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Title: Strong morphological and crystallographic texture and resulting yield strength anisotropy in Selective Laser Melted tantalum.

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Abstract

Selective Laser Melting (SLM) makes use of a high energy density laser beam to melt successive layers of metallic powders in order to create functional parts. The energy density of the laser is high enough to melt refractory metals like Ta and produce mechanically sound parts. Furthermore, the localized heat input causes a strong directional cooling and solidification. Epitaxial growth due to partial remelting of the previous layer, competitive growth mechanism and a specific global direction of heat flow during SLM of Ta result in the formation of long columnar grains with a $\langle 111 \rangle$ preferential crystal orientation along the building direction. The microstructure was visualized using both optical and scanning electron microscopy equipped with electron backscattered diffraction and the global crystallographic texture was measured using X-ray diffraction. The thermal profile around the melt pool was modeled using a pragmatic model for SLM. Furthermore, rotation of the scanning direction between different layers was seen to promote the competitive growth. As a result, the texture strength increased to as large as 4.7 for rotating the scanning direction 90° every layer. By comparison of the yield strength measured by compression tests in different orientations and the

averaged Taylor factor calculated using the visco-plastic self-consistent (VPSC) model, it was found that both the morphological and crystallographic texture observed in SLM Ta contribute to yield strength anisotropy.

Keywords: Additive Manufacturing; Tantalum; laser deposition; directional solidification; texture.

1. Introduction

Selective Laser Melting (SLM) is an Additive Manufacturing (AM) process in which complex functional parts can be produced directly by selectively melting successive layers of powder by the interaction of a laser beam. SLM offers several advantages compared to the conventional production techniques such as a high geometrical freedom, a near net shape production, an efficient use of material and a high production flexibility. Thanks to the high energy density of the laser beams, even refractory metals like tantalum may be processed by this technique [1]. For more details about the setup of the SLM process, the influencing parameters, advantages and disadvantages and various applications, the reader is referred to [2,3].

Tantalum (Ta) is a metal which is, due to the specific process of extraction, most often made available as ingots or powder for further processing [4,5]. The melting point of Ta is fairly high, i.e. 2996°C, and so is the density of 16.69g/cm³. In general Ta is also known as a very hard and ductile material [6]. The mechanical properties of pure Ta processed by various production techniques are summarized in Table 1. The ultimate tensile strength is seen to vary from 200 to 1400 MPa depending on the applied production technique.

Table 1: Overview of the mechanical properties of pure Ta processed by various production techniques.

Ta grade ^(a)	Commercial pure Ta EB ^(a) [7]	Commercial pure, P/M ^(a) [7]	Soft annealed Ta [6]	Cold worked Ta [6]
Modulus of elasticity (GPa)	185	185	186	186
Vickers Hardness (Hv)	110	120	60-120	105-200
Ultimate tensile strength (MPa)	205	310	200-390	220-1400
Yield strength (MPa)	165	220	na	na
Elongation (%)	40	30	20-50	2-20

(a) EB, electron beam furnace melted; P/M, powder metallurgy.

Tantalum is mainly used for the excellent corrosion resistance, biocompatibility and dielectric properties of its protective passivating oxide layer. This Ta₂O₅ oxide layer is formed when Ta comes in contact with oxygen at high temperatures. This oxide layer stays stable over the whole pH range in the stability region of water, which explains the good corrosion resistance [4]. The high melting point of Ta makes it suitable for heat shields or furnace applications. Ta is commonly applied as coatings onto common base metals or as rolled sheets.

Thanks to the high geometrical freedom offered by the layer wise addition of material during the SLM process, very complex parts can be produced. One example is the incorporation of porous tantalum bone scaffolds in orthopedic or dental implants for improved bone ingrowth [4]. Furthermore, due to the near-net-shaping, the costly material is efficiently used. The geometrical freedom and processing cost reduction by using SLM for tantalum processing could lead to a real breakthrough for tantalum applications.

The ability of SLM to produce porous Ta coatings and porous Ta scaffolds was already demonstrated by Fox et. al. [1]. These authors also could prove a significant improvement on the biocompatibility of porous Ta coatings compared to porous Ti coatings. Tantalum was previously also used in the Laser Engineered Net Shaping (LENSTM) process to deposit Ta coatings on titanium substrates and to produce porous Ta parts. The compressive 0.2% proof strength was measured to vary from 100-746MPa for porous samples with relative density ranging from 45.7% to 73.2% [7].

