Crack-free in situ heat-treated high-alloy tool steel processed via laser powder bed fusion: microstructure and mechanical properties

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GRAPHICAL ABSTRACT

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In this study, high-alloy tool steel S390 was processed crack-free and dense for the first time using laser powder bed fusion (LPBF). The resulting mechanical properties and microstructure of the LPBF steel parts were investigated. High-alloy tool steels, such as high-performance high-speed Boehler S390 steel (containing 1.64 wt% C and W, Mo, V, Co, and Cr in the ratio 10:2:5:8:5 wt%), are prone to cracking when processed using LPBF because these steels have high carbon and carbide-forming alloying elements content. Cracks are induced by thermal stresses and solid-phase transformation, combined with weak grain boundaries caused by segregated primary carbides. Substrate plate heating reduces thermal stresses and enables in situ heat treatment, thus modulating solid-phase transformation and carbide precipitation and preventing cracking during cooling. The resulting microstructure, precipitations, and mechanical properties of the as-built LPBF specimens, which were in situ heat-treated at 800 °C, and the conventionally post-heat-treated specimens were assessed using optical microscopy, scanning electron microscopy, transmission electron microscopy, energy-dispersive X-ray spectroscopy, electron backscatter diffraction, X-ray diffraction, hardness testing, bending testing, and density measurement. In situ heat treatment impacts microstructure, precipitation behavior, and solid-phase transformation, causing a change in the microstructure of the material along the build direction due to different thermal histories. The as-built specimens exhibit a hardness gradient along the build direction of 500 Hv1 to 800 Hv1 in the top layer. The average bending
1. Introduction

Tools for the aerospace, automotive, and medical technology industries have high requirements for strength, hardness, toughness, temperature, and wear resistance. High-speed steels (HSS) such as the Boehler S390 Microclean meet these requirements, exhibit good hot hardness, bending strength, and machinability, and are frequently used to produce milling cutters and drills. Tools made of S390 are currently produced conventionally via powder metallurgy (PM). The established conventional PM manufacturing process is complex and typically involves powder production, hot isostatic pressing (HIP), forging, and subsequent heat treatment with quenching and triple tempering [1, 2, 3, 4, 5]. With additive manufacturing (AM), near-net-shaped tool parts can be fabricated directly from powder raw material without time-consuming or costly intermediate steps. Furthermore, new functions and geometries can be implemented, such as internal cooling channels that extend the lifetime of tools or tool components and/or improve performance. The two most prominent powder bed-based AM processes for tools are electron beam melting (EBM) and laser powder bed fusion (LPBF). High-alloy tool steels such as the S390 (containing 1.64 wt% C and W, Mo, V, Co, and Cr in the ratio 10:2:5:8:5 wt%) are considered non-weldable and prone to cracking. To process HSS using LPBF, adjustments must be made and framework conditions considered. To reduce the thermal gradient and thus avoid crack formation induced by thermal stresses, substrate plate heating is applied during the LPBF fabrication of various materials. Several studies have been performed using substrate plate temperatures of 200–400 °C on HSS steels with relatively low carbon content, e.g., HSS M2 [6, 7, 8] and the HSS M50 [9], all yielding similar defect patterns, i.e., cold cracks. Furthermore, substrate plate heating has been demonstrated to reduce crack density, defect frequency [10, 11], and residual stresses in H13 hot work tool steel [12]. Only a few tool steels can be successfully processed using LPBF without defects [13, 14], not least because of the cold cracking problem [15]. Feuerhahn et al. [16] and Geenen et al. [17] have processed high carbon steel via LPBF with substrate plate heating. Geenen et al. were the first to produce and investigate an HSS (i.e., HSS 1.3344), with a C content of 1.2 wt%, using LPBF at 300 °C [17]. Using LPBF at 350 °C, Saewe et al. managed to process an HSS composed of W, Mo, V, and Co (6:5:3:8 wt%), which already has a high content of carbide formers and carbon (1.2 wt%), without cracking [18] and investigated the influence of the increased C content on this alloy [19]. A recent study by Galbusera et al. investigates the processability and cracking behavior of similar high-alloy tool steels fabricated using LPBF, exploring various analytical and empirical models (e.g., cooling rate, the carbon equivalent, martensite start temperature, solidification interval, and energy density) for shortening the experimental effort invested in alloy design [20]. They conclude that conventional LPBF machines may be inadequate for mitigating the characteristic defects.

