Influence of laser powder bed fusion process parameters on the microstructure and cracking susceptibility of nickel-based superalloy Alloy 247LC

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1. Introduction

Alloy 247LC is a γ precipitation-strengthened nickel-based superalloy. It is castable and suitable for manufacturing gas turbine blades because it has good creep and high-temperature oxidation properties [1]. Recently, there has been interest in processing this alloy using laser powder bed fusion (L-PBF). L-PBF, a layer-by-layer additive manufacturing (AM) process, is preferred over casting because intricate cooling channels with complex geometries can be printed using L-PBF by employing improved process parameters.

Microstructures of material conditions of nickel-based superalloy Alloy 247LC fabricated using laser powder bed fusion (L-PBF) were investigated. Experiments designed in a prior study revealed the L-PBF process parameters for which the material conditions displayed a reduced susceptibility to cracking. Certain process parameters produced material conditions with an increased susceptibility to cracking. In this study, the material conditions were investigated in detail to reveal their microstructure and to determine the cause of cracking. The reason for the transition between a reduced to an increased susceptibility to cracking was examined. The results revealed solidification cracking occurred at high-angle grain boundaries. Solidification cracking may have been promoted at high-angle grain boundaries because of the undercooling contribution of the grain boundary energy. Furthermore, Si segregation was observed in the cracks. Thus, the presence of Si most likely promoted solidification cracking. It was observed that a high crack density, which occurred in the high energy density material condition, was associated with a large average grain size. The fact that certain combination of process parameters produced microstructures with a low susceptibility to cracking, indicates that reliable Alloy 247LC material may be printed using L-PBF by employing improved process parameters.

More recently, [4] have reported that printing Alloy 247LC by L-PBF leads to cracking mechanisms similar to those found in welding and are described by Lippold [5]. Post-processing by hot isostatic pressing (HIP) was reported to successfully close the cracks [6]. However, Carter et al. [4] indicated that crack closure by HIP is limited to internal cracks and that surface cracks are not amenable to HIP. The limitations of HIP were also highlighted by Zhao et al. [7], who found that local brittle regions were present along the macrocracks because of incomplete diffusion bonding during HIP. Such local brittle regions are detrimental to mechanical properties of the final product. Therefore, suppressing crack formation, rather than relying entirely on post-processing by HIP, appears to be a more viable approach. Strategies implemented to print Alloy 247LC by L-PBF have been directed toward modifying the alloy and/or improving the process conditions (process parameter development, scanning strategy improvement, etc.). The main strategies reported in the literature for printing Alloy 247LC by L-PBF are summarized in Table 1, where the different strategies produce
varying crack densities.

In Table 1, process parameter development indicates that the laser power, scanning speed, hatch distance, and layer thickness suitable for manufacturing dense samples (and sometimes low crack density samples) were either designed as required or selected from available data. In some studies, the main printing strategy was not process parameter development as described above but involved designing an appropriate laser focus diameter or scanning strategy (processes F/G and I, respectively). Other process conditions were also modified, for example, the application of in situ laser shock peening (process H). Alloy development indicates that the composition of the alloy was modified. Griffiths et al. [8] removed Hf from the composition of Alloy 247LC to form a Hf-free Alloy 247LC. Two process conditions in Table 1 (A and B) were applied to print both Alloy 247LC and the Hf-free Alloy 247LC. For both alloys, process B resulted in a lower crack density. This was attributed to the lower aspect ratio (depth/width) of the melt pool, which produced lower strains. The corresponding crack densities for processes A and B are displayed in Table 1. Although the investigators reported that removing Hf reduced the crack density, this effect is uncertain. The crack densities of the samples produced by process C [9] showed that increasing the Hf content of Alloy 247LC above the nominal composition (2.5 wt% Hf in the Hf-plus Alloy 247LC) yielded a lower crack density (0.25 mm/mm²) compared with that of the Hf-free Alloy 247LC (8.2–9.3 mm/mm²). This reduction was attributed to the improved liquid feeding of the Hf-plus Alloy 247LC. Tang et al. [3] developed a new alloy, ABD 850AM, using physics-based modeling, which facilitated the design of printable alloys. The alloy design was based on calculating an acceptable solidification temperature range, a reduced susceptibility to strain-aging cracking, and an adequate creep resistance. The ABD 850AM alloy was judged to be amenable to LPBF processing and resistant to cracking. In contrast, Alloy 247LC, which the investigators processed with optimized process parameters (process E in Table 1), cracked severely, and the cracks were more predominant at the surface of the samples than in the bulk. The investigators claimed that the ABD 850AM alloy could replace Alloy 247LC. This is conceivable, given that the cracking susceptibility of the latter is undesirable. However, the low crack densities achieved by some of the processes shown in Table 1 indicates that the cracking problem in Alloy 247LC is not severe if appropriate process conditions are used. Examples of this are displayed in Table 1 and summarized as follows. Gerstgrasser et al. [10] reduced cracking by using a stable laser spot diameter instead of a shifting laser spot diameter. The former condition produced a deeper melt pool that healed the previously formed cracks. Hagedorn et al. [15] printed Alloy 247 (a composition with approximately twice the C content compared to that of Alloy 247LC) samples on a substrate preheated to approximately 1200 °C. Preheating reduced the thermal gradient and consequently the strains, which in turn reduced the amount of cracking. Basak and Das [16] also demonstrated that Alloy 247 could be repaired by L-PBF with low cracking susceptibility. A low energy density was applied, which enabled shallow remelting, which produced a low amount of strain. Another study used 3D laser shock peening during L-PBF processing of Alloy 247LC to remove tensile strains and to introduce compressive strains, which subsequently resisted crack formation [11]. Process conditions that combine the appropriate process parameters produce the lowest crack density. Catchpole-Smith et al. [12] showed that printing with an improved scanning strategy (a short laser scan length between 100 and 300 μm) in combination with a suitable laser power and scanning speed could reduce the crack density.