In this article, the microstructure and compressive yield strength of fully dense Ta parts produced by SLM are investigated. The influence of the scanning strategy on the microstructure of SLM Ta is studied. The microstructure resulting when no rotation of the scanning direction between consecutive layers is applied will be compared to the microstructure when a rotation of 60° and 90° rotation is applied. The grain structure was visualized by light optical microscopy (LOM) and scanning electron microscopy (SEM) equipped with an electron backscatter diffraction (EBSD) module. The global crystallographic texture was measured by X-ray diffraction. Based on this texture description, the visco-plastic behavior of SLM Ta was simulated using the visco-plastic self-consistent (VPSC) model [8,9]. The compression yield strength was determined for Ta samples with 60° rotation of the scanning direction between the layers to verify the effect of the unique SLM microstructure of Ta on the mechanical properties of this material and to compare with the VPSC model calculations. Finally, the formation of this microstructure will be explained based on the temperature profile around the melt pool during SLM of Ta.

2. Material and experimental procedures.

2.1. Production of parts by SLM

In this work, pure Tantalum powder with particle size ranging from 13 μm to 26 μm was used. The SLM production was performed in an inert atmosphere to avoid the pick-up of interstitial oxygen and parts were cut from the substrate using wire-EDM after production.

The selective laser melting process parameters were optimized to produce almost fully dense parts. The density was checked using the Archimedes' density method relative to the theoretical density of 16.6 g/cm^3 and amounts to $99.6 \pm 0.1 \%$. To take into account the influence of the laser path on the resulting material structure, three different scanning strategies were taken into account. In each part, long bidirectional scanning vectors are used, while the angle of the scanning direction between the successive layers α is chosen to be 0° , 60° and 90° . The samples will be named respectively Ta 0° , Ta 60° and Ta 90° in the text below. The applied scanning strategy and the different cross section views for microscopy are illustrated in Figure 1. Every layer, the laser movement started at another sample corner.

Figure 1: Illustration of the coordinate system with indication of the scanning direction SD, transverse direction TD and building direction BD and the different cross section views for microscopy. The arrows indicate the movement of the laser and α the rotation angle of the scanning direction between two consecutive layers n and n+1.

2.2 Microscopy

Samples Ta 60° and Ta 90° are cut along the XY and XZ planes for respectively a top and side view cross section. For sample Ta 0° , the same cross sections were prepared and also a cross section along the YZ plane was prepared to obtain information from the front view as well. Polished samples were etched by immersing during 60 s in a mixture of 20ml HNO_3 , 20ml HF and 60ml H_2SO_4 and viewed under a Leica DMILM

12V/100W microscope. The top surface of the as built parts was viewed using secondary electrons in a PHILIPS SEM XL40 equipped with a LaB₆ gun.

2.3 Texture measurements

X-ray measurements were performed on a SIEMENS D500 using a Cu tube at 40kV and 40mA. The diffraction pattern of the powder and Ta60° SLM part are shown in Figure 2. All peaks result from the body centered cubic Ta phase. The differences in intensities of the different peaks between the powder and the SLM part point to the presence of a crystallographic texture in the SLM part.

Figure 2: X-ray diffraction pattern of Ta powder and Ta60° SLM part on the surface perpendicular to the building direction. Intensities of the diffracted peaks relative to the highest peak are shown. The intensities of the powder material are shifted upward for visibility.

Pole figures with (110), (200), (211) and (310) reflections of the (Ta) phase were measured to explore this texture further. The rotation β of the sample around the intersection line of the sample surface with the plane of the incident and diffracted X-rays is thus measured between 0° and 80° in steps of 5°. At each β , a complete circle was measured in steps of 4°. Measuring the diffracted beam was executed during 1s for each point in space. The X-ray measurements were performed on polished cross sections perpendicular to the building direction mounted in a goniometer with the scanning direction (SD) parallel to the conventional rolling direction (RD). Also measurements on virgin powder were used to correct the measured pole figures. The MTM-FHM texture processing software [10] and MTEX Software Toolbox [11] were used for the calculations.

Electron backscattered diffraction (EBSD) is using a PHILIPS SEM XL30 equipped with a W gun and a TSL orientation imaging microscope system. The step size used for the EBSD analysis shown in this article ranges from 1 to 1.5 μm . The average confidence index of the measurements is around 0.70. The relation between the colors in the EBSD images and the corresponding crystal orientation is represented in Figure 3. In general, the orientation of the crystals is shown with respect to the building direction (BD) unless differently stated. On the image quality picture of the EBSD scans, the boundaries between zones with a misorientation of minimum 2° and maximum 5° are shown in red, a misorientation between 5° and 15° in green and a misorientation exceeding 15° in blue.

Figure 3: The inverse pole figure representing the relation between colors in the EBSD images and crystal orientations along the building direction (unless when stated differently in the text).

