $\text{Al}_{18}\text{Mg}_3\text{Cr}_2$ (black), and η' precipitates (red). (b) Darkfield image with an η precipitate and (c) brightfield image with an $\text{Al}_{18}\text{Mg}_3\text{Cr}_2$ precipitate. Their corresponding selected area electron diffractions are shown in the insert taken from the circled area. (d) Dislocations that form helices are highlighted by black arrows.

microstructural evolution in all zones:

- As mentioned in Section 4.2.2, the plastic strain of area A1 is expected to be lower than that of area A3. Although this was not predicted by the model, it has been established by Rahmati *et al.* [25]. This occurs because the feedstock increases the shearing on top of the layer during deposition [46].
- According to the thermal model of this study, area A1 exhibited a lower temperature and cooling rate than those of area A3 across all layers (see the representative layers in Fig. 6d and Fig. 6e).
- L1 (the bottom layer) exhibited a lower temperature than that of L6 (the top layer) across all areas (Fig. 6d).
- After six layers were deposited, the temperature profile of each layer did not attain a constant value.

4.2. Grain size evolution

The grain size is dictated by the CDRX and grain growth during the severe plastic deformation of the 7075 alloy [47]. The mechanisms of CDRX during friction surfacing can be summarised by the following steps according to Huang and Logé [48]: (1) increasing dislocation density, followed by sub-boundary formation during hot deformation; (2) the pinning of grain boundaries by small particles; (3) the formation of new grains via continuous dislocation trapping into these sub-boundaries; and (4) the gradual conversion of LAGBs into HAGBs. The grain size in A1 gradually increased from the bottom to the top layer, whereas the grain size in A3 exhibited an opposing trend (Fig. 9). The concluding bullet points in Section 5.1 will now assist with the explanation of these phenomena.

When the strain rate was relatively low (in A1), lower deformation levels were attained during processing. As stated by Sun *et al.* [47], the grain size was largely controlled by grain growth. Grain growth is

