Microstructure characterization and mechanical behaviour of laser additive manufactured ultrahigh-strength M54 steel

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Abstract
Ultra high-strength M54 steel blocks were fabricated by laser metal deposition. The microstructure and mechanical behavior of the material were investigated systematically. The microstructure of the as-deposited M54 steel is anisotropic; the cross-section (XOY plane) has a cellular structure, whereas the longitudinal section (XOZ and YOZ planes) shows a mixture of alternating cellular and columnar forms. Compositional segregation is present at the cell walls (interdendritic regions) in the as-deposited state, resulting in retained austenite at the cell walls. The cross-sectional XOY plane contains 10.08% austenite, whereas the XOZ and YOZ planes contain 24.59% and 22.4% austenite, respectively. The retained austenite at the cell wall (interdendritic region) has low thermal and mechanical stability and disappears after the cryogenic treatment or is transformed into martensite during a tensile test. The as-deposited samples show anisotropic mechanical properties. The transverse samples exhibit stronger transformation-induced plasticity (TRIP) and work hardenability with a lower yield strength of 662 MPa and higher ultimate strength of 1982 MPa, corresponding to a higher amount of retained austenite in this direction. The longitudinal ultimate strength and yield strength are 1832 MPa and 997 MPa, respectively. The ductility and toughness are also largely anisotropic, and their reduction in the transverse direction is only 1/3 of that in the longitudinal direction. The Vickers hardness of the microstructure increases slightly from the bottom to the middle and upper part of the sample due to less thermal cycling in the upper part.

1. Introduction

Ultra high-strength steel (UHSS) is an essential material in the aerospace industry due to its high strength, toughness, and ductility. The recently developed UHSS grade Ferrium® M45 steel partially compensates for the reduction in the Co content by increasing the Mo and W contents [1]. The conditions of use are typically over-aging (310 °C for 5 h), and the source of strength is mainly lath martensite and martensitic packages with high dislocation density, fine and uniformly dispersed M$_2$C (where M refers to chromium, molybdenum, or iron) alloy carbides, and thin film-like reverse austenite with a martensitic lath interface [2–5].

M54 steel components are typically manufactured by forging followed by machining; however, some components with complex or asymmetric geometries are difficult to forge and machine. Furthermore, due to insufficient hot workability, the dimensions of M54 steel forgings are generally no larger than 300 mm in diameter, limiting their use in large components [1, 6–9]. In addition, the non-uniformity of the microstructure and the mechanical properties of large forgings represent problems.

Laser metal deposition (LMD) is an advanced additive manufacturing technology that uses a high energy density laser as a heat source to melt the powdered materials and produces dense metal parts in near-net shapes according to a predetermined trajectory [10, 11]. It has many attractive advantages over conventional
manufacturing processes for producing difficult-to-process high-performance large and critical metallic components (UHSS), e.g., no mold required, little waste and post-deposition machining, high material buy-to-fly ratio, short production cycle, low production costs, few limitations of the component geometry, excellent design flexibility [12–17], high material utilization, high-efficiency, and one-shot shaping [18, 19].

In recent years, LMD has been widely used for manufacturing components made from titanium alloys, high-temperature alloys, and stainless steels [20–24]. In particular, the comprehensive mechanical properties of laser deposited specimens are comparable to those of forged parts due to the dense, fine, and uniform microstructure. Therefore, LMD is a promising method for manufacturing M54 steel components with large dimensions and complicated configurations.