2.4 Prediction of the melt pool and temperatures during SLM

The temperatures in- and outside the melt pool during the SLM processing of the Ta metal were estimated using the pragmatic model developed by Verhaeghe et al. [12]. This model calculates the temperatures based on an enthalpy formulation and accounts for laser light penetration, shrinkage and evaporation. The model, however, neglects certain important phenomena such as surface tension driven shape evolution of the liquid pool or Maragoni convection flows inside the melt pool. The model was adapted for the SLM of Ta using a density of 16.690 g/cm^3 and the fraction of laser input absorbed by the solid and liquid phase of 32% found in [13]. The heat conductivity in function of the temperature is given by Equation 1 and is based on the values from [14]. The relation between the specific enthalpy in J/g and the temperature T in $^\circ\text{C}$ for pure Ta was calculated using the FactSage database.

Equation 1: Heat conductivity in function of the temperature for pure Ta based on the values from [14].

$$\lambda(T) = 57.216 + 4.5 \cdot 10^{-3} T - 4 \cdot 10^{-7} T^2 \text{ for } T \in [0 \dots 2985^\circ\text{C}] \quad (1)$$

A time step of 10^{-8} s and a grid step size of 2 μ m were used. The domain size was chosen $1000 \times 500 \times 400 \mu\text{m}^3$, which appeared large enough to be considered infinite and a total of 25000 time steps was needed to ensure the simulation of a steady state condition.

2.5 Mechanical testing

Solid cylindrical compression test samples with a diameter of 7 mm and a length of 18 mm were produced by SLM using process parameters identical to Ta0° and post-machined by milling and centerless grinding to the final dimensions of 6 mm diameter and 12 mm length. The compression tests were carried out on an INSTRON 5582 test system with a maximum load of 100kN and a deformation rate of 0.007 min^{-1} . The full stress-strain curves were measured without extensometer. The yield strength was determined by the clear yield point in the sharp-kneed stress-strain diagrams. The given spreads are the standard deviation of the replicates in the same orientation.

In order to observe the influence of the loading direction, three different build orientations are considered. Five samples were built vertically, i.e. the compressive loading direction coincides with the building direction, five samples were built horizontally, i.e. the compressive loading direction is perpendicular to the building direction and five were built diagonally, i.e. the loading direction is oriented 45° to the building direction.

2.6 Yield strength prediction.

The visco-plastic self-consistent (VPSC) model [8,9] was used for the prediction of the anisotropy in yield strength based on the crystallographic texture and the grain shape. In [15], it is shown that this model can take into account the grain shape in a sufficient way for three packaging steel sheet qualities. The N_{effect} variant of the VPSC model with a N_{effect} coefficient of 10 and a rate sensitive exponent n of 99 on each individual slip system was used for the calculations. The experimentally measured textures by XRD were used as the initial crystallographic texture for the simulations. The grain aspect ratio was estimated based on the EBSD results. Both the $\{110\}\langle 111 \rangle$ and $\{112\}\langle 111 \rangle$ slip systems were taken into account in the simulations. It must be noted that this model is a so-called mean field model, which means that the intragrain strain heterogeneity is neglected. Furthermore, both the grain size and grain shape distribution were not taken into account. The goal is to capture the yield strength anisotropy by this model in an at least qualitative manner.

3. Results

3.1 Microstructure

3.1.1 As-built top surface

Figure 4 shows the top surfaces of the Ta0° and Ta90° as built SLM parts. The individual scan tracks and their directions can be discerned from the crescent-shaped solidification ripples that arise at the top of the solidified melt pools. Furthermore, melt pools have an elongated almost tear drop shape. The angle ϕ , i.e. the angle between the ripples and the laser movement direction, is in the order of 35° over the majority of the melt pool tail. Furthermore, more or less perpendicular to the ripples, fine hairlines corresponding to grain boundaries are seen. The surface is flat and neighboring tracks are seen to overlap almost 50%.

Figure 4: SEM pictures of the top surface of as built Ta90° (a and c) and Ta0° (b and d) SLM parts at different magnifications. The grain boundaries (i.e. fine hairlines) are indicated by white arrows. The angle ϕ is also indicated in white on picture d.

3.1.2 Global crystallographic texture

The global crystallographic texture of the three samples was measured by X-ray diffraction. The orientation distribution function (ODF) was calculated from the measured pole figures. Using this function, the (111) pole figure and the inverse pole

figure showing the crystal orientations along the building direction were calculated. The different pole figures and inverse pole figures are shown in Figure 5. Note that the scale of the intensity for the Ta0° sample is 8 times smaller than for the Ta60° and Ta90° sample.

Figure 5: (111) pole figures (top) and inverse pole figures along the building direction (bottom) for the three different samples Ta0° (left), Ta60° (middle) and Ta90° (right). Note that the scale for the Ta0° is different than for Ta60° and Ta90°.

In all pole figures, the $\langle 111 \rangle$ crystal direction is the preferred direction along the building direction. For Ta60° and Ta90°, an almost perfect $\langle 111 \rangle$ fiber texture is present. The overall texture intensity is described by the texture index, the texture strength and the fiber volume.