Regardless of the solution used to minimize cracking, the printability of an alloy is judged by its density (considering the location of voids), microstructure, and properties. These are similar to the requirements used to assess weldability [5]. All these factors should satisfy the mechanical properties and tortuous environment requirements of a gas turbine. Therefore, improving the process conditions may be more beneficial than modifying the alloy because it can retain the reliable microstructure and properties of Alloy 247LC. If the composition of the alloy is modified, the microstructure and properties may be altered. For example, the newly designed ABD 900AM alloy displayed a lower creep strength than that of Alloy 247LC [3]. To achieve a better strain-age cracking index, the Ta content in ABD 900AM was reduced. This reduced the y’ volume fraction [17], which consequently reduced the creep strength [18]. De Luca et al. [19] highlighted the influence of the alloy composition on the properties. The investigators noted that although the printed Ni–Cr–Al–Ti model alloy was free from hot cracks, the lack of grain boundary strengtheners (such as C or B) in the model alloy promoted solid-state cracking. In the present study, process parameter development was explored as a means of eliminating cracks and obtaining dense parts with the required microstructure. To achieve this purpose, the microstructure of a low crack density Alloy 247LC sample printed with the developed process parameters (corresponding to an energy density of 108 J/mm³) was analyzed. A low crack density was attained using process parameter development alone while retaining the chemical composition of Alloy 247LC. The other two process conditions investigated involved process parameter combinations (corresponding to energy densities of 196 and 71 J/mm³) that produced mainly cracks or voids, respectively. It is useful to reflect on the investigation performed by Carter [14], which was the first to show the influence of L-PBF process parameters on the crack density in Alloy 247LC. Carter claimed that it was not possible to eliminate cracks by process parameter development, given the technology accessible at the time of

Table 1
Published crack densities (mm/mm²) of Alloy 247LC (including Alloy 247LC with no Hf (Hf-free) and additional Hf (Hf-plus)) printed by L-PBF using different printing strategies.

<table>
<thead>
<tr>
<th>Process</th>
<th>Main strategy for printing the samples</th>
<th>Crack density of Alloy 247LC (mm/mm²)</th>
<th>Crack density of Hf-modified Alloy 247LC (mm/mm²)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>Process parameter development/ alloy development</td>
<td>1.27</td>
<td>0.81a</td>
<td>[8]</td>
</tr>
<tr>
<td>B</td>
<td>Process parameter development/ alloy development</td>
<td>0.12–0.14</td>
<td>0.02–0.03a</td>
<td>[8]</td>
</tr>
<tr>
<td>C</td>
<td>Alloy development</td>
<td>8.2–9.3³</td>
<td>0.25³</td>
<td>[9]</td>
</tr>
<tr>
<td>D</td>
<td>Process parameter development</td>
<td>0.7</td>
<td>6.7</td>
<td>[3]</td>
</tr>
<tr>
<td>E表面</td>
<td>Process parameter development</td>
<td>6.7</td>
<td></td>
<td>[3]</td>
</tr>
<tr>
<td>E bulk</td>
<td>Process parameter development</td>
<td>0.8</td>
<td></td>
<td>[3]</td>
</tr>
<tr>
<td>F</td>
<td>Laser focus diameter regulation</td>
<td>0.01</td>
<td></td>
<td>[10]</td>
</tr>
<tr>
<td>G</td>
<td>Laser focus diameter regulation</td>
<td>0.4</td>
<td></td>
<td>[10]</td>
</tr>
<tr>
<td>H</td>
<td>In situ laser shock peening</td>
<td>0.12</td>
<td></td>
<td>[11]</td>
</tr>
<tr>
<td>I</td>
<td>Scanning strategy improvement</td>
<td>&lt;0.5</td>
<td></td>
<td>[12]</td>
</tr>
<tr>
<td>J</td>
<td>Process parameter development</td>
<td>0.93</td>
<td></td>
<td>[13]</td>
</tr>
<tr>
<td>K</td>
<td>Process parameter development</td>
<td>0.02–0.04</td>
<td></td>
<td>[13]</td>
</tr>
<tr>
<td>L</td>
<td>Process parameter development</td>
<td>0.14</td>
<td></td>
<td>[13]</td>
</tr>
<tr>
<td>M</td>
<td>Process parameter development</td>
<td>1</td>
<td></td>
<td>[14]</td>
</tr>
</tbody>
</table>

a Hf-free.
b Hf-plus.
the study. This may have prompted the prominence of alloy development in the community. However, some of the crack densities displayed in Table 1 (particularly those corresponding to process conditions F and K) are significantly lower than that obtained in Carter’s study. Thus, process parameter development shows promise in the eventual elimination of cracks. The microstructure of L-PBF-printed Alloy 247LC was previously studied using analytical scanning and transmission electron microscopy [20]. The microstructure of L-PBF-printed Alloy 247LC consists of γ, γ', γ/γ' eutectic, and carbides, which are similar in type (but smaller in size) to those in the cast microstructure studied by Mostafaei and Abbasi [21]. The L-PBF microstructure contained 700 nm cells within the grains, and the cells and grain boundaries contained Hf-, W-, Ta-, and Ti-rich precipitates, which inhibit the movement of dislocations and thus promote high residual stresses. However, the study did not investigate cracks, which, as discussed above, are present in the microstructure. Similar phase constituents were revealed in the investigation by Divya et al. [22]. The microstructure displayed a predominance of low-angle grain boundaries (LAGBs), which were typically <4°; however, the cracking mechanism was not characterized. In the present study, the microstructures of the materials produced with the three energy densities were compared to one another. The microstructure was characterized in terms of defects (cracks and voids), elemental segregation, grains (structure, size, and texture), cell structures, phases, and microhardness. The major contributions of this study are the characterization of a low crack density material condition and clarification of the influence of the process parameters on the crack susceptibility of the microstructure.

2. Experiments

2.1. Laser powder bed fusion processing

In this study the ingoing material was a Sandvik Osprey™ Alloy 247LC VIGA powder. The powder was produced via vacuum induction melting in a furnace and was atomized using argon gas. The powder was sized into a powder particle size distribution of 15–53 μm and the resulting chemical composition is displayed in Table 2.