Many studies investigated the microstructure and mechanical behavior of martensitic steels fabricated by LMD or similar laser additive manufacturing techniques. Mohammad et al [25] investigated the mechanical behavior of laser-fused AISI 420 martensitic stainless steel and found that the specimens were anisotropic in the transverse and longitudinal directions. A one-hour heat treatment at 565 °C post-cladding eliminated the anisotropic behavior of the laser-cladded 420 stainless steel. Ran et al [26] found that the as-deposited microstructure of AerMet100 UHSS consisted mainly of grain boundary isotropic ferrite, intercrystalline irregular pre-eutectic ferrite, plate-like upper bainite, pin-like lower bainite, and retained austenite. The fracture of the longitudinal specimen consisted of penetrating cracks across the grain and intergranular cracks compared to pure intergranular cracks in the transverse specimen. In general, the effect of the heat treatment on the microstructure and mechanical behavior depends to a large extent on the original microstructure. In previous studies, the optimization of the heat treatment of M54 steel focused mostly on forgings. An ultrahigh temperature, rapid heating and cooling, and repeated thermal cycling in the micro-zone during LMD can cause non-equilibrium cyclic phase transformation, resulting in a microstructure that differs significantly from that of the conventional process [27]. It is unknown whether the properties of LMD M54 steel are comparable to those obtained from traditional forgings. The first step is to investigate the as-deposited microstructure. Accordingly, this study focuses on the microstructure and mechanical behavior of M54 steel after LMD.

2. Experimental

2.1. Laser metal deposition
The spherical shape of the M54 steel powder used in the laser additive manufacturing of the samples is shown in figure 1, and the chemical composition is listed in table 1. The 53–150 um diameter powder was prepared using a plasma rotating electrode atomization method. The laser additive equipment included a TruDiode 4006 laser, an ABB six-axis robot and indexer, a D70 coaxial powder feeding system with a cladding head, and a double cylinder powder feeder. The laser power was 1800 W, the scanning speed was 2.6 mm s$^{-1}$, the powder feeding speed was 16.21 g min$^{-1}$, and the substrate consisted of 30 CrMnSiA high-strength steel. The final size of the bulk material was
50 mm (length) × 40 mm (width) × 80 mm (height) with good densification. The hardness was determined in the deposition direction (DD), and the tensile specimens were cut in two directions, as shown in figure 2.

2.2. Microstructure characterization

Squares with dimensions of 10 mm × 10 mm × 5 mm were cut as metallographic samples using a wire cutting tool in the DD and in the transverse scanning direction (SD). Then, the blocks were ground with SiC abrasive paper and mechanically polished using a diamond polishing paste. The phase composition was determined at three cross-sections in the middle of the block, e.g., XOY, YOZ, and XOZ (illustrated in figure 2) using a GX-53 optical microscope (OM) and an FEI Quanta 650 thermal field emission scanning electron microscope (FE-SEM) after etching with 4% nitric acid alcohol. The molecular structure was identified using x-ray diffraction (XRD) with a voltage of 35 kV and a current of 40 mA. The solidified microstructure was examined by a JEOL JXA-8100 electron probe micro-analyzer (EPMA). Electron backscatter diffraction (EBSD) analysis of the samples was performed using an FE-SEM (FEI Quanta 650) equipped with Orientation Imaging Microscopy software (OIM 6.2). In addition, a 3 mm thick transmission slice was cut in the XOY plane, ground to 0.05 mm, and ion-milled using an RL-1 precision ion polishing system. The nano-scale morphology was determined by a Tecnia G2F20 transmission electron microscope (TEM) with a beam voltage of 200 kV.

2.3. Mechanical performance testing

The small tensile specimens and impact specimens were tested for the mechanical properties, as shown in figure 3. The longitudinal direction was the DD (referred to as the Z-direction), and the transverse direction was the SD (referred to as the X-direction). The samples were cut from the upper middle part of the deposited block. Samples of the same size were cut from a forged bar for the subsequent comparison with the additively manufactured specimens. The Vickers hardness was measured with an EV500–2A semi-automatic Vickers hardness tester with a load of 1000 g. The tensile properties of the depositions were tested at room temperature using a WDW-300E electronic universal testing machine, and the tensile fractures were characterized using an FEI Quanta 650 thermal FE-SEM equipped with an energy-dispersive spectrometer.