Boehler S390 steel is an outstanding model among HSS tool steels characterized by high bending strength (up to 4500 MPa), hardness (up to 68 HRC) [1], temperature resistance, and abrasion resistance, with potential for subsequent machining and heat treatment, whether produced conventionally or via AM. Consequently, it is popular in the industry and has been studied extensively. Over the years, several research projects have been devoted to the study of the S390 PM material [2, 3, 4, 21, 22, 23, 24, 25]. In 2004, Niederkofler et al. were the first to investigate the matrix of this PM-processed tool steel and found 20 nm-sized carbides using the atom probe analysis method [25]. This was followed by extensive characterization of the typical carbides (i.e., MC and M₂C) and the martensite matrix microstructure by Godec et al. in 2010 [4]. The influence of posttreatment, including cryogenic treatment and plasma nitriding, on mechanical properties such as hardness, fracture toughness, and tribology has also been investigated [3, 21]. A recent study investigates heat treatment—specifically, the variation of duration and temperature—and its effect on the mechanical properties, composition, solubility, and precipitation of carbides [2, 23]. These studies confirm that the mechanical properties resulting from heat treatment are advantageous and recommend a heat treatment process that includes quenching and triple tempering for the formation of a martensite matrix and to precipitate micron-sized carbides.

One study examines the AM of S390 using EBM at 780 °C to produce crack-free specimens, extensively characterizes the solidification structure, and investigates the microstructure, carbide precipitation behavior, and mechanical properties [26]. According to this study, dendritic austenitic grains have a size of approximately 50 μm and are the first set of grains to solidify, followed by MC carbides of V and W, which solidify in the interdendritic regions. The morphology of the matrix phase was characterized as a combination of equiaxed dendritic grains and columnar dendritic grains interspersed with primary carbides several micrometers in size, with a primary carbide network of M₂C comprising W, Fe, Mo, and Co in order of significance. At a density of 97.7%, mechanical properties similar to those of conventional PM S390 were obtained with a hardness of 65.5 HRC and a tensile strength of 2600 MPa.

The mechanical properties of steel are influenced by the size of the grains and carbides. Based on the Hall–Petch and Orowan mechanisms, the smaller the grain size, the higher the expected strength, and the smaller the distances of precipitates, the greater the expected hardness, respectively [27, 28]. Dissolved carbide-forming elements can be precipitated as secondary carbides during triple tempering in the form of MC (mainly vanadium carbide), M₂C (mainly Fe₃W₅C)a carbides, and the metastable M₇C₃ (containing V, W, Mo, Fe) in the submicrometer range. In addition, triple tempering in the subsequent heat treatment contributes to the transformation of retained austenite (RA) into martensite, with face-centered cubic (FCC) RA transforming into a body-centered (BC) Fe phase—more precisely, into body-centered tetragonal (BCT) martensite, as reported by Peng et al. for this high alloyed steel [23]. The distinction between solidification structures dendrites, cells, and grains should be mentioned here. For AISI 316L (DIN 1.4404), maraging steel (DIN 1.2709), and other materials in LPBF, cellular solidification structures with cell sizes of approx. 500 μm transverse to the extent of the cells and a local misorientation of <2° with respect to each other are reported within grains with high-angle grain boundaries [29, 30, 31]. For HSS in LPBF, a bimodal microstructure with coarse columnar grains (up to 50 μm) and fine equiaxed grains (of approx. 1 μm) exhibiting a dendritic substructure is reported [19].

This study is the first to investigate the microstructure and mechanical properties of HSS S390 specimens produced via LPBF. HSS S390 specimens produced under as-built and heat-treated conditions are compared against conventionally produced PM HSS and EBM HSS.

