

1 Wire and arc additive manufacturing of HSLA steel: Effect of Thermal Cycles 2 on Microstructure and Mechanical Properties

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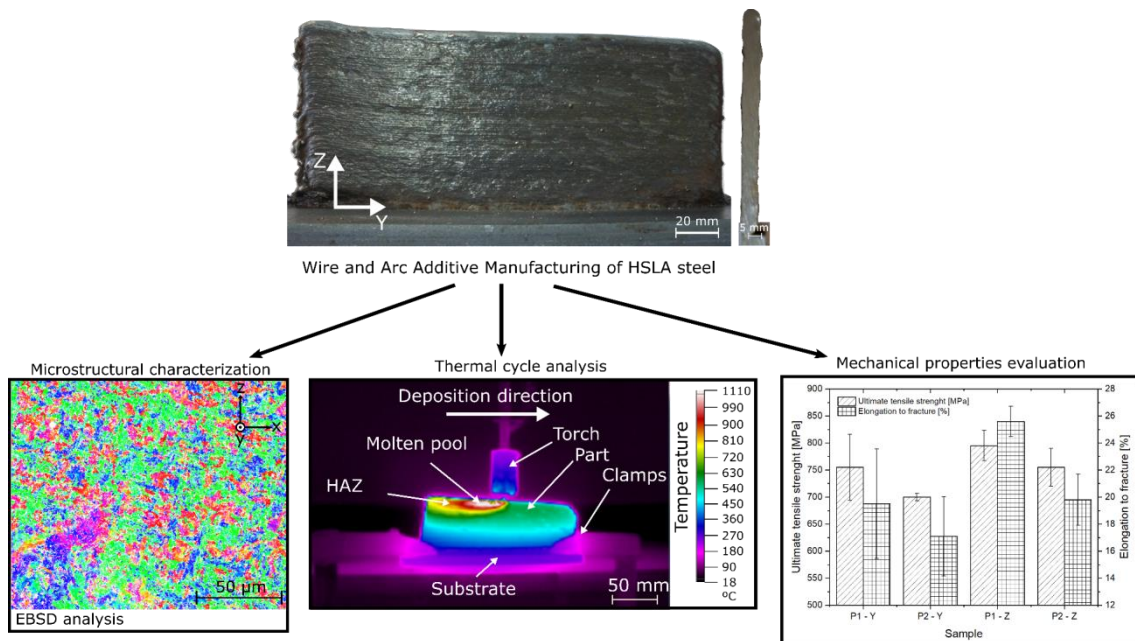
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10 Abstract

11 Wire and arc additive manufacturing (WAAM) is a viable technique for the manufacture of
12 large and complex dedicated parts used in structural applications. High-strength low-alloy
13 (HSLA) steels are well-known for their applications in the tool and die industries and as power-
14 plant components. The microstructure and mechanical properties of the as-built parts are
15 investigated, and are correlated with the thermal cycles involved in the process. The heat input
16 is found to affect the cooling rates, interlayer temperatures, and residence times in the 800–
17 500 °C interval when measured using an infrared camera. The microstructural characterization
18 performed by scanning electron microscopy reveals that the microstructural constituents of
19 the sample remain unchanged. i.e., the same microstructural constituents—ferrite, bainite,
20 martensite, and retained austenite are present for all heat inputs. Electron backscattered
21 diffraction analysis shows that no preferential texture has been developed in the samples.
22 Because of the homogeneity in the microstructural features of the as-built parts, the
23 mechanical properties of the as-built parts are found to be nearly isotropic. Mechanical testing
24 of samples shows excellent ductility and high mechanical strength. This is the first study
25 elucidating on the effect of thermal cycles on the microstructure and mechanical properties
26 during WAAM of HSLA steel.

27 **Keywords:** Wire and arc additive manufacturing; high-strength low-alloy steels; microstructure
28 characterization; mechanical properties; additive manufacturing.

29 Graphical Abstract



30

31 Highlights

- 32 - Wire and arc additive manufacturing of HSLA steel was performed.
- 33 - Microstructure and mechanical properties were related to the thermal cycles.
- 34 - No preferential texture was developed, leading to near-isotropic mechanical properties.
- 35 - As-built parts exhibited excellent ductility and high mechanical strength.

36 1. Introduction

37 Additive manufacturing (AM) technologies allow the creation of complex parts with tailored
38 mechanical properties within short delivery times [1,2]. In the recent years, AM has gained
39 interest for the design and manufacture of prototypes, with the aerospace, automotive,
40 defense, and medical industries adopting these technologies [3].

41 Amongst the different AM technologies used for metallic alloys, wire and arc additive
42 manufacturing (WAAM) exhibits several competitive advantages over the laser and electron-
43 beam technologies, such as low capital investment and high deposition rates, and it has also
44 overcome some of the difficulties associated with processing specific alloys [4]. WAAM can be
45 described as a technology combining an electric arc, used as a heat source, and a wire, used as

46 feedstock material, to produce large and complex parts [5]. It uses the fundamental concepts
47 of automatized or robotized processes and arc-welding processes such as gas metal arc
48 welding, gas tungsten arc welding, or plasma arc welding.

49 Arc-based technologies have been used successfully in AM [5–8]. Even though arc-welding
50 technologies are well known, their use in AM is rather complicated because several
51 phenomena occur simultaneously and a wide range of process parameters must be controlled
52 to fabricate high-quality parts. WAAM is a developing technology, and several challenges
53 remain to be addressed. These include residual stresses and distortions arising from excessive
54 heat input, optimization of process parameters and deposition strategies, poor surface quality
55 of WAAM parts, removal of parts from the substrate, and standardization.

56 Typically, WAAM uses heat inputs ranging from tens to hundreds of J/mm, and this heat is
57 usually dissipated by conduction through the components and substrates, forced convection
58 through the shielding gas, or radiation to the surrounding environment [9]. However, cooling
59 through conduction becomes difficult as the number of layers increases, as the large area of
60 the substrate causes heat dissipation in the layers deposited first, thereby decreasing the heat
61 accumulation [10]. This heat transfer to the already deposited layers is of major concern, as it
62 affects the cooling rate and thermal cycles of both the previously and currently deposited
63 layers, which can lead to microstructural changes along the part. Therefore, there is a need for
64 process add-ons that allow the control of the thermal cycles acting upon the material. One
65 method of achieving microstructural control is based on the inter-layer temperature, which is
66 defined as the temperature of the previously deposited layer upon the deposition of a new
67 layer [11,12]. The interlayer temperature is considered one of the most important aspects
68 related to the surface waviness and temperature distribution of the parts during fabrication;
69 therefore, it should be selected carefully to avoid undesirable contamination from foreign
70 elements belonging to the previously deposited layers [12,13]. However, process control based
71 on interlayer temperature is difficult, as some of the suitable dwell times may be quite long,
72 reducing the deposition productivity, or very small, urging the use of multiple welding torches,
73 for example. Numerical models have been successfully developed to predict the temperature
74 gradients and their distributions along WAAM walls [14] and, more recently, a model that
75 could accurately predict bead geometry was presented [15].

76 In the literature, several techniques have been described to regulate the heat dissipation and
77 thermal cycles during WAAM. Mughal et al. [16] investigated the effects of different deposition
78 sequences on the residual stress distribution, stating that layers should be deposited starting

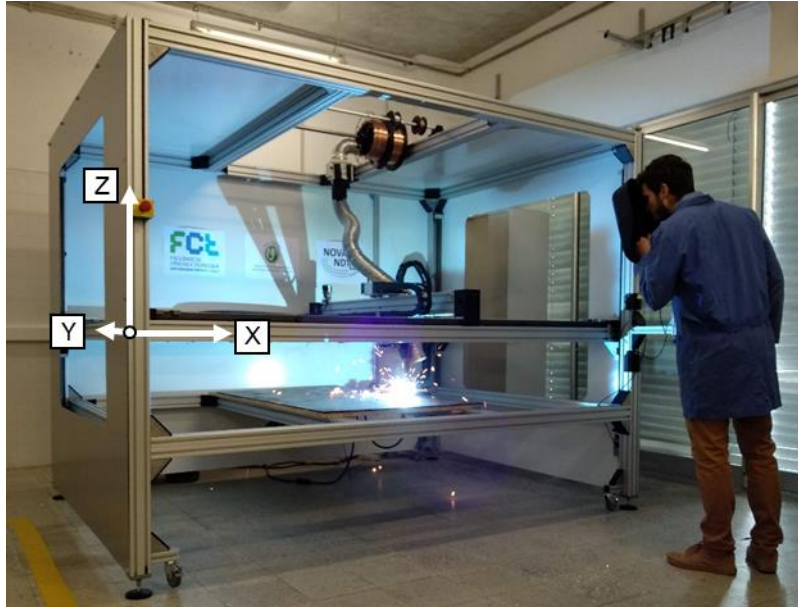
79 from the outside to the center of the part, to reduce potential detrimental effects on the as-
80 built material. Concerning in situ techniques for process control, a finite element analysis of
81 the effect of a coolant hose attached to a welding torch to increase the cooling rate of the
82 underlying beads using air jets was described by Montevicchi et al. [17]. Thermoelectric
83 cooling devices were placed on both sides of the walls being produced, in order to eliminate
84 heat-dissipation differences between the top and bottom layers [18]. A compressed CO₂ jet
85 was used to regulate the interlayer temperature, which was controlled by a pyrometer, to
86 study its effects on bead geometry, surface oxidation, microstructure, and mechanical
87 properties of deposited bead [19]. These types of process developments are beneficial for
88 WAAM, as they can increase the process efficiency and allow the control of microstructure,
89 thereby improving its potential for use in relevant engineering applications.

