

Effects of heat treatment and build orientation on the evolution of ϵ and α' martensite and strength during compressive loading of additively manufactured 304L stainless steel

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Abstract

The effect of heat-treatment and build orientation on martensitic phase transformation in additively manufactured (AM) 304L stainless steel is studied and compared with conventionally produced wrought material. The relationships between observed martensitic transformations and material microstructures and their effects on mechanical strength are established through experimental observations. *In situ* high-energy X-ray powder diffraction measurements were performed to monitor the evolution of ϵ and α' martensite during compressive loading of stainless steel. Electron backscatter diffraction (EBSD) was used to provide insight on initial grain morphology, crystallographic misorientation within grains, and crystallographic texture. Heat

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treatment alters the microstructure of AM samples creating different initial conditions. This difference in starting microstructure resulted in variability in martensitic transformation during compressive deformation. The rate of martensitic transformation decreased for samples treated with temperatures up to 1100°C, after which the AM microstructures recrystallized, resulting in increased rate of martensitic transformation for those samples treated at higher temperatures. It was also observed that aligning the axis of compression with the AM build direction resulted in a lower rate of strain-induced martensite formation as opposed to aligning the compression axis perpendicular to it. More favorable distribution of crystal orientations in the latter loading orientation promoted martensitic transformation. These and additional experimental observations from EBSD in terms of kernel average misorientation, mean grain orientation spread, and mean crystallite size reveal strong microstructural effects on strength of additively manufactured metallic materials.

Keywords:

martensite, plastic deformation, additive manufacturing, 304L stainless steel, *in situ* high-energy X-ray diffraction

1. Introduction

Austenitic stainless steels are a large family of alloys with wide-ranging applications due to their high corrosion resistance, high mechanical strength over a large range of temperatures, high ductility and formability, and exceptional weldability [1, 2]. Some of these steels are metastable at room temperature and will undergo solid-state phase transformations if plastically

7 strained, which can greatly increase material strength and formability. The
8 transformations observed in these steels are from the initial face-centered cu-
9 bic (fcc) austenite (γ) directly to a body-centered tetragonal (bct) martensite
10 (α') or from γ to α' through an intermediate hexagonal close-packed (hcp)
11 martensite (ϵ) [3, 4]. These transformations are shown in equations 1 and 2.



12 Martensitic transformations in steels occur spontaneously upon cooling
13 to temperatures below the critical martensite start temperature M_s ; however,
14 with the aid of plastic strain, these transformations can be induced at temper-
15 atures above M_s , up to a critical temperature M_d at which point the austenite
16 becomes thermodynamically stable in the presence of plasticity [2]. Marten-
17 site formed during plastic deformation occurs via an athermal, diffusionless
18 shear transformation and is commonly referred to as mechanically-induced
19 martensite [1]. At low plastic strains, *strain-induced* martensite transforms
20 according to Eq. 2; however, at high plastic strains when the hcp phase has
21 been depleted, bct martensite will transform directly from the fcc austenite
22 [2, 4, 5]. This understanding is based on observations of ϵ martensite in
23 low carbon steels at low plastic strains, but not at high strains even when α'
24 martensite continues to transform [2, 4]. ϵ martensite nucleation occurs when
25 partial dislocations separate beyond some critical distance and the faulted
26 hcp zone within the austenite matrix becomes thermodynamically stable [3].
27 This transformation is thought to occur when shear bands containing a high
28 stacking fault density intersect other slip bands, grain boundaries, or twin
29 boundaries [2, 5, 6, 7]. At higher levels of plastic strain, α' martensite has

30 been observed to transform directly from austenite in low carbon steels from
31 dislocation pileups on $\{111\}\gamma$ planes [8] and at shear band intersections [9].
32 Factors that influence austenite stability are chemical composition (alloying
33 additions) and temperature, and factors that affect the extent and rate of
34 strain-induced transformations are strain rate, stress state, grain size, grain
35 orientation, stacking fault energy (SFE), and degree of plastic strain [2, 3, 10].
36 Mechanically-induced martensite has been observed to form under applied
37 stresses in metastable stainless steels such as 304L and increases mechanical
38 strength due to the high strength of martensite.

39 Multiple studies have observed the evolution of strain-induced marten-
40 site in uniaxially loaded 304L stainless steel [3, 11, 12, 13], but the extent
41 to which of these transformations occur in additively manufactured (AM)
42 304L stainless steel (SS) at room temperature remains poorly quantified.
43 AM is a rapidly developing processing pathway due to its ability to produce
44 end-use parts with complex geometries that would otherwise be impossible
45 or very expensive and time consuming to make with traditionally cast or
46 wrought alloys [14]. Despite the weld-like microstructures commonly seen
47 in AM materials, the quasi-static mechanical properties of these alloys often
48 compare favorably to their cast or wrought counterparts [15, 16]. As a result,
49 considerable research is being conducted to establish empirical connections
50 between AM processing parameters, microstructure, phase composition, and
51 their mechanical properties [15, 16, 17, 18, 19, 20, 21, 22]. The current work
52 focuses on understanding how different initial microstructures produced by
53 AM and subsequent heat treatments affect quasi-static mechanical properties
54 with respect to martensite transformations during deformation in a 304L-SS

55 fabricated via laser powder bed fusion.

56 Over the past two decades, *in situ* high-energy X-ray diffraction tech-
57 niques to monitor microstructural evolution in materials during deformation
58 have been widely used [23, 24, 25]. In particular, the ability to accurately
59 model Bragg peak intensity and breadth through crystallographic texture,
60 crystallite/grain size, and lattice parameter gradient (microstrain) measure-
61 ments in low weight fraction phases during deformation has enabled the *in*
62 *situ* quantification of material phase composition. In this study, *in situ* high-
63 energy X-ray powder diffraction measurements were performed to monitor
64 the phase transformation in AM-304L-SS as a function of imposed macro-
65 scopic compressive strain. Compression samples were fabricated from one of
66 two regions in a plate of this steel, and with one of two orientations with
67 respect to the AM build direction. The intent was to test for mechanical
68 heterogeneity and anisotropy, respectively, and see if there was significant
69 influence on martensite evolution. Moreover, samples were heat treated to
70 observe the effect of different initial microstructures on martensite evolution.
71 Electron backscatter diffraction (EBSD) images of samples pre and post de-
72 formation were taken to measure local crystallographic misorientations to
73 quantify crystallographic recovery and static recrystallization. The results
74 are also intended to direct future model development of strain-induced phase
75 transformations in 304L-SS. Combining polycrystal plasticity modeling with
76 experimental datasets containing a materials phase evolution during defor-
77 mation can offer profound insight into the deformation micro-mechanics of
78 the observed system [16, 26, 27, 28, 29, 30].

2. Materials and methods

2.1. Sample preparation

Detailed descriptions of the additive manufacturing of the AISI 304L grade SS studied here and the baseline wrought materials are reported elsewhere [31, 32, 33, 34], and only a brief description of the AM build process will be given here. An AM-304L-SS plate ($86 \times 50 \times 10 \text{ mm}^3$) was deposited using the laser powder bed fusion (LPBF) method with the starting powder size ranging between $15\text{-}45 \mu\text{m}$. The LPBF processing parameters used were the Electro Optical Systems (EOS M290) [35] developed PH-1 $20 \mu\text{m}$ settings and the build pattern comprised of layers deposited at 67° from each other. The initial state characterization of these materials is also reported elsewhere [32, 34].

Schematics of the AM-304L-SS plate, locations, and orientations of the compression specimens are shown in Fig. 1(a). All AM samples used in this study were obtained from one of two excised blocks (B2 and B7) that originated from the top and bottom of the as-built AM plate (this varied block height above the substrate) and is indicated in Figure 1(a). A set of compression samples with dimensions $2 \text{ mm} \times 1.9 \text{ mm}$ (sides) $\times 4 \text{ mm}$ (height) were fabricated from the steel plate using electro-discharge machining (EDM). The specimens were machined such that their loading axis aligned with either the build direction (BD) or the through-thickness direction (TD). The wrought compression sample had an identical geometry as the AM samples, where the loading axis (the elongated axis) parallel to the rolling direction is referred to as the axial direction, and the other two axes are in-plane (IP) axes.

In situ compression samples obtained from two separate regions of the

104 AM-304L-SS plate were used to test for material heterogeneity and mechan-
105 ical uniformity. Samples originated from the same region of the AM plate,
106 but with different loading axes with respect to the AM build direction were
107 used to test the influence of texture on martensite evolution and mechanical
108 anisotropy. To study the effect of microstructure on martensitic transforma-
109 tion, AM samples originated from the same region of the AM plate and with
110 identical loading axes with respect to the AM build frame were heat-treated
111 at different temperatures. Prior to loading, these samples underwent one of
112 several heat treatments. Each sample was heated in an argon atmosphere to
113 either 850°C, 950°C, 1000°C, 1100°C, 1200°C, or 1300°C and held for 1 hour
114 at temperature and then air cooled. The post-processing heat-treatment
115 created samples with varying microstructures and therefore different initial
116 conditions for compression.