Figure 2. Schematic diagram of the LMD M54 thick steel plate sampled at different locations.

Figure 3. (a) Size of the tensile samples (mm) tested at room temperature; (b) size of the impact samples (mm).
3. Results and discussion

3.1. As-deposited microstructure of LMD M54 steel

The OM images of the cross-section XOY and the longitudinal cross-sections XOZ and YOZ of the M54 steel after laser deposition are shown in figures 4(a)–(c). The solidification microstructure of the as-deposited M54 steel mainly consists of rapidly directionally solidified columnar grains with interior well-aligned cellular dendrite structures, showing several deposited layers. As shown in figures 4(b) and (c), laminar bands occur between the adjacent deposited layers, and the XOY cross-section in figure 4(a) has a honeycomb-like cellular structure (20–30 μm). The non-equilibrium microstructure with the characteristic of directional solidification is formed by the continuous epitaxial growth of solidified crystals of adjacent deposited layers. In figure 4(c), the microstructure of the deposited M54 steel in the YOZ plane consists of alternating cellular and columnar crystals. Since the heat accumulation and the temperature gradient are the highest near the bottom of the molten pool, columnar crystals are most likely to form, and cellular crystals are located in the middle of the molten pool. A transition from cell to columnar crystals is observed between the lamellar channels in figure 4(c). Bright and dark etched bands occur at the melt pool boundary, and the columnar crystal growth direction is consistent with the DD.

The boundary between the layers is black, and the inner area of the layer is gray. The reason is that the layer of solidified deposits is partially remelted when the next layer is deposited. The microstructure at the bottom of the molten pool is refined due to rapid solidification. In contrast, the microstructure in the heat-affected zone is coarser due to repeated tempering and cyclic transformation. The difference in the microstructure in different
parts of the samples results in different degrees of corrosion and the formation of a layered structure with light and dark layers, as shown in figure 4(b).

The microstructures of the deposited M54 steel in the three observation planes are shown in figure 5. The XOY and XOZ planes are composed of a cellular structure, which is typical of additive manufactured steels. The evolution of the microstructure also depends on the thermal history during LMD, as shown in figure 6(a). Columnar crystals are formed in the initial stage of solidification, as the material undergoes extensive temperature fluctuations with rapid heating and cooling, resulting in a high temperature gradient and directional solidification. As the deposition progresses, the range of temperature fluctuation decreases, and the solidified block gradually warms up to an almost constant temperature, forming a multilevel microstructure. Meanwhile, the first solidified parts constrain the deformation of the latter parts, leading to increased internal stress and promoting dislocation density. The dislocations eventually aggregate by sliding due to the elevated temperature to minimize the energy of the system, leading to the formation of cell walls (figure 6(b)) [28]. Figures 5(d) and (e) show that defects such as cracks and holes are located at the cell walls, suggesting the formation of a suitable cell structure, although the dislocations cannot be characterized. The secondary microstructure inside the cell is the upper bainite and martensite; however, the cell structure is unclear in figure 5(f).

Although the rapid cooling during the additive manufacturing process limits microscopic segregation, solute redistribution still occurs in some cases, and some second phases can precipitate at the cell walls (interdendritic regions) [29]. Figure 7 shows the EPMA micrographs of the samples after deposition. It can be
seen that the cytosolic microstructure is inhomogeneous, and Mo is concentrated at the cell walls while the austenite former Ni is not significantly segregated.

Previous studies have shown [30] that $\text{M}_2\text{C}$ carbides (M is Mo, W, etc) are excellent strengthening precipitates that can improve the mechanical properties of ultrahigh-strength secondary hardening steels. However, the XRD results in figure 8 show that the main phases of the M54 after deposition are bcc-structured $\alpha$-Fe and fcc-structured $\gamma$-Fe; no diffraction peaks of other precipitated phases (e.g., carbides) are detected. The

Figure 8. XRD spectrum of M54 steel after deposition in the three observation planes (XOY, XOZ, YOZ). A refers to austenite, and M refers to martensite.