90 This work studies the mechanical properties, effects of the heat input on the thermal history,
91 and the subsequent changes induced in the microstructures at various locations in the
92 deposited walls. This investigation also points out the role of the complex thermal history in
93 WAAM, highlighting the importance of controlling the different cooling rates throughout the
94 deposition process.

95 2. Materials and methods

96 2.1 Experimental setup

97 The experimental apparatus consisted of a customized welding torch mounted on a three-axis
98 positioning system, with a working envelope of 2760 × 1960 × 2000 mm (Figure 1). A welding
99 machine from *KEMPY*, with a power source *Pro MIG 3200*, wire feeder, and control unit *Pro*
100 *MIG 501*, was used to deposit the material over the substrate. The feedstock material was a
101 commercial low-carbon high-strength steel AWS A5.28 ER110S-G wire electrode with a
102 diameter of 1 mm. The chemical composition of the wire is presented in Table 1. The parts
103 were built on mild steel substrates with dimensions of 190 × 100 × 10 mm, which was cleaned
104 and dried prior to the experiment.



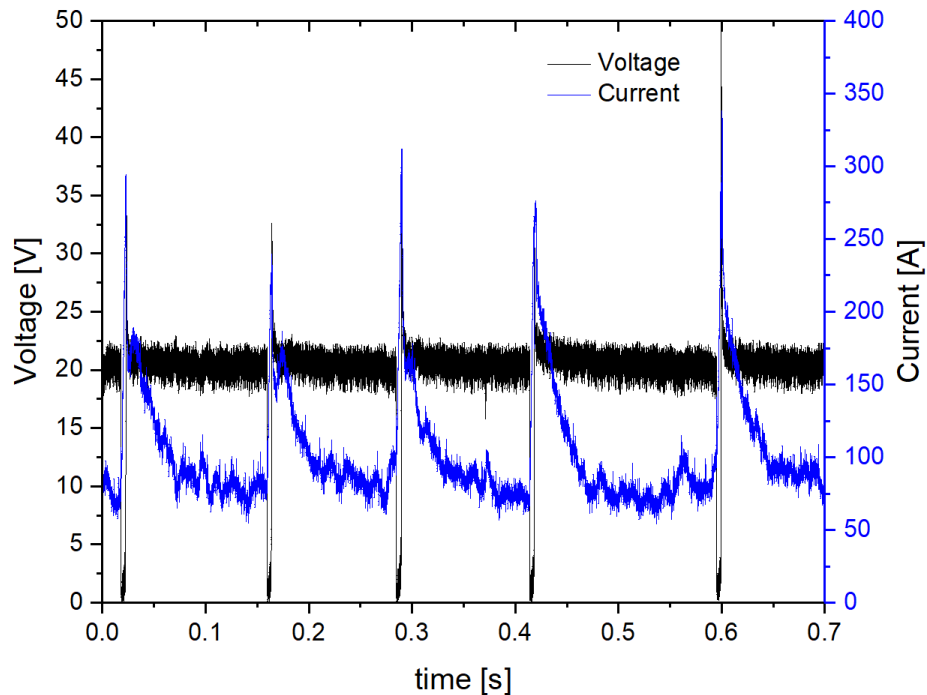
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106 *Figure 1 – Working envelope of the wire and arc additive manufacturing (WAAM) machine.*

107 *Table 1- Chemical composition of the ER110S-G wire electrode [wt.%].*

C	Mn	Si	Ni	Cr	Mo	V	Cu	Fe
0.08	1.70	0.44	1.35	0.23	0.30	0.08	0.25	Balance

108 The deposition process was instrumented to evaluate the changes in current and voltage
 109 during buildup (Figure 2). A short-circuit transfer mode was observed to occur for both
 110 depositions studied in this work.



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112

Figure 2 – Voltage and current variations during deposition.

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A Fluke TI400 thermographic infrared camera monitored the temperature of the parts during

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the fabrication; the maximum temperature was limited to 1200 °C. The camera had a

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measurement accuracy of $\pm 2\%$, an infrared spectral band up to 14 μm , a refresh rate of 9 Hz,

116

and a resolution of 320×240 pixels. The emissivity of an object depends on the temperature,

117

especially, at temperatures as high as that observed in fusion welding. Therefore, an emissivity

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of 0.84 was used, considering the existing literature [13], which was previously validated with

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thermocouples. The data were processed using the acquisition software SmartView to

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measure the temperature at any point during the buildup.

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2.2 Sample buildup

122

The length of the produced walls was set to 170 mm, the contact-tip-to-work distance was

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7 mm, and the dwell time between layers was kept constant at one minute. A continuous-

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wave mode with the electrode connected to the positive terminal (DC+) was used in all

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experiments. Deposits were made with heat inputs of 511 and 221 J/mm by varying the travel

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speed from 3.9 to 9 mm/s for samples P1 and P2, respectively. The voltage, current, and wire

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feed speed were kept constant at 21 V, 95 A, and 3 m/min for both specimens. The shielding

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gases used were pure Ar (99.999%) and a mixture of Ar + 1% CO₂ + 18% He at a flow rate of 8

129 and 16 l/min for sample P1 and P2, respectively. The process parameters are summarized in
130 Table 2.

131 *Table 2 - Summary of process parameters.*

Sample	Voltage [V]	Current [A]	Wire feed speed [m/min]	Travel speed [mm/s]	Heat input [J/mm]
P1	21	95	3	3.9	511
P2	21	95	3	9	221

132 According to the deposition strategy, after depositing one layer, the torch ascended a height
133 equal to a bead height and automatically returned to the same starting point. This process was
134 repeated until a height of approximately 100 mm was reached. It is known that an excessive
135 heat sink effect at the beginning of the deposition decreases the weld penetration, resulting in
136 a height increase [20]. However, at the end of the deposition, the increasing difficulty of heat
137 dissipation results in a contrary phenomenon. To overcome this problem, the selected travel
138 speed was lowered by 30% for the first 10 mm and increased by 30% for the last 10 mm, in
139 each deposition. As such, within these regions of the walls, the heat input varied between 730
140 and 395 J/mm for sample P1 and 315 and 170 J/mm for sample P2, at the beginning and at the
141 end of each deposited layer, respectively. It must be noticed that other deposition strategies
142 can be used in WAAM to compensate the geometrical irregularities as described in [21].

143 2.3 Characterization techniques

144 Surface waviness is one of the most important parameters that evaluate the quality of
145 depositions in arc-based AM. As defined in the literature [5], it is the maximum peak-to-valley
146 distance measured from a profile of a given area in the deposited wall. The surface was
147 measured by employing the image treatment software *Adobe Photoshop CS6* and the method
148 described by Geng et al. [22]. For each fabricated sample, five measurements of width, height,
149 and waviness were performed, and the respective average and standard deviation values were
150 determined.