The texture index was calculated from the orientation distribution function $f(g)$ using Equation 2. For isotropic materials, this texture index equals unity, anisotropic materials have a texture index larger than 1 [16]. The texture strength is the square root of this texture index. The texture strength is more meaningful to describe the texture since it has the same units as the measured intensities [16]. The fiber volume is here defined as the ratio of mass of the orientation distribution function that has an orientation within 20° from the $\langle 111 \rangle$ fiber along the building direction. Ta60° and Ta90° both show a very strong texture. Their texture index amounts respectively to 41 and 42 and their texture strength 4.4 and 4.7. The fiber volume for Ta60° and Ta90° equals respectively to 89% and 93%. Ta0° shows a much less strong texture. It has a texture strength of only 1.3 and a fiber volume of 46%.

Equation 2: Calculation of the Texture Index of a sample based on its orientation distribution function f in function of the Euler space coordinates g .

$$\text{Texture Index} = TI = \int_{\text{eulerspace}} (f(g))^2 dg \quad (2)$$

From the (111) pole figure for Ta0° at the top left in Figure 5, it can be seen that the texture of Ta0° is characterized by two different texture components. The two components have one <111> parallel to the building direction, but the two other <111> directions can take two different positions. Those two different positions are indicated by either square or triangular markers in Figure 5. It must be noted that the position of these components shows a large spread. A closer look to the (111) pole figure for Ta90° also shows some local maxima of 6.5 at the same positions as the triangles in the pole figure of Ta0°.

3.1.3 Grain structure and local texture

The grain structure and local texture was visualized by light optical microscope (LOM) and electron backscatter diffraction (EBSD) images. Figure 6 shows the top view images of Ta0°.

On these images, the individual tracks to fill the cross section of the part at a certain layer (i.e. filling tracks) can be recognized. The width of the tracks, i.e. 82 µm equals the scan spacing which was applied during processing. Most grains are seen to be confined to the track width and have in general a S shape which is mirrored for neighboring tracks. Some grains are seen to have only a low angle border along the track border. As a result, those grains are seen to overlap several neighboring tracks. Grains, which are defined by a misorientation angle larger than 15°, are seen to have an irregular shape and a large size distribution. The grain size ranges from several microns to several hundreds of microns. Furthermore, it can be seen that at the border of the tracks, small zones confined by low angle borders, i.e. with a misorientation between 2° and 5° or 5° and 15°, are present.

Near the border of the sample (within 300 µm from the sample edge), grains have random orientation. At the bulk of the parts, a <111> preferential orientation which is represented in blue, and to a lesser extent also a <100> preferential orientation which is represented in red, along the building direction is seen.

A comparison between the grain structure observed on the LOM and EBSD pictures explains the contrast seen in LOM. In LOM, grains with a <100> orientation along the building direction are seen to step out of the grains with a <111> orientation. Therefore,

it can be concluded that the grains with their $\langle 100 \rangle$ direction oriented out of the cross section plane are less attacked during polishing than the grains with a $\langle 111 \rangle$ orientation. Looking back to the LOM picture in Figure 6, it can be seen that over the whole LOM picture, some $\langle 100 \rangle$ oriented grains within a matrix of $\langle 111 \rangle$ oriented grains are present.

Figure 6: LOM and EBSD images of top view of Ta0°. a) LOM picture showing the position of EBSD inverse pole figure images b and c. Picture d shows the point to point image quality of map c and the rotation angle between misoriented zones of the top half of the picture.

On the images in Figure 6, also the contour track which is scanned along the borders of the cross section can be distinguished. The contour tracks are indicated by the black double sided arrows in Figure 6b and 6c. The grains inside the contour tracks are equiaxed and have a random orientation.

The effect of rotating the scanning direction every layer by 90° can be determined from the LOM and EBSD pictures of sample Ta90° in Figure 7. The individual solidified filling and contour tracks are again distinguishable. Here, however, a checkerboard pattern arises due to the 90° rotation.

Figure 7: LOM and EBSD images of top view of Ta90°. a) LOM picture showing the position of EBSD inverse pole figure images b and c. Picture d shows the point to point image quality and rotation angles of the misorientation borders of map c.

Within 350 µm from the border of the sample, grains have again a random orientation. This can be seen from the variety of colors in this zone. Outside this border area, in the bulk of the part, almost all grains are (close to) blue. This means that they have their $\langle 111 \rangle$ direction preferentially oriented along the building direction.

Again, the grains are mainly confined to the checkerboard square which is defined by the track width and these squares are decorated by thin elongated low, intermediate and high angle confined zones.

