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**Graphical Abstract**

SLM-processed 304L

**Tensile and neutron diffraction**

26% true strain

42% true strain

- $\gamma$-austenite favors $\varepsilon$-martensite
- $\gamma$-austenite does not favor $\varepsilon$-martensite
- $\varepsilon$-martensite
- $\alpha'$-martensite

True strain vs. True stress (MPa)

Work hardening rate (MPa)

*Graphical Abstract*
High ductility and transformation-induced-plasticity in metastable stainless steel processed by selective laser melting with low power

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Abstract

A metastable austenitic Fe-Cr-Ni steel was processed by selective laser melting with low power, resulting in nearly random crystallographic texture. In-situ tensile loading and neutron diffraction experiments were undertaken and the results indicate high ductility, despite the presence of porosity, and pronounced strain-induced martensitic transformation. A secondary hardening is observed in the mechanical data due to the martensite carrying significant load, upon its formation. The pronounced martensitic transformation is discussed with respect to the initial and the evolving deformation texture and its affinity for forming martensite under uniaxial tensile loading.

Keywords: additive manufacturing; stainless steels; tension test; martensitic phase transformation; neutron diffraction
Selective laser melting (SLM) is an additive layer-by-layer manufacturing process where the component is built by a laser heat-source that melts the powder, which is spread on the base plate by a recoater [1]. This technique is well established for processing complex component geometries and different metallic alloys, amongst which austenitic stainless steels are used extensively. Austenitic stainless steels, such as 316L and 304L exhibit a combination of high strength and ductility and good corrosion resistance. Due to their additional good SLM processability, they are ideal candidates for medical applications profiting e.g. from the possibility of additive manufacturing to produce complex geometries [2,3].

The austenitic steel 304L, due to its relatively low stacking fault energy (SFE), i.e. 18 mJ/m² as reported in [4], is known to exhibit the transformation induced plasticity (TRIP) effect upon deformation of its wrought form [5–7], where the face-centered-cubic (FCC) parent phase transforms to the hexagonal-closed-packed (hcp) ε-martensite and the cubic-body-centered (BCC) α'-martensite. The TRIP effect has been shown to result in a high rate of macroscopic strain hardening [8,9]. The extend of strain-induced martensite depends on the loading state in combination with the crystallographic texture [10–13]. It was seen that uniaxial loading favors the martensitic transformation in randomly textured, low SFE austenitic steel following the sequence γ →ε → α', where at low strains ε-martensite is the precursor of α'-martensite. The relationship of texture/loading direction is rationalized by comparing the Schmid factor (SF) of the leading partial dislocation to the SF of the trailing partial dislocation. For crystallographic orientations for which the SF of the leading partial dislocation is higher than the SF of the trailing, extensive stacking faults form allowing the formation of ε-martensite [10,11].

The SLM processing parameters can largely influence the microstructure, porosity, surface roughness and eventually the mechanical properties of the components. The crystallographic texture has been also shown to be sensitive to the SLM processing parameters and can vary from random using low energy input [14–16] to strong texture [14,16] during SLM processing. Extensive
studies have been carried out on the optimization of the SLM process parameters for the 316L steel [17,14,18–25], with respect to porosity minimization, mechanical properties optimization and occurrence of residual stress. However, not as many studies exist for SLM-processing of the 304L steel [26–29].

In a recent study, a good combination of strength and ductility was obtained in the as-SLM processed material due to the optimization of the SLM process parameters which resulted in the presence of cellular structures, small grain size and martensite, all of which are attributed to the fast cooling rates [29]. However, the deformation mechanisms, which gave rise to the combination of high strength and good ductility were not investigated in depth. In more recent studies on 304L processed by direct energy deposition (DED), it was shown that the transformation is suppressed, due to high nitrogen content of the powder used for additive manufacturing processes [30,31]. Hence, the dominant deformation behavior of materials processed with some additive manufacturing methods can be different to that of wrought materials.