Cubes of 15 mm × 15 mm × 15 mm were manufactured using an EOS M290 machine with different combinations of laser power, scanning speed, and hatch distance, as shown in Table 3. These different combinations of process parameters were part of 30 combinations produced from a design of experiments investigation performed by the authors [13]. The study in Ref. [13] showed that out of the 30 combinations of process parameters, process J recorded the highest crack density while process L recorded the largest void content. Process K produced a desired low crack density and void content material condition. Hence, in line with the objectives of the current study, the microstructure of the three process parameter combinations was investigated to determine the likely reason for the varying defect content. Here, the reason for the increased susceptibility to cracking in process J compared to K was of paramount interest.

Energy density was calculated as laser power/(scanning speed × hatch distance × layer thickness). The layer thickness applied during the process was constant at 20 μm. A continuous-wave Yb fiber laser was used, and the spot size was 70 μm. The optical system comprised an F-theta lens and a high-speed scanner. Argon was used as the shielding gas. A stripe laser scanning strategy was applied, as described by Leicht et al.

Table 3

<table>
<thead>
<tr>
<th>Sample number/ process identification</th>
<th>Power (J/s)</th>
<th>Speed (mm/s)</th>
<th>Hatch distance (μm)</th>
<th>Energy density (J/mm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3/Process J</td>
<td>220</td>
<td>2800</td>
<td>20</td>
<td>196</td>
</tr>
<tr>
<td>30/Process K</td>
<td>195</td>
<td>3000</td>
<td>30</td>
<td>108</td>
</tr>
<tr>
<td>22/Process L</td>
<td>170</td>
<td>3000</td>
<td>40</td>
<td>71</td>
</tr>
</tbody>
</table>

*This is the volumetric energy density. It was calculated using a layer thickness of 20 μm.*

[23]. The scanning direction of each new layer was rotated by 67° relative to the direction of the previous layer.

2.2. Metallography

Cross sections of the samples were cut and were mounted, ground, and carefully polished into a mirror finish using colloidal silica suspension with size 0.02 μm in the final step. The samples were characterized using optical microscopy (Zeiss Axio light optical microscope) and scanning electron microscopy (SEM, Zeiss Gemini 450 field-emission gun scanning electron microscope). Secondary electron (SE) and backscattered electron (BSE) images were acquired using SEM. Electron backscatter diffraction (EBSD) data were acquired using an Oxford Symmetry system attached to the SEM system. EBSD data were processed in Aztec software from Oxford and were subsequently analyzed using Aztec crystal software. Time-of-flight secondary ion mass spectrometry (TOF SIMS) was performed on the samples using a TOF SIMS 5 instrument and was similar to the experiment performed in Ref. [24]. The primary ion source was a 25 kV Bi cluster ion gun and a 10 kV C60 ion source was utilized for dislodging secondary particles from the surface of the samples. A pulsed primary ion beam (Bi3+ at 0.2 pA) was used to analyze the samples in the delayed extraction mode with a focus of approximately 400 nm. The spectra were obtained and processed using Surface Lab software. The spectra were internally calibrated to signals of [C]+, [CH3]⁺, [CH2]⁺ and [Cr]⁺ for the positive ion mode and [C]⁻, [CH]⁻, [C2H]⁻, [C3H]⁻ for the negative ion mode. The powder was also analyzed. 5 mg of the powder was applied to a tape and the excess powder was removed using N2 gas. The powder was analyzed in positive and negative ion mode in 4 areas ranging from 100 × 100 μm² to 500 × 500 μm². Microhardness measurements were performed using a Struers Duramin-40 Vickers microhardness tester. Microhardness was investigated by making indents with 1, 0.5, 0.3, 0.1, and 0.05 kgf loads. This was performed to evaluate the variation in the microhardness of the samples with the load. For every indentation load, 10 measurements (approximate spacing distance is 1.2 mm) were taken from the top to the bottom of the sample.

3. Results and discussion

3.1. Defects

Fig. 1 displays representative light optical micrographs of the microstructure close to the surface and in the bulk of samples 3, 30, and 22. The number of defects varied between the samples and between locations in each sample. Fig. 1a–f are regions in mounted cross-sections

Table 2

| Chemical composition of the Sandvik Osprey™ Alloy 247LC VIGA powder used in this study, the oxygen and nitrogen content were below 200 ppm, respectively. |
|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| C               | Cr              | Ni              | Co              | Mo              | W               | Ta              | Ti              | Al              | B               | Zr              |
| 0.07            | 8.5             | 8.7             | 0.6             | 9.2             | 2.5             | 0.7             | 5.5             | 0.012           | 0.01            | 1.24            |
| Si              | Mn              | S               |
| <0.01           | <0.01           | <0.003          |
of the cubes which was cut at approximately 9 mm from the surface of the cubes. The bulk areas b, d and f are located at a minimum of approximately 3 mm from the surface of the cross sections.

Overall, all samples contained cracks and voids (LOF defects and pores). Cracks in sample 3 shown in Fig. 1a were close to the surface, and the number of cracks increased closer to the bulk shown in Fig. 1b. Sample 30 displayed significantly lower crack density than sample 3, and there were no cracks close to the surface, as shown in Fig. 1c, while about two small cracks were noticeable in the bulk in the micrograph shown in Fig. 1d. As mentioned in the Introduction, internal cracks (in the bulk) can be closed by HIP, but surface cracks cannot be closed by HIP. Thus, the small cracks in the bulk of sample 30 may not be problematic if an appropriate HIP is performed. In general, crack susceptibility reduced significantly in sample 30. The micrographs are representative of other areas in the sample and testify to the low crack content produced by process K in Table 1. Sample 22 (Fig. 1e and f) predominantly displayed voids (mainly LOF defects but also some pores). While cracks were mainly associated with the LOF defects, some other cracks occurred independently of the LOF defects. It was previously proposed that the energy density (given in Table 3) revealed certain relationships with the quantity and type of defects. High energy density produces cracks, while low energy density produces voids [13].

Fig. 1. Cracks and voids observed on the cross sections of the samples. (a, c, e) Micrographs close to the surface of samples 3, 30, and 22, respectively. (b, d, f) Micrographs of the bulk of samples 3, 30, and 22, respectively. Red arrows indicate cracks, while blue arrows indicate voids (lack of fusion (LOF) defects or pores). The black arrow points to the build direction in all the micrographs. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)
The grains in all samples were columnar. Sample 3 appeared to have the largest grain size, whereas that in sample 22 was the smallest. The grain structure and sizes are further discussed in Section 3.2 using more detailed micrographs. Fig. 2a–c displays SE-mode SEM micrographs of typical defects, including LOF defects, pores, and cracks, respectively. Fig. 2d–e displays high magnification micrographs of cracks.