A comparison between the EBSD maps and the LOM confirms that the contrast is linked to the grain orientation. From the EBSD pictures, but also from the LOM picture which covers a larger area, it can be seen that in the bulk of the part mostly $\langle 111 \rangle$ oriented grains are present. Compared to Ta0°, fewer $\langle 100 \rangle$ oriented grains are left. This is in correspondence to the more intense $\langle 111 \rangle$ texture measured for the Ta90° than for the Ta0° sample.

The formation of a checkerboard pattern indicates that the grain structure of the previous layer has an influence on the grain structure of the currently added layer.

The LOM and EBSD pictures of the Ta60° in top view are shown in Figure 8. The orientation of the grains along the building direction is shown in Figure 8b and the orientation of the grains along a scanning direction is shown in Figure 8c. From the latter picture, individual tracks can be distinguished and the rotation of the scanning direction between the layers is seen to equal 60°. The $\langle 111 \rangle$ direction is preferentially

oriented along the building direction. However, also few grains with a $\langle 100 \rangle$ direction along the building direction are present.

Figure 8: LOM (a) and EBSD images (b and c) of top view of Ta60°. The EBSD inverse pole figure pictures are represented with the crystal orientations along the building direction BD (b) and along the scanning direction (c). The double-sided arrows and the dashed lines in c show the scanning directions of different layers.

Due to the layer wise addition during the process, anisotropy in the grain structure is expected. Therefore the side and front view cross sections are investigated as well. Figure 9 shows the different LOM and EBSD pictures in side and front view of Ta0°.

In the front and side view LOM pictures in respectively Figure 9a and 9d, no individual layers can be distinguished. On the contrary, elongated grains are seen to grow across the layers more or less parallel to the building direction. In the front view in Figure 9a, the width of the grains is restricted by the track width. The grains are seen to be several hundred micrometers long. Intermediate and high misorientation angle borders are seen along the track borders and perpendicular to the track in a cup-like shape. In the side view in Figure 9c, grains have a more capricious shape.

The EBSD orientation map of the front view shown in Figure 9b shows a preferential orientation of the grains with their $\langle 111 \rangle$ direction along the building direction. Furthermore, crystals rotate gradually inside the grains. This is observed in for example the second columnar grain from the left in Figure 9b.

The EBSD orientation map of the side view in Figure 9e shows the random orientation of grains within 350 µm from the edge of the part and the $\langle 111 \rangle$ direction preferentially oriented along the building direction for grains in the bulk of the part.

Figure 9: LOM and EBSD images of front (a, b and c) and side (d, e and f) views of Ta0°. a) LOM picture of the front surface, b) EBSD inverse pole figure image and c) image quality picture. The LOM side view picture d insert is showing the position of EBSD inverse pole figure in image e. The image quality picture of picture e is shown in picture f.

Rotation of the scanning direction by 60° or 90° breaks up the defined columnar structure from the front view in Figure 9a. It becomes much more difficult to distinguish the individual track as seen on the side view pictures of Ta90° in Figure 10a, 10b and 10c. Not only are the vertical columns disturbed when rotating the scanning direction, but also the $\langle 111 \rangle$ direction of the grains is more preferred along the building direction as shown by the overall blue color in Figure 10b.

Figure 10: LOM (a) and EBSD images (b and c) of side view of Ta90°. The EBSD inverse pole figure picture is represented with the crystal orientations along the building direction BD in picture b and along the scanning direction in picture c.

3.2 Temperature around melt pool.

In order to gain more insight in the solidification conditions during the SLM processing of the tantalum metal, the temperatures in- and outside the melt pool were estimated using the pragmatic model developed by Verhaeghe et al. [12]. In the simulation, the condition of the filling vectors was simulated. A powder layer is present on top a solid substrate next to an already solidified part of that layer. The laser beam moves above the powder-solid interface such that an overlap of 50% was obtained. The geometry setup is illustrated at the top in Figure 11.

Figure 11: Transverse (left) and longitudinal (right) cross sections of the temperature profile around the melt pool simulated using the pragmatic model for SLM by [1]. The temperature is shown in Kelvin.

Figure 11 shows the calculated temperatures between 300 and 3290K in the transverse and longitudinal cross sections. In the transverse cross section, the heat is dissipated radially from the melt pool center. The melt pool, in the longitudinal cross section, however is elongated in the direction of the laser movement. As a result, heat is not dissipated radially, but flows downwards. The value and angle towards the scanning SD, transverse TD and building direction BD ω_{SD} , ω_{TD} and ω_{BD} of the thermal gradient $\vec{G}$ are determined along the border of the melt pool for different cross sections. Only the thermal gradient value G and angle ω_{BD} for the tailing part of the melt pool border in the longitudinal cross section are plotted here in Figure 12.

Figure 12: Value (left) and angle to building direction (right) of the thermal gradient at the melt pool border in the tail of the melt pool of the longitudinal cross section shown in Figure 11.