2. Materials and methods

2.1. Powder characterization

The material used is the as-atomized powder of PM Boehler S390 Microclean steel. The chemical composition of the powder was determined by the manufacturer by combustion analysis (C, S), carrier gas hot extraction (O), and inductively coupled plasma optical emission spectrometry (remaining elements). The results are listed in Table 1. The atomized S390 powder was sieved through a 63-μm sieve, resulting in particle size distributions (PSD) of D₁₀ 15 μm, D₅₀ 37 μm, and D₉₀ 66 μm.
determined with the Mastersizer 3000 (Malvern Instruments, UK). The sieved powder was then vacuum-dried. The solid density of this material is 8.1 g/cm$^3$, according to Boehler Edelstahl [1]. The bulk density, tap density, and flowability of the powder were determined to be 4.57 g/cm$^3$, 5.21 g/cm$^3$ and 18.77 g/s, respectively. Figure 2 presents a scanning electron microscope (SEM) image of the predominantly spherical powder.

2.2. LPBF production and heat treatment

A customized Aconity3D LPBF system (Aconity3D GmbH, Germany) equipped with a 400 W IPG Nd-YAG laser (IPG Laser GmbH, Germany) and a substrate plate that can be heated to a temperature of 800 °C was used for the production of specimens. Argon was used as a process gas, and the oxygen level in the building chamber was kept below 20 ppmv. A high-resolution image of a powder particle surface taken in secondary-electron (SE) mode (Figure 2(c)) indicates that the grains grew distinctly during solidification. A high-resolution image of a polished as-delivered condition, the heat-treated specimens were performed via X-ray diffraction (XRD) using an Empirean Panalytical diffractometer (Malvern Panalytical Ltd., UK) in a Bragg-Brentano configuration, with Cu Kα radiation between 15° and 110°. The relative density of all cube-shaped specimens (10 × 10 × 10 mm) was measured using Archimedes’ principle, and also confirmed for some specimens using metallographic density determination.

2.4. Mechanical testing

The three-point bending test was performed at ambient temperature in line with the DIN EN ISO 7438:2020 standard on polished as-built specimens (5 × 5 × 35 mm) on a Shimadzu AGS-10kNX (Shimadzu, Japan) universal testing machine. The testing force was applied in the direction opposite to the build direction, and the test was performed four times for each parameter configuration. The Vickers hardness test (HV1) was performed in compliance with the DIN ISO 6507 standard using the Shimadzu HMV-G microhardness tester device (Shimadzu, Japan). When measuring the hardness gradient across the building height, three hardness values were averaged in each case.

3. Results

3.1. Powder properties

The as-atomized S390 PM powder has a wide size distribution, with larger particles than the powder typically used in LPBF. Sieving with a mesh size of 63 μm separated the unwanted large particles, thus establishing a PSD suitable for LPBF. Figure 1 shows the size PSD of the powder before and after sieving, measured using laser diffraction. Table 3 lists the associated characteristic diameters D10, D50, and D90, and the bulk density, tap density, Hausner ratio, and flowability. Both the bulk density and the tap density were reduced by sieving the coarse fractions. The Hausner ratio did not change. However, compared to the as-delivered condition, the flowability of the powder improves significantly after sieving. Figure 2 shows SEM images of the sieved powder. The majority of particles possess spherical morphology; however, there is a small fraction of deformed particles and particles with satellites (Figure 2(a)). In the powder cross section presented in Figure 2(b) and Figure 2(d), only a few small pores with diameters of <10 μm are visible. A high-resolution image of a powder particle surface taken in secondary-electron (SE) mode (Figure 2(c)) indicates that the grains grew dendritically during solidification. A high-resolution image of a polished cross section taken in backscattered electron (BSE) mode (Figure 2)
Table 2. Optimized process parameters determined experimentally in a preliminary parameter study.

| Parameter                      | Value          |
|--------------------------------|----------------|
| Laser power, $P_\text{L}$ (W)  | 100–400        |
| Scan velocity, $v_\text{s}$ (mm/s) | 400–2000     |
| Hatch distance, $h_\text{L}$ (μm) | 60–120        |
| Nominal layer thickness, $t$ (μm) | 30            |
| Volumetric energy density, $E_v$ (J/mm$^3$) | 16.7–333.3   |
| Scan strategy                  | Lines/Islands/Stripes |
| Scan vector length (mm)        | 2.5–10         |
| Substrate plate temperature ($^\circ$C) | 500, 600, 800 |