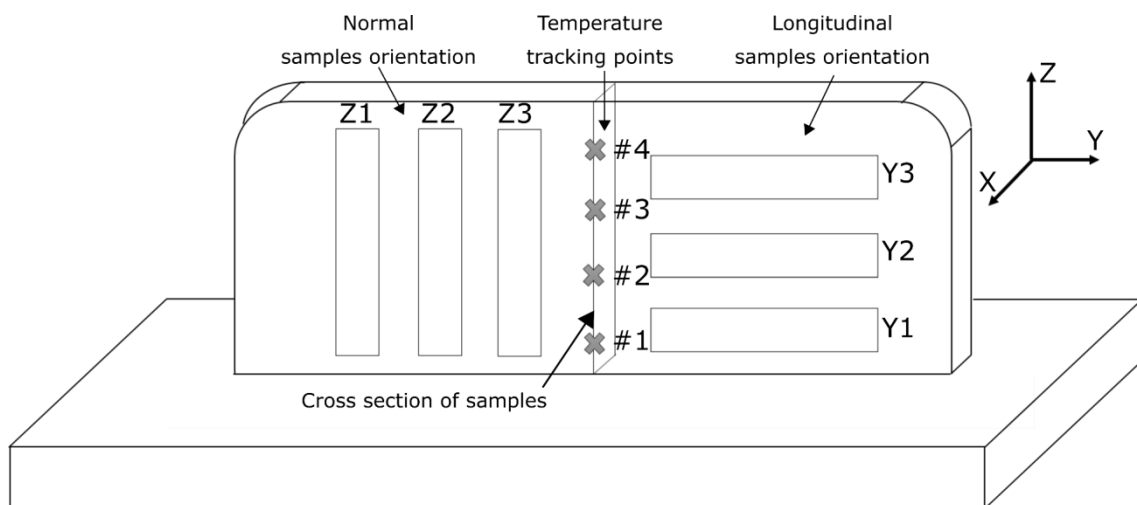
151 For microstructural characterization, the samples were cut, polished, and etched with Nital
152 (3%). Metallographic analysis was conducted using a *Leica DMI 5000 M* inverted optical
153 microscope. To obtain a deeper insight on the fine microstructural features of the as-built
154 samples, scanning electron microscopy (SEM) coupled with electron backscattered diffraction

155 (EBSD) was performed. The measurements were performed using a Quanta 650 FEG SEM and
156 a high-speed EBSD system.

157 Microhardness indentations along the sample's height were made using a *Mitutoyo HM-112*
158 *Micro-Vickers Hardness Testing Machine* for a load of 0.5 kg over 10 s, with the distance
159 between indentations being 200 μm . These measurements allowed us to analyze the effects of
160 the thermal cycles on the hardness behavior along the deposited material.

161 To investigate the evolution of the cooling rate along a wall being deposited, the temperature
162 was monitored at four different tracking points along the height of the wall, at the center of
163 each part as schematically depicted in Figure 3. At these locations, cross sections were
164 removed to pay closer attention to the microstructure and hardness properties.

165 The mechanical properties were assessed by uniaxial tensile testing and Charpy impact tests,
166 according to ISO 148-1:2016. Thus, two sets of specimens were removed from each produced
167 part, at the zones depicted in Figure 3. The uniaxial tensile and reduced Charpy V notch
168 samples were machined to dimensions of 55 \times 8 \times 2 mm and 55 \times 10 \times 2.5 mm, respectively.



169

170 *Figure 3 - Schematic representation of the temperature measurement points and location of*
171 *specimens used for uniaxial tensile and impact testing, and cross section of samples.*

172 3. Results and discussion

173 3.1 Macroscopic characterization

174 Figure 4 a) and b) present the overview of the manufactured samples named P1 (high heat
175 input) and P2 (low heat input), respectively. The corresponding transverse cross sections are
176 depicted in Figure 4 c) and d).



177

178 *Figure 4 – Aspects of produced parts: a) sample P1 (high heat input); b) sample P2 (low heat*
 179 *input). Corresponding transverse sections: c) sample P1; d) sample P2.*

180 Figure 4 c) and d) depict a narrow bead width in the first layers of each cross section, which is
 181 especially evident for sample P2. This occurs because of the rapid cooling to the cold substrate
 182 and the small amount of heat accumulated at the beginning of the process. However, with the
 183 deposition of the subsequent layers, the substrate temperature increases, which prevents the
 184 tightening of the deposited material. Table 3 presents the average and standard deviation
 185 results of waviness, width, and height per layer of each sample.

186 *Table 3- Results of the average and standard deviation of waviness, width, and height for each*
 187 *sample.*

Sample identification	Heat input [J/mm]	Waviness [μm]	Width [mm]	Height of each layer [mm]
P1	511	356 ± 16	8.8 ± 0.6	1.3 ± 0.1
P2	221	546 ± 66	5.6 ± 0.3	0.9 ± 0.1

188 Sample P1 was built with a lower travelling speed, resulting in a higher volume of material
 189 being deposited per layer. P1 is also the sample with the lowest surface waviness because of
 190 the higher interlayer temperature resulting from the heat-dissipation conditions. Its lower
 191 waviness is explained by the higher heat input used to deposit the wire feedstock, which

192 increases the interlayer temperature, improving the wettability of the subsequent layers.
 193 Additionally, the presence of CO₂ in the shielding gas improves the bead penetration [23]. In
 194 order to reduce the surface waviness, a potential approach would require decreasing the
 195 interlayer time, which would result in an increase in the interlayer temperature. However,
 196 such an approach cannot be viewed from an aesthetic aspect alone, as it can also lead to solid-
 197 state transformations that may change the resulting microstructure following a change in the
 198 imposed thermal cycles.

199 As explained previously, the travel speed varies during the deposition of each layer, to
 200 compensate for the heat-transfer differences during the building of parts. This is of extreme
 201 importance for depositing materials to a similar height in each layer, throughout the process.
 202 As shown in Table 4, for sample P1, a difference of approximately 10 mm was measured in the
 203 height, between the beginning of the arc striking (78.2 mm) and its extinction (67.3 mm).
 204 However, when the heat input was reduced (sample P2), the height difference between the
 205 start and finish of a given deposited layer was less than 4 mm, resulting in uniform distribution
 206 of material. These results emphasize the critical role of heat input in the control and stability of
 207 the molten pool and its solidification: a higher heat input promotes a more significant increase
 208 in the temperature of the substrate or the previously deposited layer, which, in turn, hinders
 209 heat dissipation, making it more difficult to control the height of the material deposited along
 210 a given layer.

211 *Table 4 – Comparison of heights at arc start and end, with travel-speed compensation.*

Sample	Height at arc striking [mm]	Height at arc extinguishing [mm]
P1	78.2	67.3
P2	65.4	61.7

212 While comparing the effects of heat input on the layer waviness and height, it is clear that the
 213 effects on part roughness and layer height are contrary. For example, for sample P1, which had
 214 a higher heat input, a potential solution to control the differences in the height of each
 215 deposited layer could cause an increase in the interlayer time, thus reducing the temperature.
 216 However, as discussed before, an increase in the interlayer time results in a rougher surface.
 217 Hence, depending on the application and/or subsequent need for post processing (namely
 218 machining processes), a trade-off must be made between the roughness and bead height.

219 3.2 Thermal analysis

220 In AM, the thermal cycle acting upon the material will significantly influence the
221 microstructure, that is, the solid-state transformations upon cooling and the grain size [24].
222 Therefore, microstructure-induced changes in the material should be reasoned based on the
223 thermal-cycle analysis of the material. As evidenced before, the thermal cycle acting on the
224 first deposited layers is different from that of the middle or upper layers, because of the heat
225 buildup during production. Figure 5 a) and b) depict the cooling rates due to the subsequent
226 passes above the tracking points for samples P1 and P2, respectively. These cooling rates were
227 measured by calculating the average temperature gradient from 800 to 500 °C ($t_{8/5}$), as it is
228 well-known that, within this temperature range, the cooling rates have a significant impact on
229 the microstructure that can be obtained in the steel [25]. Some of the subsequent passes did
230 not introduce sufficient energy to heat the previous layers to produce temperatures above 500
231 °C. In those cases, the cooling rates were not calculated.