117 2.2. *In situ high-energy X-ray diffraction during compressive loading*

118 The schematic of the experimental setup is shown in Fig. 1(b). *In situ*
119 high-energy X-ray powder diffraction experiment was performed at the Ad-
120 vanced Photon Source (APS) 1-ID-E beamline. The X-ray energy was 80.725
121 keV (wavelength of 0.015359 nm), the sample to detector distance was ap-
122 proximately 650 mm, and the size of the incident beam was 0.1 mm $\times$ 0.1
123 mm. The instrument was calibrated using a standard CeO₂ specimen [36].
124 Two-dimensional WAXS patterns were converted into intensity vs. d-spacing
125 using GSAS-II [37]. Monotonic, displacement-controlled compression tests
126 were carried out at room temperature at a strain rate of $4 \times 10^{-4} \text{ s}^{-1}$ using an
127 MTS servo-hydraulic load frame. The wide angle X-ray scattering (WAXS)
128 data were collected on a single panel silicon-based integrating area detector

129 [38]. Both the WAXS and the MTS load frame data acquisition rates were
130 set to 10 Hz, making it possible to obtain WAXS data as a function of sample
131 displacement.

132 2.3. Microstructural characterization

133 Samples were prepared for scanning electron microscopy by grinding with
134 silicon carbide (SiC) abrasive paper, beginning with medium grits and end-
135 ing with fine grit. Subsequent mechanical polishing took place using a 5:1 by
136 volume mixture of an aqueous 0.3 μm alpha alumina (Al_2O_3) suspension and
137 concentrated hydrogen peroxide (H_2O_2). Mechanical preparations concluded
138 with a final polish of a 5:1 by volume mixture of an aqueous 0.04 μm colloidal
139 silica (SiO_2) suspension and concentrated H_2O_2 . Just prior to characteriza-
140 tion, samples were cleaned and coated with a carbon-based conductive paint.
141 EBSD patterns were collected using a ThermoFisher ScientificTM Apreo scan-
142 ning electron microscope (SEM) with an accelerating voltage of 20 kV and
143 step size of 0.75 μm . Scan area was 500 x 500 μm^2 per sample and provided
144 information on crystallographic texture and grain morphology in the as-built
145 and heat-treated material for both initial and deformed specimens.

146 EBSD was performed on all samples to collect crystallographic orienta-
147 tion data from which sample microstructures were reconstructed and image
148 quality (IQ), kernel average misorientation (KAM), grain orientation spread
149 (GOS), and crystallite size information were calculated. EBSD data was an-
150 alyzed using the EDAX TSL Orientation Imaging Microscopy (OIM) Anal-
151 ysis software in addition to the MTEX [39] toolkit in MATLAB. EBSD data
152 with a confidence index (CI) less than 0.1 was discarded due to those values
153 being associated with low orientation indexing accuracy [40]. Additionally,

154 a grain and a crystallite are defined as a group of 5 or more pixels with
155 less than 5° and 0.75° of crystallographic misorientation with each other,
156 respectively. This definition incorporates both sub-grains and grains while
157 preventing unintentional crystallite classification that could result from in-
158 tragranular orientation gradients, indexing errors, etc. Crystallites bordering
159 the edge of the EBSD scans were not incorporated into crystallite size calcu-
160 lations. Pole figures were also constructed from EBSD data, but it should be
161 noted that scans of the AM samples heated at 1200°C and 1300°C , as well
162 as the wrought sample, were not reported because the mapped area did not
163 include enough grains for statistically relevant pole figure calculations.

164 2.4. X-ray data analysis

165 A step-by-step guide to the phase analysis performed in GSAS-II us-
166 ing *in situ* X-ray diffraction data is provided in Appendix A, and only a
167 brief description of the procedure exists here. In order to determine the
168 lattice parameters, microstrain (lattice parameter gradient contributing to
169 peak broadening), and crystallographic texture (12^{th} order spherical har-
170 monics) for the γ phase in each sample, a series of Rietveld refinements was
171 performed on the deformed X-ray diffraction datasets. This was not done
172 for ϵ and α' phases because all deformed AM-304L-SS datasets had low in-
173 tensity peaks corresponding to the martensite phases, which were difficult
174 to fit robustly during Rietveld analysis [41]. The deformed wrought mate-
175 rial, however, had higher intensity peaks corresponding to the martensite
176 phase; therefore, microstructural parameters for the martensite phase were
177 determined by refining the deformed wrought 304L-SS dataset, which were
178 then applied to all other datasets. Mean crystallite/grain sizes were also de-

179 terminated through refinement of the deformed wrought dataset for ϵ , and α'
180 while γ crystallite/grain sizes were measured in each sample via EBSD. Sub-
181 sequent sequential Rietveld refinements were performed for each *in situ* test
182 where the diffraction histogram corresponding to each X-ray exposure was fit,
183 sequentially, in GSAS-II. These refinements incorporated γ , ϵ , and α' phases
184 and calculated phase weight fractions for each histogram. The martensite
185 and austenite lattice parameters and crystallite sizes (also martensite mi-
186 crostrain) were fixed for each phase during the sequential refinements, while
187 γ microstrain, γ hydrostatic strain (to account for any net change in lattice
188 parameter during deformation), γ texture, and phase fractions were refined.

189 Atomic displacement parameters, (often referred to as Debye-Waller fac-
190 tors or B_{iso}) which describe the attenuation of X-ray scattering caused by
191 thermal atomic motion, were based on values calculated by Peng et. al in
192 [42] and fixed in all refinements. This assumes that the crystallite sizes
193 and atomic displacement parameters for each martensite phase are identi-
194 cal across all samples and do not evolve under macroscopic elastic or plastic
195 strains. This is a reasonable assumption for undeformed martensite because
196 the mechanisms causing it to transform produce crystallites with a specific
197 morphology [7]. Additionally, dynamic recrystallization, which would alter
198 crystallite size, is unlikely in any phase because mechanical tests were only
199 carried out to small total plastic strains. Refining crystallite size is also
200 unnecessary because small changes in value would have a negligible effect
201 on peak breadth given the length scale of crystallites in the material. B_{iso}
202 will rise with increased elastic strain [43], but was not refined because slight
203 changes in its value would also have little effect on peak intensities at high

d-spacings. Moreover, because the martensite phases have low-intensity or absent peaks at low plastic strains, which are difficult to fit, simultaneously refining B_{iso} , crystallite size, microstrain, and texture would not have resulted in converged values [41]. Profile fitting uncertainty is calculated during each refinement in GSAS-II and is reported with phase weight fractions. These values are large for the 1200°C and 1300°C sample datasets due to large grain size, which resulted in under-sampled, suboptimal diffraction patterns with low signal/noise that were difficult to fit using the Rietveld method. These two datasets were plotted using moving averages because of the high degree of noise.

3. Results

3.1. Mechanical Response

Macroscopic true stress versus true strain curves for specimens loaded under uniaxial compression are shown in Fig. 2. Tests were performed until samples reached a maximum true strain of approximately 11%. Fig. 2(a) shows compression curves that correspond to samples originating from blocks B2 and B7 both with their loading axes oriented along BD. Both samples exhibit yield strengths (YS) of about 450 MPa and nearly identical flow stresses. This was also seen in a study by Wang et al. who observed similar yield and ultimate tensile strengths for BD and TD samples in AM-304L-SS produced by directed energy deposition (DED) [20]. The compression curve corresponding to a sample originating from block B2, but with its loading axis along the through-thickness axis (TD) is also shown. Although this sample shares a YS with the former two (~ 200 MPa greater than the

228 wrought material), it has a smaller hardening rate resulting in a lower flow
229 stress, which begins to manifest around 5% true strain. However, because
230 these curves each represent a single *in situ* test, it is likely this flow stress
231 discrepancy is statistical scatter.

232 Fig. 2(b) shows the macroscopic true stress-strain responses during com-
233 pressive loading of AM samples that were subjected to different HTs prior to
234 testing. The compression samples were all cut in the same orientation with
235 respect to the AM build frame (BD parallel to loading). All HT samples
236 had lower YS and flow stress than the as-built AM-304L-SS. The highest
237 YS of the HT samples (approximately 375 MPa) corresponds to the sample
238 held at the lowest temperature during HT (850°C), and the lowest YS (~125
239 MPa) corresponds to the sample held at the highest temperature (1300°C).
240 Moreover, increasing or decreasing the HT soak temperature by 100-150°C
241 resulted in a respective drop or gain in YS by ~50 MPa. The one exception
242 is the nearly 150 MPa difference in YS between the AM-304L-SS heated at
243 1100°C and 1200°C. Also of note is that the wrought material had a YS (250
244 MPa) between that of the AM samples HT at 1100°C (300 MPa) and 1200°C
245 (150 MPa). Concerning flow stress, the hardening rate of the 850°C sample
246 is like that of the as-built material, but samples heated at or above 950°C
247 had slightly higher hardening rates, which were more similar to that of the
248 wrought material.

