

# Pore evolution mechanisms during directed energy deposition additive manufacturing

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## Abstract

Porosity in directed energy deposition (DED) deteriorates mechanical performances of components, limiting safety-critical applications. However, how pores arise and evolve in DED remains unclear. Here, we reveal pore evolution mechanisms during DED using *in situ* X-ray imaging and multi-physics modelling. We quantify five mechanisms contributing to pore formation, migration, pushing, growth, removal and entrapment: (i) bubbles from gas atomised powder enter the melt pool, and then migrate circularly or laterally; (ii) small bubbles can escape from the pool surface, or coalesce into larger bubbles, or be entrapped by solidification fronts; (iii) larger coalesced bubbles can remain in the pool for long periods, pushed by the solid/liquid interface; (iv) Marangoni surface shear flow overcomes buoyancy, keeping larger bubbles from popping out; and (v) once large bubbles reach critical sizes they escape from the pool surface or are trapped in DED tracks. These mechanisms can guide the development of pore minimisation strategies.

## 32 **Introduction**

33 Directed energy deposition (DED) <sup>1</sup> is a promising layer-by-layer additive  
34 manufacturing (AM) technology that fabricates complex geometries for high-value-  
35 added products <sup>2</sup>. DED is also applied to repair applications, such as the repair of  
36 damaged turbine blades <sup>3</sup>. However, the industrialisation of the DED process for  
37 applications in automotive, marine, aerospace and biomedical fields has been limited  
38 by porosity introduced during the process, as porosity can be detrimental to a  
39 component's final mechanical performance, especially fatigue life <sup>4</sup>.

40 Porosity is a common feature in DED-produced components and has been observed in  
41 various alloys <sup>5-7</sup>, including titanium alloys <sup>6,8,9</sup>, nickel-based superalloys <sup>7,10</sup> and  
42 aluminium alloys <sup>11</sup>. Porosity mainly consists of gas porosity and lack of fusion features,  
43 categorised by their formation mechanisms <sup>12</sup>. Gas porosity can originate from  
44 feedstock, entrapment of shielding gas <sup>12</sup>, and the evolution of gases such as hydrogen  
45 which are less soluble in the solid than the liquid metal <sup>13,14</sup>. A lack of fusion porosity  
46 can be formed due to insufficient energy input <sup>15</sup>. Porosity in DED is generally  
47 investigated with *ex situ* observation techniques including metallographic observation  
48 and X-ray computed tomography <sup>16-19</sup>. However, these techniques fail to capture either  
49 the phenomena by which pores form, or the dynamics of their growth and migration.  
50 To develop high-performance DED components with minimal porosity, it is necessary  
51 to gain a clear understanding of pore evolution and dynamics mechanisms using *in situ*  
52 observations.

53 Many *in situ* X-ray imaging studies have been conducted to investigate dynamic  
54 phenomena during solidification<sup>13,20-25</sup>, including the molten pool behaviour in laser  
55 powder bed fusion (LPBF)<sup>26-36</sup>, but only few have been performed on DED<sup>5,6,10,37</sup>.  
56 Pore formation was studied during LPBF by combining *in situ* synchrotron X-ray  
57 imaging and multi-physics modelling, and it was found that the high thermocapillary  
58 force can eliminate pores from the melt pool<sup>31</sup>. Pores were also found to be formed at  
59 the end of the scan vector during laser turning due to the formation and subsequent  
60 collapse of deep keyhole depressions, such that pockets of inert shielding gas are  
61 trapped by the solidification front<sup>38</sup>. Two studies systematically investigated pore  
62 formation during LPBF using high-speed X-ray imaging<sup>30,33</sup>. It was found that pore  
63 formation can be caused by a critical instability at the bottom of the keyhole<sup>30</sup>. However,  
64 this mechanism does not apply to the DED process which has a larger laser spot size  
65 and a lower energy density than LPBF. Hence DED is normally in conduction mode  
66 with no keyhole<sup>2,39</sup>, has a much larger molten pool<sup>40,41</sup>, and includes powder  
67 bombardment<sup>42</sup> which can contribute to different bubble evolution and melt pool  
68 dynamics. In DED, Wolff *et al.*<sup>5</sup> reported pore formation mechanisms as a result of  
69 powder delivery, keyhole dynamics, melt pool dynamics and shielding gas in Ti-6Al-  
70 4V using a piezo-driven powder delivery DED system; however, the energy density  
71 used was much greater than many industrial-scale DED builds with a keyhole formed,  
72 and hence some of the phenomena observed were more typical of the LPBF process.  
73 Therefore, there is a strong demand in getting results with industrial-relevant conditions

74 and at a high temporal resolution to explain these pore mechanisms and physics  
75 involved in DED.