In the present work, the SLM processing parameters are selected such that the as-SLM processed 304L steel exhibits nearly random crystallographic texture. Based on previous observation this crystallographic texture is favorable for exhibiting pronounced TRIP effect under uniaxial tension, for low SFE wrought austenitic steels [11]. The SLM-processed material is investigated using in-situ uniaxial deformation with neutron diffraction to reveal the deformation mechanisms of the SLM-processed material. The mechanical properties are discussed in relation to the residual stress that is present in the SLM-processed material, the evolving deformation texture and the TRIP effect.

For the SLM process, gas-atomized 304L powder with maximum particle size of 45 μm, was purchased from Goodfellow, UK. The SLM-fabrication was performed using a Sisma MySint 100 with a spot size of 55 μm, laser power 175 W and laser scan velocity of 1200 mm/s. A “chess-board” scanning pattern (squares of 4×4mm²) for each layer was rotated 90 degrees and shifted 1 mm in x and y with respect to the previous layer. The obtained density is ~98%, as characterized by optical
microscopy on sample cubes produced with the same conditions (cf. Fig. S1 of the supplementary materials). Cylinders of 14 mm in diameter and 86 mm in height were built with their long direction parallel to the building direction. “Dogbone” specimens for uniaxial tension tests were then machined from the as-built cylinders, the geometry is shown in Fig. S2 of the supplementary material.

Residual stress characterization by neutron diffraction of the SLM processed material was undertaken on the POLDI instrument at SINQ at the Paul Scherrer Institute, Switzerland, using a 3.8×3.8×3.8 mm³ gauge volume. The residual strain was measured along the radial and axial direction along a line in the center of the cylinder scanned with 5 steps of 12 mm step size over +/-24 mm from the center of the as-built cylinder and only at the central point for the machined dogbone specimen (shown in Fig. S3 of the supplementary material). For the residual stress characterization, the lattice strain was calculated using the {311} lattice plane family as it best represents the bulk elastic properties for FCC materials [32,33]. Reference measurements were undertaken on an annealed cylinder at 450°C for 5 hours for obtaining the strain free interplanar lattice spacing, \( d_{0}^{[311]} \). Based on the fact that the interplanar lattice spacing of the reference measurement shows values very close to the ones obtained from a measurement on the as-received gas-atomized powder, it can be considered “strain-free”. The value of \( d_{0}^{[311]} \) did not vary significantly from the top to the bottom of the cylinder, implying no considerable chemical variations, and therefore an average value was used for calculating the residual strain for all measurement points. The residual elastic strain, \( \varepsilon_{[311]} \), was then calculated from the interplanar lattice spacing, \( d_{[311]} \), of each point along an SLM-built cylinder, as follows:

\[
\varepsilon_{[311]} = \frac{d_{[311]} - d_{0}^{[311]}}{d_{0}^{[311]}} \tag{1}
\]

Residual stress is then calculated using Hooke’s law. For the diffraction elastic constants, the lattice strain evolution of the {311} lattice plane family obtained from the (longitudinal) in situ deformation
test was fitted in the elastic regime by a line, the slope of which is equal to the Young’s modulus, i.e. 
\[ E_{[311]} = 175 \text{ GPa} \] (cf. Fig. S4 of the supplementary materials). The ratio of the lattice strain in the elastic 
regime between the transverse and longitudinal measurements was used to calculate the Poisson’s 
ratio of the [311] lattice plane family, i.e. \( v_{[311]} = 0.3 \) as show in Fig. S4 of the supplementary material. 
It is seen that the as-SLM-processed material exhibits high compressive stress along the building 
direction, which is in good agreement with the reports in literature [34]. The machining process of 
the dogbone sample relieves approximately 80% of the residual stress, as shown in Fig. S3 of the 
supplementary material.