249 3.2. Phase evolution

250 Fig. 3 shows X-ray diffraction profile corresponding to a single exposure,
251 both prior to loading and at a compressive strain of 11% for *in situ* tests
252 performed on wrought, as-built AM, and HT AM 304L samples. The profile

253 corresponding to the undeformed material show no evidence of martensite in
 254 the initial state (true strain = 0%), apart from the wrought material, which
 255 displays low-intensity α' reflections. However, profile in Fig. 3 (true strain
 256 = 11%), which corresponds to the deformed material, display both ϵ and α'
 257 reflections for all tested samples with the greatest ϵ and α' peak intensities
 258 being in the wrought data. This observation in PBF 304L-SS is different
 259 from studies by Brown et al. [31] and Wang et al. [20], where strain-induced
 260 martensite was not observed in deformed DED 304L-SS. Two-dimensional X-
 261 ray diffraction patterns for the deformed wrought and AM as-built material
 262 are also provided in Appendix B, which clearly shows continuous intensity
 263 contributions from both austenite and martensite phases present in the de-
 264 formed steels.

265 The martensite evolution during the *in situ* testing of **as-built** samples
 266 manufactured from different blocks with varied loading axis orientations (B7-
 267 BD, B2-BD, B2-TD) are shown in Fig. 4. The initial and final phase weight
 268 percentages are shown in Table 1. All as-built samples began *in situ* testing
 269 with no detectable ϵ or α' martensite, whereas the wrought sample had no
 270 detectable ϵ martensite, but consisted of 0.26 wt. % α' martensite. Similar
 271 to the true stress-strain response, there is negligible difference in material
 272 behavior between the two BD samples cut from the same block; both have
 273 indistinguishable discrete ϵ and α' martensite volume percentages, relative
 274 to their respective uncertainties, during loading. The sample from block B2
 275 with loading axis along TD, however, evolved more ϵ and α' martensite at
 276 11% applied macroscopic strain than the sample from block B2 with loading
 277 axis along BD. The former had final ϵ and α' weight percentages of 4.21%

278 and 1.26%, whereas the latter had 5.44 % and 1.95 %, respectively. The
279 wrought material had ϵ and α' martensite weight percentages of 7.01% and
280 2.80%, respectively, at 11% applied macroscopic strain.

281 Since B2 and B7 samples showed close to identical macroscopic load-
282 ing and martensitic transformation behaviors, only heat-treated samples ob-
283 tained from the B2 block were used for *in-situ* compressive loading measure-
284 ments. Fig. 5 shows martensite evolution as a function of strain for HT AM
285 samples that underwent *in situ* testing. The initial and final phase weight
286 percentages of the martensite phases are shown in Table 2. Similar to previ-
287 ous tests, no martensite was detected in undeformed AM material. Weight
288 percentages of ϵ and α' at 11% applied macroscopic strain were 2.87% and
289 0.45% in the AM sample heated to 850°C and final ϵ quantity was similar in
290 the deformed 950°C sample while α' was slightly lower. The 1100°C sample
291 had the lowest weight percentages of final ϵ and α' martensite at 2.00% and
292 0.23%, respectively. The 1200°C sample, however, at 11% applied macro-
293 scopic strain had increased levels of α' almost to that of the as-built sample
294 and increased levels of ϵ to similar levels as the 850°C and 950°C samples.
295 The 1300°C sample had an even higher weight percent of α' (5.83%), which
296 was between that of the as-built and wrought samples at 11% applied macro-
297 scopic strain.

298 4. Discussion

299 4.1. Effect of crystallographic recovery on martensite evolution

300 A **qualitative** microstructural analysis with emphasis on KAM, austenite
301 GOS, and austenite crystallite size was performed on undeformed samples

302 to provide insight into the extent to which crystallographic recovery and re-
 303 crystallization occurred in the undeformed 304L samples during HT. This
 304 allowed for correlations amongst recovery, recrystallization, and martensite
 305 evolution to be made. High KAM often highlights the presence of disloca-
 306 tion structures within grains [44], which form when materials with relatively
 307 high dislocation content and high SFE undergo a period of crystallographic
 308 recovery. During recovery, dislocations rearrange themselves to reduce total
 309 lattice energy by creating slightly misoriented sub-grains [45]. Therefore, a
 310 decrease in crystallite size will also be an indicator of recovery.

311 Mean KAM and mean crystallite size measurements are shown in Table
 312 3 while low angle ($<5^\circ$ misorientation) sub-grain boundaries are overlaid in
 313 red on GOS maps in Fig. 6 for all undeformed samples. Mean crystallite
 314 sizes were $23.51\ \mu\text{m}$ and $16.13\ \mu\text{m}$ for the as-built and 1000°C AM samples,
 315 while mean KAM values were 0.563° and 0.738° , respectively. This increase in
 316 mean KAM and decrease in mean crystallite size, combined with an observed
 317 increase in sub-grain boundaries as seen in Fig. 6, is strong evidence of
 318 crystallographic recovery occurring as a result of heat-treatment.

319 It has been reported that the decrease in crystallite size as a result of
 320 recovery would decrease the chance of shear band formation and would
 321 lower stacking fault probability[46] and would therefore reduce strain-induced
 322 martensite nucleation [2, 47, 48]. This was observed during *in situ* compres-
 323 sion. Martensite transformation rates were high in the as-built sample, lower
 324 in the 850°C and 950°C AM samples, and lowest in the 1100°C AM sample
 325 (see Fig. 5). Note that the sample heated at 1000°C was not measured during
 326 *in situ* loading experiment at the beamline but was used for EBSD measure-

327 ments, due to lack of samples heat treated at 850°C and 950°C. The AM
328 sample heated at 1100°C had lower mean KAM (0.630°) and greater mean
329 crystallite size (20.97 μm) than the AM sample heated at 1000°C because it
330 had begun to recrystallize.

331 4.2. Effect of recrystallization on martensite evolution

332 Both ϵ and α' martensite began transforming at lower plastic strains and
333 at higher rates in the wrought sample than any AM sample, as seen in Fig.
334 5. Moreover, strain-induced martensite began transforming at higher plas-
335 tic strains and at slower rates with increased HT temperature until 1200°C
336 when rates began rising again. This rise is coincident with the recrystal-
337 lization of the AM microstructure in samples heated to 1200°C and 1300°C.
338 Recrystallization is evident due to the decrease in mean GOS, which is shown
339 in Table 3. GOS is a measure of the range of crystallographic orientations
340 within a grain and often highlights the presence of recrystallized grains [48].
341 Mean GOS values remain high for as-built, 1000°C, and 1100°C samples,
342 but drop significantly for 1200°C and 1300°C specimens. Newly recrystal-
343 lized grains are strain-free with little-to-no orientation gradient, have low
344 dislocation content, and if given sufficient time at elevated temperatures can
345 coarsen significantly. Such grains are seen in Fig. 6 with dark blue coloring,
346 corresponding to a low GOS. Due to the presence of strain-free grains and
347 lack of sub-grain dislocation structures, it is clear the 1200°C and 1300°C
348 microstructures fully recrystallized while the 1100°C microstructure did so
349 partially. This also explains the slight drop in mean KAM and GOS between
350 1000°C and 1100°C and the large drop between 1100°C and 1200°C. More-
351 over, it explains the slight increase in mean crystallite size between 1000°C

and 1100°C and the large increase between 1100°C and 1200°C. Because an increase in austenite crystallite size increases the probability of strain-induced martensite nucleation (due to increased chance of shear band and stacking faults formation) [2, 46, 47], we see an increase in the rate of martensite transformation during loading in recrystallized microstructures.

Concerning the wrought material, martensite transformation rate is higher than the non-recrystallized AM samples primarily due to having a much larger mean crystallite size. The difference in martensite evolution between recrystallized AM material and the wrought sample is also likely in part due to a larger wrought crystallite size; however, differences in crystallographic texture and solute homogeneity between AM and wrought material may also play a role.

4.3. Effect of crystallographic texture on martensite evolution and mechanical strength

As-built AM samples originating from the same block (B2) of material were machined with their loading axes parallel to the build (BD) and transverse (TD) directions. It was found that while both samples exhibited similar YS, the flow stress was slightly higher in the TD sample at true strains >5%. Additionally, both ϵ and α' martensite transformed at higher rates and resulted in larger final weight percentages in the TD sample versus the BD sample. The orientation relationship between austenite and incipient martensite adjacent lattices is $\{111\}\gamma \parallel \{0001\}\epsilon$ for γ and ϵ crystals and $\{001\}\gamma \parallel \{001\}\alpha'$ for γ and α' crystals [13, 49]. Additionally, the orientation relationship between ϵ and incipient α' crystals is $\{0001\}\epsilon \parallel \{110\}\alpha'$ [50]. This implies that if the austenite has a bulk crystallographic texture, then

so would the transformed martensite. Inverse pole figures showing α' and ϵ martensite texture calculated from X-ray diffraction patterns (post-loading) are shown in Appendix C. Moreover, it has been reported in other works [11, 30, 45] that while under compressive load, the γ to α' transformation via intermediate ϵ stacking faults is facilitated in 304L-SS when austenite grains are oriented with 001γ plane-normal to the axis of loading. This is made possible due to the crystallographic orientation relationships amongst these phases [13, 50, 51].