3.1.1. Lack of fusion defects
As discussed previously, LOF defects were prevalent in sample 22, which was manufactured with the lowest energy density. LOF defects are mainly due to a low energy density, which causes insufficient melting of the substrate or solidified powder (in a previously fused layer) to ensure epitaxial solidification from the latter [25]. Hence, LOF defects were formed between the solidified powder layers and had irregular shapes with sharp edges, as shown in Fig. 2a. Cracks may propagate from these sharp edges because of their high stress concentration [26].

3.1.2. Pores
Pores form during melting when the melt pool entraps the shielding gas [27]. They can also form when the powder entraps gases during atomization [27]. Gas pores have a characteristic spherical shape, as shown in Fig. 2b.

3.1.3. Cracks
Fig. 2c display an SEM micrograph of cracks (enclosed in the rectangle) in sample 3. As mentioned in the introduction, Alloy 247LC is prone to weld cracks, which is a major factor responsible for the non-printability of the alloy. In these samples, the solidification cracking mechanism was identified based on the surface appearance of the cracks. The crack was rough, and the surface had the characteristic dendritic appearance of solidification cracks, as shown in Fig. 2d [5]. The dendritic surface is also present in the cracks displayed in Fig. 2e. Because a large solidification temperature range increases the susceptibility to solidification cracking [5], the composition of the Alloy 247LC powder

Fig. 2. SE-mode SEM micrographs displaying (a) an LOF defect, (b) a pore, (c) cracks, (d) the dendritic surface of the cracks in (c) at a high magnification and (e) another example of cracks. The arrow points to the build direction in all the micrographs.
was used as input to Thermo-Calc (TCNi8 database) and the equilibrium and Scheil–Gulliver solidification temperature ranges were calculated. Scheil–Gulliver analysis is a non-equilibrium solidification calculation that assumes infinitely fast diffusion in the liquid but no diffusion in the solid [28]. The analysis also assumes that equilibrium conditions occur at the solid/liquid interface. The Scheil–Gulliver plot is based on equation (1) [28]:

\[ X_S = kX_L(1 - f_S)^{(k-1)} \]

where \( k \) is the partition coefficient, given as \( k = X_S/X_L \). \( X_S \) and \( X_L \) represent the equilibrium mole fractions of the solute in the solid and liquid at a particular temperature, respectively. \( X_0 \) is the initial mole fraction of the solute in the liquid at the beginning of solidification. \( f_S \) is the mole fraction of the solid. The solidification temperature ranges are shown in Fig. 3.

A large solidification temperature range is obtainable in Alloy 247LC. From the Scheil–Gulliver analysis, the solidification temperature range is 1380–750 \( ^\circ \)C (\( \Delta T = 630 \) \( ^\circ \)C), whereas from the equilibrium analysis, it is 1380–1239 \( ^\circ \)C (\( \Delta T = 141 \) \( ^\circ \)C). These ranges are comparable to a similar calculation performed for a difficult-to-print nickel-based superalloy [2]. The large Scheil–Gulliver solidification temperature range of Alloy 247LC promotes the stability of liquid between the dendrites at low temperatures. Subsequently, the high strains in the process may promote the separation of these dendrites at the most susceptible stage of solidification, where continuous liquid is available, but filling is limited [29]. The fraction of solid that is susceptible to cracking is uncertain and depends on the material and process conditions. Wang et al. [30] assumed that the material was most susceptible to cracking at 0.9 \( f_S \)–0.94 \( f_S \). Consistent with the models given by Wang et al. [30], it is useful to note that even though the Scheil–Gulliver analysis predicts a large solidification temperature range, it may not satisfactorily explain the cracks because the cracks form at high-angle grain boundaries (HAGBs), where the solidification temperature range is larger than that modeled in the Scheil–Gulliver analysis. Thus, considering the fraction of solid most susceptible to cracking (0.9 \( f_S \)–0.94 \( f_S \)), the Scheil–Gulliver temperature range of interest is 1235–1137 \( ^\circ \)C (\( \Delta T = 98 \) \( ^\circ \)C), where 1235 \( ^\circ \)C is the temperature at 0.9 \( f_S \) and 1137 \( ^\circ \)C is the temperature at 0.94 \( f_S \). Below 0.9 \( f_S \), there is sufficient liquid to fill and suppress cracks. Above 0.94 \( f_S \), there is sufficient solid bridging to resist the stresses that cause cracking. However, the grain boundary energy of the HAGBs provides additional undercooling that extends this range to >98 \( ^\circ \)C. This reasoning relies on the theory of Rappaz et al. [31] and the experiments and calculations by Wang et al. [30]. Fig. 4a shows an inverse pole figure (IPF) of cracks at HAGBs in the present study which agrees with the findings of Rappaz et al. and Wang et al. that solidification cracks occur at HAGBs. The cracks in Fig. 4a are the same cracks earlier displayed in Fig. 2e.

The cracks at approximately 11 and 37 \( \mu \)m along the thin horizontal arrow in Fig. 4a occurred at HAGBs of 44° and 37°, respectively, as plotted in Fig. 4b. Similar results were observed for all cracks examined. These observations conform to typical results for PBF of \( \gamma' \) nickel-based superalloys; for example, Chauvet et al. [2] showed that cracks propagate along HAGBs (\( >15° \)) during PBF of a difficult-to-print nickel-based superalloy. However, even if cracks appeared at HAGBs, LAGBs were more prevalent in the microstructure (as displayed in Fig. 5), which is similar to the results reported in a previous study [22]. High relative frequencies were observed at a misorientation of 1°, above which, the relative frequencies began to decrease and were very low at 15°. The HAGBs from 15° to 54° had relative frequencies close to those at 15°.