Table 3. Powder characterization with laser diffraction and bulk density, tap density, and flowability measurements.

| Material                  | $D_90$ (μm) | $D_10$ (μm) | $D_50$ (μm) | Bulk Density (g/cm$^3$) | Tap Density (g/cm$^3$) | Hausner ratio | Flowability (g/s) |
|---------------------------|-------------|-------------|-------------|-------------------------|------------------------|---------------|------------------|
| As-atomized PM S390 Powder| 20.6 ± 0.2  | 69.7 ± 2.7  | 292 ± 14.4  | 5.06 ± 0.08              | 5.97 ± 0.08            | 1.18          | 12.02 ± 1.10     |
| Sieved S390 Powder        | 15.5 ± 0.3  | 36.7 ± 0.1  | 66.3 ± 0.2  | 4.57 ± 0.02              | 5.21 ± 0.06            | 1.14          | 18.77 ± 6.45     |
with a red square (38 wt% and 35 wt%, respectively), with elevated levels of the other carbide-forming elements and carbon. The green square marks the phase, which is dominated by V (38 wt%) and has a high Cr and C content (7 wt% and 6 wt% respectively). W, Fe, and Mo are also present in the area. The black phase is marked in blue as an impurity and shows a segregation of Mn (14 wt%) associated with S (10 wt%) and Fe.

Phase analysis of the powder for LPBF, the as-built parts in the top layers and bottom layers, and the heat-treated specimens is presented in Figure 10. The XRD patterns of the specimens show characteristic peaks for the dominant Fe phases: austenite FCC, martensite BCT, and ferrite BCC, and the carbides MC and M6C are also identifiable. For the powder, peaks of austenite and MC are visible. For the as-built top layer, austenite peaks, martensite double peaks, and MC are visible. In the as-built bottom layer, BCT patterns are visible, in addition to a weak MC peak. In the heat-treated condition, prominent peaks are observed for BCT and MC, accompanied by a slight MC peak.

### 3.3. Mechanical properties

The results of the bending tests are plotted in Figure 11, which shows that the strength of the specimens increases after heat treatment. The gradient of hardness along the height of the specimen is presented in Figure 12. For the as-built specimens, a gradient of hardness is observed, with the increasing height measured in mm increments and the last built top layers having the highest hardness values. The heat-treated specimens show no hardness trend. Table 3 presents two mechanical properties: bending strength and Vickers hardness (HV1) of the as-built and heat-treated specimens. The mean values, including the maximum and minimum of both the bending strength and hardness, are also presented in Table 3. The as-built specimens exhibit a mean bending strength of 2535 MPa, with a maximum value of 2775 MPa and a minimum of 2330 MPa. The hardness of the as-built specimens ranges from 499 HV1 in the lower layers to 798 HV1 in the top layer. The heat-treated specimens exhibit a mean bending strength of 2799 MPa, with a maximum value of 3504 MPa and minimum of 2613 MPa. The mean hardness of the heat-treated specimens is 611 HV1, with a 663 HV1 maximum and 561 HV1 minimum, without a trend along the height of the specimen.

### 4. Discussion

The Boehler S390 Microclean powder used for the tests in this study has a low content of S, P or O impurities (Table 1). The powder particles show few satellites, no internal porosity, a spherical particle shape (Figures 2(a) and 2(b)), and a PSD in the typical LPBF size range (Figure 1). Therefore, the powder exhibits high flowability, high bulk, and high tap density and is suitable for processing via LPBF (Table 3). The sieved powder’s lower theoretical packing density can explain the slight
decrease in tap and bulk density to a PSD typical for LPBF [32]. The significant increase in flowability can be explained by the changed storage conditions (vacuum drying) [33]. A preliminary study was carried out with this pre-processed powder. In the preliminary study, the process parameters and substrate plate temperature (500 °C, 600 °C, and 800 °C) were varied over a wide range, and only a substrate plate temperature of 800 °C enabled process-safe production of dense (>99.8%) and crack-free specimens (Table 2). HSS, with a high carbon content, tends to crack because of thermal stresses, embrittlement due to martensite formation, incoherent primary carbides, and the associated increase in stress concentration [24, 34, 35, 36]. This is especially true for the S390, which has a carbon content of 1.64 wt% and a high carbide-forming alloying element content.