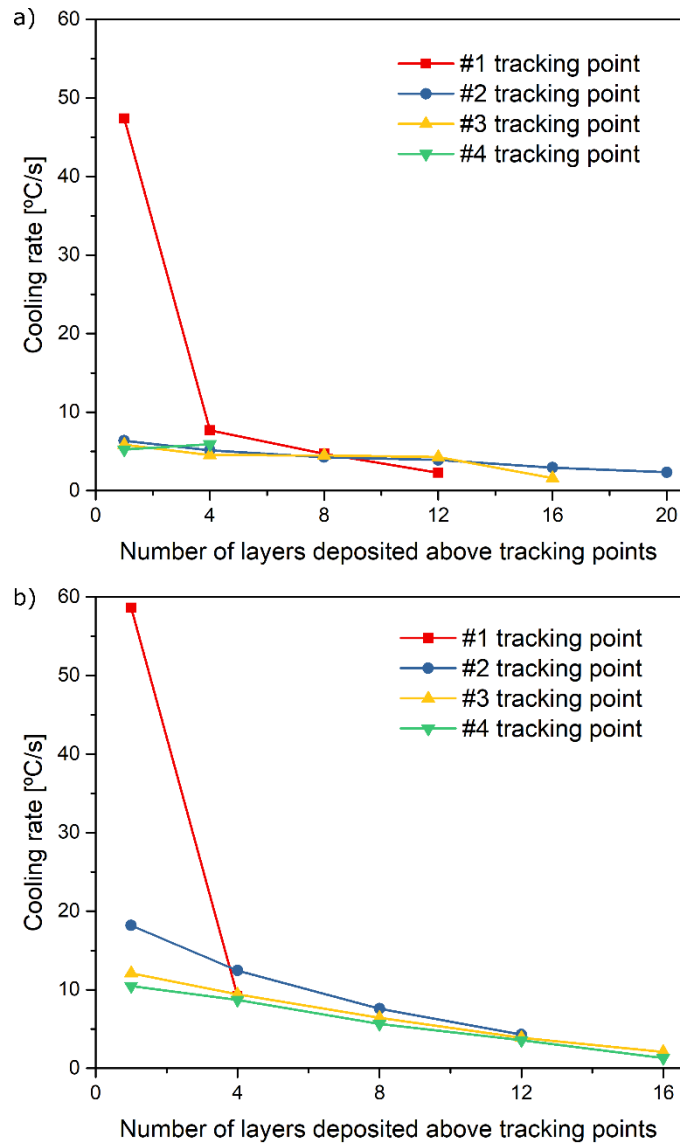


Figure 5 – Cooling rates [°C /s] of samples: a) P1; b) P2.

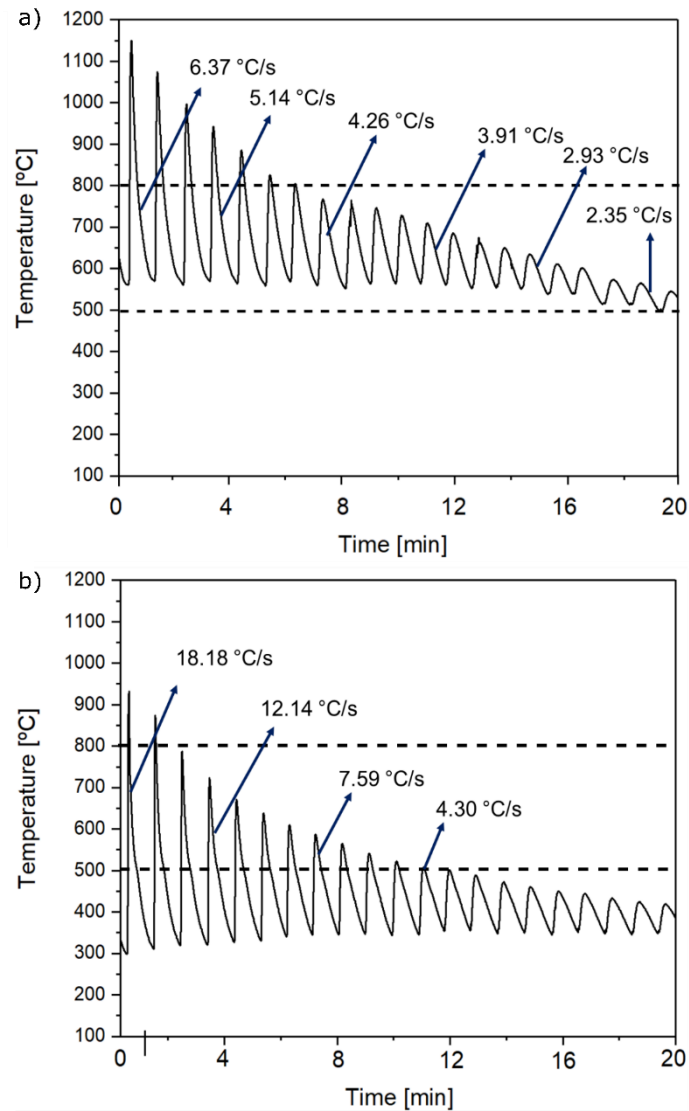
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234 As expected, the sample fabricated with a higher heat input, P1, experienced lower cooling
 235 rates than the sample P2, owing to the higher heat accumulation. Moreover, in both
 236 depositions, a large contact area with the substrate facilitated heat dissipation in the first
 237 deposited layers, resulting in higher cooling rates. After this point, the cooling rate was
 238 dominated by the thermal accumulation caused by the different heat input values used for the
 239 deposition: higher heat inputs led to higher amounts of accumulated heat in the already
 240 deposited layers, which, in turn, decreased the cooling rate of the material.

241 Figure 6 depicts the cooling rates (in °C /s) computed for the second tracking point of the
 242 analyzed samples, in which the effect of twenty sequential passes is visible. A decrease in the
 243 peak temperature and cooling rate is observed with the increase in the distance to the weld
 244 pool. On analyzing the peak temperatures after the deposition of each layer, the effect of the

245 heat input was clear: a higher heat input increased the number of times (four in total) a given
246 region surpassed the austenization temperature (approximately 800 °C according to [26]),
247 whereas, for sample P2, this occurred only twice.



248

249 *Figure 6 – Sequence of thermal cycles on the second tracking point of samples: a) P1 and b) P2.*

250 During fusion welding of steel, above the Ac_3 temperature where the material is in the
251 austenitic domain, two phenomena can occur: grain growth, at higher temperatures, leading
252 to the formation of the coarse-grain heat-affected zone (HAZ); or recrystallization, forming a
253 fine-grain HAZ. Between Ac_3 and Ac_1 , the material is in the biphasic domain of austenite and
254 ferrite; this region is the intercritical zone [27]. Below Ac_1 , tempering of the microstructure
255 occurs. From the observed thermal cycles measured between the two tracking points in
256 samples P1 and P2, a change in the microstructure is expected, and this will be discussed when
257 analyzing the microstructural characteristics of the deposited walls.

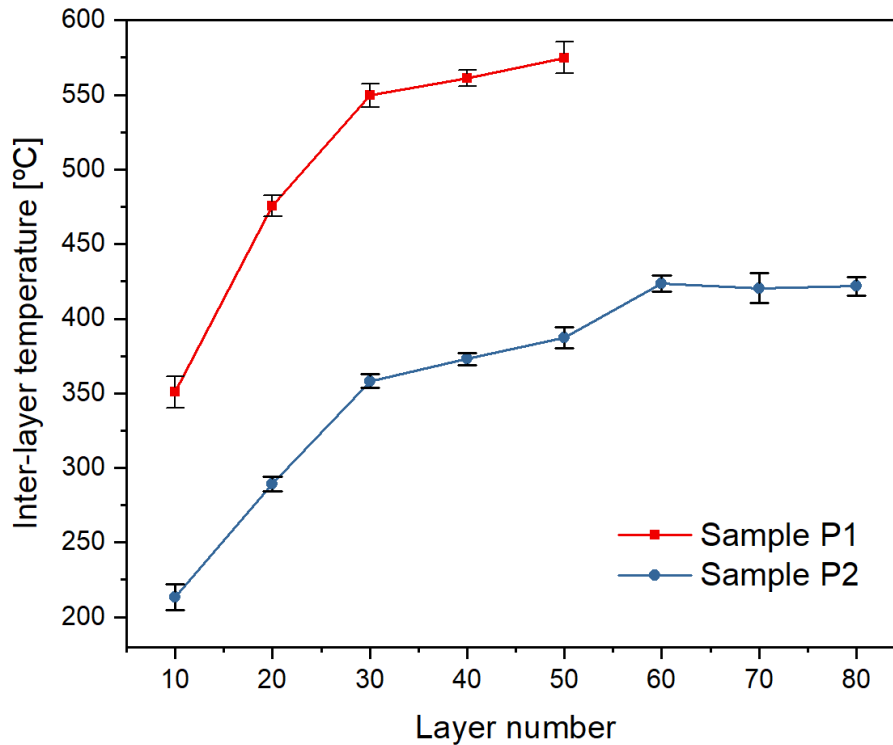
258 For steel, the residence time between 800 and 500 °C is known to be critical as it controls the
 259 solid-state transformations of the material [28]. For this reason, the residence time of the two
 260 tracking points in the 800 to 500 °C interval was determined, and these results are presented
 261 in Table 5. However, it must be noted that the residence time count was initiated only when
 262 the layer that was being deposited did not induce temperatures above the austenization
 263 temperature in the layer where the tracking point was positioned.