76 Similarly, there have been many multi-physics and multi-scale models of the LPBF  
77 process <sup>43-48</sup>, but for DED models, there have been a relatively limited number of  
78 studies using high-fidelity multi-physics models <sup>49-54</sup>. Mostly, materials deposition and  
79 layer accumulation <sup>51,52</sup> and melt pool flow field <sup>53</sup>, have been discussed. Additionally,  
80 Yang *et al.* <sup>49</sup> modelled the flow characteristics during DED, and ultrasound-assisted  
81 DED, using multi-physics modelling, coupled with high-speed optical imaging, but not  
82 X-ray. Two of the current authors developed a high-fidelity physics-based simulation  
83 to capture the chemical mixing between titanium and dissimilar refractory metals and  
84 its corresponding thermal-fluid characteristics during the DED <sup>50</sup>. However, these DED  
85 models did not include the formation, migration and release of pores, although models  
86 of these phenomena were well established in LPBF models and the wider field of  
87 solidification modelling <sup>20,48,54,55</sup>. For example, Li *et al.* <sup>48</sup> numerically investigated pore  
88 dynamics in LPBF such as coalescence and surface escape. This study is very  
89 suggestive of the pore dynamics and its effect on the product quality, but the melt pool  
90 scale was smaller (width of  $\sim 100 \mu\text{m}$ ), and the bubble buoyancy effect and the  
91 temperature dependence of thermo-physical properties were not included.

92 In DED, where the melt pool is larger for the conduction mode under representative  
93 industrial conditions, the bubble size could be larger, and the effects of melt flow and  
94 buoyancy still remain unanswered. It is worth investigating since the buoyancy force is

95 proportional to the cube of the bubble diameter. Furthermore, the effect of powder  
96 bombardment is particular to DED, which adds disturbance to the bubble dynamics.  
97 Therefore, the bubble dynamics in DED should be investigated comprehensively. In the  
98 numerical simulation, the melt pool velocity field information can be directly obtained,  
99 and the extraction of each specific effect could be possible.

100 One of the key issues is that these DED models were mainly validated with high-speed  
101 optical and thermal imaging results, and limited to the surface-based phenomena<sup>49,56,57</sup>.  
102 Importantly, these models benefit significantly from high-resolution and high-speed X-  
103 ray imaging experimental results to both determine the key physics to include and for  
104 validation. Therefore, it is critical to reveal the pore and melt pool dynamics in DED  
105 by combining high-fidelity multi-physics modelling and *in situ* X-ray imaging  
106 experiments.

107 In this work, we perform *in situ* high-speed synchrotron X-ray imaging (>20 kHz) to  
108 investigate pore evolution mechanisms during DED-AM. We quantify the pore  
109 behaviour including formation, coalescence, pushing, migration, escape and  
110 entrapment in the radiographs. We also quantify how these phenomena are correlated  
111 to key DED processing parameters. A multi-physics and high-fidelity modelling is  
112 applied to validate the hypothesised mechanisms including bubble migration,  
113 coalescence, pushing and escape. Our work contributes to an in-depth understanding of  
114 the DED additive manufacturing process, providing insights into how pore  
115 minimisation strategies may be developed.

116 **Results & Discussion**

117 **Pore behaviour in DED.** *In situ* high-speed synchrotron X-ray imaging was used to  
118 observe pore dynamics and formation during the DED-AM. The experiment was  
119 performed using a Blown Powder Additive Manufacturing Process Replicator version  
120 II (BAMPR-II) on the ID19 beamline at the European Synchrotron Radiation Facility  
121 (ESRF) (details of the BAMPR II system and experimental set up can be found in  
122 Methods and Supplementary Fig. 1 and references about BAMPR<sup>6,9,10</sup>). The powder is  
123 the gas atomised nickel-based superalloy RR1000 with a D50 size of 90  $\mu\text{m}$  (see  
124 Supplementary Information for composition and size distribution in Supplementary Fig.  
125 2 and Supplementary Table 1).