For the in situ uniaxial deformation and neutron diffraction experiments, the dogbone specimens 
were deformed with 0.01 mm/min displacement rate. The neutron diffraction measurements were 
undertaken upon stopping and holding the displacement at pre-defined force values (in the elastic 
regime) and pre-defined strain values (in the plastic regime). The in-situ diffraction neutron 
measurements were only undertaken at the center of the dogbone sample. For both residual stress 
and in situ deformation tests, the neutron data were analyzed and fitted using Mantid [35]. The data 
were analyzed qualitatively in terms of appearance of new reflections due to the presence of 
martensite. The evolution of the elastic lattice strain \( \varepsilon_{hkl} \) was also determined by the relative change 
of the interplanar lattice spacing \( d_{hkl} \) with respect to \( d_{0hkl} \) which is the initial value prior to 
deformation:

\[
\varepsilon_{hkl} = \frac{d_{hkl} - d_{0hkl}}{d_{0hkl}} \tag{2}
\]

Using \( d_{0hkl} \) which is the initial value for austenite, prior to deformation, does not account for the 
lattice strain, already present due to the low residual stress in the material. However, it does not 
affect assessing the slope in the elastic regime for calculating the diffraction elastic constant, or for 
assessing which phase accumulates more strain once martensite forms. For martensite, \( d_{0hkl} \) is 
considered the interplanar lattice spacing of the martensite reflections \( \{hkl\} \) upon martensite 
formation.
The as-SLM-processed and the deformed materials were further characterized by electron backscattered diffraction (EBSD). Samples were cut from the top (away from the base plate), bottom (close to the base plate) and the middle of the deformed dogbone sample. The samples were ground with 1200 grit SiC paper and then electropolished for 12 s with a 16:3:1 (by volume) ethanol, glycerol and perchloric acid solution at 42 V. A field emission gun scanning electron microscope (FEG SEM) Zeiss ULTRA 55 equipped with EDAX Hikari Camera operated at 20 kV in high current mode with 120 μm aperture was used. The EBSD raw data was post-processed using the EDAX OIM Analysis 7.3 software.

The as-SLM-processed material is fully austenitic, as seen in the inverse pole figure (IPF) map shown in Fig. 1 and in the neutron diffraction pattern shown in Fig. 2-b, which is in contrast with previous works where the as-SLM-processed material exhibited a small fraction of martensite, the presence of which was attributed to the fast cooling rates [29,36]. Fig. 1 shows the microstructure of the as-SLM-processed material, parallel to the building direction and the inverse pole figures (IPFs) parallel to the three principal directions (i.e. building direction-BD, transverse direction-TD and normal to the sample surface-ND). The crystallographic texture is seen to be nearly random, as previously seen for low energy SLM-processed stainless steels [14,17] and when the laser scanning pattern of each layer is rotated 90 degrees with respect to the previous layer [37].

Fig. 2-a shows the true stress versus true strain mechanical data and the work hardening rate obtained from a continuous (uninterrupted) test performed ex-situ. The mechanical data from the in-situ test coincide very well with the data obtained from the ex-situ test. The yield stress, \( \sigma_{0.2} \) is 450 MPa which is slightly lower than for the SLM-processed 304L alloy reported in [27]. However, such variations can be due to different magnitude of residual stress and/or microstructural differences (e.g. grain size, grain morphology, crystallographic texture etc.). Upon deformation, a secondary strain hardening regime is observed after approximately 0.23 true strain as shown in Fig. 2-a. The strengthening is also apparent in the work hardening rate also plotted in Fig. 2-a. This
hardening coincides with the onset of the martensitic transformation, as apparent by the appearance of the (110) reflection of $\alpha'$-martensite shown in Fig. 2-b. Two very weak reflections, i.e. (10$\overline{1}$0) at 2.9 Å and (10$\overline{1}$1) at 3.2 Å, corresponding to $\epsilon$-martensite also appear at true strain higher than 0.23. The latter observations indicate the formation $\epsilon$-martensite, additional to of $\alpha'$-martensite, as previously seen for low SFE austenitic steels [38,11]. During the initial stages of plastic deformation, diffraction peak broadening is observed as indicative of plasticity by dislocation density increase, slip, or stacking fault formation as seen in Fig. S5 of the supplementary material [38,11]. Significant amount of deformation induced $\alpha'$-martensite is observed in the SLM-processed 304L material in contrast to previous observations on 304L processed by DED, where strain-induced martensite was suppressed [30,31].