264 *Table 5 - Residence time [s] between 800 °C and 500 °C.*

Tracking Point	$t_{800-500\text{ °C}}$ [s] for sample:	
	P1	P2
1	947	48
2	1452	362
3	792 (until cooling of last layer)	516
4	90 (until cooling of last layer)	549

265 It can be observed that higher heat input depositions resulted in longer residence times, with
 266 the exception of the fourth tracking point where the subsequently deposited layers always
 267 increased the temperature above 800 °C. Thus, in this case, the residence time between 800-
 268 500 °C was only initiated when the last layer was deposited. This result demonstrates the
 269 importance of controlling the heat and dwell times of each deposition, as well as the proper
 270 selection of process parameters, in order to achieve a given desired microstructure.

271 The interlayer temperature is critical to the deposition strategy in WAAM, as it controls the
 272 cooling rate of the deposited layer. Figure 7 depicts the interlayer temperatures measured via
 273 thermal camera measurements for both samples.



274

275 *Figure 7 – Variation of interlayer temperature [°C] for P1 and P2 samples.*

276 From the interlayer temperature analysis, it can be observed that the temperature continues
 277 to increase, and then stabilizes, presenting evidence of the effect of the substrate temperature
 278 on the first-deposited layers. For sample P2, a steady-state condition, that is, with near
 279 constant interlayer temperature, was reached approximately at the 60th deposited layer, in
 280 contrast to P1 sample that reached a stable interlayer temperature after 30 deposited layers.

281 The interlayer temperature increased faster for sample P1 than for sample P2, because of the
 282 heat accumulation effects caused by the higher heat input. To provide the same interlayer
 283 temperature for both samples, the dwell time should be increased in sample P1. However,
 284 from a management perspective, such adjustments would render increasing lead times.

285 The thermal field analysis during material deposition can also be used to determine some of
 286 the effects of the heat input on the material. Figure 8 depicts a thermographic image of the
 287 last-deposited layer for both samples. It can be observed that the width and depth of the HAZ
 288 during a single deposition is higher for the higher input sample. Furthermore, the higher heat
 289 input clearly increases the temperature in the already-deposited material, and the heat
 290 accumulation is noticeable.

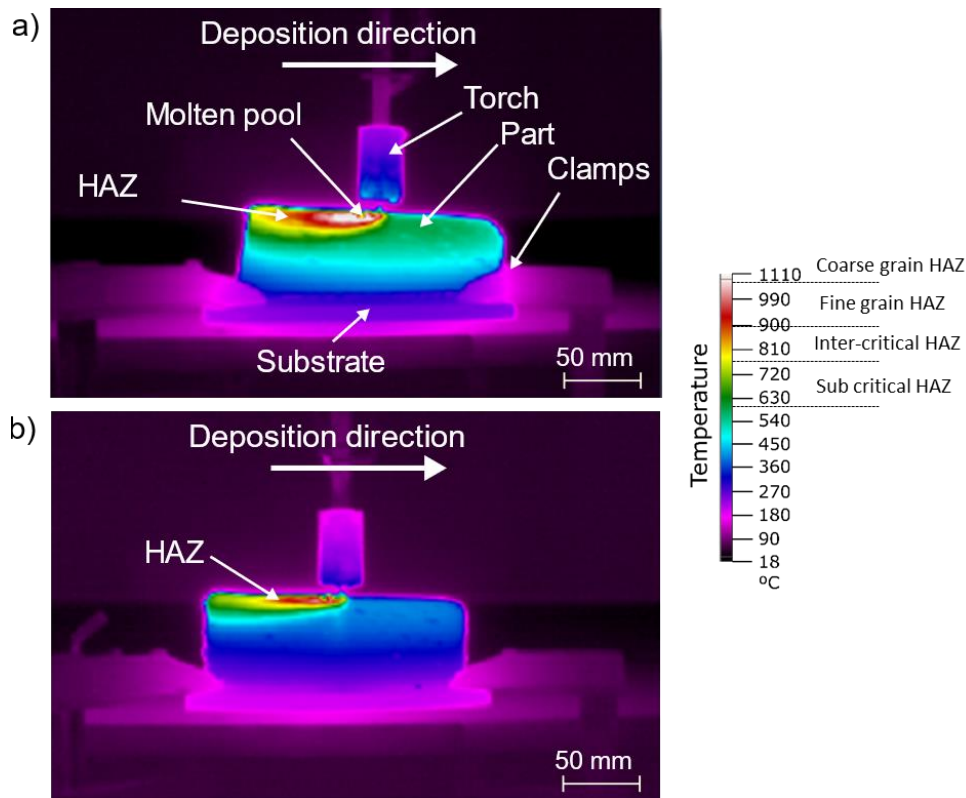


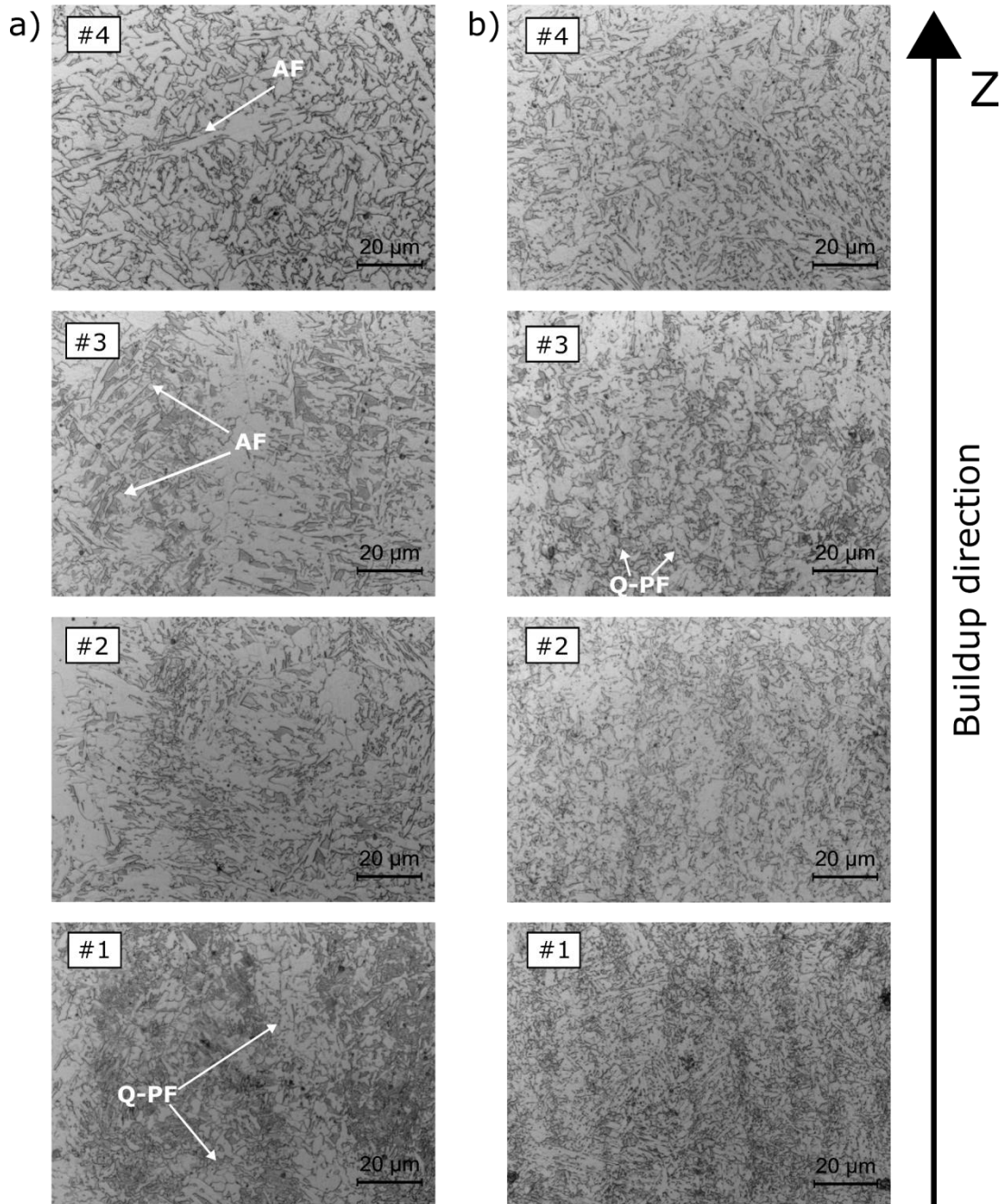
Figure 8 – Thermographic analysis of the last deposited layer for samples: a) P1; b) P2.