In situ heat treatment (HT) of HSS, and specifically S390, is dependent on temperature and duration [27]. Because of the in situ HT via substrate plate heating at 800 °C during the LPBF process, layers at different heights of the specimen were heat-treated for different durations. This results in differences in the material with respect to phase transformations and the precipitation of carbides, which are reflected in specific mechanical properties [1, 27].

4.1. In situ heat treatment and microstructural evolution

The HT of HSS, and specifically S390, is dependent on temperature and duration [27]. Because of the in situ HT via substrate plate heating at 800 °C during the LPBF process, layers at different heights of the specimen were heat-treated for different durations. This results in differences in the material with respect to phase transformations and the precipitation of carbides, which are reflected in specific mechanical properties [1, 27].
For 10 mm high cube specimens produced in these experiments, the lowest layer was exposed to the 800 °C substrate plate heating for 3 h. The higher the layer in the specimen, the shorter the duration of exposure to in situ HT via substrate plate heating. In addition to the in situ HT at 800 °C via substrate plate heating, each solidified melt track underwent a brief, repeated annealing from heat exposure to adjacent and overlying melt tracks. Consequently, the top layer differs from the other layers because, first, it was annealed exclusively by laterally adjacent melt tracks and not by the fabrication of overlying layers. Second, it was exposed to in situ HT via substrate plate heating at 800 °C for a relatively short time. All layers in the as-built specimens were subjected to different durations of in situ HT via substrate plate heating. In addition, the top layer has a unique position because it is the only layer that is not reheated (i.e., annealed) to high temperatures. The resulting differences along the buildup direction are shown in the LOM image in Figure 3(a) using an etched specimen area. The in situ HT duration is shortest for the top layer, which appears bright in the LOM image, while the underlying heat-treated layers all the way down to the bottom layer appear progressively darker. This is because the nital etching of the different phases (e.g., RA, martensite, and carbides) is rendered with a different level of brightness in LOM [40]. The hardness of the specimens decreases with increasing distance from the top layer—and thus with

![Figure 4. SEM micrographs of the center of the as-built specimen. (a) IQ (image quality) image with darker areas marked in yellow in a brighter matrix. (b) IPF mapping of the FCC phase fraction with yellow markings from 4(a). (c) IPF mapping of the BC phase fraction with yellow markings from 4(a). (d) shows a micrograph of the center area of the etched as-built specimen as an overlay of the BSE and secondary electron (SE) detectors. The Z-direction indicates the build direction. The red dashed lines indicate melt pool borders. Fine globular structures are marked in blue and columnar structures with black arrows.](image)

![Figure 5. SEM BSE image of the center of the specimen. (a) Image of a region of an as-built specimen in which large grains with columnar dendritic growth are visible. Carbides are visible as segregations in the interdendritic areas. (b) Heat-treated specimen: precipitated carbides embedded in martensite matrix.](image)
increasing in situ HT duration—from 798 HV1 to 499 HV1 (Figure 12). In the EBSD images, distinct columnar dendritic FCC grains are visible in the top layer (Figure 3(b)) and embedded in a fine-grained BC structure (Figure 3(c)). The XRD results for the as-built top layer indicate FCC austenite, FCC carbides, and BCT martensite (Figure 10). The FCC fraction in Figure 3(b) consists of austenitic columnar dendritic grains, which can identified as RA, and of much smaller FCC carbides. In contrast, the majority of the microstructure in the top layer BCT corresponds to martensite.