Figure 9. EBSD results for the XOY, XOZ, and YOZ surfaces of the M54 steel after deposition. XOY: (a) austenite (red) distribution, (b) austenite orientation, (c) IPF plot of martensite. XOZ: (a) austenite (red) distribution, (b) austenite orientation, (c) IPF plot of martensite [001]. YOZ: (a) austenite (red) distribution, (b) austenite orientation, (c) IPF plot of martensite.
The diffraction peaks of the \( \gamma \) \((111)\) are higher in the XOZ and YOZ directions.\( \alpha \)-Fe is the main phase composition (about 70%-90%), and the austenite contents are 10.08% in XOY, 24.59% in XOZ, and 22.4% in YOZ.

Austenite is concentrated near the cell walls/columnar boundaries (figures 9(a), (d), (g)) because of the segregation of Mo (figure 7) and other elements at the cell walls. This distribution enhances austenite stability and reduces the Ms point, resulting in an incomplete transformation of the martensite phase. Figures 9(b), (e), and (h) show the different orientations in different colors. The dispersed austenite fragments have the same colors on each side, indicating that the austenite orientations are inherited from the prior austenite grains. These results indicate that the austenite is retained rather than reversed and grows in various directions. Furthermore, figure 9(b) and (c) show two crystallographic orientations, suggesting a tilt and twist boundary between two austenite grains.

Figure 10 shows the TEM images of the as-deposited M54. The microstructure consists mainly of lath martensite (nano-sized twin martensite is also observed in figure 10(c) [31]) and thin film-like retained austenite at the interface (shown by arrows in figure 10(a)). Figure 10(f) shows the HRTEM images and fast Fourier transform (FFT) analysis results of the selected region, indicating that the lattice distortion might be M\(_2\)C carbides embedded in the laths.

Figure 11 illustrates the \textit{in situ} EBSD morphology of the same selected area before (a) and after (b) the cryogenic treatment (soaked in liquid nitrogen for 8 h). The [001] direction-oriented retained austenite at the...
columnar grain boundary has almost entirely disappeared after the cryogenic treatment. The reduction amount is about 5%–8%, which is consistent with the XRD results in figure 12. This finding indicates that the retained austenite after deposition possesses low thermal stability.

3.2. Tensile properties and fracture behavior

Figure 13 shows the engineering stress-strain curves of the M54 steel in two directions. The mechanical properties are summarized in table 2. The mechanical properties of the as-deposited samples show anisotropy in the two directions. Impact toughness of longitudinal direction is 54 J cm\(^{-2}\), 80% higher than that of transverse direction. The tensile strength is 150 MPa higher in the transverse direction than in the longitudinal direction, and the longitudinal yield strength is 335 MPa higher (33.4%) than the transverse yield strength. In other words, the transverse samples show a much longer work-hardening stage, which is attributed to the transformation-induced plasticity (TRIP) of the retained austenite. This result is consistent with the higher austenite amounts in the XOZ and YOZ samples.

Figure 14 shows the EBSD results of the cross-section and longitudinal section near a tensile fracture of the deposited M54 steel. Figures 14(a) and (b) indicate that the retained austenite at the cell wall or the columnar boundary has nearly disappeared after tensile deformation due to the TRIP effect, indicating the low mechanical
stability of the retained austenite. The ductility and toughness are largely anisotropic, and their reduction in the transverse direction is 1/3 that in the longitudinal direction.