126 Based on the observations made using high-speed synchrotron radiography, pore  
127 behaviour can be divided into five stages: 1. pore formation; 2. bubble coalescence and  
128 growth; 3. solid/liquid interface pushing of large bubbles; 4. large bubble entrainment  
129 in the molten pool; and finally, 5. bubble escape or entrapment (see Fig. 1):

130 (1) Pore formation. Pores are observed to be generated predominantly from the  
131 feedstock when using gas atomised powders. When these powders are atomised using  
132 argon gas, small bubbles of argon are entrained at the centre of many of the powder  
133 particles. These bubbles of argon are released into the molten pool when the powders  
134 melt. As shown in Fig. 1a and Supplementary Movie 1, at  $t = t_0$ , a powder particle,  
135 marked with a blue circle, hits the melt pool and is partially submerged. At  $t = t_0 + 3$   
136 ms, as the powder melts into the melt pool, and the argon bubble, marked with a yellow

137 circle, is transferred into the pool (see schematic in Fig. 1b). The second largest source  
138 of pores is from the melting of the previous track (see Fig. 1g and i, where the pore,  
139 marked with purple, is released from the previous layer into the melt pool). As will be  
140 demonstrated later, the pores in the previous tracks are feedstock argon pores that have  
141 been transferred to the molten pool and then captured by the solidification front and  
142 frozen into the track.

143 (2) Bubble coalescence and growth. We observed that small bubbles can coalesce into  
144 larger ones ( $t = t_0 + 146$  ms) (Fig. 1c). The bubbles formed by the coalescence of a  
145 couple of feedstock bubbles recirculate with the Marangoni flow in the melt pool and  
146 continue to grow by coalescing with more small bubbles (at  $t = t_0 + 150$  ms, the bubble  
147 marked in a yellow circle). The bubble, marked with a yellow arrow and circle, is  
148 observed to migrate from the recirculating flow of the front to the back of the melt pool  
149 ( $t = t_0 + 146$  ms), and then coalesce with the bubble in the back ( $t = t_0 + 150$  ms). This  
150 large bubble at the rear of the molten pool is formed by the coalescence of tens of  
151 feedstock bubbles, and surprisingly remains relatively stable in the flow for relatively  
152 long periods of time ( $\sim 0.5$  s in this condition), growing and being released in a periodic  
153 cycle, as discussed later. Fig. 1d shows the schematic of bubble circulation and bubble  
154 lateral movement. Quantification of their instantaneous circulation velocities is  
155 discussed later in the bubble migration section.

156 In Fig. 1a-d, the outward Marangoni flow is expected to occur in the melt pool, as the  
157 surface temperature is the highest under the laser than near the edge of the melt pool,

158 so the liquid flows from this low surface energy area out to the colder edges (higher  
159 surface energy) to minimise the free energy. This creates a very fast strong surface flow  
160 outward from the laser, creating recirculation flow cells at the front and back <sup>58</sup>. Bubbles  
161 are observed to follow the outward Marangoni flow in the melt pool. Based on the 2D  
162 projections of the events (Fig. 1c), in the front and back regions of the melt pool ( $t = t_0$   
163 + 146 ms and  $t_0 + 150$  ms), bubbles are observed to recirculate, driven by the Marangoni  
164 flow.

165 (3) Solid/liquid interface pushing of bubbles. *In situ* radiography has been used to show  
166 that an advancing solid-liquid interface can either push or capture bubbles <sup>13</sup>; both  
167 pushing and capture mechanisms were observed here. For the smaller bubbles (25-  
168 40  $\mu\text{m}$ ), many were captured by the solidification front at the rear of the melt pool,  
169 forming pores in the track, such as the pores marked with green circles in Fig. 1e.  
170 However, the large coalesced bubbles are pushed by the solidification front during the  
171 steady state, as shown in Fig. 1e, where a large bubble is pushed by the solidification  
172 front near the solid/liquid interface in the back of the melt pool (from  $t = t_0 + 269$  ms to  
173  $t_0 + 440$  ms). This large bubble continues to grow as smaller bubbles flow from the rest  
174 of the pool and then coalesce with it. Surprisingly, this bubble remains in the melt pool,  
175 rather than rising under a strong buoyancy force.