As seen in Fig. 7, the pole figures constructed from EBSD data of the undeformed as-built, 1000°C, and 1100°C AM material have a $\{110\}\gamma$ fiber, but virtually no $\{001\}\gamma$ texture components parallel to BD. This is indicated by the spike in pole density, represented as multiples of random distribution (m.r.d.), in the center of the as-built $\{110\}\gamma$ pole figure and the low pole densities in the centers of the as-built $\{001\}\gamma$ pole figures. $\{110\}\gamma$ fibers have been observed before in AM-304L-SS [16]. Additionally, 90° from BD (in all directions) in the $\{001\}\gamma$ pole figures is a ring component. This ring has a lower associated m.r.d. than the $\{110\}\gamma$ fiber along BD, but it does indicate some degree of preferred orientation where $\{001\}\gamma$ poles are along TD. This preferred orientation can also be seen in the undeformed austenite inverse pole figures in Appendix C. Therefore, due to the lack of grains with $\{100\}\gamma$ poles oriented along BD, as indicated by the ~ 0 m.r.d., and the relatively high presence of grains (2 to 3 m.r.d.) with $\{100\}\gamma$ poles oriented along TD, it is reasonable to conclude that the as-built TD sample evolved more martensite than the BD sample during compressive loading due to initial crystallographic texture. More precisely, the alignment of TD with the

loading axis was favorable, with respect to the crystallographic texture of the AM-304L-SS, and resulted in higher quantities of strain-induced martensite during deformation as described by Eq. 2.

4.4. *Effect of martensite evolution and heat-treatments on mechanical strength*

The effect martensite transformations had on the flow stress observed in samples is seen to be minimal if at all. YS decreases continually in AM samples with increasing HT temperature, similar to flow stress, and is unaffected by strain-induced martensite transformations, which occur after the onset of plasticity. This was also observed in a study by Brown et al.[31] who posited that the decreases in the strength of a HT AM-304L-SS fabricated via DED were due to decreases in dislocation density resulting from heat treating at higher temperatures. Brown et al.[31] also observed a significant drop in the strength of AM-304L-SS heated above 1100°C, where the as-built AM grain morphology transformed into an equiaxed structure after recrystallization, similar to what was observed in this work [31]. Crystallographic recovery reduces the dislocation content in a material by mobilizing dislocations, which annihilate at matrix defects and minimize lattice energy [52]. Recrystallization implies nucleation and growth of low dislocation density grains, which if grains become coarse will also contribute to softening according to the Hall-Petch relation [52, 53]. Therefore, the steady decline in YS observed at HT below 1100°C is due to crystallographic recovery, while static recrystallization is responsible for the drop in strength observed in AM-304L-SS heated above 1100°C. The wrought material has a YS (~ 250 MPa) between that of the 1100°C and 1200°C sample likely because of a combination of its large grain size and high dislocation content, relative to the recrystallized

427 AM samples. Similar YS for wrought 304-SS were also observed in studies
428 by Brown et al.[31] and Gray III et al.[32].

429 4.5. *Effect of material heterogeneity on martensite evolution and mechanical* 430 *strength*

431 As-built samples oriented identically with respect to the compression
432 frame (BD along loading axis) were machined from blocks B2 and B7 within
433 a single AM plate. It was found that both samples exhibited near identical
434 true stress-strain responses and phase evolution when loaded in compression.
435 The thermal profile of the as-built plate varied as a function of distance from
436 the build plate during cyclic heating [16, 31, 54]. Typically, material near
437 the build plate cools more rapidly during solidification than further from the
438 built plate. Mechanical uniformity, however, was confirmed due to the similar
439 performance and phase evolution in BD samples originating from the block
440 closest (B2) and farthest from the build plate (B7). Although AM material
441 has localized heterogeneity, at length scales corresponding to sample size the
442 plate material was either homogenous or the heterogeneities did not affect
443 mechanical performance over the range of tests conducted in this work.

444 On sub-grain length scales, it is likely that there was chemical hetero-
445 geneity in the as-built AM material in the form of solute micro-segregation
446 resulting from cellular and/or dendritic solidification where solute atoms
447 concentrated in cell walls or dendrite interfaces [52]. In micro-segregated
448 304L-SS, solute-poor cell/dendrite cores would have higher SFE due to the
449 absence of SFE-lowering elements such as Cr, Si, and Mn, which would have
450 decreased the probability of strain-induced martensite nucleation [55]. There-
451 fore, it is possible that the 1 hour soak time during the AM heat-treatments

was not long enough for significant solute diffusion to take place at temperatures lower than 1300°C HT, which resulted in unchanged, chemically inhomogeneous materials. This potential for solute diffusion along concentration gradients in the 1300°C material could explain the observed increase in martensite transformation rate.

5. Conclusions

This work has examined the effect of sample build orientation and heat-treatment on the evolution of martensite and strength in an AM-304L-SS alloy. In situ high-energy X-ray diffraction was performed during compressive loading to examine martensite evolution as a function of macroscopic strain. Macroscopic stress-strain response was also recorded during in situ tests and EBSD was used to provide insight on austenite grain/crystallite morphology, intragranular crystallographic misorientation, and crystallographic texture. The main conclusions of this study are:

- Static recrystallization of an AM-304L-SS microstructure as a result of heat-treatment increases the rate at which strain-induced martensite transforms during compressive loading. This is primarily due to an increase in mean austenite crystallite size, with respect to the initial microstructure, and facilitates martensite transformation by means of increasing the probability of shear band and stacking fault formation, which raises the number of potential martensite nucleation sites.
- Crystallographic recovery in an AM-304L-SS microstructure as a result of heat-treatment decreases the rate at which strain-induced martensite

transforms during compressive loading. This is due to the formation of low-angle, intragranular dislocation structures during a period of recovery that reduce mean austenite crystallite size. This hinders martensite transformation by reducing probability of shear band and stacking fault formation, which decreases the number of potential martensite nucleation sites.

- Crystallographic texture influenced the rate at which strain-induced martensite formed in as-built AM-304L-SS. Samples with loading axis parallel to BD had lower final weight percentages of martensite than samples with loading axis parallel to TD. This was due to the lack of grains with $\{100\}\gamma$ pole oriented along BD and the relatively high presence of grains with $\{100\}\gamma$ pole oriented along TD.
- Phase composition had no appreciable effect on the flow stress of AM (as-built and heat-treated) and wrought 304L stainless steel. YS decreased for AM samples as HT temperature increased, which was caused by crystallographic recovery below 1100°C and recrystallization above 1100°C.
- At length scales corresponding to compression sample size, the AM-304L-SS plate was either homogenous or the heterogeneities did not affect mechanical performance over the range of tests conducted in this work. It is, however, plausible that on sub-grain length scales, solute heterogeneity created localized regions within the austenite matrix that had high SFE, which lowered the probability of shear banding and consequently strain-induced martensite nucleation.

499 The current work will be complemented with modeling in the future.
500 Complementary neutron diffraction measurements will be used to extract
501 the initial bulk textures for crystal plasticity model instantiation. Along
502 with the martensite phase evolution information, additional analysis of the
503 in situ X-ray data will be performed to extract *hkl* reflection specific lattice
504 strain evolution during compression and will be used for model validation.

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518 Appendix A

519 Phase analysis procedure performed in GSAS-II:

520 *I) CeO₂ calibration*

- 521 1. Import summed two-dimensional (2D) CeO₂ data (diffraction pattern)
- 522 files.
- 523 2. Calibrate → Integrate 2D data (full integration) → 1D → Full Rietveld
- 524 with CeO₂
- 525 3. Save sample, instrument, and image control parameters.

526 *II) 304L 2D data integration*

- 527 1. Import summed 2D data files.
- 528 2. Load CeO₂ image control parameters → Integrate 2D data (full inte-
- 529 gration) → one dimensional (1D) histograms.
- 530 3. Save 1D data.

