

A microstructural modification strategy to improve thermal conductivity of tool steels produced by laser powder bed fusion

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ABSTRACT

Laser additive manufacturing is a promising technology for producing tools with complex geometry. The tools for hot working applications typically require high thermal conductivity to reduce thermal stresses. However, the relationship between thermal conductivity and the unique microstructure created by Laser Powder Bed Fusion (LPBF) is not yet fully understood.

In this study, we investigated the thermal conductivity of AISI M50 in the as-built and different heat-treated conditions. As-built samples exhibited anisotropy and low thermal conductivity. The low thermal conductivity can be improved after heat treatments by modifying the microstructure. Moreover, the influence of each microstructural characteristic was interpreted. Thereby, a correlation between thermal conductivity and microstructure can be addressed. With this knowledge, we proposed a suitable heat treatment concept for improving the thermal conductivity of additively manufactured tool steels.

1. Introduction

Laser Powder Bed Fusion (LPBF), also known as Selective Laser Melting (SLM) or Powder Bed Fusion–Laser Beam (PBF-LB), is one of the most commercially used additive manufacturing methods for the manufacturing of metallic components [1]. The LPBF technology has been applied and developed for creating industrial tools to meet the requirements, such as fast product development, low cost, and high efficiency [2]. Specifically, for the manufacturing of molds and dies, LPBF enables the fabrication of complex conformal cooling channels, which improve the lifetime of tools and the process productivity by efficient heat removal [3–6].

Limited tool steel grades are suitable for the LPBF process. Only high speed steels M2, M3:2, and M50 and hot work steels H11 and H13 are popularly studied [7–17]. The as-built microstructure of these tool steels consists of a martensitic matrix with retained austenite, and a cellular/dendritic solidification structure formed in the matrix due to the rapid cooling [9,10,15–17]. The standard conventional heat treatment, comprising quenching and tempering, is normally performed to achieve a fully martensitic matrix with finely distributed carbides [8,10,14,16]. A novel heat treatment strategy, namely direct tempering, can also

achieve a fully martensitic matrix without cellular solidification structures [14,18–20]. Corresponding mechanical properties, such as hardness, tensile strength, toughness, and fatigue strength, which are critical for the tool-making industry, were also studied [16].

Tools for hot working applications should have not only good mechanical properties but also high thermal conductivity [21]. For instance, the lifetime of a press tool can be extended by sufficient thermal conductivity, since thermal shock or thermal fatigue are the most influential failure modes for hot working dies [22,23]. High thermal conductivity of the die material can reduce thermal gradients efficiently, thereby, thermal stresses generated from temperature differences are decreased [24]. Moreover, high thermal conductivity benefits the productivity of press hardening, injection molding, and die casting processes, since a shorter cooling stage is necessary [4,21,25].

The physical theory of thermal conductivity of metals has been well established in crystal lattice scale. In metallic materials, mainly electrons and phonons contribute to thermal conductivity, and their contributions to thermal conductivity are assumed to be independent [26,27]. Electrons can absorb thermal energy and move through the material, thus transferring heat. Phonons basically contribute through lattice vibration waves. Therefore, the thermal conductivity of steels λ is

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the sum of phonon thermal conductivity λ_{ph} and electron thermal conductivity λ_e : $\lambda = \lambda_{ph} + \lambda_e$. For pure iron, the heat conducts predominantly via electrons. For steels, phonons contribute up to around 40% to the thermal conductivity [28]. In both cases, the heat is transported by mobile carriers, and the thermal conductivity from each carrier can be expressed by the form: $\lambda_i = \frac{1}{3}C_i v_i l_i$, where C_i is the contribution of carriers of type i to the specific heat per unit volume, v_i is the average speed of carrier, and l_i is the mean free path. This equation shares the same expression for the thermal conductivity in the kinetic theory of gases and can be derived from the Boltzmann Eq. [26].

Deviations from the ideal periodic structure of the lattice cause scattering of electrons and phonons, thereby reducing the free path of heat carriers and increasing the resistance to heat transport. Therefore, lattice defects, such as dislocations, vacancies, and solid solution atoms, can significantly decrease the thermal conductivity [26,27].

At the microstructure level, the thermal conductivity of steels is affected by microstructural characteristics. Austenite exhibits the lowest thermal conductivity, compared to ferrite or martensite, because the ferritic or martensitic lattice provides a higher concentration of free conduction electrons and also causes less scattering to heat carriers [29–31]. A high carbon content in the matrix significantly reduces the thermal conductivity [29]. Hence, the precipitation of carbides in the matrix improves thermal conductivity due to less scattering from the distorted lattice and the high thermal conductivity of carbides can also contribute to the overall thermal conductivity [31–33].

The effect of heat treatment on conventionally produced martensitic steels has been studied. Increasing the austenitization temperature leads to a decrease in thermal diffusivity, caused by the higher content of dissolved alloying elements in the iron matrix [34]. After hardening, the steels present low thermal conductivity. During tempering, the various microstructural evolution leads to the increase in thermal conductivity. The alloying elements precipitate in the form of carbides from the solid solution, reducing scattering for heat carriers. Additionally, recovery and recrystallization during tempering lower the dislocation density [31,32,35].

The thermal conductivity of metals produced by LPBF has rarely been studied so far. Simmons et al. [36] firstly investigated the thermal conductivity of 316 L produced by LPBF, illustrating that porosity reduces thermal conductivity. Džugan et al. [37] evidenced the anisotropic thermal conductivity of H13 by LPBF. However, the reason for this anisotropy was not explained in detail. Nevertheless, both results showed a lower thermal conductivity than conventionally produced material, which has not been yet, but should be illustrated with the different microstructure generated in the LPBF process.

In the present study, the authors systematically investigated the influence of each microstructural characteristic from LPBF on thermal conductivity of M50. The concept for improving thermal conductivity was revealed. With this knowledge, the authors also proposed an economical and practical heat treatment concept for additive manufactured tool steels.

2. Material and methods

The material in this investigation is the tool steel AISI M50 (80MoCrV42–16, Mat. Nr. 1.3551). The corresponding chemical composition is shown in Table 1, together with the ASTM A600-92a standard [38]. Argon-atomized powder was supplied by Thyssenkrupp (Essen, Germany) with a size distribution from 10 μm to 45 μm .

All samples were produced under an argon atmosphere by a Realizer SLM 100 machine (DMG MORI GmbH, Bielefeld, Germany). Optimal process parameters with minimized porosity were found with a laser power of 160 W, a laser speed of 1 m/s, and a hatch distance of 100 μm . The layer thickness was 30 μm , and the laser scan direction rotated 67° for each layer. The substrate was preheated to 250 °C or 500 °C respectively, to reduce the thermal stress. Only the samples with 500 °C preheating were used for heat treatment. 1 cm³ cubic samples were fabricated for microstructure characterization, hardness test, and density measurement. To determine the thermal expansion coefficient, rectangular dilatometric samples were produced with a dimension of 20 × 5 × 5 mm³. Cylindrical disc samples for specific heat measurement were produced with a diameter of 4 mm and a thickness of 2 mm. The samples for thermal diffusivity measurement were manufactured with the dimension of 10 × 10 × 2 mm³. All samples for the thermal physical experiments were built-up in two directions: (1) parallel to the building direction (vertical) and (2) perpendicular to the building direction (horizontal).

In this study, two types of heat treatment were applied. One is the conventional heat treatment for tool steels, namely quenching and tempering. The samples were austenitized at either 1050 °C or 1180 °C for 20 min in a vacuum furnace, followed by nitrogen gas quenching under 5 bar pressure. Subsequent triple tempering was performed at 580 °C for 3 h each in a vacuum furnace, as shown in Fig. 1a. The second type of heat treatment is named as direct tempering. As presented in Fig. 1b, the as-built samples were directly tempered at 600 °C, 650 °C or 700 °C for 4 h, with temperatures set slightly higher than the secondary hardening peak to promote the precipitation of carbides and dissolve cellular solidification structure from LPBF. Based on previous study, the relative high direct tempering temperature is also necessary for achieving a fully martensitic matrix [14].