HAGBs are characterized by a high grain boundary energy, which influences the nucleation of solidification cracks. Rappaz et al. [31] initially considered a pure metal, where the critical coalescence undercooling (\( \Delta T_c \)) is given by equation (2).

\[ \Delta T_c = \left( \gamma_{\delta} - 2\gamma_{sl} \right) / \Delta S_{gb} \delta \]

where \( \gamma_{\delta} \) is the grain boundary energy, \( \gamma_{sl} \) is the solid–liquid interfacial energy, \( S_{gb} \) is the entropy of fusion per unit volume, and \( \delta \) is the thickness of the diffuse interface. Rappaz et al. defined a repulsive grain boundary energy (\( \gamma_{\delta} > 2\gamma_{sl} \)), which promotes a large undercooling \( \Delta T_c \) that can sustain a continuous liquid film down to a coalescence temperature \( T_c \). Such conditions are typically satisfied by HAGBs owing to their high grain boundary energy. It is known that the grain boundary energy increases with increasing misorientation (\( \theta \)) and eventually remains constant above a certain \( \theta, \theta_m \approx 10°–15° \) [28]. Although equation (2) is defined for a pure metal, it can approximate the behavior of alloys by compensating for back diffusion in the solid perpendicular to the

Fig. 3. Scheil–Gulliver (blue arrow) and equilibrium (black arrow) plots showing the larger solidification temperature range of the former compared to that of the latter. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)
solid–liquid interface and diffusion in the liquid parallel to the solid–liquid interface. Subsequently, an attractive grain boundary energy is defined by $\gamma_{gb} < 2\gamma_{sl}$ which is typically satisfied by LAGBs. In this case, the undercooling owing to the grain boundaries is zero, which makes the continuous liquid unstable and promotes early solid bridging of the dendrite at a relatively high temperature $T_0 (T_0 > T_1)$. The transition from attractive to repulsive grain boundary actually occurs at a critical angle $\theta_c$ when $\gamma_{gb} = 2\gamma_{sl}$. Following the approach of Wang et al. [30], $T_0$ of an alloy undergoing non-equilibrium solidification corresponds to the temperature at 0.94 $f_S$ in the Scheil–Gulliver analysis. Hence, from the Scheil–Gulliver analysis in Fig. 3, $T_0 = 1137$ °C. Following this, $T_1$ at a misorientation $\theta = \theta_m$ is given by $T_1 = T_0 - \Delta T_x(\theta_m)$. Where $\Delta T_x(\theta_m)$ can be estimated by ascribing or computing the relevant parameters in equation (2). For example, $\gamma_{gb}(\theta)$ can be calculated using the Read–Shockley equation [32]. However, this is beyond the scope of the present study. The implication of this is that the vulnerable region with a continuous liquid film will remain down to $T_1$ and is more susceptible to strains than at $T_0$. Thus, it is plausible that solidification cracking is more likely to occur at $T_1$. As mentioned above, $T_1$ is not only be influenced by $\gamma_{gb}$ but also by diffusion in the liquid and back diffusion in the solid. Furthermore, another practical implication of this line of reasoning is that the elemental segregation profile may be different because of possible variations in solute diffusion in the liquid and solid states at these grain boundaries. For example, Hariharan et al. [33] observed that the segregation profiles of HAGBs (with or without cracks) and LAGBs are different in 1-PBF-printed Alloy 738LC. An investigation of directionally solidified Alloy 247LC showed that the segregation at HAGBs was stronger than that at LAGBs, which increased the number of cracks.

Fig. 4. (a) IPF (produced parallel to the build direction). The thick arrows point to cracks on HAGBs of 44° and 37° located at two points on a thin horizontal arrow. (b) Plot showing the misorientation angles of the grain boundaries along the thin horizontal arrow. At the crack surface, the misorientation angles indicate that the grain boundaries are HAGBs of 44° and 37° and are significantly lower at other points along the thin horizontal arrow where there are no cracks. The large undercooling in the HAGB promotes the cracking.
Fig. 5. Relative frequencies of grains, in samples 3, 30 and 22, with various misorientation angles.

Fig. 6. TOF SIMS maps showing segregation of Si to cracks. The maps on the left-hand side display the pictures of the analyzed cracks; an example of a crack is indicated by the white arrow. The maps in the right-hand side are the TOF SIMS maps that display the Si segregation to the cracks; an example is indicated by the green arrow. The scale bars, displaying the distance of the analyzed points in μm, are located on the edges of the TOF SIMS maps. The bars on the right-hand side of the TOF SIMS maps are the heat maps indicating increasing Si content from the bottom to the top of the bars; the lighter the color the higher the Si content. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)
in the former [34]. Thus, any experimental investigation of elemental segregation, which possibly influences solidification cracks, should be performed around the crack surface. This ensures that the enrichment at the HAGBs responsible for the cracks is tracked. Thus, the energy-dispersive X-ray spectroscopy (EDX) elemental profile of the LAGBs cells performed by Griffiths et al. [8] may not represent the segregation profile causing cracks. Furthermore, the result may not provide the vital segregation information necessary to justify alloy modification (reduction of Hf) to reduce cracking susceptibility. In the present study, elemental segregation was investigated by performing TOF SIMS in and around the crack surface. The analysis revealed that the samples contained the elements listed in Table 2. It was observed that Si segregated to the cracks. Fig. 6 displays representative TOF SIMS maps of Si segregation to cracks in sample 3.

Pictorially, the analyzed cracks are displayed on the left-hand side of Fig. 6, while the Si ion maps indicating Si segregation to the cracks are displayed on the right-hand side of the figure. The white arrow indicates a crack, and the green arrow indicates high Si content in the region of the crack close to the tip of the white arrow. The other map also displays the segregation of Si to the cracks. The readings in the heat maps indicate that there was just a slight segregation of Si to the cracks. The Si content in the regions of the cracks can be visually enhanced by comparing Si to other elements, for example Cr and Ni, which are not present in the regions of the cracks. This is illustrated in Fig. 7 where Cr, Si and Ni are represented with red, green and blue respectively as indicated in the first column of micrographs.