The more extended in situ heat-treated areas in the center and bottom layer of an as-built specimen appear dark in the LOM image in Figure 3(a). In the EBSD images of these areas, almost only BC phases are detected, both in the center of the specimen (Figure 3(e)) and in the bottom layer (Figure 3(g)). Examining the XRD patterns of the bottom layer presented in Figure 10, clear martensitic double peaks are visible. Therefore, the BC phases in the bottom layer can be identified as martensitic BCT phases [41].
In the EBSD image of the as-built specimens in the center and in the bottom layer (Figures 3(d) and 3(f)), at the given magnification of the 250 μm square, the FCC fraction is visible as finely distributed phases in each case. Coarse residual austenite grains as in the top layer are not found in these two areas. This is confirmed by the XRD results, which show no austenite peak for the bottom layer (Figure 10). At the higher magnification in the 50 μm square in Figure 4(b) at the center area of the as-built specimen the detected FCC phases have sizes in the sub-micrometer range, are finely distributed, and are arranged in equally oriented clusters. The dimensions and spacing of these FCC phases, as well as their location in the grain interior, match the interdendritic segregations observed in the SEM BSE image in Figure 5(a). In the TEM image in Figure 6, interdendritic segregations from the specimen center of the as-built specimens are shown in high resolution and show enrichments of C in combination with the carbide-forming elements V, W, Mo, and Cr in the element mapping. This composition has been reported in the literature for the primary FCC MC carbides [23, 26, 39]. The elongated shape and cellular arrangement in Figure 6 indicate that the carbides are segregated in the interdendritic spaces during solidification. The FCC MC carbides can also be detected in the bottom layer in situ heat-treated via XRD (Figure 10). Furthermore, a publication by Jin et al. [26] reports that interdendritic MC primary carbides with similar sizes and spacing are observed during EBM processing of S390. Similar equiaxial and columnar dendritic structures have been reported for EBM of S390, but the structures are much larger than in LPBF [26]. A differentiation between small equiaxial grains with high angle grain boundaries, dendrite arms and solidification cells with small angle grain boundaries will be the task of further investigations and will be presented in another publication.

Based on the identified and localized phases, the microstructural evolution of built-up specimens can now be traced. In the top layer, columnar dendritic RA grains are located at the edges of former LPBF melt pools (Figure 3(b)). The grain growth of the columnar dendritic grains occurs along the temperature gradient in the melt pool, progressing radially from the melt pool edge inward to the melt pool center, as shown in Figure 4(d). During the grain formation process, alloying elements dissolved in the melt are rejected at the solidification front (i.e., at the austenitic dendrite), and these segregate at the interdendritic interstices, as shown in Figure 6, or at the grain boundaries. Fischmeister et al. report that MC carbides can form both eutectically and as primary grains from the melt [42]. In the melt pool center, columnar dendritic grains are no longer observed due to a locally reduced temperature gradient, causing a transition to equiaxed solidification [26, 27]. Once solidified, dendritic regions can be partially remelted by adjacent or subsequent melt traces or heat treated at temperatures close to liquidus. This results in structures similar to those shown in Figure 4(d) and Figure 5(a), consisting of columnar dendritic grains, equiaxial grains, and segregations at the grain boundaries and interdendritic interstices. Similar equiaxial and columnar dendritic structures have been reported for EBM of S390, but the structures are much larger than in LPBF [26].

In essence, after solidification of the meltpool and cooling to 800 °C, FCC phase austenite is present in columnar and equiaxed grains...
accompanied by primary carbides. Martensite formation from austenite occurs only during the eventual cooling from 800 °C and the temperature falling below $M_s$. As mentioned earlier, the duration of in situ HT at 800 °C increases from the top to the bottom layers. The in situ HT of the rapid solidified austenite at 800 °C leads to the reduction of internal stresses and a high dislocation density on the one hand, and on the other hand, to the precipitation of the carbon C and other alloying elements in the form of carbides at the grain or dendrite boundaries [26]. Thus, with increasing in situ HT duration, the carbon content in the austenite decreases toward the bottom layer. The local composition of the austenite grains determines their $M_s$ [43, 44]. Thus, the annealing of austenite, in situ HT at 800 °C, and the resulting precipitation of alloying elements increases the $M_s$ and promotes the transformation of austenite to martensite [44, 45, 46]. Ultimately, upon cooling, this forms a softer, less
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In the in situ HT duration [23, 47], similar to the tetragonally distorted martensite with a hardness gradient corresponding to the in situ HT duration [23, 47], similar to the autotempered martensite as reported by Jin et al. [26]. Thus, large dendritic structures indicate prior austenite grains (Figures 4(a) and 4(b)), which can be identified after cooling as a fine-grained martensitic microstructure in EBSD images (Figure 4(c)) and XRD patterns (Figure 10).