The microstructure of samples in as-built and heat-treated conditions was characterized. The cross sections along the building direction were prepared by standard metallographic methods with the final step of polishing using colloidal silica suspension. To reveal the microstructure, the samples were etched by V2A etchant for 10 s. The microstructure was observed by Optical Microscopy (OM) and Scanning Electron Microscopy (SEM) (Helios Nanolab G3 CX, FEI Company) assembled with Secondary Electron (SE), Energy-Dispersive X-ray Spectroscopy (EDS), and Electron Backscatter Diffraction (EBSD) detectors. For high-resolution microstructure characterization, the samples were prepared by Focused Ion Beam (FIB) for Scanning Transmission Electron Microscopy (STEM). EBSD was performed with a step size of 200 nm, and Transmission Kikuchi Diffraction (TKD) was with a step size of 7 nm. These data were analyzed by the TSL OIM Analysis™ software (7.3.1, EDAX Inc., Mahwah, NJ, USA). The parent austenite grains were reconstructed from the EBSD data by the ARPG software [39]. The X-ray Diffraction System 3003 PTS (GE Inspection Technologies, New York, NY, USA) with a Cr X-ray tube was applied for the XRD phase analyses. The phase fraction was analyzed by Materials Analysis Using Diffraction (MAUD) software (2.91 version) for phase analysis (ICDD/PDF 00–006-0696 for ferrite and ICDD/PDF 00–023-0298 for austenite).

Rockwell hardness tests were performed according to the ASTM E18–22 standard [40], and the Rockwell C hardness was averaged from 5 measurements.

The thermal conductivity was measured using an indirect method by the determination of the thermal diffusivity α , the specific heat C_p , and the density ρ at different temperatures. The thermal conductivity λ can be calculated from the equation:

Table 1
Chemical composition of M50 powder (wt%) according to the supplier's specification and ASTM A600-92a [38].

Elements [wt%]	Fe	C	Cr	Mo	V	Mn	Si
Powder	Bal.	0.78	4.21	4.49	0.98	0.18	0.44
ASTM A600-92a	Bal.	0.78–0.88	3.75–4.50	3.90–4.75	0.8–1.25	0.15–0.45	0.2–0.6

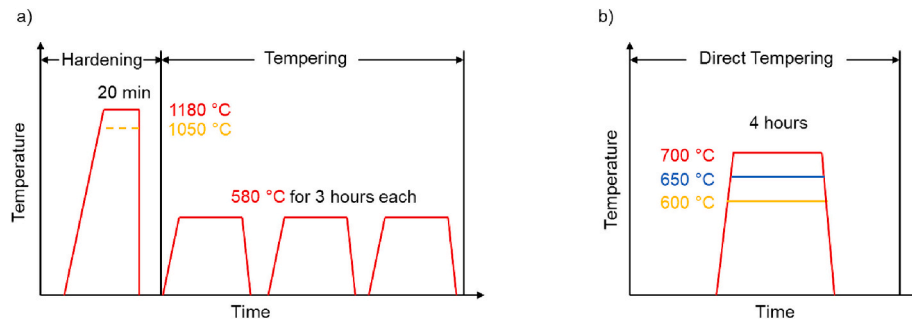


Fig. 1. Schematic thermal profiles of (a) conventional heat treatment and (b) direct tempering.

$$\lambda = \alpha \times C_p \times \rho$$

The density at room temperature ρ_0 was determined by a helium pycnometer and a high-precision laboratory balance. A dilatometer (Netzsch DIL 402C, Germany) was used to measure the Coefficient of Thermal Expansion (CTE) with a heating rate of 10 K/min. Then the density at different temperatures can be derived from the equation:

$$\rho(T) = \frac{m}{L^3} = \frac{m}{((CTE \times (T - T_0) + 1) \times L_0)^3} = \frac{\rho_0}{1 + 3 \times CTE \times (T - T_0)}$$

where m is mass, T is temperature and L is sample length. Thermal diffusivity was measured by a laser flash analyzer (Netzsch LFA 427, Germany) using samples coated with a thin layer of graphite for better laser absorption. At each temperature, the thermal diffusivity was measured 8 times to have an average value. The specific heat measurement was carried out in a Differential Scanning Calorimeter (DSC) machine (Netzsch DSC 404C, Germany) with a heating rate of 10 K/min. Both measurements were conducted under argon atmosphere. The thermal diffusivity and specific heat were directly calculated by the software Netzsch Proteus Analysis.

3. Results

3.1. Microstructure

The typical as-built microstructure of M50 tool steel produced by LPBF with preheating temperatures of 250 °C and 500 °C is presented in Fig. 2a and b, respectively. A cellular/dendritic solidification structure is visible, formed by rapid solidification and strong constitutional supercooling. The direction of cellular grains and dendritic strains is mainly along the building direction, which is determined by the thermal gradient. Fig. 2c reveals the local chemical composition of these solidification structures, where cellular walls are depleted of iron and enriched with carbon and other alloying elements, such as Mo, V and Cr.

The dark-field STEM image depicts more microstructural characteristics of as-built M50 sample with 500 °C preheating (Fig. 3a). Martensite is within the cells, where the martensitic laths can be observed. Nano-scale carbides, as white spots, precipitated in the matrix. Fig. 3b presents the corresponding phase map. The unindexed black area during the EBSD measurement is caused by the high lattice distortion due to the strong alloy segregation at cell walls. The retained austenite, which is stabilized by carbon, is located near the segregation in the interdendritic regions.

Fig. 4 exhibits the microstructure after conventional heat treatment.

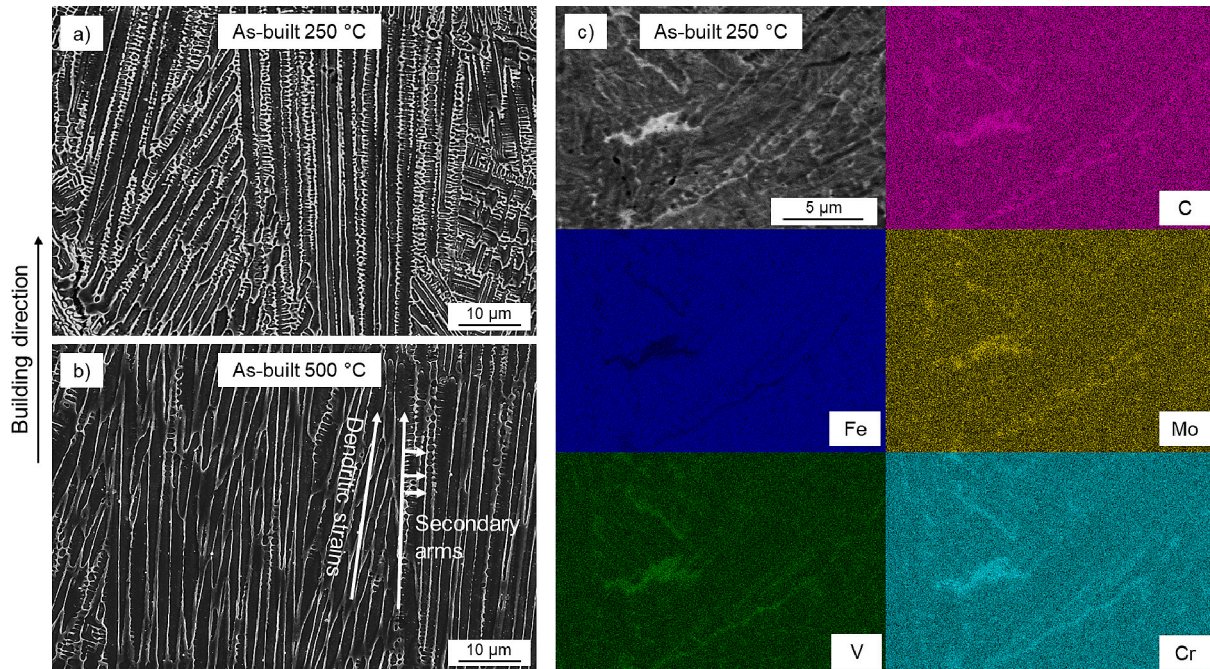


Fig. 2. Microstructure in the as-built condition. SE micrographs of solidification microstructure from the samples produced with 250 °C (a) and 500 °C (b) preheating. EDS-mapping (c) presenting the enrichment of alloy elements at solidification walls.