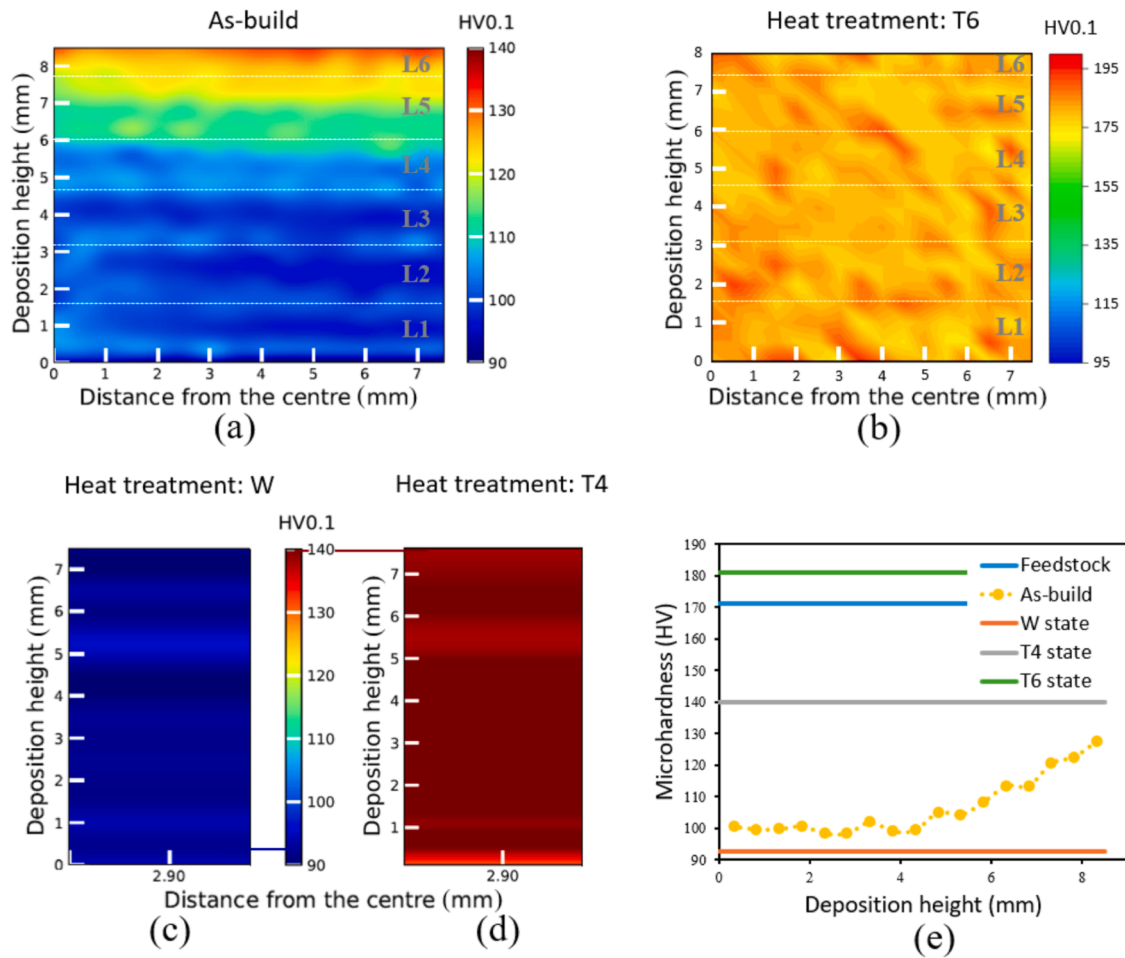


Fig. 13. Microhardness analysis for various heat treatments. (a) Microhardness map after multi-layer friction surfacing. The microhardness is homogeneous in layers 1–3, then it increases from layer 3 until the top layer. (b) More homogenous microhardness map after T6 heat treatment, (c) W heat treatment, and (d) T4 heat treatment (with 1-week ageing). The zero (0) on the y-axis corresponds to the centre of the deposit. (e) Overview of the mean microhardness values at different states and for the as-build sample along the deposition height highlighting the trends.

sensitive to temperature; therefore, a higher temperature in area A1 of L6 enhances grain growth. The higher heat accumulation in L1 compared with that of L6 can also exhibit a marginal effect on grain growth when the temperature facilitates grain boundary migration. However, increasing the number of layers exhibited a reduced impact on the grain growth of the base layer because the temperature of L1 decreased for each new cycle. Therefore, the grain size of L1 can be affected by L2 deposition; however, only marginally by the deposition of additional layers. This was observed because the temperature was lower than 300 °C during L3 deposition. Based on the observations made in this study, the grain growth due to multiple heat cycles was negligible because area A1 of L6 exhibited a larger grain size than that of area A1 of L1 [48].

At high strain rates (A3), higher deformation levels were attained during processing. The dislocations induced by these deformations do not have sufficient time to participate in dynamic recovery. Varying levels of recovery were observed because of the different temperatures and cooling rates (related to the time of exposure) in A3 of the layers. Therefore, the accumulation of dislocations was faster than their consumption, leading to an increase in the dislocation density. The grain size was mainly controlled by CDRX when the strain rate and temperature were high, corresponding to a low Zener-Hollomon parameter; indeed, dynamic recovery was low [47]. Thus, because area A3 of L6 exhibited a higher temperature than that of area A3 of L1, CDRX was enhanced in A3 of L6, which resulted in a smaller grain size. Moreover, the cooling rate observed in area A3 of L6 increased as proven on Fig. 6e,

confirming the expected reduction in the grain size for that area, as grain growth is inversely dependant on cooling rates [17]. Another explanation to the decrease in grain size in the A3 areas from bottom to top layer, could be an increase in the Zener-Hollomon parameter due to an increase in strain rate. Indeed, the temperature increase reduces the layer height (see Section 2.1), which could subsequently affect the strain rate.