4.2. Heat treatment

The conventional HT of HSS, performed as a separate subsequent process, serves to dissolve the alloying elements during austenitizing, obtaining a supersaturated martensitic matrix via quenching. Then, during triple tempering, HT relaxes the martensite, transforming the RA, and finely precipitating the secondary carbides [1, 2, 48]. In Figure 5, the SEM BSE images show a comparison of the as-built and heat-treated specimens. The interdendritic segregations visible in the as-built specimen in Figure 5(a) are no longer visible after HT (Figure 5(b)). Similarly, the dendritic columnar and equiaxed grains of the matrix are no longer visible after HT. Figure 5(b) shows a fine-grained martensitic structure and equiaxial precipitates, which is a similar microstructure to that reported by Peng et al. [23] for heat-treated PM S390. In the XRD in Figure 10, it is clear that in the HT state, there is a preponderance of martensite, and no RA peaks are visible. In the EBSD images in Figure 7, fine BC phases of a few μm in size can be seen, matching the description of martensite in the literature [23]. The BC matrix is homogeneously interspersed with precipitates, as shown in Figure 7 and Figure 8. These precipitates differ in brightness in the BSE images (Figures 5(b) and 8(c)). In the EDX element mapping in Figure 8 and in the breakdown of the mass fractions of the alloying elements in Figure 9, different types of precipitates can be distinguished:

i. One type, which appears bright in the BSE, is rich in W and Fe and has a high C content in the element mapping. The EDX mapping of this particle in Figure 9 shows the mass fractions with approximately equal amounts of Fe (35 wt%) and W (38 wt%), and a high C content. In the XRD patterns, it can be identified by its characteristic peaks as M6C—more precisely, as Fe3W2C. However, it should be mentioned that atomic percentages do not correspond to the stoichiometry of the phase with 14.3 at% W and 42.5 at% Fe.

ii. The gray phase in the BSE image has a high V content (38 wt%) in the element mapping (Figure 9), accompanied by high proportions of C, W, Cr, and Mo. The vanadium-rich particle is probably a MC vanadium carbide (VC) mixed with MC in various proportions. The MC peak is superimposed on the martensite peak in the XRD spectra (Figure 10), making it difficult to distinguish one from the other [26].

iii. The black phase in the BSE image is an impurity rich in Mn (13.6 wt%) and S (10.3 wt%). Such impurities occur only occasionally.

It has been reported in the literature that the carbides in heat-treated S390 specimens are primarily Fe3W2C and VC, comparable in size and distribution to those observed in this study [3, 4, 22, 23, 39]. Subsequent HT compensates for differences in the as-built specimens and transforms the primary microstructure into a homogenized, fine-grained martensitic matrix with precipitated secondary carbides (Figure 5 and Figure 7).

4.3. Mechanical properties compared to state-of-the-art

The as-built specimens have a bending strength of 2535 MPa, taking into account that the area of the bending specimen with tensile load was in situ heat treated for a longer duration, and the area with compressive load only for a short time. The hardness values of the as-built specimens range from 798 HV1 in the top layer to 499 HV1 in the longer in situ heat-treated layers toward the bottom (Figure 12). Because the carbide precipitates do not change significantly across the specimen height, the hardness gradient across height is primarily due to differences in martensite hardness. Subsequent HT can increase bending strength to about 2800 MPa, with an average hardness of 611 HV1 (Table 4). According to the manufacturer, a hardness of 63 HRC can be achieved with this HT via HIP, but these values could not be achieved with the LPBF specimens treated with a furnace without inert atmosphere [1] HT homogenizes the microstructure and hardness of the specimen and causes the dissolution of alloying elements, the reformation of fine martensite grains, and the precipitation of secondary carbides at small intervals (micrometer to submicrometer in size) at all positions in the specimen.