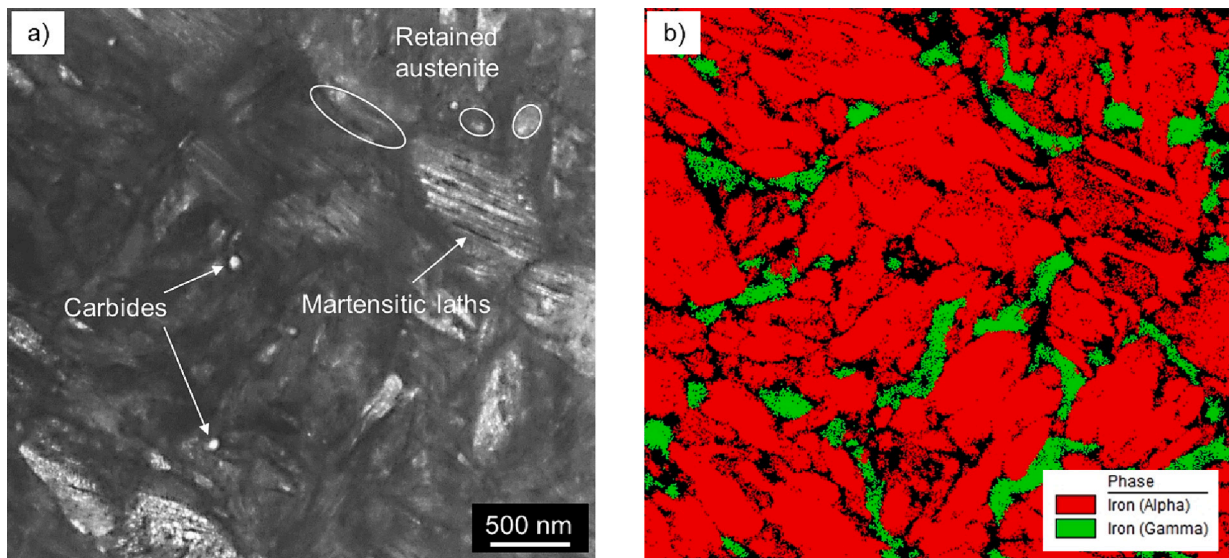


Fig. 3. (a) Dark-field STEM image of as-built M50 sample produced with 500 °C preheating and (b) corresponding TKD phase map.

Fig. 4a and b present the martensitic matrix, where the martensite laths can be observed from the contrast. According to the parent austenite grain boundaries in both samples, austenite grains grew more significantly during the austenitization at 1180 °C compared to 1050 °C. The corresponding tempered carbides of these samples are shown in Fig. 4c and d. Both conventional heat treatments lead to carbides with similar morphology and size of about 100 nm, regardless of the selected austenitization temperature. Therefore, through both conventional heat treatments, the microstructural features from the LPBF process were completely removed by the strong recrystallization, and a martensitic matrix with tempered carbides was achieved. However, the contents of alloying elements in the matrix vary with the austenitization temperature. As present in Fig. 4e, the martensitic matrix quenched from 1180 °C exhibits higher higher contents of carbide forming alloying elements. Although the carbon content cannot be measured accurately by EDS, a qualitative comparison is still valid. Higher austenitization temperatures dissolve more alloy elements into the matrix, leading to a higher concentration of alloy solid solution atoms, or in other words, more lattice defects.

Fig. 5 demonstrates the microstructure at different direct tempering temperatures. During direct tempering, the cellular structure from LPBF process is dissolved, and carbides start to precipitate. After direct tempering at 600 °C, only the martensitic matrix and no solidification structure can be observed, as shown in Fig. 5a. Moreover, the carbides are at the nanoscale and can only be characterized by STEM. The homogenous carbon distribution in EDX map Fig. 5d confirms the dissolution of the cellular/dendritic solidification structure. Fig. 5b and e show the microstructure and carbon distribution of the sample directly tempered at 650 °C. At this direct tempering temperature, carbides mainly precipitated at the grain boundaries along the building direction. The higher direct tempering temperature of 700 °C leads to strong carbide precipitation (Fig. 5c and f), where carbides are connected with each other forming carbide networks. These carbide networks present the same morphology as the solidification structure from the LPBF process.

Fig. 6 illustrates Kernel Average Misorientation (KAM) maps of samples tempered at varying temperatures. Theoretically, KAM maps represent local grain misorientation, where lattice defects, such as local lattice distortion and dislocation density, contribute to high KAM values. Elevated direct tempering temperatures prompt carbide precipitation and matrix recovery, resulting in a matrix with fewer dislocations and solid solution alloy atoms, that is, a matrix exhibiting fewer lattice defects.

The crystal grain orientation and texture are revealed by the EBSD measurements. When comparing the Inverse Pole Figures (IPF) and Pole Figs. (PF) of the martensitic matrix in the directly tempered sample (Fig. 7a) and the conventionally heat-treated sample (Fig. 7b), a stronger texture exists in the conventional heat treatment condition. This result is contrary to the theory: After direct tempering, the texture from the LPBF process should be retained since no strong recrystallization occurred, while after conventional heat treatment the matrix should be homogenized due to the strong recrystallization and grain growth. This contradiction can be explained with the help of reconstructed parent austenite. Fig. 7c presents the parent austenite of the directly tempered sample, where the elongated grains orientate along the building direction, and the typical $\langle 100 \rangle \parallel \text{BD}$ (Building Direction) texture can be specified from the pole figures. The coarse parent austenite grains after conventional heat treatment were reconstructed in Fig. 7d. Since the martensite laths within a single austenite grain share certain same orientations according to the Kurdjumov—Sachs relationship, these laths show preferred orientations in the pole figures, whereas the fine martensite after direct tempering seems to contain no strong texture. However, for the directly tempered samples, the reconstructed parent austenite allows us to specify the typical $\langle 100 \rangle \parallel \text{BD}$ texture from epitaxial grain growth during the LPBF process.

The microstructural characteristics in different conditions can be summarized with the XRD results, as presented in Fig. 8. As-built samples consist of the martensitic matrix with cells/dendrites and retained austenite, located in the intercellular/interdendritic area. These solidification cells/dendrites orientate mainly along the building direction. Based on the XRD result, higher preheating temperature leads to lower fraction of retained austenite (42.3% at 250 °C preheating and 38.1% at 500 °C preheating). During direct tempering, the retained austenite transforms to martensite, and the $\langle 100 \rangle \parallel \text{BD}$ texture from the as-built condition is retained. The direct tempering temperature determines the size and morphology of carbides. After conventional heat treatment, a homogenous microstructure was achieved with a martensitic matrix and tempered carbides. With higher austenitization temperature, the martensitic matrix contains more solid solution alloy atoms. After all post treatments, no retained austenite can be detected by the XRD, indicating a fully martensitic matrix.