176 (4) Large bubble entrainment in the molten pool. We observed that the large bubbles at  
177 the rear are pushed along ahead of the solidification front, and surprisingly do not rise  
178 to pop at the melt surface, remaining entrained in the molten pool (see bubble marked



179 with a yellow circle in Fig. 1g). We hypothesise that the large pores are kept in the pool  
180 by the downward force exerted on them by the very fast-moving Marangoni surface  
181 shear flow that gets compressed above the bubble as it flows outwards over it (see  
182 schematic in Fig. 1h). The bubble appears to be maintained stably in the flow by the  
183 equal and opposite forces, until it exceeds a critical size.

184 (5) Bubble escape or entrapment in the solid. We also captured what happened to the  
185 bubbles in the end. Some bubbles escape from the melt pool. When the large bubbles  
186 grow beyond a critical size ( $\sim 120 \mu\text{m}$  in diameter) by coalescence, the upward  
187 buoyancy force overcomes the downward Marangoni shear flow force, and the bubbles  
188 rise to the melt pool surface and burst, as shown in Fig. 1i and j. Using fast 20 kHz  
189 frame rate imaging (Supplementary Fig. 3), the large bubble escape process is clearly  
190 observed, namely, the large bubble moves close to the melt pool surface, coalesces with  
191 the melt pool surface and then bursts. Interestingly, many of the recirculating small  
192 bubbles burst as they reach the surface (detailed calculation can be found in the bubble  
193 escape section), perhaps due to the reduced blockage of the Marangoni shear flow as  
194 compared to larger bubbles.

195 As already discussed, the small bubbles are often entrapped in the solid-liquid interface,  
196 while the larger bubbles are usually pushed during the steady state. However, when the  
197 laser is turned off at the end of the track (Fig. 1k and l), both small and large bubbles  
198 are often captured by the solidification front towards the end of the track, as the front  
199 becomes less planar (and often more dendritic as the thermal gradient reduces). This

200 observation explains the propensity of large pores being found at the end of the track

201 <sup>59</sup>, a phenomenon confirmed by our tomography results (Supplementary Fig. 4).

202 These five stages of bubble behaviour depict the life cycle of bubbles in DED AM, and

203 we observed that they repeat periodically during the building process, as discussed