Fig. 3-a shows the evolution of the lattice strain of austenite with increasing strain. Until the macroscopic yield point, $Y_{S0.2}$, a typical mechanical behavior for polycrystalline FCC materials is seen, where the {111} family of planes is the stiffest and the {200} family is the most compliant. The {311} family exhibits an intermediate mechanical behavior, which justifies the choice of this family of lattice planes for the residual stress characterization. The secondary hardening can be explained by comparing the austenite and martensite lattice strain beyond 0.23 true strain. As shown in Fig.3-b, the martensite lattice planes accumulate strain at higher rate than austenite. For instance, by comparing the lattice strain in the {311}$_{\text{fcc}}$ and {211}$_{\text{bcc}}$ lattice plane families, which are typically considered as representative of the bulk material properties, it is apparent that martensite carries more load than austenite.

It is known that the crystallographic orientation changes by deformation and a strong <111>-texture evolves parallel to the loading direction in uniaxial tensile deformation [11]; the evolved texture at 0.26 and 0.42 true strain is shown in Fig. S6 of the supplementary material. The low magnification EBSD maps in Fig 4-a and 4-b show the austenite grains which have crystallographic orientations favorable for the splitting of the partial dislocations in blue and the austenite grains which have
crystallographic orientations not favorable for the splitting of the partial dislocations in red. The α’-
martensite grains are shown with yellow. Figures 4a and -b confirm that the majority of the
austenite grains (at 0.26 and 0.42 true strain) have crystallographic orientations that favor the
formation of ε-martensite (shown with blue color in Fig. 4-b, -c and -d). A significant amount of α’-
martensite is observed at 0.26 true strain and even more is formed at 0.42 true strain (Fig. 4-d),
which is in good agreement with the neutron diffraction results. The presence of ε-martensite is
evidenced in the high resolution EBSD map at high magnification presented in Fig. 4-c. It shows an
austenite grain with favorable crystallographic orientation for the formation of ε-martensite. ε-
martensite plates shown with green color form on the (111) slip plane of this grain. Furthermore, α’-
martensite is seen to form inside the bands of ε-martensite rather than at the ε-martensite
intercepts as is frequently reported [11]. According to Tian et al. [39] α’-martensite can form in
individual ε-martensite in very low SFE steels, indicative of the instability of austenite. It is therefore
seen that the martensitic transformation occurs via the path γ→ε→α’.

In conclusion, good ductility, despite the observed porosity, and pronounced martensite formation
was observed under uniaxial deformation of SLM-processed 304L material produced with low laser
power and chessboard laser scanning pattern. The crystallographic texture of the as-SLM-processed
material is nearly random while upon deformation a strong <111>-texture evolves with increasing
deformation. A majority of grains are favorably oriented so that the leading partial dislocation
experiencing higher stress than the trailing partial dislocation, allowing the formation of ε-
martensite, while α’-martensite forms in single bands of ε-martensite. The appearance of strain-
induced martensite results in a secondary work-hardening regime as martensite carries more load
than austenite.
Figures and captions

Fig. 1 IPF map of the austenite phase of the as-SLM-processed 304L steel in the direction parallel to the building direction (BD), obtained at the center of the cylinder. The inverse pole figures show the crystallographic orientation with respect to BD, transverse direction (TD) and normal direction (ND).