3.2. Thermal conductivity

The thermal conductivity was measured indirectly from the thermal diffusivity α , the specific heat C_p , and the density ρ . Fig. 9 presents these

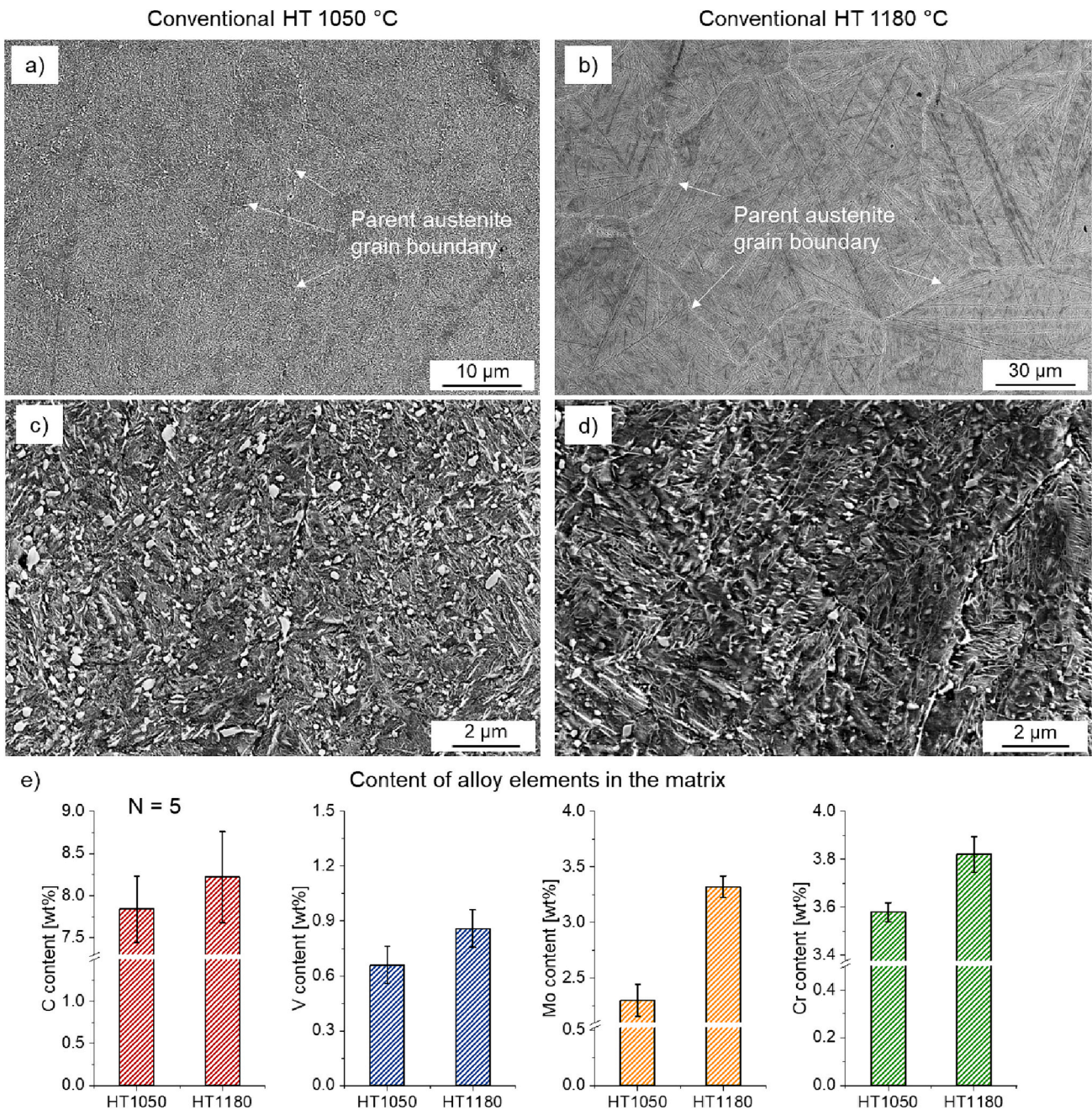


Fig. 4. Microstructure of samples after conventional heat treatment at the austenitization temperature 1050 °C and 1180 °C. (a) and (b) presenting micrographs of martensitic matrix with parent austenite grain boundaries; (c) and (d) showing tempered carbides; (e) alloy contents of martensitic matrix averaged from 5 EDS point measurements.

results and calculated thermal conductivity in different conditions.

The dependency of density on temperature was calculated from the density at room temperature and the Coefficient of Thermal Expansion (CTE), which was determined by the dilatometer measurement. As demonstrated in Fig. 9 (a-c), the relative elongations among various conditions are almost identical, and the CTEs between various conditions deviate slightly in the range of measurement uncertainty, which illustrates that the microstructure does not influence much the temperature dependency of density.

The specific heat was measured by the DSC method, as shown in Fig. 9 (d-f). For all conditions, the specific heat increases with the temperature. However, the as-built samples present lower specific heat than the heat-treated samples, which may result from the retained austenite. The fluctuation of as-built curves can be caused by

ascribed to the phase transformations during tempering, such as carbides precipitation and decomposition of retained austenite. Regarding the directional dependency, the specific heat of vertical samples (along the building direction) overlaps with that of the horizontal samples (perpendicular to the building direction), indicating that the specific heat is isotropic.

Thermal diffusivity describes how quickly a material reacts to a change in temperature. For the as-built samples, the thermal diffusivities along the building direction are always higher than perpendicular to the building direction (Fig. 9g). This anisotropy was removed after direct tempering at 600 °C (Fig. 9h). However, with the higher direct tempering temperature, the anisotropy appears again. After conventional heat treatment, the thermal diffusivities along the two directions are identical (Fig. 9i). The thermal diffusivities of as-built samples are

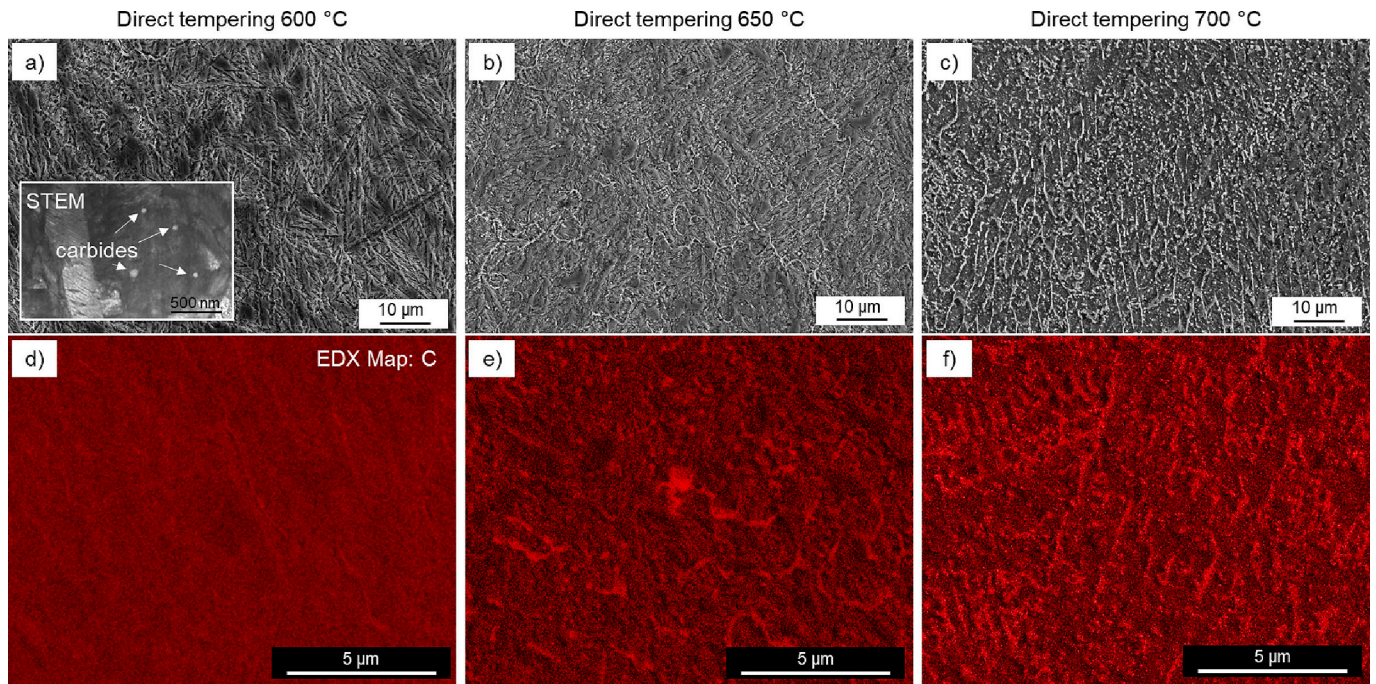


Fig. 5. Microstructure of directly tempered samples. (a-c) SEM micrographs showing the characteristics of matrix and carbides. (d-f) EDS map revealing the distribution of carbon.