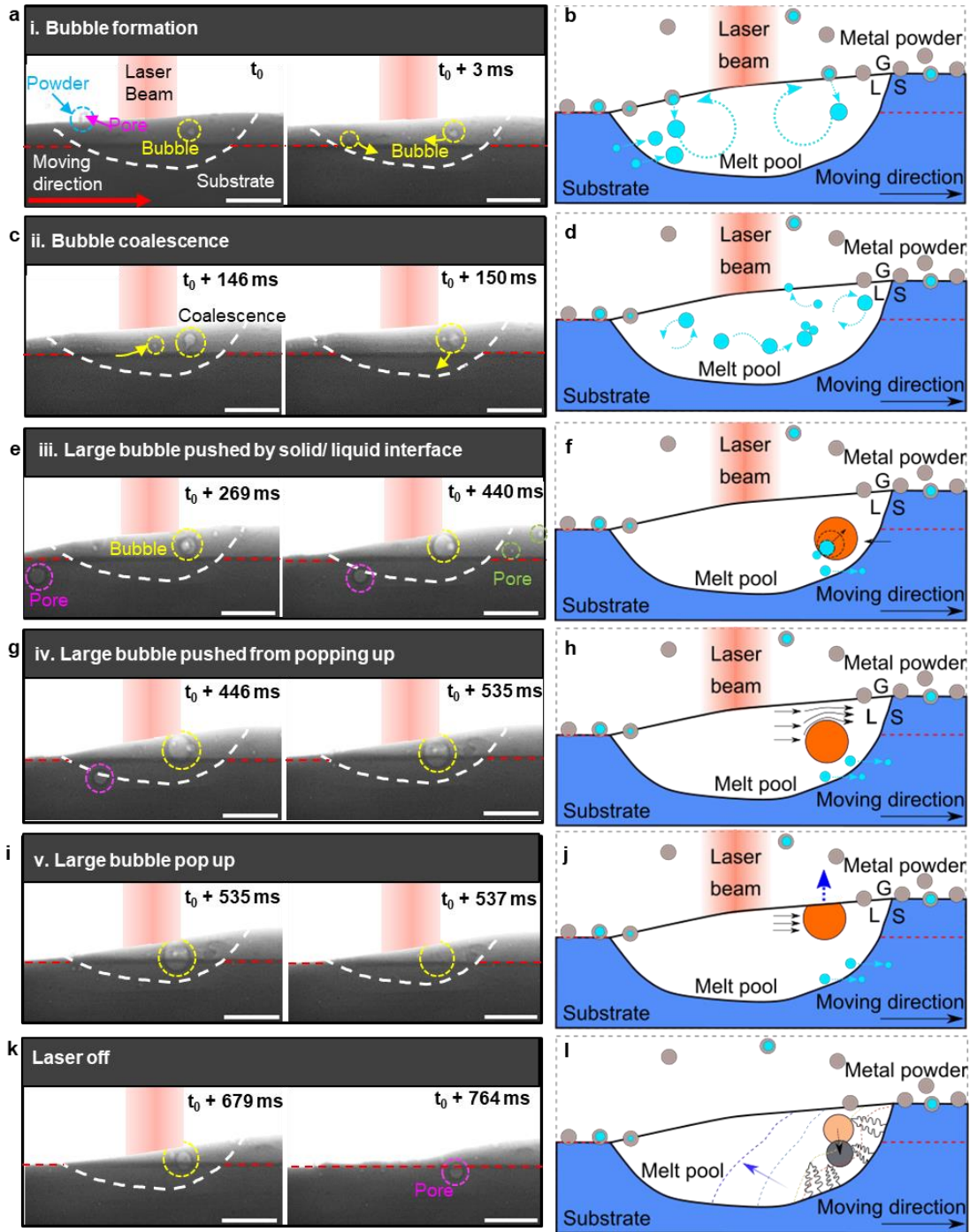
204 below in the bubble growth section.

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210 **Fig. 1 Dynamic bubble behaviour and mechanisms during DED.** **a, b** Radiographs with  
 211 associated schematic showing a bubble formed from an argon pore inside a powder particle.  
 212 Small bubbles are entrained in the recirculating flows in the melt pool. G represents gas, L  
 213 represents liquid, S represents solid in the schematic. **c, d** Radiographs with associated  
 214 schematic showing small bubbles coalescing into a larger bubble. Small bubbles often migrate  
 215 from the front to the rear of the recirculating flows in the melt pool. **e, f** Radiographs with

216 associated schematic showing a large bubble pushed by solid/liquid interface, growing as small  
217 bubbles coalesce into it. **g, h** Radiographs with associated schematic showing a large bubble  
218 entrained in the melt pool, prevented from bursting at the surface by the squeezed Marangoni  
219 shear flow. **i, j** Radiographs with associated schematic showing the large bubble (yellow circle)  
220 bursting at the melt pool surface after it reaches a critical size. **k, l** Radiographs with associated  
221 schematic showing the large bubble trapped by the solidification front when the laser is turned  
222 off. The substrate traverse speed is  $2 \text{ mm s}^{-1}$ , the laser power is 160 W, layer 1. The laser beam  
223 in the X-ray radiographs and corresponding schematics are shown in red colour, and the laser  
224 beam location is nearly symmetrical to the melt pool geometry, while it is slightly in the forward  
225 of the centre due to the advection of heat. See the video corresponding to **a, c, e, g, i, k** in  
226 Supplementary Movie 1. Scale bars in radiographs:  $300 \mu\text{m}$ .