In this first column, it can be observed that although Cr and Ni were present in regions of the micrographs, they were absent in the cracks. This was evidenced by the darkness of the cracks. Si can be observed to show a slight increase in the cracks which was similar to Fig. 6. The larger maps (areas of 156 μm x 156 μm and 168 μm x 168 μm) in the second column combine all the three colors of Cr, Si and Ni (red, green and blue respectively). Here, it can be clearly observed that only Si was present in the cracks; this was evidenced by the green color. The higher Si content observed in the cracks most likely promoted solidification cracking. Note that the Si detected was not due to contamination from external sources. A common source of Si is surface contamination from PDMS (Polydimethylsiloxane). However, the main peak from PDMS was not present on these surfaces as observed in Fig. 8, i.e., the Si signal was not likely a contamination from external sources, which confirmed the authenticity of the Si signal.

For the same reason, the Si did not come from the SiO2 content of the colloidal silica suspension used to polish the samples. To further demonstrate that Si was authentically present in the samples, the powder utilized to build the samples was re-analyzed by TOF SIMS. The analysis showed that the powder contained Si. Si has previously been linked to the occurrence of grain boundary cracking during the directional solidification (DS) of Alloy 247 blades [1]. Notably, Si was reduced from 0.10 wt% in crack-susceptible DS Alloy 247 to 0.03 wt% in crack-resistant DS Alloy 247LC. In another study, the Si content was shown to have the highest statistical correlation with solidification cracking during L-PBF processing of Alloy 738LC [35]. The present TOF SIMS experiment observation of Si in the cracks strengthens the finding in Engeli et al. [35] that Si was the main element possibly responsible for the cracking. Note that Alloy 247LC demonstrates a higher susceptibility to solidification cracks than Alloy 738LC but the detrimental influence of Si could be worse in the former alloy. Engeli et al. suggested that Si possibly increased the grain boundary energy or reduce the solid–liquid interfacial energy thereby promoting a large undercooling as described in equation (2). Furthermore, Microprobe and Auger spectroscopy revealed that residual elements such as Si can segregate to the grain boundaries during solidification and deteriorate the properties of superalloys [36]. The composition of Alloy 247LC powder given in Table 2, indicated that the Si content was <0.01 wt%. Although it is lower than the recommended 0.03 wt% in DS Alloy 247LC, it may not be sufficiently low to resist solidification cracking in L-PBF. Note that the thermal strains in L-PBF are higher than in DS. Thus, even though the solidifying dendrites in L-PBF could have lower amount of segregating Si compared to DS, the dendrites in L-PBF could still be susceptible to the effect of the higher thermal strains which caused cracking. Thus, the higher thermal strains may intensify the potential for the smaller amount of Si to cause cracking. It was also mentioned in Engeli et al. [35] that the rapid solidification process in L-PBF could amplify the detrimental effect of Si. In Carter [14], the Si content was 0.04 wt%, and it was assumed that this should be acceptable because it did not significantly deviate from the recommended 0.03 wt% composition. However, based on the high crack density observed in that study, the Si content may not be acceptable. Thus, it is suggested that Si could be reduced significantly in Alloy 247LC powder to eliminate the solidification cracking susceptibility. It is also worth noting that B and Zr, which typically cause solidification cracking [37], did not display segregation in the TOF SIMS analysis performed in the present study. Hf, which was also proposed in Ref. [8] as a possible detrimental element, did not display evidence of segregation in the presently investigated cracks.

The effects of stresses and strains at HAGBs and LAGBs should also be considered. Rappaz et al. [29] proposed a hot-cracking model termed the RDG model, which was used by Wang et al. [30] to calculate the
cavitation pressure drop ($\Delta P$) in the liquid along the grain boundaries of solidifying dendrites. The HAGBs demonstrated a significantly higher $\Delta P$ than that of the LAGBs. $\Delta P$ consists of stress contributions from shrinkage and deformation, and a higher value above a certain threshold promotes the nucleation of cracks [30].

In this study, solidification cracks were prominent in sample 3, but they were also observed, albeit to a lesser degree, in samples 30 and 22. The crack densities for samples 3, 30, and 22 are given for processes J, K, and L, respectively, in Table 1, which shows the higher crack density in sample 3 than in samples 30 and 22. The crack density in sample 30 was low, which was mainly due to the choice of process parameters (process K in Table 3). Details about the influence of the process parameters on

![Fig. 8. Close up spectra of Si peak (left) and lack of PDMS peak (right).](image)

![Fig. 9. SEM micrographs displaying the grain structures of samples (a) 3, (b) 30, and (c) 22.](image)
the quantity of cracks were discussed by Adegoke et al. [13]. In this study, the connection between the process parameters and the microstructure was the main concern. To determine the reason the process parameters influence the crack density, it is necessary to further investigate the grain structure, which is discussed in the next section.

3.2. Grain structure

Fig. 9a–c displays SEM micrographs of the grain structures of samples 3, 30, and 22, respectively. Columnar grains that grew epitaxially through several layers in the build direction are visible in the micrographs. Epitaxial growth occurs in L-PBF in a manner similar to that observed in welding when a layer of material is deposited on a material of a similar composition [38]. Here, the energy barrier required for heterogeneous nucleation \( \Delta G_{het}^* \), given by equation (3), is small.

\[
\Delta G_{het}^* = 16\phi \times \phi_S / \Delta G_f^2 S(\phi) \tag{3}
\]

\( \phi_S \) is the solid-liquid interfacial energy, \( \Delta G_f \) is the free energy per unit volume of liquid minus free energy per unit volume of solid, and \( \phi \) is the wetting angle. \( S(\phi) = (2 + \cos \phi)(1 - \cos \phi) / 4 \). Because the powder was melted on a base of similar composition, \( \phi \approx 0 \); therefore, \( \Delta G_{het}^* \) is negligible. Hence, new grains readily nucleated from the preexisting grains at the boundary of the melt pool. The grains then grew epitaxially in the direction opposite to the direction of heat flow. Recently, it was proposed that in addition to heterogeneous nucleation by epitaxial growth, other types of heterogeneous nucleation can occur within the melt pool away from the melt pool boundaries [39]. For example, nucleation can occur at the surface of a partially melted powder. This may explain some of the complex grain structures with different sizes found in this study (even within the same sample), as shown in Fig. 9 and other micrographs presented later. Other factors that may have contributed to this heterogeneity include the remelting and retained solidification mechanism of the previously solidified layers. These factors promote new solidification and grain growth mechanisms [39]. The rotation of the laser scans by 67° between every layer also altered the solidification and grain formation mechanisms. The grain boundaries are not readily discernible in the SEM micrographs in Fig. 9. Thus, it is necessary to employ EBSD to produce micrographs with identifiable grain boundaries and misorientation angles, which will help discern the complex grain structure highlighted above. Fig. 10a, c, and e display IPFs of samples 3, 30, and 22, respectively. The misorientation angles displayed in the figure are >10°. Pole figures are also displayed in Fig. 10b, d, and f in the same order as the IPFs.