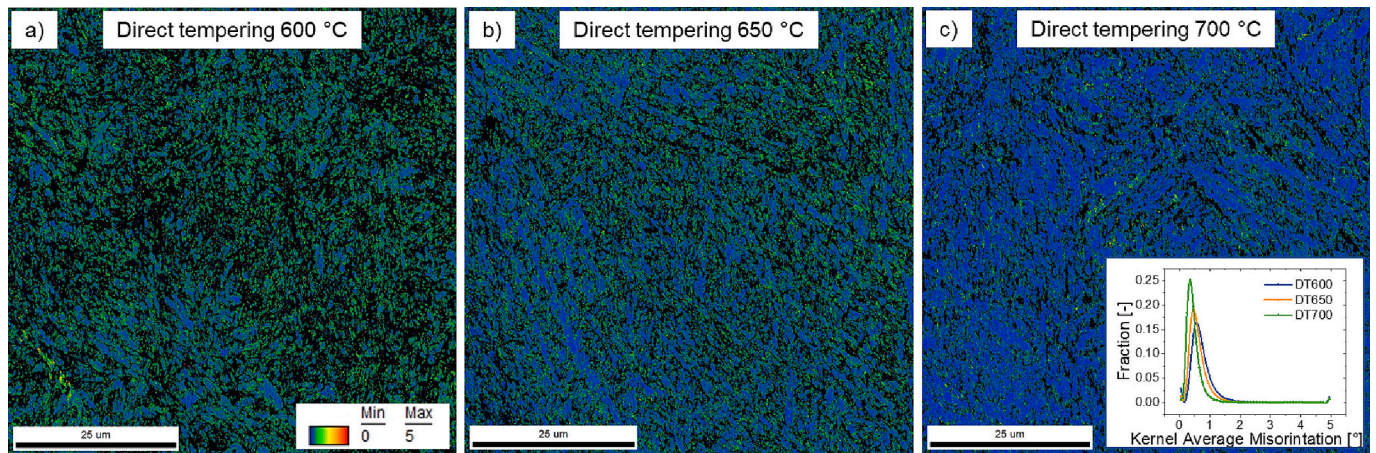


Fig. 6. KAM maps derived from EBSD results in different direct tempering conditions.

generally lower than heat-treated samples.

Fig. 9 (j-l) presents the thermal conductivities in different conditions. All thermal conductivities of as-built samples show a similar trend with an increase up to 400 °C, followed by a decrease. The thermal conductivities in the as-built condition are anisotropic and higher along the building direction (Fig. 9j). The preheating temperature also influences the thermal conductivity, and the 250 °C preheating shows better thermal conductivity. After direct tempering, the thermal conductivity is significantly improved (Fig. 9k). The highest thermal conductivity is achieved in the condition of 650 °C direct tempering along the building direction. The direction dependency does not exist when directly tempered at 600 °C, but this directional difference becomes stronger with a higher temperature of direct tempering. In the conventional heat treatment group, vertical and horizontal thermal conductivities overlap, indicating no anisotropy (Fig. 9l). The samples quenched from 1050 °C show a better thermal conductivity than those from 1180 °C.

The relationship between thermal conductivity and hardness is presented in Fig. 10. Here the vertical thermal conductivities at room

temperature in each condition are presented. In as-built conditions, even though high hardness (over 50 HRC) can be achieved, the thermal conductivities are lower than 15 W/mK. A higher preheating temperature leads to harder samples with lower thermal conductivity. The direct tempering can significantly improve the thermal conductivity. The best thermal conductivity was achieved by direct tempering at 650 °C with a value of 32 W/mK. Due to the carbide coarsening at a higher temperature of direct tempering, the hardness drops from 57.7 HRC to 38.1 HRC. The conventional heat treatment can also improve thermal conductivity respect to as-built condition, though not as effectively as the direct tempering. A higher austenitization temperature leads to higher hardness and lower thermal conductivity. Following a quench from 1180 °C coupled with tempering, the reference group, conventionally produced M50, exhibits similar microstructure and material properties to those of additively manufactured M50. These attributes are outlined in the Appendix. In all conditions, there is a general trend that harder material has a lower thermal conductivity, except direct tempering at 700 °C, which will be discussed in text next section.

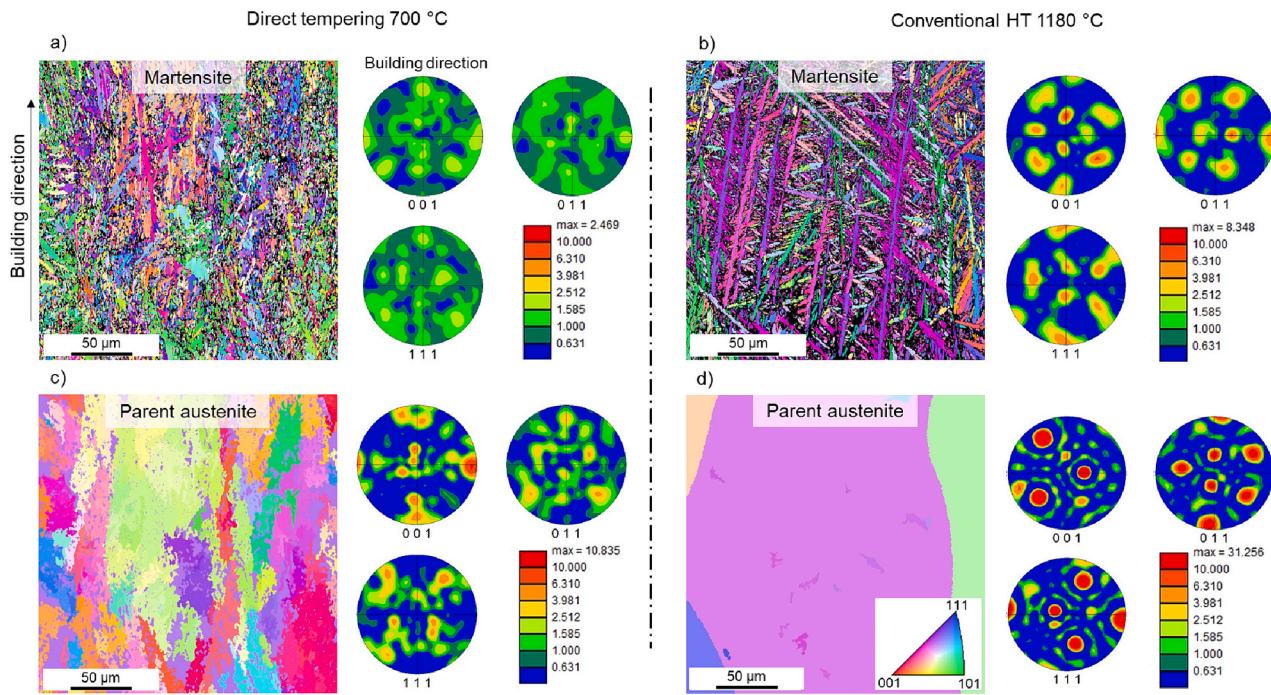


Fig. 7. IPF map and PF of (a) the sample directly tempered at 700 °C, and (b) the sample conventionally heat treated at 1180 °C; (c) and (d) are IPF map and PF of corresponding reconstructed parent austenite (all measured cross sections are parallel to the building direction).