For each of the analysed layers, the grains in area A2 were larger than those in A1 and A3. This phenomenon was not observed in a similar study of Wu *et al.* [46] on AFSD. Kallien *et al.* [49] stated that the main differences between AFSD and MLFS were based on the strain rate and temperature profile. Consequently, the grain size gradients were due to these variables in combination or individually. Depending on the strain rate, CDRX or grain growth exhibited a dominant effect on grain size. Grain growth was favoured at lower strain rates, whereas the size of the grains was mainly influenced by CDRX at higher strain rates [47]. EBSD analysis showed that area A2 contained coarser grains than that of the other areas in each layer (Figs. 7, 8, and 9), indicating that the grain growth effect was dominant, as observed for other Al samples by Kallien *et al.* [49]. The coarseness of the grains increased as the temperature increased. Thus, based on the numerical model predictions, the temperature was higher in area A2 than in area A1. Higher temperatures induced enhanced grain growth in area A2 compared with area A1. In addition, the grain size in A2 did not increase in the top layers. This may have been induced by the concomitant increase in the cooling rate (Fig. 6e). A higher cooling rate limits the time required for grain growth.

However, the maximum temperature of A2 in L1, and A1 in L4 were similar; additionally, the maximum temperature of A2 in L4 and A1 in L6 were similar (Fig. 6d). These areas contained varying grain sizes but were mainly affected by grain growth during microstructural transformation. Consequently, these observations may be associated with different strain rates in areas that experience similar maximum temperatures.

4.3. Coarse precipitation and hardness

The softening of the friction-surfacing material was associated with precipitation coarsening. The as-deposited precipitation state was influenced by the temperature, dislocation density, and grain size. During friction surfacing, the alloying elements are dissolved before the recrystallisation of areas A1–A3. Subsequently, coarse precipitates tend to nucleate on the HAGBs during the cooling stage according to Lezaack and Simar [19]. As shown in Fig. 6a, most of the grains in A2 contained HAGBs. Moreover, the fraction of HAGBs in all areas decreased with increasing deposition height (z-direction). The higher fraction of HAGBs in L1 (Fig. 10) favoured an increase in the surface fraction of the coarse precipitates (Fig. 11c).

The accumulated heat (Fig. 6f) due to subsequent deposition favoured the growth of the precipitates. This explained the increase in the size of the precipitates from A2 of L6 to A2 of L1 (Fig. 11c).

The temperature evolution in association with the repetition of thermal cycles exhibited an impact on the fraction of coarse precipitates, which decreased from the bottom to the top layers (see Fig. 11). This effect was consequently exhibited on the microhardness and size of the grains. TEM analysis confirmed that coarse precipitates of $\text{Al}_{18}\text{Mg}_3\text{Cr}_2$ were present in L6, as well as in the suspected solid solution and GP zones in that area. These precipitates, as well as η and η' , were observed in the other layers (L1 and L4). The microhardness results shown in Fig. 13 suggest that the coarse precipitates do not contribute to deposition strengthening. However, hardening of the top layers likely occurred because of solid-solution strengthening or GP zones. This strengthening is well-documented in Al 7075 after T4 heat treatment by Sabard *et al.* [50]. To verify this hypothesis, T4 heat treatment was performed on the deposited wall. Microhardness maps were constructed immediately after quenching (Fig. 13c) and room temperature aging (Fig. 13d). Natural aging led to strengthening of the alloy, with an increase in hardness from an average of 92 HV to 139 HV. The culminating hardness corresponded to the hardness of the last layer (Fig. 13a). This suggested that the last layer corresponded to a T4-state where the alloy was strengthened by the solid solution and GP zones. After successive depositions, heat accumulation in the other layers resulted in the nucleation and coarsening of GP zones, then the η' and η precipitates. Therefore, the overgrowth of the η precipitates in the bottom layer resulted in its reduction in microhardness (Fig. 13a). AlCu_2Mg precipitates, and Cr-rich and Fe-rich precipitates were observed in all layers. These precipitates originated from the feedstock because they cannot be dissolved during the process [39]. After SHT is applied to the deposits, η precipitates are completely dissolved. Subsequently, a homogeneous T6 precipitation state was attained throughout the deposition section via low temperature aging treatment, which resulted in the uniformity of the hardness in T6 state (Fig. 13b). Finally, the microhardness evolution is not fully linear from bottom to top, see Fig. 13e. At the scale of a layer, a plateau is observed for the first three layers prior to an increase of the hardness towards the top layer. Looking inside a layer, a cyclic drop of microhardness is measured in area A2 of each layer. This observation could, among others, be attributed to grain size effects.