3.3 Microscopic observations

Microstructural characterization by means of optical microscopy of the as-built samples, depicted in Figure 9, revealed fully dense parts without cracks, porosity, or lack of fusion. AM techniques where melting occurs have several similarities with fusion-based welding [24]. In the available welding literature, there are multiple procedures such as pre or post-heating, to mitigate cold cracking, which is a very common discontinuity in HSLA steel [29]. However, the deposition of material in a layer-by-a layer process increases the temperature of the previous layers, resulting in a reduction in the cooling rate, thus mitigating the cold-cracking susceptibility. Additionally, the nonexistence of mechanical constraints in WAAM, as it occurs during welding, allows the material to accommodate some of the thermal stresses that are developed during building, which further contributes to reducing the probability of cold cracking.

A more comprehensive view of the microstructure removed from the midsection of both P1 and P2 samples, previously illustrated in Figure 3, is depicted in Figure 9. The grain size is larger in the high heat input sample owing to the heat buildup, which favors grain growth. Additionally, the grain size increases along the height of the produced walls. Ferrite

309 morphologies, acicular and granular morphologies, are observed in Figure 9. Other
310 microconstituents can also be observed; however, their low dimensions hinder their
311 identification by optical microscopy.



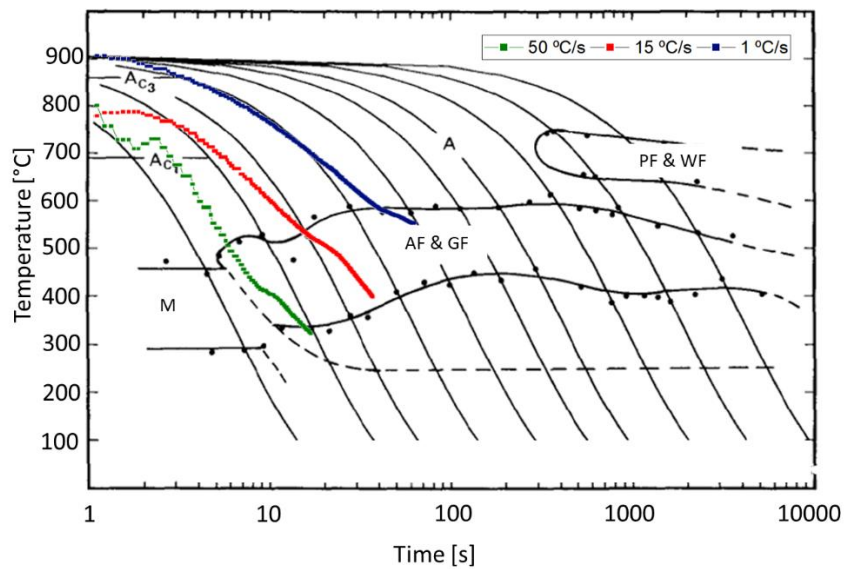
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313 *Figure 9 – Cross section micrographs of samples: a) P1 and b) P2 along the height (AF: acicular*
314 *ferrite; Q-PF: quasi-polygonal ferrite).*

315 The resolution of optical microscopy prevents accurate determination of all the
316 microconstituents that were formed along the built parts. For that reason, SEM aided by EBSD
317 was performed.

318 Owing to the multiple reheating cycles that any layer experiences (from subsequent
319 depositions), the microstructures of the as-built parts were similar to that of a HAZ in HSLA
320 steel. In such steel, the solid-state transformations can be classified within two temperature
321 regimes: 1300 to 800 °C and 800 to 500 °C. From 1300 to 800 °C, significant austenite grain
322 growth occurs, while in the 800 to 500 °C range, austenite transforms to distinct ferrite
323 morphologies and bainite [26]. Upon cooling from the high-temperature regime, the
324 decomposition from austenite to ferrite occurs, with the formation of allotriomorphic ferrite at
325 the prior austenite grain boundaries. Then, nucleation of the side-plate ferrite may occur at
326 the austenite/ferrite boundaries and extend into the untransformed austenite grains. Acicular
327 ferrite formation is usually associated with oxide inclusions, weld-metal hardenability, and
328 cooling conditions. In the absence of potent inclusions, bainitic ferrite might form upon further
329 cooling [30]. If bainitic ferrite is formed without the presence of carbides, the remaining
330 austenite will be enriched in carbon, which promotes its stability. During the final cooling to
331 room temperature and depending on the carbon content, which influences the M_s and M_f
332 transformation temperatures in carbon steels [31], the remaining austenite may transform
333 fully or partially into martensite. In the latter case, the so-called martensite–austenite (M–A) is
334 formed.

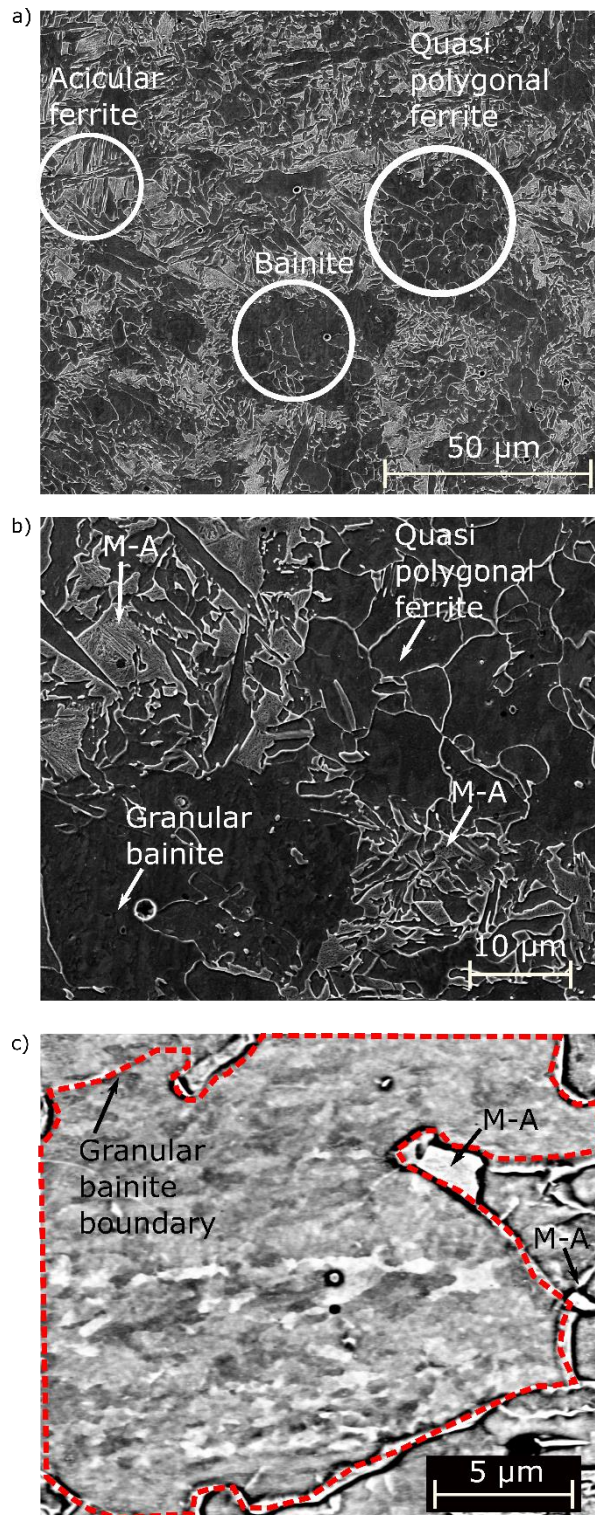
335 Etching with Nital solution preferentially attacks ferrite. Therefore, other phases such as
336 bainite and M–A constituents are elevated relative to the polygonal ferrite. All these
337 microconstituents are clearly observed in the SEM image of sample P1 in zone #3 within the XY
338 plane in Figure 11 a). Special attention is given to sample P1, as the same microconstituents
339 are observed in sample P2. Figure 11 a) depicts an SEM overview image highlighting the three
340 distinctive microstructures: acicular ferrite (AF), quasi-polygonal ferrite (Q-PF), and bainite. All
341 these phases are predicted by the continuous cooling transformation (CCT) diagram of HSLA
342 steel [32] with a similar chemical composition of the feedstock material used in this work, and
343 are the main constituents of the as-built parts. Fully austenite and martensitic structures could
344 be achieved only at cooling rates higher than 100 °C/s and without reheating cycles, as
345 foreseen by the CCT diagram. Figure 10 depicts the superimposition of some experimental
346 cooling curves on the CCT diagram of the alloy used in this work. It is observed that all
347 reported microstructures are predicted by the CCT diagram, and these depend on the cooling
348 rates experienced by the material. No fully martensitic microstructure was observed in the as-
349 built walls owing to the need to achieve very high cooling rates (> 100 °C/s) which were not
350 obtained during the buildup.