227 **Pore formation mechanisms in DED-AM.** We observed from the radiographs that  
228 pores mainly form from two sources. The first and dominant source is the gas atomised  
229 powder feedstock. Argon pores present in the powder feedstock are transferred into the  
230 molten pool as the powders melt. Fig. 2a captures the phenomenon in detail as a powder  
231 particle hits the molten pool surface and the pore transfers into the melt pool after about  
232 1 ms after the particle melts. Similar phenomena can be observed in the pore formation  
233 process Aii in Fig. 2a. The second source of porosity is the track material which is laid  
234 down on, initially a substrate machined from a DED-AM produced block, and after the  
235 first build, prior tracks. The substrate and prior tracks contain small pores that are  
236 clearly visible in the radiographs (Fig. 2a), and they are released into the melt pool  
237 when the laser beam remelts the substrate / prior track (Fig. 2b).

238 For the conditions used in this study, namely a gas atomised powder and conduction

239 mode laser power, feedstock porosity is the dominant source of porosity. This was  
240 quantified by counting the newly formed pores over 100 ms of the build for each source  
241 (Fig. 2d), with the argon pores in the feedstock powder introducing 2 to 4 times as many  
242 as pores enter from all other sources. The only other source of bubbles we observed  
243 was those entering from the prior tracks (Fig. 2d). However, reference <sup>5</sup> suggested that  
244 during DED-AM of Ti-6Al-4V porosity can be generated from the feedstock, keyhole  
245 collapse, and by entraining gas when the powder particles enter the pool. For their  
246 conditions, using plasma atomised powders and laser conditions creating a keyhole,  
247 they concluded that feedstock porosity is a relatively insignificant contribution to the  
248 process with a contribution ratio of 0.22% <sup>5</sup>. Our results show that for the more  
249 industrial conditions used here, feedstock porosity becomes the major source of pores  
250 rather than a negligible one. This would be the one of major differences in pore  
251 formation between this work and the prior study <sup>5</sup>.

252 The pore formation rate, defined here as the number of pores formed in the melt pool  
253 per milli-second, is shown in Fig. 2e. For the two build velocities used, this graph shows  
254 the pore formation rate from feedstock powder is 2 to 4 times higher than the bubble  
255 uptake from previous layers. It also shows the pore formation rate from powders is  
256 similar for both 1 and 2 mm s<sup>-1</sup>, as expected since the powder feed rate, and hence the  
257 source of pores, is the same. However, a higher pore formation rate from porosity in  
258 previous layers is observed at 1 mm s<sup>-1</sup> than 2 mm s<sup>-1</sup>. This is probably due to the smaller  
259 pool size at the higher speed, and hence less remelted material entering the pool. Further,

260 the porosity in the previous layers is greater at  $1 \text{ mm s}^{-1}$ .

261 Unlike the reference <sup>5</sup>, we did not observe any keyhole porosity in our experiment as

262 we operated in a ‘conduction’ mode, with an energy density closer to industrial

263 standards. Nor did we observe any pores formed from the delivery gas or entrained gas

264 on particle bombardment. Powder particles are observed to gradually melt into the melt

265 pool after they hit the melt pool surface and create ripples, see Supplementary Movies

266 2 and 3. Note, keyholes normally occur in the laser powder bed fusion process rather