5. Conclusions

Numerical simulations and experiments were combined to investigate the effects of thermomechanical processing on microstructural and hardness heterogeneities in multi-layer friction surfacing (MLFS) addi-

tively manufactured AA7075. Multiple thermal cycles in the lower layers caused heat accumulation and an increase in the maximum temperature in the upper layers. The T6 post-treatment on the multi-layer friction surfacing specimens was investigated. Main conclusion points can be listed:

- (1) The grain sizes of the A1 and A3 areas of a given layer were smaller than those of area A2 because of the presence of a higher local strain rate and varying temperatures near the interfaces with other layers or the substrate. The grains in A3 of layer 6 were the smallest across all layers because no additional thermo-mechanical cycles were induced by another layer.
- (2) The grain size of area A1 of a layer increased from the bottom to the top layer. This was due to an increase in the maximum temperature of the first thermal cycle from the bottom to the top layer (predicted by the thermal model) under a lower level of plastic deformation.
- (3) The grain size of area A3 of a layer decreased from the bottom to the top layer. Compared with area A3 of the bottom layer, a higher temperature at a high strain rate enhanced the continuous dynamic recrystallisation (CDRX) of area A3 of the top layer. When subjected to a higher strain rate, the grain size was mainly controlled by CDRX.
- (4) The size and volume fraction of the precipitate decreased from the bottom to the top layers. This can be directly related to the higher quantity of accumulated heat (resulting from multiple thermal cycles) in the lower layers than that of the upper layers. The higher maximum temperature in the top layers may also have increased the quantity of coarse precipitates.
- (5) The deposition exhibited a microhardness reduction compared to the initial feedstock owing to the overgrowth of the η precipitates. The higher microhardness of the top layer was caused by the strengthening of the solid solution and Guinier-Preston zones. Coarse η' and η precipitates were formed in the bottom layers via heat accumulation, leading to strength reduction.
- (6) Homogenous microhardness can be restored by applying T6 treatment. However, this study showed that avoiding secondary recrystallisation during the solution heat treatment component of T6 treatment is challenging.

Managing the abovementioned factors will improve the understanding of the MLFS process in AA7075. In addition, MLFS can be used to construct large components with moderate complexity [51]. This is ideal for structural components machined for a net-shaped finish. Because MLFS combines the information on material effects with the freedom of component production (materials and complexity), it exhibits great potential for future aerospace applications [52]. The mechanical strength of an as-built configuration is not optimal. Owing to T6 heat treatment, homogeneous hardness can be restored, which facilitates high-strength and large-scale additive manufacturing 7xxx series components.

CRediT authorship contribution statement

Jichang Xie: Writing – review & editing, Methodology, Formal analysis. **Matthieu Lezaack:** Writing – review & editing, Supervision, Methodology. **Thanesan Sapanathan:** Writing – review & editing, Supervision, Formal analysis. **Mohamed Rachik:** Writing – review & editing, Supervision, Resources. **Aude Simar:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Investigation, Funding acquisition, Conceptualization. **Matthieu Jadot:** Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Jishuai Li:** Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Romain Gautier:** Writing – review & editing, Methodology,

Formal analysis, Data curation.

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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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jmatprotec.2024.118661](https://doi.org/10.1016/j.jmatprotec.2024.118661).