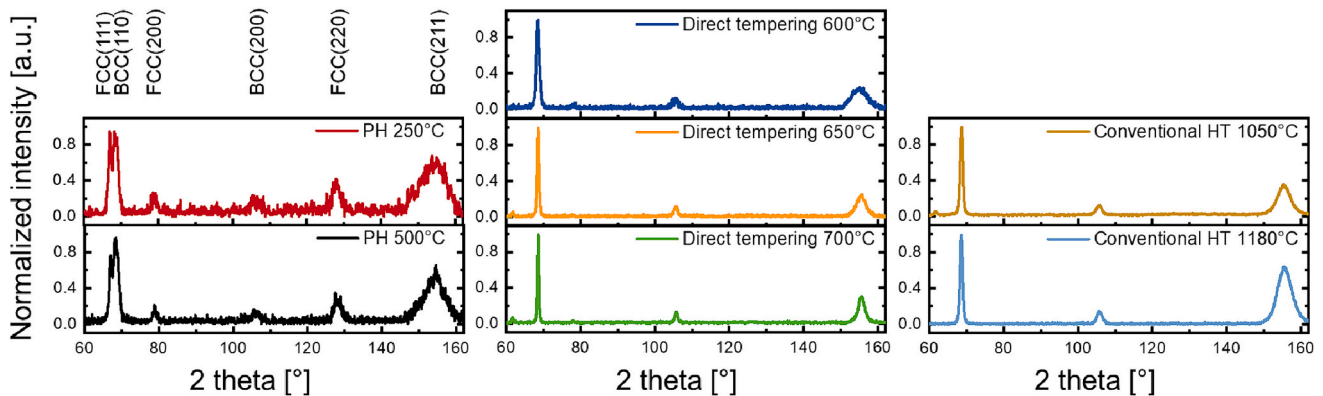


Fig. 8. XRD patterns of samples in different conditions: preheating (PH) at 250 °C and 500 °C; direct tempering at 600 °C, 650 °C and 700 °C; and conventional heat treatment (HT) at 1050 °C and 1180 °C.

4. Discussion

This section discusses first the anisotropy of the thermal conductivity and then the influence factors on thermal conductivity. Summarily, a mechanism for improving thermal conductivity is illustrated, and a post treatment concept is proposed for achieving better thermal conductivity of tool steels or even for more alloys produced by LPBF.

4.1. Anisotropy of thermal conductivity

The thermal conductivity is anisotropic in the as-built conditions and in two directly tempered groups. This anisotropy can be caused by the texture of the matrix or/and the anisotropic distribution of secondary phases.

The influence of texture can be perfectly discussed with the sample directly tempered at 600 °C. As illustrated in Fig. 7a and c, after direct tempering, the matrix did not recrystallize, and only the retained austenite transformed into martensite. Therefore, the $\langle 100 \rangle \parallel \text{BD}$

texture from epitaxial grain growth remained in the fully martensitic matrix. The secondary phase is shown in Fig. 5a and d, where the cellular/dendritic solidification structure was dissolved, proved by the homogeneous distribution of carbon, and nano-scale carbides precipitated in the matrix. In this condition (direct tempering at 600 °C), the thermal conductivity is isotropic, indicating that the texture in the fine martensitic matrix has no pronounced influence on thermal conductivity. Even though the orientation of a single-crystalline ceramic show an anisotropic thermal conductivity [41], the influence of the texture of the fine martensite is negligible.

The anisotropy of thermal conductivity appears again with higher direct tempering temperatures at 650 °C and 700 °C. In these conditions, the matrix mirrors the one formed after the 600 °C direct tempering. However, the distribution of the secondary phase (carbides) shows variation. After direct tempering at 650 °C, carbides precipitated at parent austenite grain boundaries along the building direction (Fig. 5b and e). With this microstructure, the thermal conductivity along the building direction is higher than that perpendicular to the building

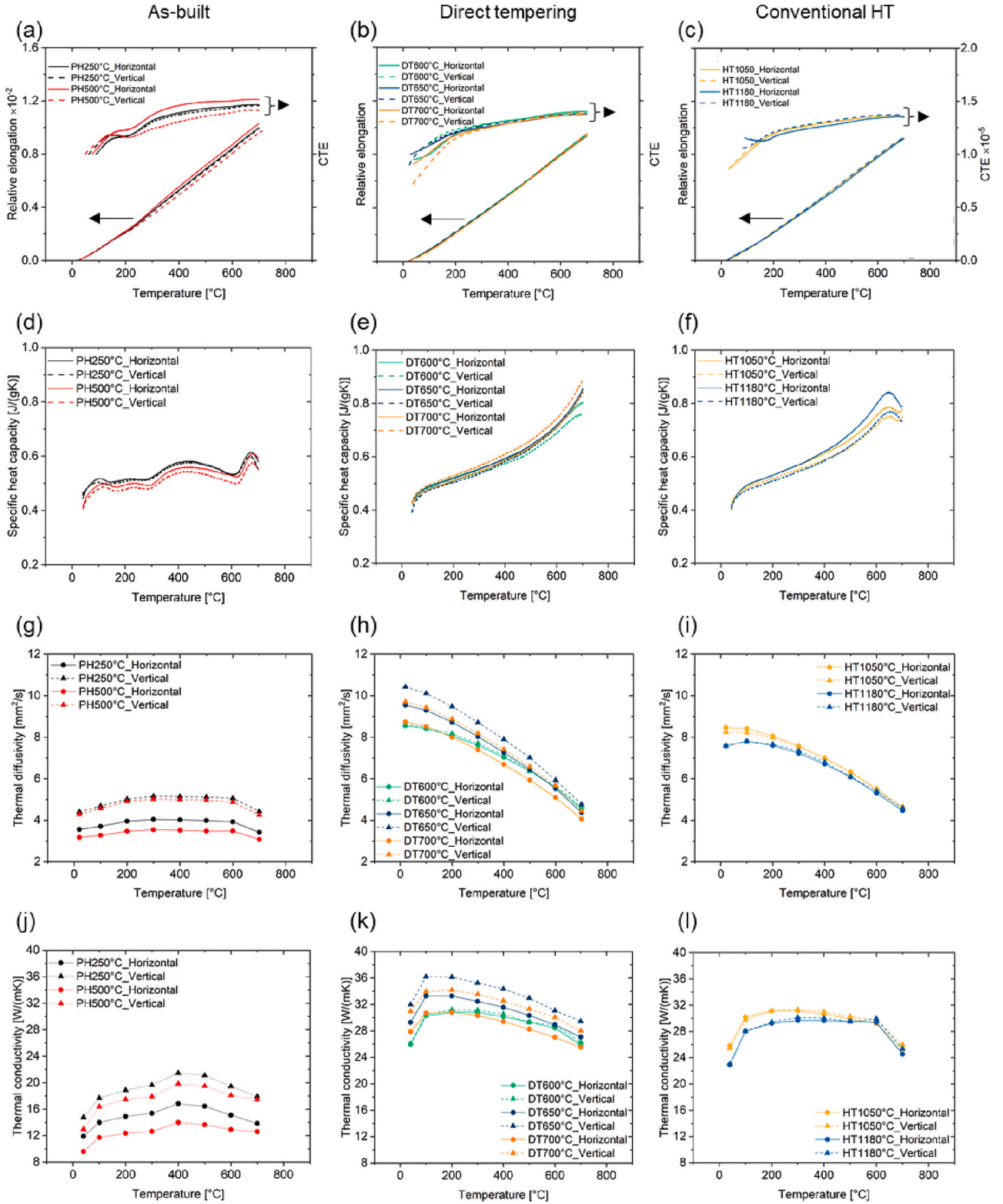


Fig. 9. Thermal material properties in as-built, direct tempering, and conventional heat treatment conditions: (a-c) relative elongation and Coefficient of Thermal Expansion (CTE), (d-f) specific heat capacity, (g-i) thermal diffusivity, (j-l) thermal conductivity. Vertical shows material properties along the building direction, while horizontal is perpendicular to the building direction.

direction, indicating that carbides are the barriers to heat transfer and the inhomogeneous distribution of carbides leads to the anisotropy of thermal conductivity. After direct tempering at 700 °C, numerous carbides precipitated and connected with each other as carbide networks in the morphology of the solidification structure (Fig. 5c and f). This

pronounced carbide precipitation leads to a stronger anisotropy of thermal conductivity.

On the basis of the discussion above, it can be concluded that the inhomogeneous secondary phases (solidification cellular structure or carbides) are responsible for the anisotropy of the thermal conductivity.