531 *III) Martensite phase calibration*

- 532 1. Import γ , ϵ , and α' .cif files and last 1D dataset (highest strain with
- 533 most martensite).
- 534 2. Set Biso values for all phases based on the literature.
- 535 3. Set γ crystallite sizes based on EBSD measurements.
- 536 4. Import instrument and sample parameters.
- 537 5. Refine hist scale, background (bk), weight fractions (VF), γ lattice
- 538 constants, γ crystallite size, γ microstrain.
- 539 6. Refine hist scale, bk, WF, γ lattice constant, γ microstrain, sample X
- 540 + Y coordinates.
- 541 7. Refine hist scale, bk, WF, γ lattice constant, γ microstrain.
- 542 8. Refine hist scale, bk, WF, ϵ lattice constants.

- 543 9. Refine hist scale, bk, WF, ϵ lattice constants, ϵ microstrain.
- 544 10. Refine hist scale, bk, WF, ϵ lattice constants, ϵ microstrain, ϵ crystallite
- 545 size.
- 546 11. Refine hist scale, bk, WF, ϵ texture (spherical harmonics=12).
- 547 12. Refine hist scale, bk, WF, α' lattice constants.
- 548 13. Refine hist scale, bk, WF, α' lattice constants, α' microstrain.
- 549 14. Refine hist scale, bk, WF, α' lattice constants, α' crystallite size.
- 550 15. Refine hist scale, bk, WF, α' texture (spherical harmonics=12).

551 *IV) 304L 1D data refinement/analysis*

- 552 1. Import Austenite and Martensite cif files and first steel 1D dataset.
- 553 (a) Import sample and instrument parameter files from CeO₂ calibra-
- 554 tion.
- 555 (b) Add constraint that phase weight fractions sum to unity.
- 556 (c) Manually enter phase parameters that were determined in Step 3.
- 557 (d) Locate the first histogram where ϵ peaks begin forming and man-
- 558 ually fix ϵ WF to zero in all previous histograms. Repeat for α' .
- 559 (e) Refine: bk, histogram scale parameter, γ lattice constant.
- 560 (f) Refine: bk, histogram scale parameter, γ lattice constant, γ mi-
- 561 crostrain.
- 562 (g) Refine: bk, histogram scale, γ microstrain, sample X + Y dis-
- 563 placement.
- 564 (h) Refine: bk, histogram scale, γ microstrain, γ texture (spherical
- 565 harmonics = 12).
- 566 2. Import the rest of the 1D datasets.

- 567 (a) Perform sequential refinement on all profiles with freed parameters.
568
- 569 3. Sequential refinements
- 570 (a) Controls: SVD zero tolerance = 0.0001, Max cycles = 15, copy
571 results to next histogram, begin at last histogram.
- 572 (b) Refine: histogram scale parameter, bk, VF, γ hydrostatic strain,
573 γ microstrain.
- 574 (c) Refine: histogram scale, bk, VF, γ hydrostatic strain, γ microstrain,
575 γ texture (spherical harmonics =12).
- 576 4. Export sequential refinement phase information as csv.

577 Appendix B

578 Fig. 8 shows two-dimensional X-ray diffraction patterns for the de-
579 formed wrought and AM as-built material, where diffraction signals from
580 both austenite and martensite phases are visible. Resolvable γ , α' , and ϵ
581 Debye-rings are labeled at higher d-spacings (toward the center of the diffraction
582 patterns), and all phases appear textured apparent through azimuthal
583 variation in Debye-ring intensities. Note that the intensity scales are different
584 for these diffraction patterns due to varying degree of attenuation used
585 during data acquisition. The intensity scale has arbitrary unit.

586 Appendix C

587 Inverse pole figures (IPFs) for undeformed and deformed γ phase are
588 shown in Fig. 9, and the deformed (IPFs) for the α' and ϵ phases are shown
589 in Fig. 10. From single 2D X-ray diffraction patterns collected before and/or

590 after loading for each sample, orientation distribution functions were cal-
 591 culated using the E-WIMV algorithm in the diffraction software Maud and
 592 refined using the Rietveld method to model the texture of γ , α' , and ϵ phases.
 593 This procedure is explained in further detail in Wenk et al. [56]. No sample
 594 symmetry was assumed during the texture refinements and from the final
 595 orientation distribution functions, inverse pole figures were plotted. Because
 596 no sample rotations were performed about the loading axis during experi-
 597 mentation, it is possible there were many grains in the sampled volume of
 598 material oriented such that they never met the Bragg criteria for diffraction
 599 and were therefore not included in texture calculations. This increases the
 600 likelihood of calculating an orientation distribution function that describes
 601 non-physical texture components. That being said, the as-built austenite
 602 inverse pole figures (undeformed samples) are in good agreement with the
 603 textures reported in [34]. Because of this, we assume the X-ray diffraction-
 604 produced textures to be reasonable approximations. Martensite textures are
 605 not reported for undeformed samples because martensite was either not ob-
 606 served or had too few peaks (and of low intensity) to fit a texture model.
 607 Various 1200°C and 1300°C plots are missing because of low quality powder
 608 data, which made texture refinements infeasible.

609 AM process conditions create a $\{220\}$ fiber-like texture in the as-built
 610 state. This has been previously reported for AM-304L-SS, where the as-built
 611 texture resembled that of the wrought 304L steel subject to compressive
 612 deformation [34]. Heat treatment does not evolve the initial γ texture, at
 613 least up to 1100°C, and the initial features are preserved. Compression of
 614 this austenite phase seems to align the $\{220\}$ crystal planes with the loading

axis and also creates rings in the previously spotty diffraction patterns (as seen in 1200°C and 1300°C samples). We observe these features in the inverse pole figures, but only slightly, because the macroscopic true strain is small ($\sim 11\%$). Because of small macroscopic strain levels, we do not see major γ texture evolution. α' and ϵ textures are products of the strain-induced phase transformations.

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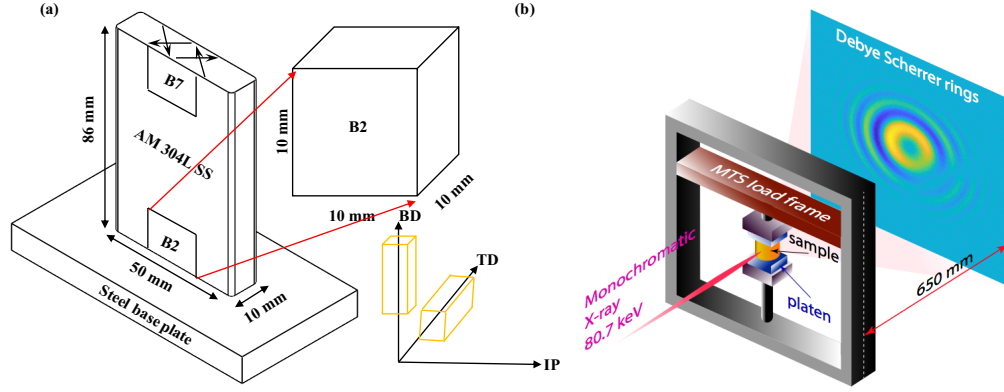


Figure 1: (a) Build geometries of the AM-304L-SS plate and regions/blocks within the plate (B2 and B7) from which compression samples were excised. Arrows on top of the AM-304L-SS plate indicate rotation (67.5°) of the laser path every after every pass. **Sample orientation with respect to build frame is indicated where BD is the build direction, TD is the through-thickness direction, and IP is the in-plane direction.** (b) Schematic of the experimental apparatus for X-ray diffraction measurements during in situ compressive loading. **MTS servo-hydraulic load frame was used, where the sample was held between two die, which exert a compressive force on the sample while allowing unobstructed path for the incident high-energy monochromatic X-rays and outgoing diffracted X-rays.**

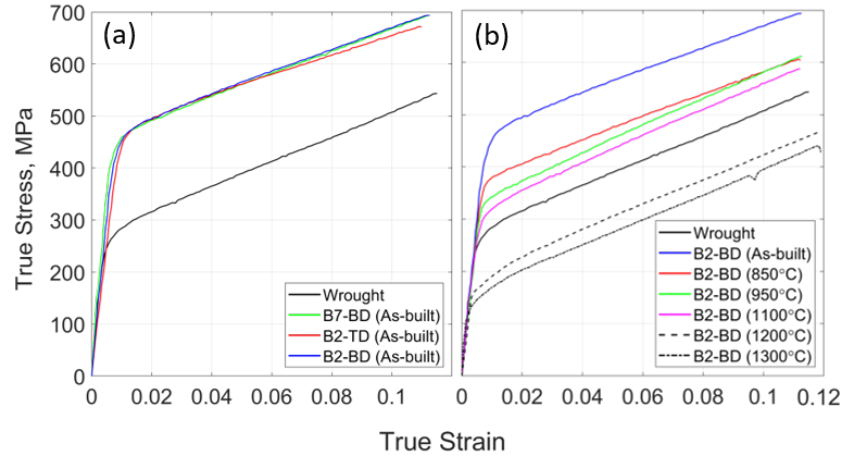


Figure 2: True stress-strain behavior of wrought and AM-304L-SS in simple compression. AM samples were oriented with BD or TD parallel to the loading axis during compression and were cut from one of two different regions (B2 or B7) on an AM plate (a). AM samples underwent one (or none) of several heat-treatments prior to loading, were oriented with BD parallel to the loading axis during compression and were cut from the B2 region on the initial AM plate (b).