References

- [1] Dursun, T., Soutis, C., 2014. Recent developments in advanced aircraft aluminium alloys. *Mater. Des.* (1980–2015) 56, 862–871. <https://doi.org/10.1016/j.matdes.2013.12.002>.
- [2] Martin, J.H., Yahata, B.D., Hundley, J.M., Mayer, J.A., Schaedler, T.A., Pollock, T.M., 2017. 3D printing of high-strength aluminium alloys. *Nature* 549, 365–369. <https://doi.org/10.1038/nature23894>.
- [3] Zhou, B., Liu, B., Zhang, S., 2021. The advancement of 7xxx series aluminum alloys for aircraft structures: a review. *Metals* 11. <https://doi.org/10.3390/met11050718>.
- [4] Mauduit, A., Pillot, S., Gransac, H., 2017. Study of the suitability of aluminum alloys for additive manufacturing by laser powder bed fusion. *Bull., Ser. B* (79).
- [5] Phillips, B.J., Mason, C.J.T., Beck, S.C., Avery, D.Z., Doherty, K.J., Allison, P.G., Jordon, J.B., 2021. Effect of parallel deposition path and interface material flow on resulting microstructure and tensile behavior of Al-Mg-Si alloy fabricated by additive friction stir deposition. *J. Mater. Process. Technol.* 295, 117169. <https://doi.org/10.1016/j.jmatprotec.2021.117169>.
- [6] Seidi, E., Miller, S.F., Carlson, B.E., 2021. Friction surfacing deposition by consumable tools. *J. Manuf. Sci. Eng.* 143. <https://doi.org/10.1115/1.4050924>.
- [7] Karmakar, S., Swarnkar, R., Pal, S.K., 2023. Effect of multi-layer deposition in solid-state friction stir surfacing-based additive manufacturing for fabrication of large-scale metal product. *J. Mater. Process. Technol.* 320, 118107. <https://doi.org/10.1016/j.jmatprotec.2023.118107>.
- [8] Yoder, J.K., Hahn, G.D., Zhao, N., Brennan, R.E., Cho, K., Yu, H.Z., 2023. Additive friction stir deposition-enabled upcycling of automotive cast aluminum chips. *Addit. Manuf. Lett.* 4, 100108. <https://doi.org/10.1016/j.ADDLET.2022.100108>.
- [9] Gandra, J., Krohn, H., Miranda, R.M., Vilaça, P., dos Santos, J.F., 2014. Friction surfacing—a review. *J. Mater. Process. Technol.* 214, 1062–1093. <https://doi.org/10.1016/j.JMATPROTEC.2013.12.008>.
- [10] Lezaack, M.B., Hannard, F., Zhao, L., Orekhov, A., Adrien, J., Miettinen, A., Idrissi, H., Simar, A., 2021. Towards ductilization of high strength 7XXX aluminium alloys via microstructural modifications obtained by friction stir processing and heat treatments. *Materials* 20, 101248. <https://doi.org/10.1016/j.MTLA.2021.101248>.
- [11] Yu, H.Z., Jones, M.E., Brady, G.W., Griffiths, R.J., Garcia, D., Rauch, H.A., Cox, C.D., Hardwick, N., 2018. Non-beam-based metal additive manufacturing enabled by additive friction stir deposition. *Scr. Mater.* 153, 122–130. <https://doi.org/10.1016/j.SCRIPATAMAT.2018.03.025>.
- [12] Prabhakar, D.A.P., Shettigar, A.K., Herbert, M.A., Patel G C, M., Pimenov, D.Yu., Giasin, K., Prakash, C., 2022. A comprehensive review of friction stir techniques in structural materials and alloys: challenges and trends. *J. Mater. Res. Technol.* 20, 3025–3060. <https://doi.org/10.1016/J.JMRT.2022.08.034>.
- [13] Suhuddin, U., Mironov, S., Krohn, H., Beyer, M., Dos Santos, J.F., 2012. Microstructural evolution during friction surfacing of dissimilar aluminum alloys. *Met. Mater. Trans. A Phys. Met. Mater. Sci.* 43, 5224–5231. <https://doi.org/10.1007/s11661-012-1345-8>.