351

352 *Figure 10 – CCT diagram for the alloy used in this investigation with superimposition of*
 353 *experimental cooling curves (adapted from [32]).*

354 Higher SEM magnifications reveal the presence of M–A (Figure 11 b and c) with a maximum
 355 grain size of approximately 10 μm, distributed sparsely along the material. The granular
 356 bainite, as shown in Figure 11 c) in a high-contrast image, is represented by a large bainite
 357 packet (> 20 μm) enclosed by a red dashed line, with sub grains of sizes less than 1 μm. These
 358 sub grains usually present similar crystallographic orientations [33].



359

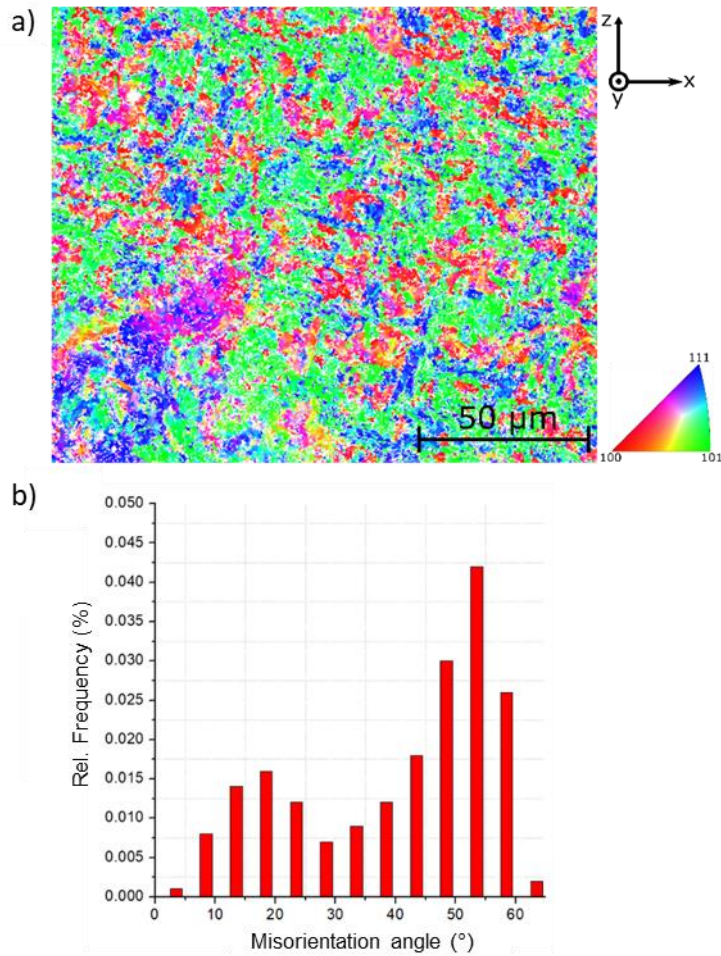
360 *Figure 11 – Scanning electron microscopy (SEM) image of the typical microstructure of the as-*
 361 *built samples (with martensite–austenite (M-A), bainite, acicular ferrite, and quasi-polygonal*
 362 *ferrite): a) overview; b) high-magnification image; c) close-up of a granular bainite packet*
 363 *obtained using a high-contrast detector.*

364 M-A particles are typical microstructures in HSLA steels and are observed for cooling rates
 365 between 10 to 40°C/s and stinger-type above these rates [34]. In the case of this research, due

366 to the subsequent heating and cooling of the deposited material it is difficult to track the M-A
367 transformation through the fabrication process, however, mostly blocky-type structures are
368 depicted and these coincide with the morphology reported in the literature [34–36]. The
369 formation of M–A microstructures in low-alloyed steel is attributed to an incomplete
370 transformation from austenite to martensite in the HAZ after reheating to intercritical
371 temperatures [35]. Enrichment of C and Mn have been reported in M-A constituents of a
372 intercritically reheated coarse-grained HAZ (ICCGHAZ) of a HSLA steel [36]. In this work,
373 multipass beads favored the formation of M-A microconstituents, since each following bead
374 caused a reheating at lower temperature and allow elements to stabilized new blocky-type M-
375 A. The presence of necklace-type M–A in ICCGHAZ is typically undesirable as it can impair
376 fracture toughness [36], nevertheless, no necklace-type M–A were observed in the deposits.

377 Bainite is formed at intermediate cooling rates, between that of martensite (high cooling rate)
378 and perlite (low cooling rate). The presence of bainitic microstructures usually increases the
379 mechanical strength of the material [37]. Bainite formation is a diffusionless transformation, in
380 which tiny plates known as “sub-units” can be supersaturated in carbon [38]. This leads to
381 carbon partitioning to residual austenite right after the growth of the plates. Such carbon
382 partitioning further complicates the transformation of residual austenite to martensite upon
383 cooling, and favors the formation of M–A regions. The presence of bainite in steel plays a
384 critical role in crack initiation and propagation [35].

385 To further determine the influence of thermal cycles on the solidification conditions, stability
386 of phases, and developed textures, EBSD measurements were performed. Owing to the large
387 number of steel microstructures, identification of the microconstituents is often problematic
388 [39]. As stated in [39], one of the shortfalls of EBSD, in the quantification of steel
389 microstructures, is the fact that the different morphologies of ferrite have the same
390 crystallographic structure, which may lead to erroneous interpretations. Bainitic
391 microstructures also have lower confidence indexes owing to the large number of dislocations,
392 which is responsible for the high strength and low ductility typical of this phase. EBSD
393 measurements of sample P1 in zone #3, depicted in Figure 12 a), revealed that no significant
394 preferential orientation was developed during the production of the samples. In addition,
395 bainitic areas are often limited by high-angle boundaries, providing another distinct feature to
396 identify such microstructures using EBSD, as shown in Figure 12 b). When the misorientation of
397 these high-angle boundaries is above 40°, the boundaries of the bainite packets can stop the
398 propagation of brittle cracks [33].

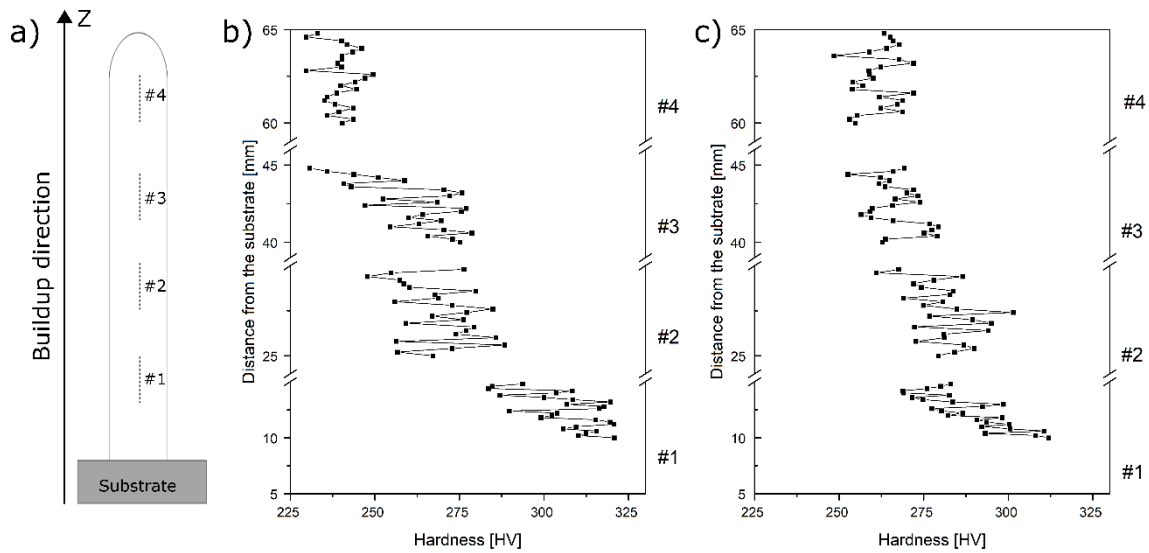


399

400 *Figure 12 - a) Electron backscattered diffraction (EBSD) inverse pole figure measurements of*
 401 *sample P1; b) misorientation angle.*

402 3.4. Microhardness results

403 Figure 13 depicts the microhardness distributions near the four tracking points on the cross
 404 sections of samples P1 and P2, at a center line along the building height. Globally, for both
 405 samples, because of the repeated reheating and thermal accumulation, the grain size was
 406 coarser along the height of the sample (building direction), leading to a decrease in hardness.
 407 Additionally, the fast cooling rate in the first deposited layers, aided by the cold substrate and
 408 its capacity for heat dissipation, justified the higher hardness observed in this region.