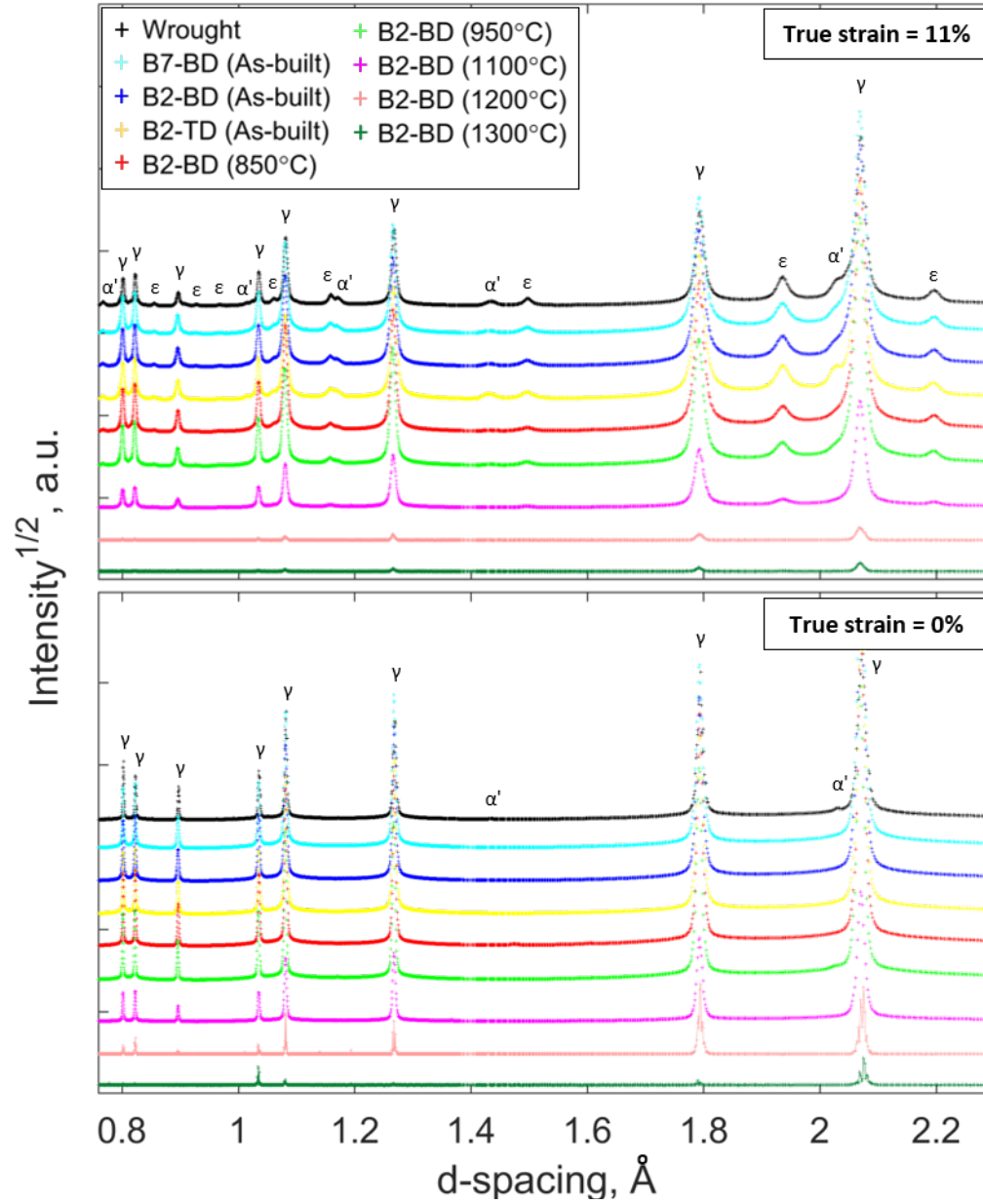


Figure 3: Diffraction profile created from single exposures prior to (true strain = 0%) and during (true strain = 11%) *in situ* loading of wrought, as-built AM, and heat-treated AM 304L stainless steel samples. Bragg reflections for austenite (γ), α' martensite, and ϵ martensite phases are also indicated. Y-axis units are arbitrary (a.u.).

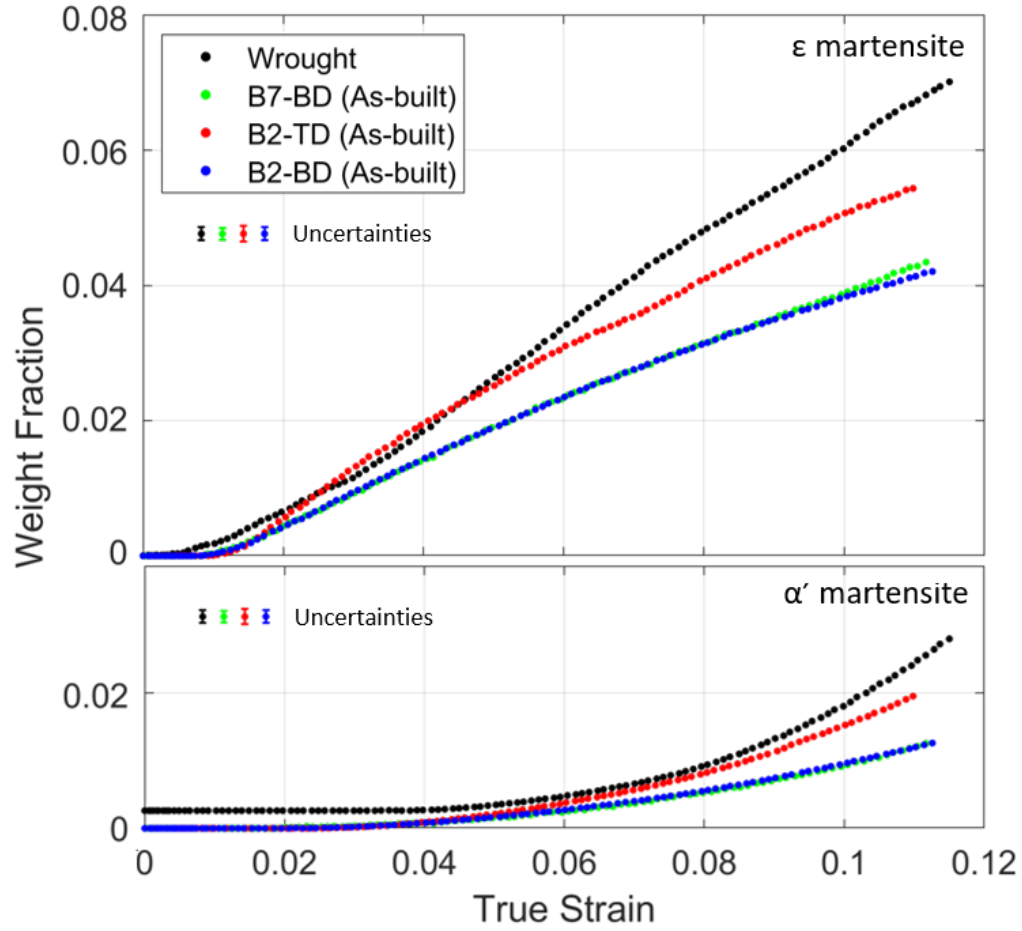


Figure 4: Evolution of ϵ and α' martensite in wrought and AM-304L-SS during monotonic compression as determined by *in situ* x-ray diffraction. AM samples were oriented with BD or TD parallel to the loading axis during compression and were cut from one of two different regions on an AM plate (B2, B7). The error bars below the legend indicate the average uncertainty in each dataset.