Fig. 2 a) True stress-strain and work hardening rate of an ex-situ continuous test showing hardening after approximately 0.23 true strain (indicated with a blue dashed line). (b) Evolution of neutron diffraction patterns showing the martensite formation (appearance of the 110_{BCC}, 1010_{HCP} and 1011_{HCP} reflections) after approximately 0.23 true strain (indicated with a black dashed line).
Fig. 3 (a) Lattice strain evolution in austenite. (b) Lattice strain evolution at 0.23 true strain and with progressing the deformation showing that martensite accumulates lattice strain at higher rate than austenite. The lines in (b) are liner fits to the experimental data for guiding the eye.

Fig. 4 EBSD maps showing with blue the preferred orientations along the building direction (parallel to the loading direction-LD) that favor the formation of ε-martensite under uniaxial tension along the given LD at a) 0.26 true strain and b) 0.42 true strain. The orientations that do not favor the formation of ε-martensite are given in red, α’-martensite is given in yellow and ε-martensite in green. c) Detail of a grain which is favorably orientated for the formation of ε-martensite showing ε-martensite plates being formed along the ⟨111⟩ slip plane traces, while α’-martensite forms within single bands of ε-martensite.
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References

[1] D. Herzog, V. Seyda, E. Wycisk, C. Emmelmann, Acta Mater 117 (2016) 371–392.

[2] F. Bartolomeu, M. Buciumeanu, E. Pinto, N. Alves, O. Carvalho, F.S. Silva, G. Miranda, Addit Manuf 16 (2017) 81–89.

[3] D. Kong, X. Ni, C. Dong, X. Lei, L. Zhang, C. Man, J. Yao, X. Cheng, X. Li, Mater Des 152 (2018) 88–101.

[4] R.E. Schramm, R.P. Reed, Metall Mater Trans A 6 (1975) 1345.

[5] G.B. Olson, M. Cohen, Metall Mater Trans A 6 (1975) 791.

[6] F. Lecroisey, A. Pineau, Metall Mater Trans B 3 (1972) 391–400.

[7] A.K. De, D.C. Murdock, M.C. Mataya, J.G. Speer, D.K. Matlock, Scripta Mater 50 (2004) 1445–1449.

[8] J.B. Leblond, Int J Plast 5 (1989) 573–591.

[9] R.G. Stringfellow, D.M. Parks, Int J Plast 7 (1991) 529–547.

[10] S. Martin, C. Ullrich, D. Rafaja, Mater Today: Proceedings 2 (2015) S643–S646.

[11] E. Polatidis, W.-N. Hsu, M. Šmíd, T. Panzner, S. Chakrabarty, P. Pant, H. Van Swygenhoven, Scripta Mater 147 (2018) 27–32.

[12] M. Zecevic, M.V. Upadhyay, E. Polatidis, T. Panzner, H. Van Swygenhoven, M. Knezevic, Acta Mater (2019).
[13] E. Polatidis, M. Smid, W.-N. Hsu, M. Kubenova, J. Capek, T. Panzner, H. Van Swygenhoven, Materials Science and Engineering: A Under review (n.d.).

[14] T. Niendorf, S. Leuders, A. Riemer, H.A. Richard, T. Tröster, D. Schwarze, Metall Mater Trans B 44 (2013) 794–796.

[15] Y.M. Wang, T. Voisin, J.T. McKeown, J. Ye, N.P. Calta, Z. Li, Z. Zeng, Y. Zhang, W. Chen, T.T. Roehling, R.T. Ott, M.K. Santala, P.J. Depond, M.J. Matthews, A.V. Hamza, T. Zhu, Nat Mater 17 (2018) 63–71.

[16] Z. Sun, X. Tan, S.B. Tor, C.K. Chua, NPG Asia Mater 10 (2018) 127–136.

[17] T. Niendorf, F. Brenne, Mater Charact 85 (2013) 57–63.

[18] B. Zhang, L. Dembinski, C. Coddet, Mater Sci Eng A 584 (2013) 21–31.

[19] A.S. Wu, D.W. Brown, M. Kumar, G.F. Gallegos, W.E. King, Metall Mater Trans A 45 (2014) 6260–6270.