From the value G , it can be seen that the SLM process is characterized by cooling gradients larger than 10^7 K/m or 10^7 K/s taking into account laser movement speed typically in the order of 1 m/s . Furthermore, the thermal gradient increases from the back of the melt pool to the bottom of the melt pool. The angle ω_{BD} is seen to vary between 80° at the back of the melt pool to around 10° at the bottom of the melt pool. Based on the results shown in Figure 12, also the growth rate R of the solidification front can be calculated. Under steady state condition, R equals the scanning speed times the cosines of the angle ω_{SD} [17]. Since solidification is both dependent on the growth rate and the thermal gradient, an average cooling gradient orientation can be estimated weighting the angle $\omega(i)$ with the normalized product of the thermal gradient G and the growth rate R as shown in Equation 3.

Equation 3: Calculation of the average orientation angle of the thermal gradient along the melt pool border.

$$\bar{\omega} = \sum_{border} \omega(i) \cdot \frac{G(i) \cdot R(i)}{\sum_{border} G(i) \cdot R(i)} \quad (3)$$

For the melt pool border of the longitudinal cross section, the average $\bar{\omega}_{SD}$ is calculated to be 57° and $\bar{\omega}_{BD}$ to be 33° . For the melt pool border in a transverse cross section the $\bar{\omega}_{TD}$ equals 41° and $\bar{\omega}_{BD}$ 49° .

3.3 Mechanical properties of Ta60°

To determine the effect of the strong $\langle 111 \rangle$ fiber texture and the elongated grains along the building direction on the yield strength, 15 compression samples were built using process parameters identical to sample Ta60° and oriented in three different directions. Five horizontally, i.e. loaded along the SD, five diagonally, i.e. loaded along 45° to the BD and 45° to the SD, and five vertically, i.e. loaded along the BD, oriented cylinders were tested. The resulting yield strength was measured to be $528 \pm 7 \text{ MPa}$ for the horizontally, $464 \pm 3 \text{ MPa}$ for the diagonally and $654 \pm 25 \text{ MPa}$ for the vertically oriented samples. These values are higher than all strengths, except for cold worked Ta, given in Table 1. The spread between the samples of a certain orientation is small, especially for the horizontal and the diagonal orientations. The differences in yield strength between the orientations, however, are significant. As a conclusion, the texture of the material has a large influence on the yield strength.

3.4 VPSC model prediction of yield strength anisotropy of Ta60°

The anisotropy in yield strength can be determined by calculating the Taylor factor M for a strain free material with a given texture and grain shape.

If the critical resolved shear stress τ_{crss} is equal and evolving with isotropic hardening for each slip system, the averaged Taylor factor can be calculated as following [18]. For one grain, the Taylor factor M_g is the sum of the total slip $\Delta\gamma_i^g$ of all slip systems S in the grain during the strain increment $d\varepsilon$ as shown in Equation 4. This Taylor factor depends on the imposed strain mode and grain orientation. For a polycrystalline material, a weighted average for a certain number of grains N , each with a weight factor w_g is calculated according to Equation 5. In this paper, the initial texture has been discretized into 5000 grain orientations using the ‘statistical’ method described by [19].

Equation 4: Taylor factor for one grain.

$$M_g = \frac{\sum_{i=1}^S \Delta\gamma_i^g}{d\varepsilon} \quad (4)$$

Equation 5: Weighted average Taylor factor for a set of grains.

$$\overline{M} = \frac{\sum_{g=1}^N w_g M_g}{\sum_{g=1}^N w_g} \quad (5)$$

This averaged Taylor factor $\overline{M}$ then relates the macroscopic flow stress σ_{33} of a polycrystalline material along a compressive direction -3 to the averaged critical resolved shear stress τ_{crss} in the polycrystal according to Equation 6 [16]. The physical meaning of this equation is that the increment of plastic work imposed by compressive stress σ_{33} is equal to the work accumulated by dislocation slip at the micro scale level for each slip systems in an average sense during this strain increment.

Equation 6: Relation between the macroscopic flow stress, the averaged Taylor factor and the critical resolved shear stress in the polycrystal.

$$\sigma_{33} d\varepsilon_{33} = \overline{M} \cdot \tau_{crss} d\varepsilon_{33} \quad (6)$$

So for a given strain increment, a larger Taylor factor $\overline{M}$ will correspond to a larger initial yield stress σ_{yield} , since at the beginning of the plastic deformation the averaged critical resolved shear stress τ_{crss} in the polycrystal is identical in different orientations of the sample.