| Mean Bending Strength (MPa) | Max Bending Strength (MPa) | Min Bending Strength (MPa) | Mean Hardness (HV1) | Max. Hardness (HV1) | Min. Hardness (HV1) |
|-----------------------------|---------------------------|---------------------------|---------------------|---------------------|---------------------|
| As-Built                    | 2,535                     | 2,775                     | 2,330               | 798                 | 499                 |
| Heat-Treated                | 2,799                     | 3,504                     | 2,613               | 611                 | 561                 |
The solid solution strengthening is caused primarily by the alloying elements Co (12 wt%), Cr (5 wt%), and C (1 wt%) dissolved in the martensitic matrix (Figure 9). Temperature-resistant primary carbides, which are not dissolved during HT (e.g., Mo carbides), hinder the grain growth of the austenite grains during solution annealing [39]. Figure 7 shows fine martensite grains contributing to grain boundary strengthening via the Hall–Petch mechanism [27]. In Figure 7, and in the SEM BSE images (Figure 8(a)), uniform fine precipitated carbide grains approximately 1 μm size are visible; these cause precipitation hardening via the Orowan mechanism [27, 28].

The mechanical properties of the specimens can be further improved by HT under HIP conditions, as has already been demonstrated for EB MB specimens in the Jin et al. study [26]. Such a treatment is also standard for PM S390. However, because LPBF specimens have not yet been subjected to HT under HIP conditions, a comparison with conventionally produced PM S390 is not yet possible.

5. Conclusions

This is the first study to investigate the processing of PM HSS S390 via LPBF, and it provides a detailed analysis of dense and crack-free as-built specimens, as well as heat-treated specimens. The conclusions of the study are as follows:

- HSS S390 can be processed via LPBF crack-free, with a relative density of 99.8% achieved through in situ HT up to 800 °C.
- As-built specimens exhibit changing microstructure and phases depending on temperature history and specimen height, with distinguishable columnar dendritic and equiaxed grains. Segregated intergranular carbides (several microns) and interdendritic carbides (submicron) are visible. Interdendritic MC carbides are identifiable and contain V, W, Cr, and C.
- Heat-treated specimens show homogenous microstructure and evenly distributed micron- and submicron-sized secondary carbides. Due to HT, MC (predominantly VC) and M2C (predominantly Fe3W2C) are precipitated.
- The achieved mechanical properties are as follows:
  - As-built: a mean bending strength of 2535 ± 148 MPa, and a hardness gradient along the specimen height of 500 HV1 to 800 HV1.
  - Heat treated: a mean bending strength of 2799 ± 278 MPa, with a mean hardness value of 610 ± 50 HV1.

LPBF in situ HT is sufficient to suppress crack mitigation in HSS S390. Subsequent conventional HT improves mechanical properties and homogenizes the microstructure of as-built specimens, which are similar to PM [1] and EBM [26] specimens. The results indicate an evident influence of thermal history on the microstructure and resulting mechanical properties of the manufactured specimens In-situ-HT in the LPBF build chamber, but post-process might overcome the gradients of hardness and microstructure along the building direction of the as-built specimens. Future research may experiment with alternative heating concepts for comparison against the substrate plate heating approach investigated in this study (e.g., radiation-based heating for the top layer of the specimen using infrared heating modules or large area diodes) and evaluate the effect on the resulting microstructure [49, 50, 51, 52].

Declarations

Author contribution statement

Simon Bergmüller: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Lukas Kasner: Conceived and designed the experiments; Analyzed and interpreted the data.

Lorenz Fuchs: Performed the experiments. Jakob Braun: Analyzed and interpreted the data.

Nikolaus Weinberger: Contributed reagents, materials, analysis tools or data.

Ise Letofsky-Papst: Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data.

Gerhard Leichtfried: Analyzed and interpreted the data; Wrote the paper.

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Data availability statement

Data will be made available on request.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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