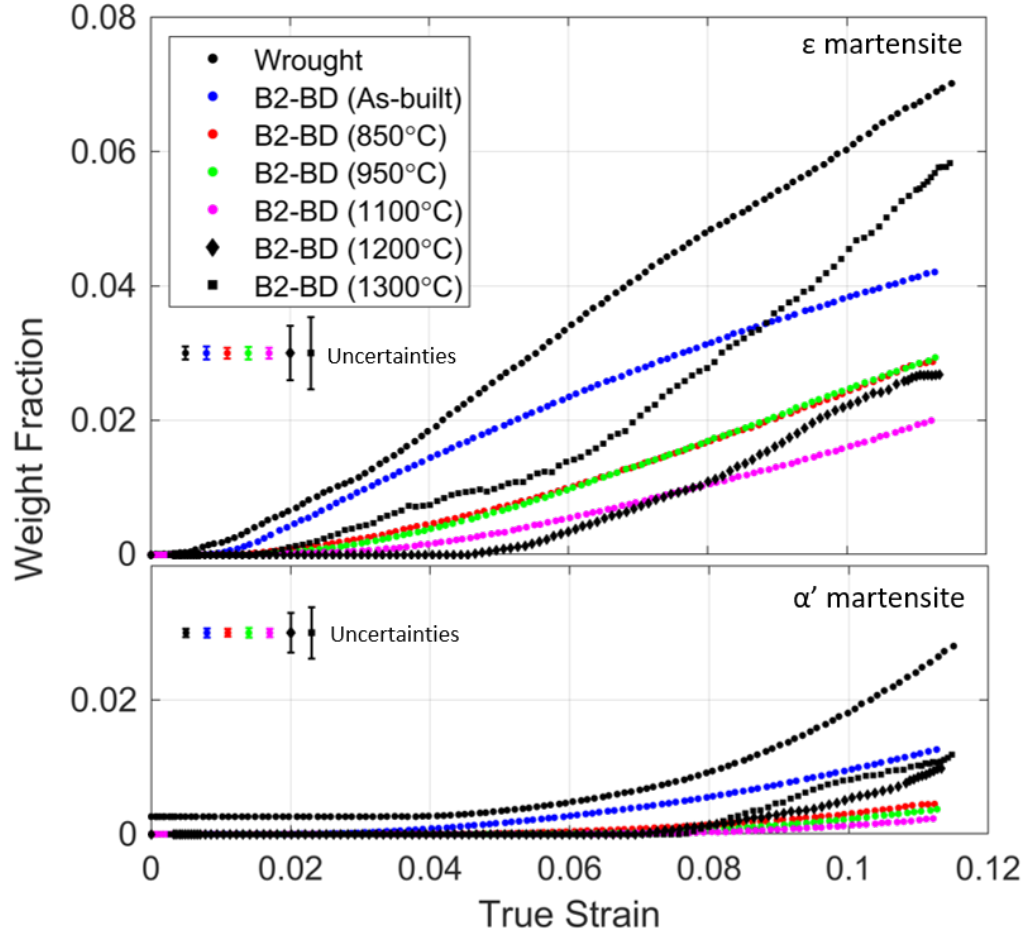


Figure 5: Evolution of ϵ and α' martensite in wrought, as-built-AM and heat-treated-AM 304L-SS during monotonic compression as determined by *in situ* x-ray diffraction. AM samples underwent one (or none) of several heat-treatments prior to loading. The error bars below the legend indicate the average uncertainty in each dataset.

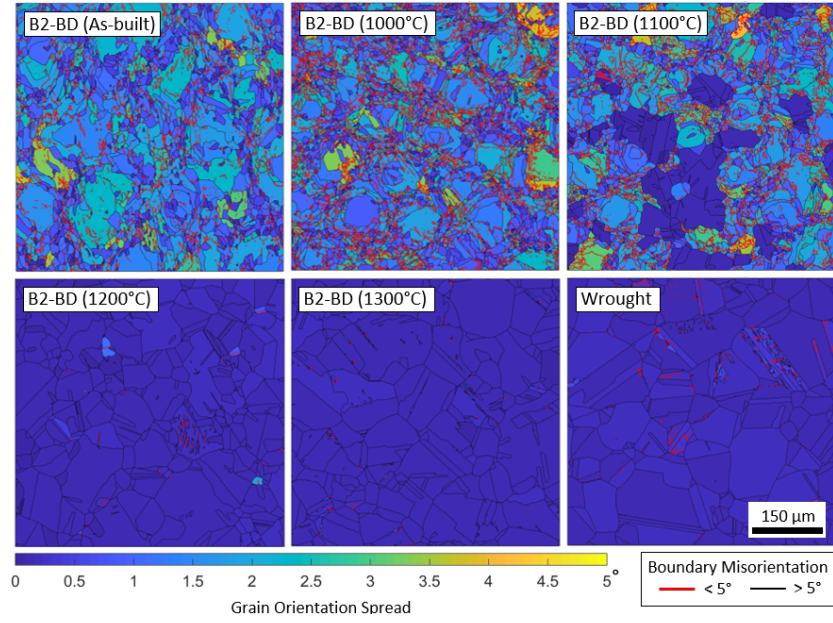


Figure 6: Grain orientation spread (GOS) maps created from EBSD data for undeformed wrought and AM (as-built and heat-treated) 304L-SS samples. Black lines highlight grain boundaries and red lines highlight sub-grain boundaries. All maps have 500 μm view fields, and the axis perpendicular to the maps for the AM sample is BD.

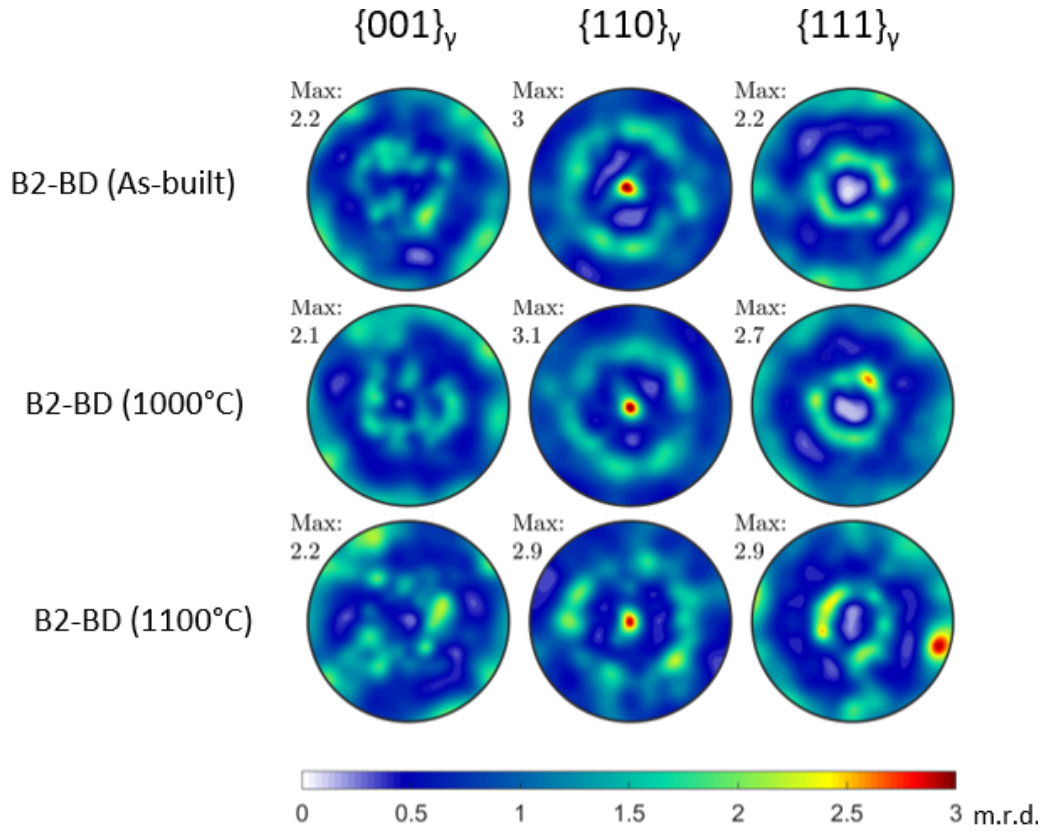


Figure 7: Pole figures created from EBSD data which show austenite texture for selected AM (as-built and heat-treated) 304L-SS samples. BD is out of plane for all pole figures. Intensities are measured in multiples of random distribution (m.r.d.).

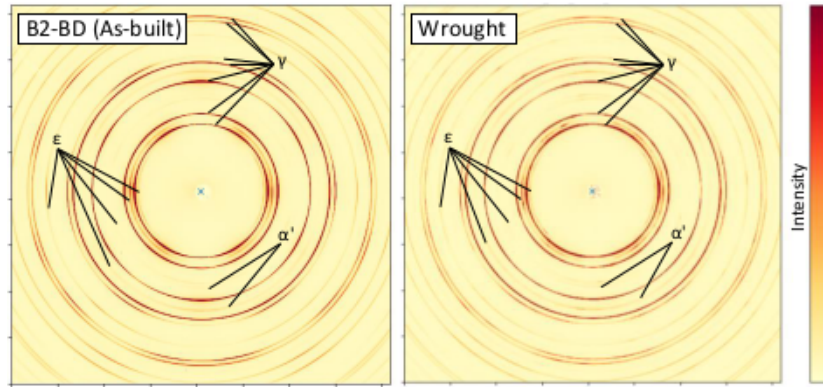


Figure 8: Raw detector images of the as-built (left) and the wrought (right) 304L-SS after 11% compressive strain.