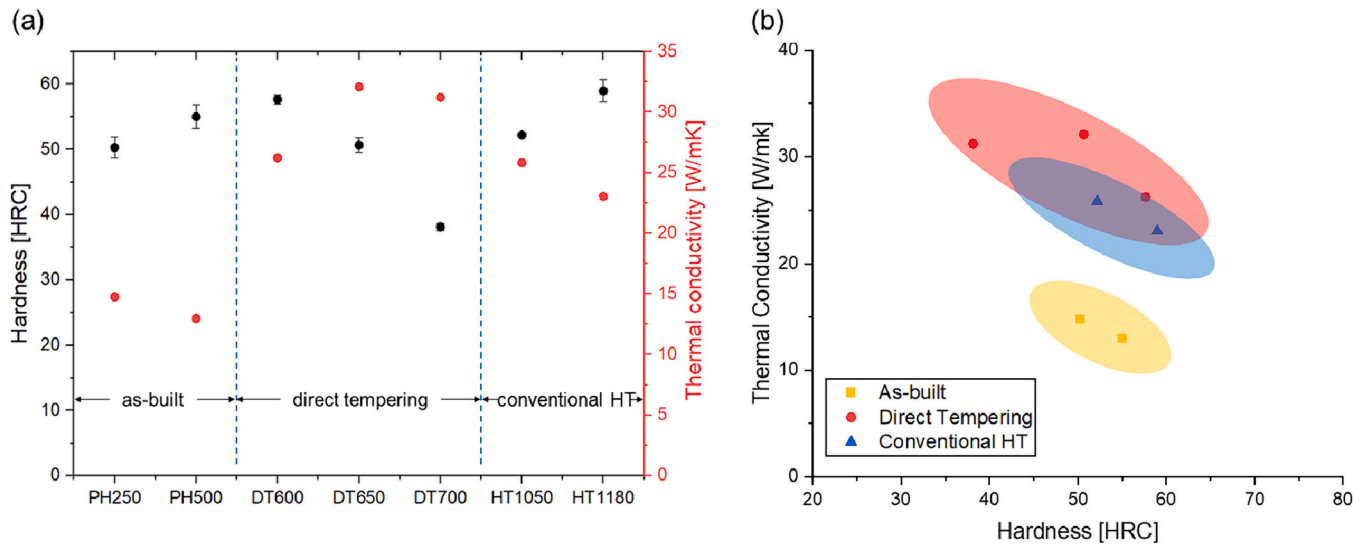


Fig. 10. (a) Thermal conductivity along building direction at room temperature and hardness in different conditions: Preheating (PH) at 250 °C, and 500 °C; Direct Tempering (DT) at 600 °C, 650 °C, and 700 °C; Conventional Heat Treatment (HT) at the austenitization temperature of 1050 °C and 1180 °C. (b) The relationship between thermal conductivity and hardness in different heat treatment conditions.

The thermal conductivity is higher along the building direction since the solidification structure or coarsen carbides, orientating mainly in this direction, can hinder heat transfer.

4.2. Factors influencing thermal conductivity

For focusing on the influence of each microstructural characteristic, the thermal conductivity of the conventionally heat-treated group is first discussed, regarding of the lattice defects (solid solution atoms) in the matrix. After quenching and tempering, all samples contained a homogeneous martensitic matrix with tempered carbides. However, quenching from 1180 °C leads to a higher hardness than quenching from 1050 °C, indicating that more carbides from the as-built condition were solved into the matrix during austenitization at 1180 °C. These alloy atoms in the matrix contribute to the solid solution hardening, convinced by EDX results (Fig. 4e) and high hardness of sample quenched from 1180 °C conditions (Fig. 10). Meanwhile, these solid solution atoms act as lattice defects, leading to lattice distortion, which shortens the mean free length of the path of heat carriers. Therefore, the sample austenitized at 1180 °C presents lower thermal conductivity. And lattice defects in the matrix can significantly reduce the thermal conductivity.

Besides lattice defects in the matrix, the amount and morphology of secondary phase (carbides) also influence the thermal conductivity in the direct tempering group. As convinced by KAM results in Fig. 6, the higher direct tempering temperature leads to a lower content of lattice defects in the matrix, by consuming the alloy atoms and reducing dislocation density. Therefore, the higher direct tempering temperature leads to a matrix with higher thermal conductivity. But, the higher thermal conductivity of matrix does not necessarily mean a higher overall thermal conductivity of the material. More specifically, after direct tempering at 650 °C, carbides precipitated and coarsened at parent austenite grain boundaries (along the building direction) by consuming the solid solution alloy atoms (Fig. 5b), which leads to a lower hardness and a higher thermal conductivity compared to the sample directly tempered at 600 °C. These carbides are the barriers to heat transfer, since thermal conductivity is higher along the building direction. After direct tempering at 700 °C, due to the severe carbon precipitation and recovery, the sample with fewer lattice defect shows the lowest hardness and should have the best thermal conductivity. However, the overall thermal conductivity of the sample directly

tempered at 650 °C decreased compared to the sample directly tempered at 600 °C. This reduction of overall thermal conductivity is caused by the carbide network (Fig. 5c), which becomes more effective barriers to heat transfer. In addition, despite the cellular carbide structure, the thermal conductivity after the 700 °C direct tempering is still higher than after the 600 °C direct tempering, which illustrates that crystal lattice defect in a matrix is a more dominant factors than secondary phases. Summarily, a matrix and its lattice defects determine the thermal conductivity, but the morphology of secondary phases also significantly influences the thermal conductivity: isolated elongated carbides slightly influence the overall thermal conductivity, and the overall thermal conductivity along the elongated carbides is higher, while connected carbides become more critical and can effectively decrease the overall thermal conductivity.

For the as-built group, the influence factors are more complicated. There are lattice defects, retained austenite, texture, and dendritic/cellular solidification structure. The high carbon content of the matrix contributes to the solid solution hardening, leading to a higher hardness of the as-built sample with 500 °C preheating compared to the sample produced at 250 °C [17]. This high amount of solid solution atoms also leads to a lower thermal conductivity in the martensitic matrix of the as-built sample with 500 °C preheating. Austenite presents lower thermal conductivity than martensite due to the stronger scattering on heat carriers by austenitic lattice. The as-built sample produced at 500 °C has a lower fraction of retained austenite but simultaneously a lower thermal conductivity, indicating that the matrix plays a critical role in the thermal conductivity. As discussed in section 4.1, the texture of fine martensite does not have significant influence on thermal conductivity. The influence of the solidification structure is comparable to that of the carbide network: the solidification structure acts as barrier to heat transfer and leads to an anisotropic thermal conductivity due to the directional distribution of the solidification structure.

Fig. 11 summarizes the importance of microstructure characteristics to the thermal conductivity. The crystal structure of the steel matrix determines the thermal conductivity. The martensite and ferrite present higher thermal conductivity than austenite, because the martensitic/ferritic lattice provides a higher concentration of free conduction electrons and also causes less scattering of conduction electrons [31]. A fully martensitic matrix is critical to achieve a high thermal conductivity for tool steels. Lattice defects, such as solid solution atoms and dislocations, can significantly decrease the thermal conductivity due to the strong

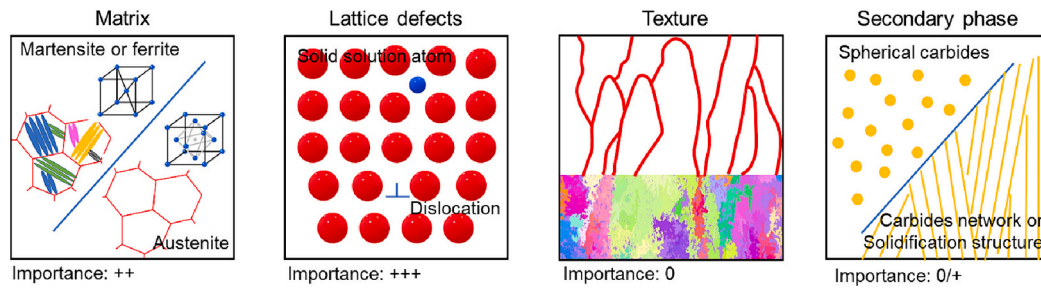


Fig. 11. The influence of microstructure characteristics on thermal conductivity.

scattering to heat carriers, which are electrons and phonons. Thus, these lattice defects are the determinantal influence factor to the thermal conductivity. The influence of the texture, especially the texture in a martensitic matrix produced by LPBF, can be negligible, compared to other influences factors, even though the thermal conductivity differs along different crystal orientations [42]. Secondary phases include carbides, retained austenite, solidification structure or even pores and cracks. These secondary phases can be generally considered as barriers to heat transfer. Isolated spherical secondary phases, such as tempered carbides, have no significant influence on overall thermal conductivity. In addition, isolated elongated secondary phases, such as coarsened carbides along grain boundaries, lead to an anisotropic thermal conductivity. However, when the secondary phase is connected as a network, this network can reduce the overall thermal conductivity. Thus, the carbides network and solidification structure shall be avoided.