- [14] Mukhopadhyay, A., Saha, P., 2020. Mechanical and microstructural characterization of aluminium powder deposit made by friction stir based additive manufacturing. *J. Mater. Process. Technol.* 281, 116648. <https://doi.org/10.1016/j.jmatprotec.2020.116648>.
- [15] Ehrlich, J., Roos, A., Klusemann, B., Hanke, S., 2021. Influence of Mg content in Al alloys on processing characteristics and dynamically recrystallized microstructure of friction surfacing deposits. *Mater. Sci. Eng. A* 819. <https://doi.org/10.1016/j.msea.2021.141407>.
- [16] Yu, P., Wu, C.S., Shi, L., 2021. Analysis and characterization of dynamic recrystallization and grain structure evolution in friction stir welding of aluminum plates. *Acta Mater.* 207. <https://doi.org/10.1016/j.actamat.2021.116692>.
- [17] Bararpour, S.M., Jamshidi Aval, H., Jamaati, R., 2019. Modeling and experimental investigation on friction surfacing of aluminum alloys. *J. Alloy. Compd.* 805, 57–68. <https://doi.org/10.1016/j.jallcom.2019.07.010>.
- [18] Dilip, J.J.S., Janaki Ram, G.D., 2013. Microstructure evolution in aluminum alloy AA 2014 during multi-layer friction deposition. *Mater. Charact.* 86, 146–151. <https://doi.org/10.1016/J.MATCHAR.2013.10.009>.
- [19] Lezaack, M.B., Simar, A., 2021. Avoiding abnormal grain growth in thick 7XXX aluminium alloy friction stir welds during T6 post heat treatments. *Mater. Sci. Eng.: A* 807, 140901. <https://doi.org/10.1016/J.MSEA.2021.140901>.
- [20] Shen, J., Hanke, S., Roos, A., Santos, J.F.D., Klusemann, B., 2019. Fundamental study on additive manufacturing of aluminum alloys by friction surfacing layer deposition. In: *Conf Proc. A.I.P. (Ed.)*, American Institute of Physics Inc. <https://doi.org/10.1063/1.5112691>.
- [21] Rath, L., Kallien, Z., Roos, A., Santos, J.F. dos, Klusemann, B., 2023. Anisotropy and mechanical properties of dissimilar Al additive manufactured structures generated by multi-layer friction surfacing. *Int. J. Adv. Manuf. Technol.* <https://doi.org/10.1007/s00170-022-10685-3>.
- [22] Elshaghoul, Y.G., El-Sayed Seleman, M.M., Bakkar, A., Elnekhaily, S.A., Albajian, I., Ahmed, M.M., Reda, R., 2023. Additive friction stir deposition of AA7075-T6 alloy: impact of process parameters on the microstructures and properties of the continuously deposited multilayered parts. *Appl. Sci.* 13, 10255. <https://doi.org/10.3390/app131810255>.
- [23] Vitanov, V.I., Javald, N., 2010. Investigation of the thermal field in micro friction surfacing. *Surf. Coat. Technol.* 204, 2624–2631. <https://doi.org/10.1016/j.surfcoat.2010.02.003>.
- [24] Kallien, Z., Klusemann, B., 2022. Combined experimental-numerical analysis of the temperature evolution and distribution during friction surfacing. *Surf. Coat. Technol.* 437. <https://doi.org/10.1016/j.surfcoat.2022.128350>.
- [25] Rahmati, Z., Jamshidi Aval, H., Nourouzi, S., Jamaati, R., 2019. Modeling and experimental study of friction surfacing of AA2024 alloy over AA1050 plates. *Mater. Res Express* 6. <https://doi.org/10.1088/2053-1591/ab255a>.
- [26] Li, H., Qin, W., Galloway, A., Toupis, A., 2019. Friction surfacing of aluminium alloy 5083 on DH36 steel plate. *Metals* 9. <https://doi.org/10.3390/met9040479>.
- [27] Li, J., Jadot, M., Xie, J., Lezaack, M.B., Sapanathan, T., Rachik, M., Simar, A., 2024. Process parameters selection for multi-layer friction surfacing of 7075 aluminium alloy. *Sci. Technol. Weld. Join.* <https://doi.org/10.1177/13621718241275092>.