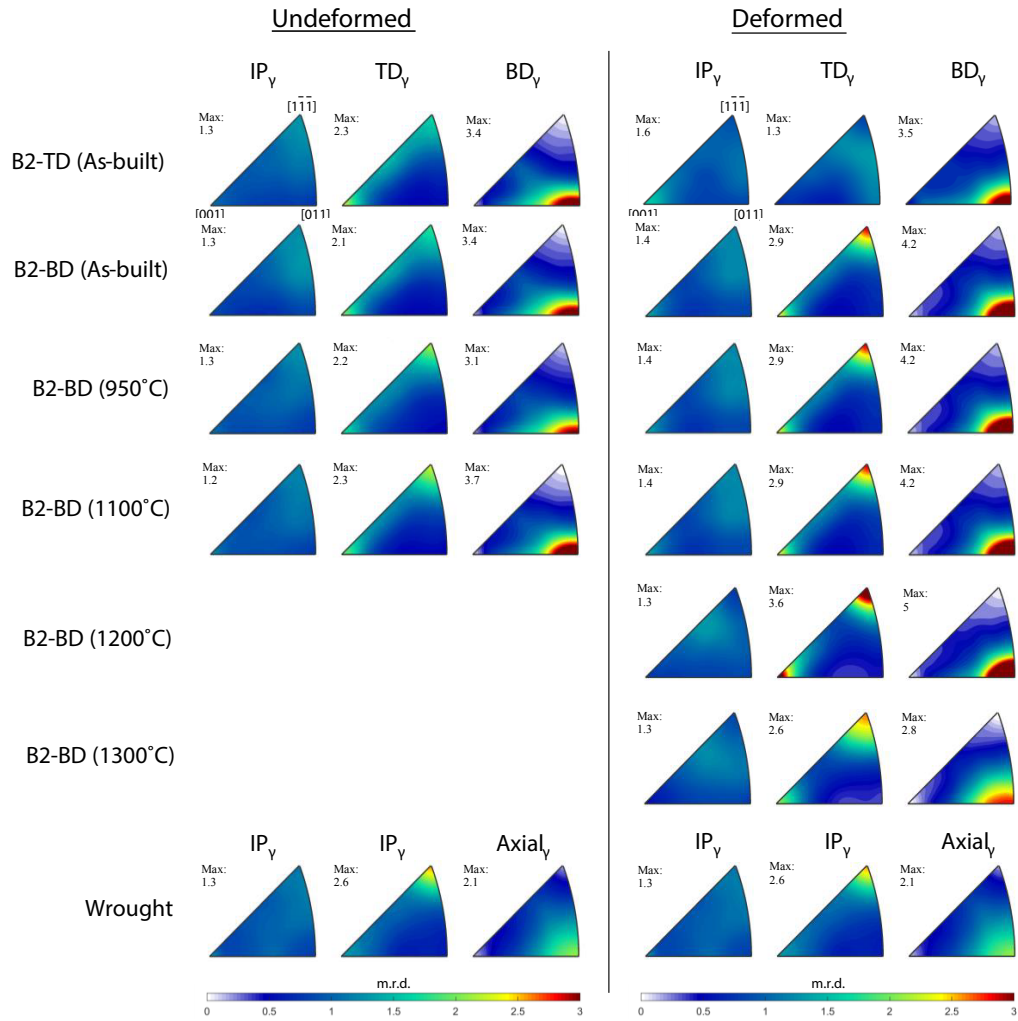


Figure 9: Inverse pole figure for the γ phase of 304L-SS before and after 11% compressive strain.

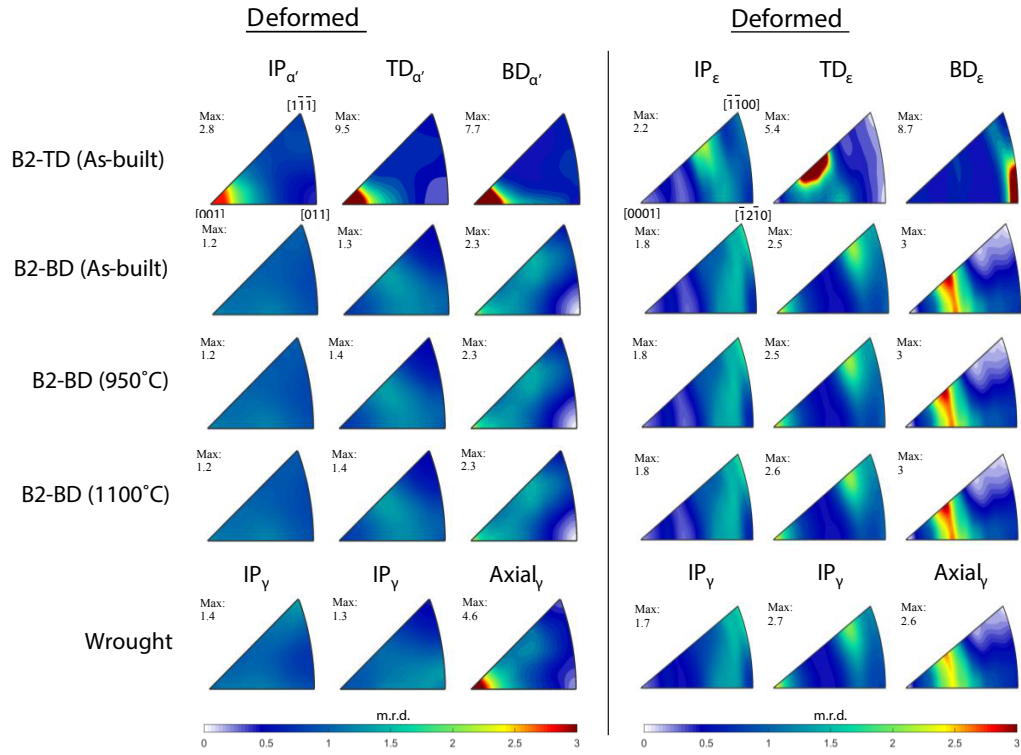


Figure 10: Inverse pole figure for α' (left) and ϵ (right) phases of 304L-SS before and after 11% compressive strain.

Table 1: The initial and final (true strain = 11%) martensite phase compositions as a result of *in situ* compressive loading of wrought and two AM 304L stainless steels. AM samples were oriented with BD parallel or normal (TD) to the loading axis during compression and were cut from one of two different regions on an AM plate (B2, B7). The units are in wt. %. Final phase value is a product of profile fitting uncertainty in GSAS-II, given in parenthesis. Note the phase compositions add to 100% with austenite as balance.

	$\epsilon_{initial}$	ϵ_{final}	$\alpha_{initial}$	α_{final}
Wrought	0.0	7.01 (0.10)	0.26	2.80 (0.06)
B2-BD (As-built)	0.0	4.21 (0.10)	0.0	1.26 (0.07)
B2-TD (As-built)	0.0	5.44 (0.12)	0.0	1.95 (0.08)
B7-BD (As-built)	0.0	4.34 (0.09)	0.0	1.25 (0.06)

Table 2: Initial and final (true strain = 11%) martensite phase compositions as a result of *in situ* compressive loading of wrought and AM (as-built and heat-treated) 304L stainless steel. The units are in wt. %. Final phase value is a product of profile fitting uncertainty in GSAS-II, given in parenthesis. Note the phase compositions add to 100% with austenite as balance.

	$\epsilon_{initial}$	ϵ_{final}	$\alpha_{initial}$	α_{final}
Wrought	0.0	7.01 (0.10)	0.26	2.80 (0.06)
B2-BD (As-built)	0.0	4.21 (0.10)	0.0	1.26 (0.07)
B2-BD (850°)	0.0	2.87 (0.09)	0.0	0.45 (0.06)
B2-BD (950°)	0.0	2.94 (0.10)	0.0	0.37 (0.07)
B2-BD (1100°)	0.0	2.00 (0.09)	0.0	0.23 (0.06)
B2-BD (1200°)	0.0	2.68 (0.43)	0.0	0.98 (0.31)
B2-BD (1300°)	0.0	5.83 (0.55)	0.0	1.18 (0.40)

Table 3: Mean kernel average misorientation (KAM), mean grain orientation spread (GOS) and mean crystallite size for undeformed wrought and AM (as-built and heat-treated) 304L stainless steels. EBSD data was used to reconstruct sample microstructures from which these values were calculated. Standard deviation assuming normal distribution is expressed in brackets.

	<KAM> [°]	<GOS> [°]	<Crystallite size> [μm]
Wrought	0.31 [0.15]	0.32 [0.10]	63.0 [31.4]
B2-BD (As-built)	0.56 [0.53]	3.11 [1.43]	23.5 [19.8]
B2-BD (1000°)	0.74 [0.69]	3.12 [1.49]	16.1 [14.3]
B2-BD (1100°)	0.63 [0.66]	2.75 [1.89]	21.0 [17.4]
B2-BD (1200°)	0.22 [0.10]	0.26 [0.17]	50.1 [26.6]
B2-BD (1300°)	0.22 [0.09]	0.24 [0.05]	57.7 [29.3]