From the IPFs, it can be visually identified that sample 22 had the smallest average grain size. This corresponds with the optical microscopy and SEM observations across the entire area of the three cross sections of each sample. This observation was verified in the Aztec crystal software by calculating the average area of all the grains in three micrographs, which spanned a total area of 0.5 mm² for each sample. The average grain sizes for samples 3, 30, and 22 were 424, 250, and 96 µm², respectively. Fig. 11 shows a chart of the grain size variation in the samples.

The trend in the plot, which is strengthened by the small standard deviation, suggests that sample 3 had a relatively large grain size, which significantly decreased in samples 30 and 22 in that order. Therefore, the grain size decreased as the power and energy density (see Table 3) decreased. Furthermore, the grain size decreased as the hatch distance increased. Thus, this result highlights the influence of the process parameters on grain size. A high power and energy density increase the melt pool size and temperature, as well as the thermal strains according to the numerical heat transfer and fluid flow models by Mukherjee et al. [40]. Consequently, the high thermal strain may increase the crack density. It is also conceivable that large melt pools, high temperatures, and the consequent slow cooling produced large grains in sample 3. Large grains are more susceptible to cracking than small grains, which was observed in directionally solidified Alloy 792 and PWA1483 [41]. It is thought that the strain is more concentrated in large grains because there are fewer boundaries. It was also suggested that the presence of a continuous eutectic melt in large grains hinders solid bridging, which increases the cracking susceptibility. According to this theory, sample 22 should have had a lower crack density than that of sample 30 because the former had a smaller grain size. However, this was not the case because the low energy density in sample 22 led to high levels of LOF defects, and the sharp edges of the LOF defects are associated with strains that cause cracks [13,42]. The cracks associated with LOF defects are not the solidification cracks discussed above, which are influenced by the grain size. An increase in cracks associated with LOF defects at low energy densities was also observed by Carter [14]. It is suggested that the process parameters applied for sample 30 produced a microstructure consisting of suitable grains with sizes that have low susceptibility to cracking and can grow epitaxially through several layers (thereby preventing LOF defects). The aspect ratios of the grains were calculated using The Aztec software and are displayed in Fig. 12. A high aspect ratio indicates strong columnar grains, while a low aspect ratio indicates a more equiaxed grain. Columnar grains are more susceptible to solidification cracking [43].

Although sample 30 appeared to have the lowest aspect ratio, that is, least columnar grain morphology, the values were close, and there was no significant trend. It is also not apparent from Fig. 10 that sample 30 had a less columnar morphology than those of the other samples. The textures of the various samples were also measured. The IPFs in Fig. 10a and c indicate that the textures of samples 3 and 30 were predominantly oriented in the <001> direction, which is also the build direction. Compared to samples 3 and 30, sample 22 displayed less texture in the <001> direction, as shown in Fig. 10e. The texture is also evident in the <001> pole figures displayed in Fig. 10b, d, and f. The contours of multiples of uniform density (MUD) (which reached 9.77, 7.22, and 5.41 for samples 3, 30, and 22, respectively) correspond to the strength of the texture in the samples. Here, sample 3 exhibited the strongest texture while sample 22 displayed the weakest. Two other micrographs were analyzed for each sample. Sample 3 and 30 displayed the strongest texture while sample 22 displayed the weakest texture.

When the microstructures of samples 3, 30, and 22 displayed in Fig. 9 were observed at higher magnification using SEM, cells with different structures and sizes were visible. The cells varied in structure and size within each sample and across different samples. Some of the cells and sub-grain structures observed in the samples are shown in Fig. 13.

Fig. 13a shows an example of the cells in sample 22. The middle of the micrograph consists of smaller cells with lower aspect ratios (indicated with a blue arrow) than those at the edge of the micrograph, which are long with high aspect ratios (indicated with a red arrow). This attests to the varying solidification conditions and cooling rates within the sample. This complex heterogeneity is further revealed in Fig. 13b. Here, equiaxed grains (an example indicated with a blue arrow) are present, which are surrounded by sub-grains that contain cells with varying sizes and aspect ratios (indicated with red arrows). The boundaries of some sub-grains are outlined in red. The equiaxied grains do not have sub-grains or cells within them, as shown in the high-magnification image in Fig. 13c. An example of the cells observed in sample 30 is shown in Fig. 13d. The cells in the micrograph have long columnar structures with bright microconstituents on the boundaries of the cells. Here, the average cell size is approximately 500 nm, but cell sizes ranging from 300 nm to 1 µm were observed in the three samples. To calculate the potential cooling rate in the cells, equation (4) was applied (Harrison et al. [44]).