- [28] Yang, Q., Zhang, P., Cheng, L., Min, Z., Chyu, M., To, A.C., 2016. Finite element modeling and validation of thermomechanical behavior of Ti-6Al-4V in directed energy deposition additive manufacturing. *Addit. Manuf.* 12, 169–177. <https://doi.org/10.1016/J.ADDMA.2016.06.012>.
- [29] Ma, F.J., Fu, D.Y., Liu, Y., Yang, D.P., Zhang, S.F., Sha, Z.H., 2020. The Numerical Simulation of Temperature Field in Friction Stir Welding of 7075 Aluminium Alloy. In: *IOP Conf Ser Mater Sci Eng.* Institute of Physics Publishing. <https://doi.org/10.1088/1757-899X/751/1/012081>.
- [30] He, M., Zhang, Z., Mao, W., Li, B., Bai, Y., Xu, J., 2019. Numerical and experimental study on melt treatment for large-volume 7075 alloy by a modified annular electromagnetic stirring. *Materials* 12. <https://doi.org/10.3390/MA12050820>.
- [31] Li, J.S., Raelison, R.N., Sapanathan, T., Hou, Y.L., Rachik, M., 2020. Interface evolution during magnetic pulse welding under extremely high strain rate collision: mechanisms, thermomechanical kinetics and consequences. *Acta Mater.* 195, 404–415. <https://doi.org/10.1016/J.ACTAMAT.2020.05.028>.
- [32] Li, J.S., Sapanathan, T., Raelison, R.N., Hou, Y.L., Simar, A., Rachik, M., 2021. On the complete interface development of Al/Cu magnetic pulse welding via experimental characterizations and multiphysics numerical simulations. *J. Mater. Process. Technol.* 296, 117185. <https://doi.org/10.1016/J.JMATPROTEC.2021.117185>.
- [33] Simar, A., Lecomte-Beckers, J., Pardoën, T., de Meester, B., 2006. Effect of boundary conditions and heat source distribution on temperature distribution in friction stir welding. *Sci. Technol. Weld. Join.* 11, 170–177. <https://doi.org/10.1179/174329306X84409>.
- [34] Karmakar, S., Swarnkar, R., Pal, S.K., 2023. Effect of multi-layer deposition in solid-state friction stir surfacing-based additive manufacturing for fabrication of large-scale metal product. *J. Mater. Process. Technol.* 320, 118107. <https://doi.org/10.1016/j.jmatprotec.2023.118107>.
- [35] Simar, A., Pardoën, T., de Meester, B., 2007. Effect of rotational material flow on temperature distribution in friction stir welds. *Sci. Technol. Weld. Join.* 12, 324–333. <https://doi.org/10.1179/174329307X197584>.
- [36] Kalinenko, A., Vysotskii, I., Malopheyev, S., Mironov, S., Kaibyshev, R., 2021. Relationship between welding conditions, abnormal grain growth and mechanical performance in friction-stir welded 6061-T6 aluminum alloy. *Mater. Sci. Eng.: A* 817, 141409. <https://doi.org/10.1016/J.MSEA.2021.141409>.
- [37] Sakai, T., Belyakov, A., Kaibyshev, R., Miura, H., Jonas, J.J., 2014. Dynamic and post-dynamic recrystallization under hot, cold and severe plastic deformation conditions. *Prog. Mater. Sci.* 60, 130–207. <https://doi.org/10.1016/j.pmatsci.2013.09.002>.
- [38] Zhao, Y., Liu, J., Topping, T.D., Lavernia, E.J., 2021. Precipitation and aging phenomena in an ultrafine grained Al-Zn alloy by severe plastic deformation. *J. Alloy. Compd.* 851, 156931. <https://doi.org/10.1016/j.jallcom.2020.156931>.
- [39] J.R. Davis, Ed., *Metals Handbook Desk Edition*. ASM International, 1998. <https://doi.org/10.31399/asm.hb.mhde2.9781627081993>.