The SEM images of the transverse and longitudinal fractures of M54 after deposition are shown in figure 15. Figure 15(a) illustrates the macroscopic fracture of the transverse sample. The fracture consists of three areas divided by two edges, where the left and right flanks are tear lips. Figure 15(b) shows the middle of the microscopic fracture, indicating a mixture of cleavage planes with many secondary cracks and small shallow dimples. Figure 15(c) shows the macroscopic morphology of the longitudinal tensile fracture with a large fiber zone and surrounding shear lips, which is typical for ductile fractures. Figure 15(d) shows the magnification of the fiber zone in figure 15(c), indicating large and deep dimples. The fracture morphology corresponds well to the anisotropic toughness in the different directions.

The low ductility of the sample in the transverse direction is attributed to the low crack resistance of the columnar grain boundary of the original austenite, which readily forms coarse cracks (figure 5(e)). In addition, the transverse tensile stress promotes crack propagation, and the stress concentration at the crack tip induces crack propagation perpendicular to the tensile stress direction [32]. Therefore, in the transverse direction, the applied tensile stress is perpendicular to the grain boundary of the sample, and the sample propagates along the austenite grain boundary with low ductility. In the longitudinal direction, the applied tensile stress is parallel to the columnar grain boundary of the specimen. The martensite microstructure inside the grain has high crack propagation resistance, which is conducive to the shrinkage of the longitudinal specimen.
The hardness from the bottom to the top was tested in the as-deposited state. The results in figure 16 show that the hardness varies from the bottom to the middle and upper parts. The top part is the hardest, and its hardness values is comparable to that of the wrought iron samples after the heat treatment. The middle and bottom parts have lower hardness values due to reheating and repeated tempering of the martensite. However, the hardness values are much higher for the steel undergoing the deposition than no heat treatment.

Table 2. Room-temperature mechanical properties of the M54 steel in two directions.

|       | UTS/MPa | YS/MPa | EL/% | RA/% | (\(\text{\(\varepsilon_{\text{tr}}\)}\)) cm\(^{-2}\) |
|-------|---------|--------|------|------|----------------------------------|
| X(TD) | 1982    | 664    | 10   | 10   | 29.5                             |
| Z(DD) | 1832    | 997    | 9.5  | 28   | 54.0                             |

Figure 15. Fracture morphology of M54 after deposition. (a) Transverse fracture and (b) enlarged fiber area. (c) Longitudinal fracture and (d) enlarged fiber area.

Figure 16. Hardness curves in the deposition direction of the as-deposited LMD M54 steel samples.
4. Conclusions

In this study, the microstructure of M54 martensitic steel fabricated by LMD was characterized using optical microscopy, XRD, electron probe microanalysis, scanning electron microscopy, EBSD, and transmission electron microscopy. The mechanical properties of the samples were assessed in the transverse and longitudinal directions. The results are summarized as follows.

1. The as-deposited structure of the M54 steel was characterized by directional solidification, i.e., columnar crystals growing in the DD. The microstructure was anisotropic; the cross-section (XOY plane) exhibited a cellular structure, whereas the longitudinal section (XOZ and YOZ planes) showed a mixture of alternating cellular and columnar forms.

2. Mo is concentrated at the cell walls (interdendritic region), resulting in lower Ms point and a higher austenite content. The cross-sectional XOY plane contained 10.08% austenite, whereas the XOZ and YOZ planes contained 24.59% and 22.4% austenite, respectively. The retained austenite at the cell walls (interdendritic region) after deposition had low thermal and mechanical stability. The austenite disappeared after the cryogenic treatment or was transformed into martensite during the tensile test.

3. The as-deposited samples showed anisotropic mechanical properties. The transverse samples exhibited a stronger TRIP effect and work hardenability with a lower yield strength of 662 MPa and higher ultimate strength of 1982 MPa, corresponding to the higher amount of retained austenite in this direction. The longitudinal ultimate strength and yield strength were 1832 MPa and 997 MPa, respectively. The ductility and toughness were also largely anisotropic, and their reduction in the transverse direction was only 1/3 of that in the longitudinal direction. The Vickers hardness increased slightly from the bottom to the top of the sample due to less thermal cycling in the upper part.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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