\[
\lambda = 9(7 \pm 5) T^{-0.36(0.01)}
\]

where \( T \) is the cooling rate, and \( \lambda \) is the cell size. Cell sizes of between
Fig. 10. IPFs (produced parallel to the build direction) displaying the variations in the texture and grain size of samples (a) 3, (c) 30, and (e) 22. The black scale bar corresponds to 100 μm. Pole figures displaying the variation in the texture of samples (b) 3, (d) 30, and (f) 22.
300 nm and 1 μm yielded cooling rates of approximately between $3.6 \times 10^5$ and $1 \times 10^7$ K/s. Therefore, cooling rates between these two values were possible. These high cooling rates are typical of those obtained by L-PBF. Harrison et al. [44] suggested that rapid cooling up to approximately $3 \times 10^5$ K/s may lead to solute trapping, which prevents microsegregation during L-PBF of superalloys. Calculations of L-PBF of Hastelloy X indicated that the solute will not diffuse fast enough to cause microsegregation and that the rapid temperature drop will further suppress diffusion. The rapid cooling rate attained in L-PBF suggests that the local equilibrium condition at the solid–liquid interface assumed in the Scheil–Gulliver analysis may not be valid. Thus, solute trapping is feasible. Contrary to Harrison et al. solute trapping did not occur in the present investigation, which is evident from the TOF SIMS microsegregation of Si presented previously. This is further supported by the microconstituents formed in the samples, which is discussed in Section 3.3. These microconstituents formed from elements that segregated to the cell boundaries. The high-resolution transmission electron microscopy (TEM)–EDX investigations by Wang et al. [20] and Divya et al. [22] also indicated that microsegregation occurred. According to Porter [28], microsegregation occur during solidification when a planar interface breaks down into cells. Such a condition is as a result of constitutional supercooling, and it occurs when equation (5) is satisfied.

$$\frac{G}{R} < \frac{\Delta T}{D}$$

where $G$ is the thermal gradient, $R$ is the solidification rate, $\Delta T$ is the equilibrium solidification temperature range, and $D$ is the diffusion coefficient. The cells are thereby characterized by a gradient of element concentration from the cell core to the cell boundaries.

### 3.3. Phases and microconstituents

When the samples were observed at a higher magnification than those of the micrographs in Fig. 13, the bright microconstituents at the cell boundaries were more visible. These microstructural features are labeled and displayed in Fig. 14.

The phases and microconstituents in Fig. 14 are representative of those observed in samples 3, 30, and 22. In this micrograph, the cells are approximately 1 μm in size, and no deleterious phases, such as the topologically close-packed (TCP) σ phase, are noticeable. Such phases could deplete the surrounding microstructure of alloying elements or serve as crack nucleation sites, which could be detrimental to the mechanical properties. The microstructure consists of cell cores and cell boundaries with bright microconstituents and even brighter spherical precipitates, which resemble the γ/γ′ eutectic and MC carbides (and/or oxides), respectively, previously identified by TEM by Wang et al. [20] and Divya et al. [22]. γ′ was not observed, although the relatively high microhardness values of the samples indicate that it was present. The microhardness values of these samples at 0.5 kgf were measured previously by Adegoke et al. [13]. The microhardness values of samples 3, 30, and 22 were re-measured at various loading conditions to investigate the variation of microhardness with load. The findings are presented in Section 3.4.

### 3.4. Microhardness

The variation in the microhardness with the load may indicate a difference in the micromechanical characteristics of the grains and cells.
under different loading conditions. Fig. 15 shows the measured microhardness values of the samples. Generally, the microhardness values were relatively high at all loads, demonstrating that the samples possessed high strength. Sample 3 generally displayed microhardness values higher than those of samples 30 and 22, suggesting that sample 3 offered improved strengthening.

Fig. 13. BSE-mode SEM micrographs showing cells and sub-grains with different sizes and structures. (a) Cells with different sizes and aspect ratios are indicated by the arrows, (b) equiaxed grains surrounded by sub-grains with cells, (c) high-magnification micrograph of the equiaxed grains, and (d) long columnar cells within sub-grains.

Fig. 14. SEM image in the BSE mode displaying cells with bright microconstituents at the cell boundaries. BD is build direction.
The samples displayed higher microhardness values at lower loads, which was possibly due to the indentation size effect (ISE) proposed by Nix and Gao [45]. A similar trend was recently revealed in L-PBF-printed Hastelloy X [46]. However, Balos et al. [47] showed that increasing the indentation load caused microhardness to increase in L-PBF of MS1 maraging steel. This phenomenon was termed reverse ISE. The fact that it was revealed that either ISE or reverse ISE was a possible mechanism makes it pertinent to clarify what occurs in the case of LPBF of Alloy 247LC. As mentioned previously, it was ISE that was observed in the present investigation. Furthermore, microhardness measurements in the literature are generally presented at different loads, which makes comparison complicated without knowledge of the indentation characteristics at different loads. Therefore, a pre-experiment is required to identify the values of the load where microhardness is load independent [47]. In the present study, it appears that above 0.5 kgf, the average microhardness was relatively independent of the applied load. Thus, it is reasonable to apply a load \( \geq 0.5 \) kgf to obtain microhardness measurements that do not significantly vary with the value of the applied load.

4. Conclusion

In the present study, the microstructure of L-PBF printed Alloy 247LC was characterized and the cracking mechanism detected. It was demonstrated that improving the process parameters can produce a good microstructure and significantly reduce the crack susceptibility. The following conclusions were made.

- The material printed with a process parameter combination corresponding to an energy density of 108 \( J/mm^3 \), which reduced crack susceptibility, produced the typically complex L-PBF grain structure but contained the desired phases without noticeable detrimental phases. This material condition is promising for post L-PBF processing to meet the demands of gas turbine applications.
- The increased cracking susceptibility of the printed material at an energy density of 196 \( J/mm^3 \), was most likely due to the larger grain size observed in the increased crack susceptible material compared to the grain size observed in the reduced crack susceptible material.
- The cracking mechanism was solidification cracking.
- The solidification cracks occurred at high angle grain boundaries where solidification cracking susceptibility was higher due to the undercooling contributions of the grain boundary energy.
- Si was present in the cracks. This means that Si segregated to several high angle grain boundaries during solidification, and most likely promoted the solidification cracking.
- The microhardness of the printed materials depends on the applied load. Low indentation load produces high microhardness and vice versa.

Data availability

The raw data/processed data required to reproduce these findings cannot be shared at this time as the data also forms part of an ongoing study.

Credit author statement

Olutayo Adegoke: Conceptualization; Formal analysis; Investigation; Methodology; Project administration; Validation; Visualization; Roles/writing – original draft; Writing – review & editing. Joel Andersson: Funding acquisition; Methodology, Project administration; Resources; Supervision; Writing – review & editing. Håkan Brodin: Resources, Project administration, Robert Pederson: Funding acquisition; Project administration; Resources; Supervision; Writing – review & editing. Peter Harlin: Resources, Writing – review & editing.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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