- [40] R.W.G. Wyckoff, Crystal Structures, 2nd ed., vol. 2. John Wiley & Sons, Inc., 1964.
- [41] Zheng, J.K., Xu, X., Luo, R., Zeng, X., Chen, B., 2018. Degradation of precipitation hardening in 7075 alloy subject to thermal exposure: a Cs-corrected STEM study. *J. Alloy. Compd.* 741, 656–660. <https://doi.org/10.1016/j.jallcom.2018.01.172>.
- [42] Su, J.-Q., Nelson, T.W., Mishra, R., Mahoney, M., 2003. Microstructural investigation of friction stir welded 7050-T651 aluminium. *Acta Mater.* 51 (3), 713–729. [https://doi.org/10.1016/S1359-6454\(02\)00449-4](https://doi.org/10.1016/S1359-6454(02)00449-4).
- [43] Narayan, J., Narayan, R., 2022. Discovery of double helix and impact on nanoscale to mesoscale crystalline structures. *ACS Omega* 7 (29), 25853–25859. <https://doi.org/10.1021/acsomega.2c03501>.
- [44] D. Caillard and J.L. Martin, Eds., Thermally activated mechanisms in crystal plasticity, vol. 8. Pergamon, 2003. [https://doi.org/10.1016/S1470-1804\(03\)80031-7](https://doi.org/10.1016/S1470-1804(03)80031-7).
- [45] G.M. Ludtka, D.E. Laughlin, The Influence of Microstructure and Strength on the Fracture Mode and Toughness of 7XXX Series Aluminum Alloys, n.d.
- [46] Wu, L.B., Peng, Y.C., Tang, H.Q., Meng, C.C., Zhong, Y.F., Zhang, F.L., Zhan, Y.Z., 2023. Improving grain structure and dispersoid distribution of nanodiamond reinforced AA6061 matrix composite coatings via high-speed additive friction stir deposition. *J. Mater. Process. Technol.*, 117983 <https://doi.org/10.1016/j.jmatprotec.2023.117983>.
- [47] Sun, Z.C., Wu, H.L., Cao, J., Yin, Z.K., 2018. Modeling of continuous dynamic recrystallization of Al-Zn-Cu-Mg alloy during hot deformation based on the internal-state-variable (ISV) method. *Int. J. Plast.* 106, 73–87. <https://doi.org/10.1016/j.ijplas.2018.03.002>.
- [48] Huang, K., Logé, R.E., 2016. A review of dynamic recrystallization phenomena in metallic materials. *Mater. Des.* 111, 548–574. <https://doi.org/10.1016/j.matdes.2016.09.012>.
- [49] Kallien, Z., Hoffmann, M., Roos, A., Klusemann, B., 2023. Correlation of microstructure and local mechanical properties along build direction for multi-layer friction surfacing of aluminum alloys. *JOM* 75, 4212–4222. <https://doi.org/10.1007/s11837-023-06046-4>.
- [50] Sabard, A., McNutt, P., Begg, H., Hussain, T., 2020. Cold spray deposition of solution heat treated, artificially aged and naturally aged Al 7075 powder. *Surf. Coat. Technol.* 385, 125367. <https://doi.org/10.1016/j.surfcoat.2020.125367>.
- [51] Li, S.S., Yue, X., Li, Q.Y., Peng, H.L., Dong, B.X., Liu, T.S., S, T., Yang, H.Y., Fan, J., Shu, S.L., Qiu, F., Jiang, Q.C., 2023. Development and applications of aluminum alloys for aerospace industry. *J. Mater. Res. Technol.* 27, 944–983. <https://doi.org/10.1016/j.jmrt.2023.09.274>.
- [52] Blakey-Milner, B., Gradi, P., Snedden, G., Brooks, M., Pitot, J., Lopez, E., Leary, M., Berto, F., Du Plessis, A., 2021. Metal additive manufacturing in aerospace: a review. *Mater. Des.* 209, 110008. <https://doi.org/10.1016/j.matdes.2021.110008>.