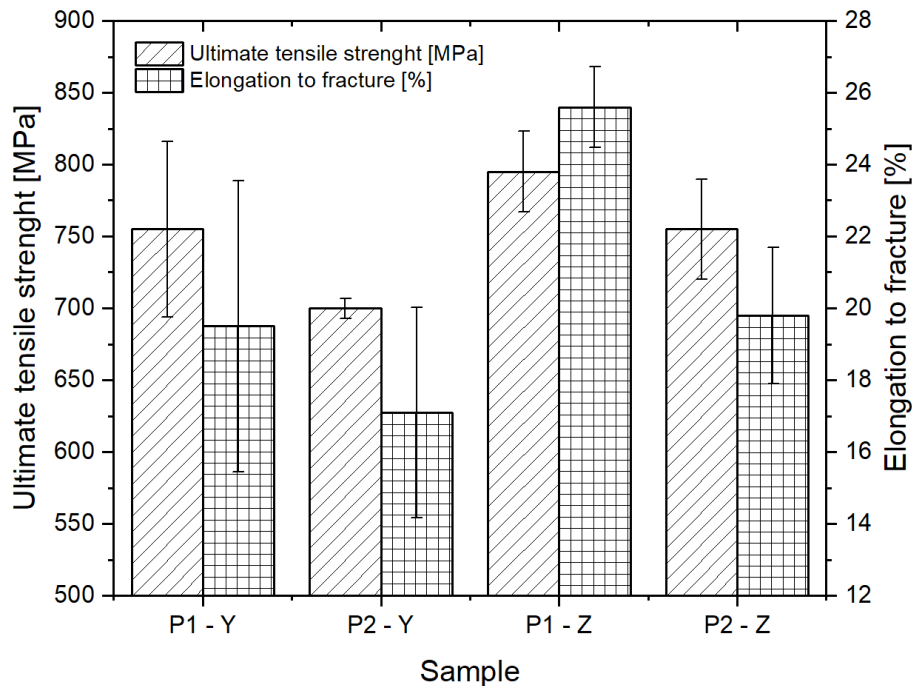
409

410 *Figure 13 - a) Schematic representation of indentations made on the cross sections of as-built*
 411 *samples. Microhardness profiles along the height of samples: b) P1; c) P2.*

412 In Figure 13, a gradient of hardness is visible along sample P1: in zone #1, the hardness value
 413 varies between 283 and 320 HV; in zone #2, between 247 and 288 HV; in zone #3, the
 414 hardness value ranges from 230 to 278 HV; and finally, in zone #4, it varies from 229 HV to 250
 415 HV. Sample P2, with a lower heat input during deposition, exhibited slightly lower
 416 microhardness along the part height. For sample P2, in zone #1, the hardness changed
 417 between 268 and 311 HV, while for zones #2, #3, and #4, it was found to vary within 268 to
 418 300 HV, 256 to 280 HV, and 247 to 277 HV, respectively. It must be noticed that the range of
 419 hardness values obtained are in good agreement with those predicted by the CCT diagram for
 420 this alloy.

421 3.5 Uniaxial tensile tests

422 To evaluate the mechanical properties of the as-built parts, tensile tests were performed along
 423 the longitudinal (Y) and normal (Z) directions of the samples obtained (schematically depicted
 424 in Figure 3). A summary of the results is presented in Figure 14.

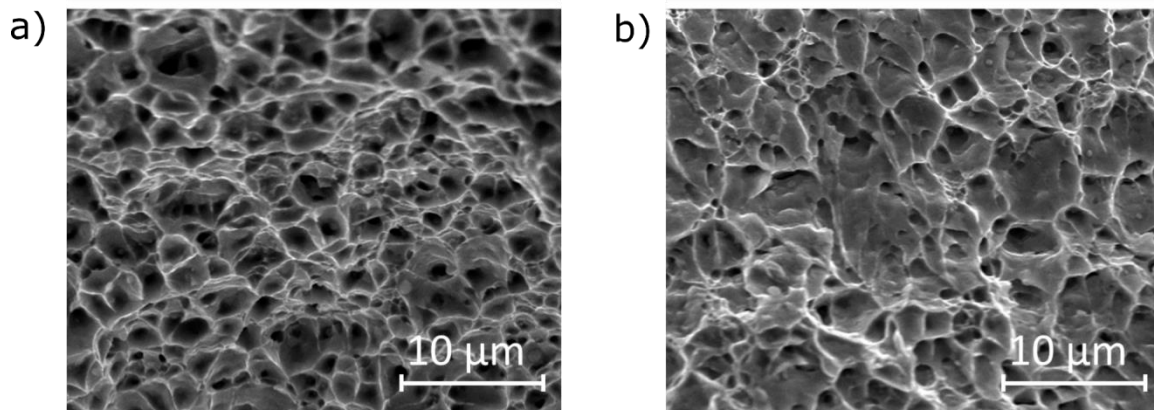


425

426 *Figure 14 – Ultimate tensile strength (UTS) and elongation to fracture of the tested samples.*

427 Both samples exhibited average values for the ultimate tensile strength (UTS), ranging
 428 between 700 and 795 MPa, while the elongation-to-fracture varied from 17.1 to 25.6%. The
 429 similar values of UTS obtained along the Y and Z directions for the samples suggested
 430 homogeneity of mechanical properties along the height of the parts, regardless of the different
 431 cooling rates. Moreover, the elongation-to-fracture values of all samples indicated the good
 432 performance of WAAM in the manufacturing of this steel. The existence of ferrite in the
 433 microstructure improves the deformation ability of the material, while the formation of bainite
 434 leads to an increase in tensile strength. Therefore, the presence of both bainitic and ferritic
 435 microstructures produces a material with excellent ductility and mechanical strength.

436 The SEM analysis of the fracture surfaces revealed a ductile fracture, as evidenced by the
 437 massive presence of dimples as depicted in Figure 15 a) and b), corresponding to samples P1
 438 and P2, respectively, when tested along the longitudinal direction. Despite the presence of M–
 439 A in the microstructure, no evidence of transgranular cleavage fracture was observed.
 440 Therefore, the presence of M–A was not considered detrimental to the mechanical properties
 441 of the as-built material, owing to its relatively low volume fraction, which is insufficient to
 442 critically change the fracture mode of the as-built parts.



443

444 *Figure 15 – Scanning electron microscopy (SEM) image of a surface fracture for a sample tested*
 445 *along the longitudinal direction, in specimen: a) P1 and b) P2.*

446 3.6 Charpy impact tests

447 The impact toughness of the reduced samples extracted along the longitudinal (Y) direction
 448 was 15 J, while for those obtained along the normal (Z) direction, the impact toughness
 449 increased slightly, to 18 J. In order to convert the acquired data to that of a normalized 10 mm
 450 thickness specimen, an empirical hyperbolic-tangent equation [40] was used. The
 451 corresponding values were 57 and 71 J, respectively. The impact toughness of the as-received
 452 feedstock material was approximately 70 J, according to the wire manufacturer. This
 453 difference was within the range of uncertainty of the measurements and the empirical method
 454 used, which led to the conclusion that homogeneity of mechanical properties existed between
 455 the vertical and horizontal samples, confirming the uniaxial tensile test results.

456 4. Conclusions

457 HSLA steel parts were fabricated by WAAM, with different process parameters, and the
 458 microstructural evolution along the height of the samples was rationalized based on the
 459 complex thermal cycles acting upon the material. The following conclusions were drawn:

- 460 • The sample with the highest heat input had less surface waviness, because of the good
 461 wettability of the layers over the previously deposited ones.
- 462 • As the height increased, the temperature gradient decreased for both depositions
 463 (high and low heat input). When a high heat input was used, lower cooling rates were
 464 observed and higher residence times occurred in the 800–500 °C temperature range.

- 465 • Despite the significantly different heat input values used to build the samples, no
466 significant microstructural changes were observed; the presence of ferrite, bainite,
467 and MA were identified by electron microscopy;
- 468 • Owing to the similar microstructures and the fact that no preferential texture was
469 developed, both uniaxial tensile test and Charpy impact test showed similar
470 mechanical properties in the longitudinal (Y) and normal (Z) directions. The as-built
471 parts presented high UTS and excellent ductility.
- 472 • Sample P2, with a lower heat input during deposition, exhibited lower and more
473 uniform microhardness along the part height compared to sample P1.

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