[20] J.A. Cherry, H.M. Davies, S. Mehmood, N.P. Lavery, S.G.R. Brown, J. Sienz, Int J Adv Manuf Technol 76 (2015) 869–879.

[21] Z. Sun, X. Tan, S.B. Tor, W.Y. Yeong, Mater Des 104 (2016) 197–204.

[22] R. Casati, J. Lemke, M. Vedani, J Mater Sci Technol 32 (2016) 738–744.

[23] E. Liverani, S. Toschi, L. Ceschini, A. Fortunato, J Mater Process Tech 249 (2017) 255–263.

[24] T. Simson, A. Emmel, A. Dwars, J. Böhm, Addit Manuf 17 (2017) 183–189.

[25] C. Qiu, M.A. Kindi, A.S. Aladawi, I.A. Hatmi, Sci Rep 8 (2018) 7785.

[26] D. I. Bourell, K. Abd-Elghany, Rapid Prototyp J 18 (2012) 420–428.

[27] K. Guan, Z. Wang, M. Gao, X. Li, X. Zeng, Mater Des 50 (2013) 581–586.

[28] D.W. Brown, D.P. Adams, L. Balogh, J.S. Carpenter, B. Clausen, G. King, B. Reedlunn, T.A. Palmer, M.C. Maguire, S.C. Vogel, Metall Mater Trans A 48 (2017) 6055–6069.

[29] Q.B. Nguyen, Z. Zhu, F.L. Ng, B.W. Chua, S.M.L. Nai, J. Wei, J Mater Sci Technol 35 (2019) 388–394.

[30] Z. Wang, T.A. Palmer, A.M. Beese, Acta Mater 110 (2016) 226–235.
[31] Z. Wang, A.M. Beese, Mater Sci Eng A 743 (2019) 811–823.

[32] B. Clausen, T. Lorentzen, T. Leffers, Acta Mater 46 (1998) 3087–3098.

[33] T. Pirling, G. Bruno, P.J. Withers, Mater Sci Eng A 437 (2006) 139–144.

[34] K. An, L. Yuan, L. Dial, I. Spinelli, A.D. Stoica, Y. Gao, Mater Des 135 (2017) 122–132.

[35] O. Arnold, J.C. Bilheux, J.M. Borreguero, A. Buts, S.I. Campbell, L. Chapon, M. Doucet, N. Draper, R. Ferraz Leal, M.A. Gigg, V.E. Lynch, A. Markvardsen, D.J. Mikkelson, R.L. Mikkelson, R. Miller, K. Palmen, P. Parker, G. Passos, T.G. Perring, P.F. Peterson, S. Ren, M.A. Reuter, A.T. Savici, J.W. Taylor, R.J. Taylor, R. Tolchenov, W. Zhou, J. Zikovsky, Nucl Instrum Methods Phys Res A 764 (2014) 156–166.

[36] C. Haase, J. Bültmann, J. Hof, S. Ziegler, S. Bremen, C. Hinke, A. Schwedt, U. Prahl, W. Bleck, Materials 10 (2017) 56.

[37] L. Thijs, K. Kempen, J.-P. Kruth, J. Van Humbeeck, Acta Mater 61 (2013) 1809–1819.

[38] A. Das, S. Sivaprasad, M. Ghosh, P.C. Chakraborti, S. Tarafder, Mater Sci Eng A 486 (2008) 283–286.

[39] Y. Tian, O.I. Gorbatov, A. Borgenstam, A.V. Ruban, P. Hedström, Metall Mater Trans A 48 (2017) 1–7.
Figure 2

(a) (b)
Figure 3

(a) Lattice strain (με) vs. True stress (MPa) for different crystallographic planes. (b) Lattice strain (με) vs. True stress (MPa) for different crystal structures.
Figure 4

(a) γ-austenite favors ε-martensite
(b) γ-austenite does not favor ε-martensite
(c) ε-martensite

LD

α′-martensite