Here, the VPSC model [8,9] was used to calculate the averaged Taylor factor $\overline{M}$ in function of the plastic strain. The grain shape aspect ratio along RD, TD and ND, indicated by RD:TD:ND was determined using the information from the EBSD maps. The irregular grain shape and broad variation in grain sizes, however, made it difficult to determine one single value of the aspect ratio which is representative to all grains. Therefore, a rough estimate of the grain shape ratio of 1:1:5 was used for the calculations. To only determine the effect of texture, also the plastic behavior for an equiaxed microstructure, i.e. with aspect ratio 1:1:1, is calculated. The calculations were performed for the three orientations corresponding to the compression test orientations. For each deformation orientation and grain shape ratio, the evolution of the average Taylor factor $\overline{M}$ in function of the plastic strain along the compressive direction is shown in Figure 13.

Figure 13: Evolution of the average Taylor factor $\overline{M}$ in function of the plastic strain. Considering equiaxed grains indicated by a grain shape ratio of 1:1:1 and elongated grains along the vertical direction indicated by 1:1:5 for three different orientations: horizontal, diagonal and vertical.

If an equiaxed microstructure is assumed, the $\overline{M}$ factor for the vertically built/loaded samples is much larger than for the horizontally and diagonally built/loaded samples.

The difference between the horizontally and diagonally built samples is small. If however, the elongated grain shape along the BD is taken into account, the difference between the horizontally and diagonally built samples becomes larger. Moreover, due to the introduction of the elongated grains along the BD, the horizontally built sample has a larger $\overline{M}$ than the diagonally built sample at the early stage of deformation. This grain shape does not affect the $\overline{M}$ value of the vertically built sample significantly.

The initial $\overline{M}$ value for the horizontally, diagonally and vertically built sample was calculated to be respectively 2.71, 2.68 and 3.6 with the aspect ratio equal to 1:1:5. Compared to the vertically built samples, the $\overline{M}$ value of the horizontally built samples is 88.7% and of the diagonally built direction 87.9% of the value for the $\overline{M}$ value of the vertically built. Based on the measured data, however, the yield strength of the horizontally built samples is 80.6% and the value of the diagonally built samples 71.0% of the yield strength of the vertically built ones. Consequently, the model is only able to calculate the correct trend, but not the absolute values in a quantitative manner. We believe that this less quantitative accuracy is mainly related to the very irregular grain shape of SLM Ta. This irregular grain shape makes the deformation in each grain inhomogeneous, while the model assumes a homogeneous strain distribution in each grain.

4. Discussion

4.1 Origin of the strong texture in Ta SLM parts

The results above show that the Selective Laser Melting process is able to melt completely the refractory Ta metal which has a melting point of almost 3000°C and to obtain nearly fully dense material (relative density of 99.6 ± 0.1 %). The unique process condition during SLM, furthermore, created a highly texturized material. The selected process parameters resulted in an almost 50% overlap of neighboring filling tracks as seen in Figure 4. Consequently, only half of the solidified melt pool tracks remained after scanning a layer.

From the temperature profiles and the SEM images of the top surfaces, it is seen that the movement of the beam caused the melt pool to elongate along the scanning direction. As a result, the thermal gradients G varies over the melt pool border and the direction of heat flux deviates from the radial direction to the melt pool center like reported previously also for welding processes [17]. The direction of the heat flux relevant for solidification during SLM of Ta is seen to point away from the laser movement. Based on the temperature modeling, the heat is seen to flow in the negative SD direction under an angle of 57° to SD in the longitudinal cross section when the laser is moved in the positive SD direction and in the positive SD direction when the laser is moved in the

negative SD. The heat flow always points in the negative TD direction when the order of the neighboring tracks of one layer is in the positive TD. The angle of the heat flow to the TD direction in the transverse cross section is 41° as mentioned above. Furthermore, the heat is always transferred in the negative BD direction. As a result, the heat flux is mirrored along TD when changing the sign of the scanning direction. Since grains grow opposite to the heat flux, this mirrored heat flux explains the mirrored grain structure observed in Figure 6 and the mirrored $\langle 111 \rangle$ components in the (111) pole figure of Ta0° in Figure 5.

During the SLM process, part of the underlying solid material, i.e. previously deposited layer or substrate, is remelted. This can be seen from the good density and from the melt pool depth calculated by the thermodynamic model. Remelting part of the previous layer eliminates the nucleation barrier for solidification and initial solidification will occur epitaxially at the partially melted grains [17]. The epitaxially solidification explains why grains grow across the layers in the micrographs above. Due to the planar solidification front, long columnar grains will arise. This epitaxial columnar growth in SLM is previously also reported for Ti6Al4V processed by SLM [20-25]. In [17], it is furthermore reported that initially, the grain growth is strongly influenced by the crystallographic orientation of the parent grain. But, due to a preferred easy growth direction of the crystals along the heat transfer direction, a competitive process among the grains will take place. Cubic metals are known to preferentially grow along the $\langle 100 \rangle$ crystal direction [26]. As a conclusion, due to the partial remelting, the crystal orientation of the previously deposited layer will be continued and will outlive under the condition that the $\langle 100 \rangle$ direction is oriented close to the optimal heat flux direction.