As discussed above, the desired microstructure is clear for achieving a good thermal conductivity of tool steels: a fully martensitic matrix is necessary, and this matrix is mainly strengthened by precipitation hardening instead of solid solution strengthening. Therefore, the concept for the heat treatment of as-built tool steels is to transform retained austenite to martensite, dissolve the solidification structure, and form solid solution atoms into carbides, which can be simply and economically realized by the direct tempering, as illustrated in Fig. 12. The size, morphology, and distribution of carbides can be tuned by the temperature level of direct tempering (Fig. 5). In addition, the direct tempering retains the fine microstructure from LPBF process, compared to the conventional heat treatment (Fig. 7a and b). This fine microstructure potentially contributes to high strength and good toughness. For instance, as in Fig. 10 presented, even though direct tempering at 650 °C and conventional heat treatment at 1050 °C lead to similar hardness ca. 50 HRC, direct tempering can significantly improve the corresponding thermal conductivity from 26 W/mK to 32 W/mK. Therefore, direct tempering emerges as an efficient and practical approach to heat treat tool steels produced by LPBF, showing promise in applications where high thermal conductivity is crucial.

5. Conclusions

In the present work, we have illustrated the mechanism of how microstructure characteristics influence the thermal conductivity of tool steels produced by LPBF. More specifically, AISI M50 was printed with two different preheating temperatures and was either directly tempered or conventionally heat treated. Due to different microstructure characteristics from the above conditions, the influence of each microstructure characteristics can be specified: Martensite exhibits higher thermal conductivity than austenite; lattice defects, like solid solution atoms, can significantly decrease the thermal conductivity; the influence of crystallographic texture in a martensitic matrix is negligible; secondary phases, such as carbides and solidification structures, are barriers to heat transfer, but only when connected as networks, the heat transfer will be impeded.

Furthermore, an efficient heat treatment concept has been proposed for high thermal conductivity. The direct tempering is a practical and economical approach for the heat treatment of tool steels produced by LPBF. After direct tempering, a fully martensitic matrix without solidification structure can be achieved. The carbide precipitation, together with material properties, can be tuned by applying various direct tempering temperatures. In absence of any strong recrystallization or grain growth during direct tempering, compared to conventional heat treatment, the fine microstructure from LPBF process is retained, potentially contributing to high strength and good toughness. Hence, this new heat treatment concept, realized by direct tempering, leads to a good combination of thermal conductivity and material properties. This approach is applicable to a wide range of laser additively manufactured tool steels.

CRediT authorship contribution statement

Siyuan Qin: Writing – original draft, Visualization, Methodology, Investigation, Conceptualization. **Simone Herzog:** Writing – review & editing, Supervision. **Anke Kaletsch:** Writing – review & editing, Supervision. **Christoph Broeckmann:** Writing – review & editing, Supervision, Project administration.

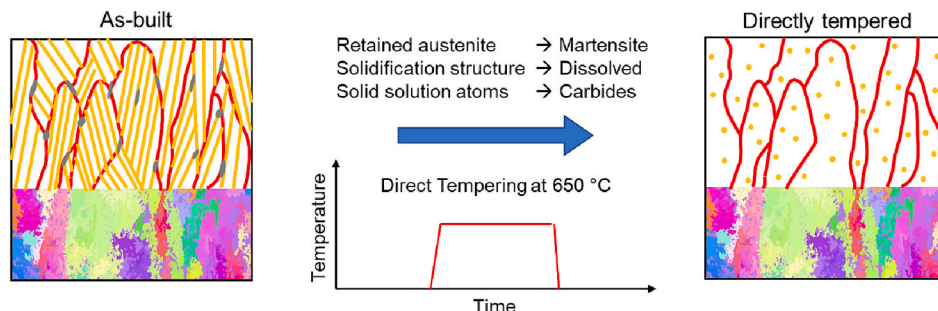


Fig. 12. Schematic illustration of microstructure evolution from the as-built condition to the direct tempering condition.

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.

Data availability

Data will be made available on request.

Appendix A. Appendix

A reference group was established using conventionally produced M50. The reference M50 was produced by Böhler (Germany) using a process of Vacuum Induction Melting followed by Vacuum Arc Remelting (VIM-VAR). Then the samples were quenched from 1180 °C and tempered at 580 °C, as outlined in Fig. 1a. Since the reference group exhibit very similar microstructure and material properties (Fig. A.1) to the LPBF produced sample with same heat treatment, these results do not offer new insights for discussion or conclusions, but comparison with LPBF produced group. As such, they are included in the appendix.

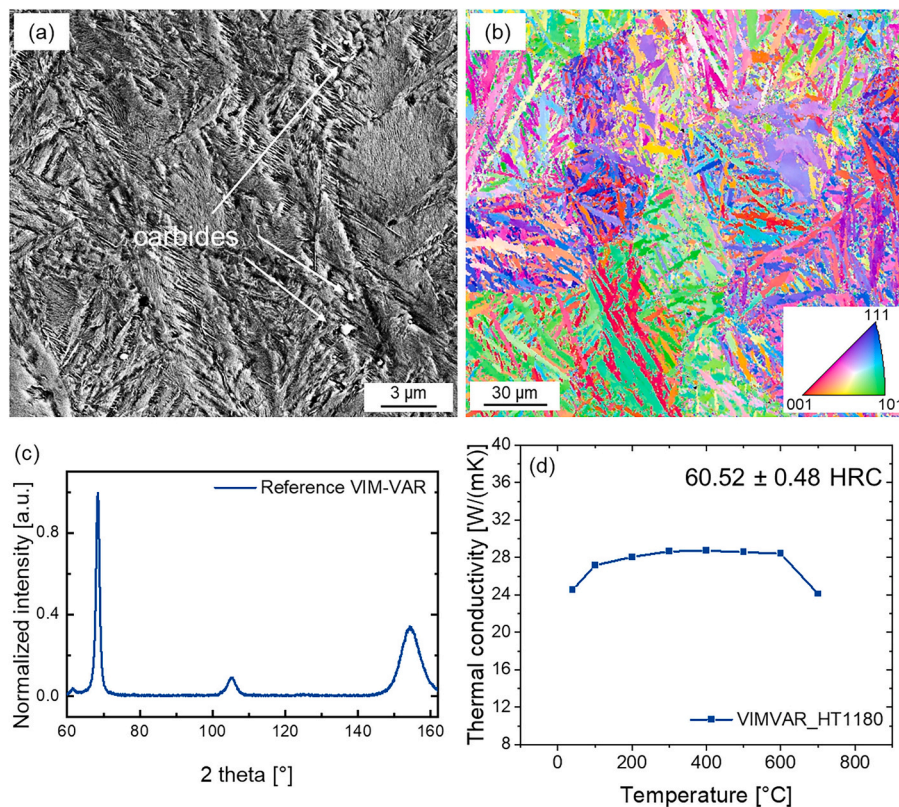


Fig. A.1. The microstructure and material properties of conventionally produced M50. (a) SEM, (b) IPF of EBSD, (c) XRD diffraction pattern, and (d) thermal conductivity and hardness.

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