The texture of Ta0° shown in Figure 5 can be explained given the overall heat flux direction and the $\langle 100 \rangle$ preferred growth direction for the Ta crystals. The $\langle 111 \rangle$ direction along the BD results from rotating a cubic crystal with $\langle 100 \rangle$ directions along the specimen coordinate system 45° around SD and subsequently rotating 54° around TD. These rotations are close to the calculated average angles of the heat flux at the melt pool border to the different directions. The two different components are caused by the alternation of the scanning direction. The texture component indicated by the triangular markers arises from scanning in the positive SD and the one indicated by the square markers from scanning in the negative direction. Due to the moving heat source, however, the heat flow direction is seen to vary along the melt pool boundary from 90° at the position of the heat source to 0° at the tail of the melt pool. As a result, grains may shift their growth direction to be more aligned with the thermal gradient. [17] This is observed by the gradual color transitions inside the grains, i.e. gradual rotation of the crystal orientation in the EBSD maps. Also the spread in the SD direction of the $\langle 111 \rangle$ component along the BD and the rotational spread of the other local maxima in the pole figure of Ta0° in Figure 5 can be explained by the variation in the heat flux direction.

Near the edge of the parts, the temperature profile around the melt pool deviates from the steady state condition. The heat flux direction will differ and more random crystallographic orientations are present.

The competitive growth mechanism becomes more important when rotating the scanning direction between the layers. In the Ta0° samples, the laser paths are the same for every layer so also the heat flow direction is similar from layer to layer. When the direction of scanning between the layers is rotated, also the heat flux direction is rotated. Due to the epitaxial solidification, a checkerboard pattern arise when rotating 90° and grains ordered in a hexagonal way when rotating 60°. The formation of a checkerboard pattern due to rotating the scanning direction 90° is also observed in SLM Ti6Al4V [22]. Furthermore, when applying a rotation, less remelted grains will have an optimal crystal orientation due to the rotation of the heat flux and the competitive grain selection will become more pronounced. As a result, a stronger texture will arise as confirmed by the calculated texture strength and fiber volume.

4.2 Effect of the strong texture on the mechanical properties

The compression test results showed good yield strengths. The yield strength of SLM Ta is higher than the values reported for the Ta grades, except cold worked Ta, listed in Table 1. Furthermore, a large influence of texture was determined. The strength in the vertical direction is much higher than the strength in the horizontal or diagonal direction. And the strength in the horizontal direction is a little, but still significantly higher than the strength in the diagonal direction. The estimation of the averaged Taylor factor $\overline{M}$ value shows that the main contribution to this anisotropy comes from the crystallographic texture. The {110}<111> and {112}<111> slip systems are seen to be oriented least favorable when loading the material in the vertical direction compared to loading in the diagonal direction. However, also the elongated grain shape along the building direction is seen to add to this anisotropy and causes the strength in the diagonal direction to drop below the strength in the horizontal direction. In [15], they found that the grain shape effect is also sensitive to certain texture component. In other words, there is an interaction between the given grain shape and certain texture components. According to the VPSC model calculations here, the <111> fiber texture in SLM Ta is insensitive to the elongated grain shape when loading along the building direction.

5. Conclusions

Selective Laser Melting of Ta is able to fully melt Ta powder and to obtain almost fully dense and strong parts. The additive nature of the process, i.e. addition of material track after track and layer after layer, and the large directional cooling creates unique solidification conditions. These unique solidification conditions result in the creation of

large columnar grains across the layers and a $\langle 111 \rangle$ crystal direction preferentially along the building direction. This preferential crystal orientation could be explained by the overall heat flux direction simulated by a pragmatic model for SLM. Alternating the scanning direction in both a positive and negative direction, give rise to alternating heat flux directions and as a result two $\langle 111 \rangle$ texture component mirrored along the TD direction. Furthermore, grain shape orientation from neighboring tracks is also seen to be mirrored along the TD direction when viewed from the top.

Rotating the scanning direction between the layers gives rise to more planar isotropy in both the crystal texture and the grain shape. In addition, due to the more pronounced selection of grains by the competitive growth mechanism when rotating the scanning direction, the crystallographic texture is seen to be intensified.

Both morphological and crystallographic texture results in large yield strength anisotropy. This is verified by both experimental and simulated compressive loading in three different directions for samples produced using the Ta60° process parameters. Using the VPSC model, it was qualitatively shown that the crystallographic texture is the dominant cause for the large difference in yield strength between the vertical direction and the diagonal and horizontal ones. The morphological texture is seen to further increase this difference and to let the yield strength in the diagonal direction drop below the one for the horizontal direction.

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