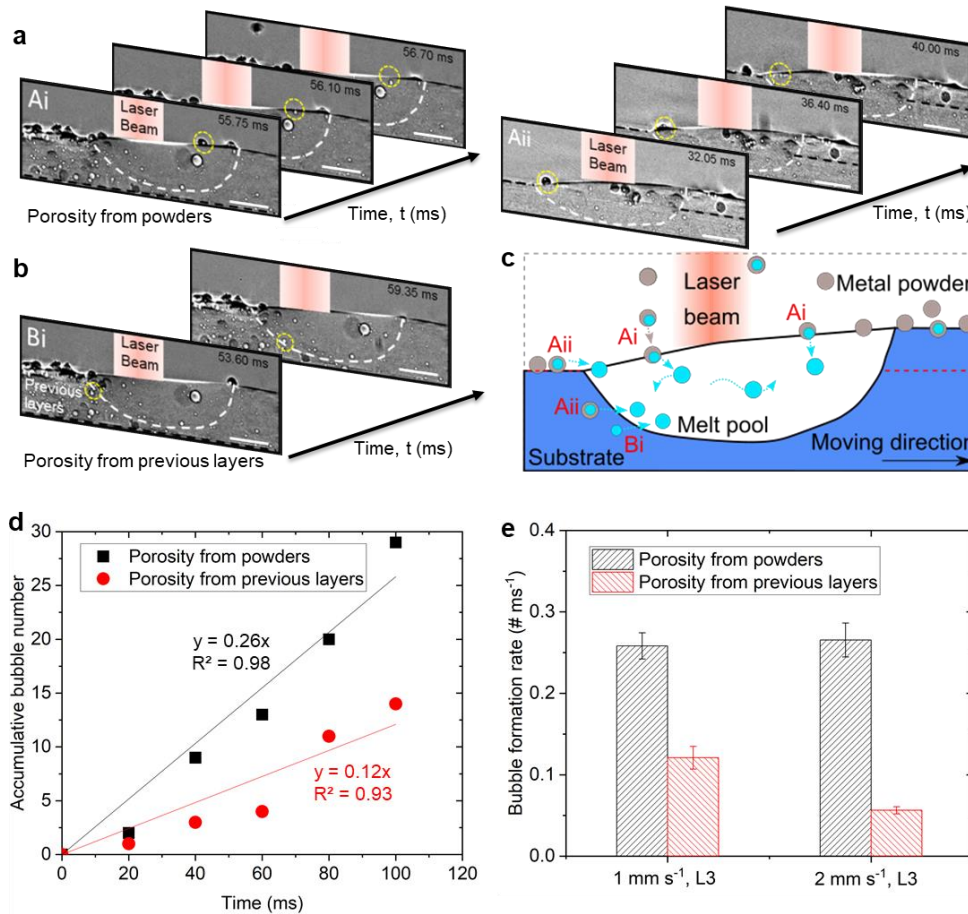
267 than DED, as LPBF is normally operated with a much higher laser power density <sup>33</sup>.

268 Further discussions about the comparison between our work with the previous work <sup>5</sup>

269 and the industrial DED can be found in Supplementary Discussion 1 and

270 Supplementary Table 2.

271



272

273 **Fig. 2 Pore formation mechanisms during DED.** **a** Pore formation dynamics. **Ai**, a pore was  
 274 captured to form in the melt pool from the porosity in the powder feedstock particle at a  
 275 substrate traverse speed of  $1 \text{ mm s}^{-1}$ , a laser power of 160 W, layer 3; **Aii**, a pore formed from  
 276 a powder when the laser melts the powders at a substrate traverse speed of  $1 \text{ mm s}^{-1}$ , a laser  
 277 power of 160 W, layer 1. **b** **Bi**, a pore formed from the porosity in the previous layers. **c**  
 278 Schematic illustration of the pore formation mechanism at a traverse speed of  $1 \text{ mm s}^{-1}$ , a laser  
 279 power of 160 W, layer 3. **d** Accumulative number of pores from powders and previous layers  
 280 with increasing time in DED at a traverse speed of  $1 \text{ mm s}^{-1}$ , a laser power of 160 W, layer 3. **e**  
 281 Pore formation rate from porosity in powders and previous layers in DED at a traverse speed  
 282 of  $1 \text{ mm s}^{-1}$ , a laser power of 160 W, layer 3; a traverse speed of  $2 \text{ mm s}^{-1}$ , a laser power of 160  
 283 W, layer 3, respectively. Error bars represent standard deviation. See the videos corresponding  
 284 to **a-c** in Supplementary Movies 2 and 3. Scale bars in **a-b** are  $300 \mu\text{m}$ .

285 **Bubble growth and pushing mechanisms.** We carefully measured the bubble diameter

286 changes against different processing conditions (including traverse speed, laser power  
287 and layers of build tracks) as an indication to understand the bubble growth mechanism,  
288 as shown in Fig. 3. Fig. 3a shows the bubble diameter changes against different  
289 substrate traverse speeds. It is observed that the large bubble behaviour is periodic, with  
290 bubbles growing to a critical size and then escaping, with a new large bubble then  
291 forming in a similar location, repeating the process over the recorded distance of the  
292 build. The sudden diameter drops in Fig. 3a indicate the time when the large bubble  
293 escapes. The phenomena are compared for two different traverse speeds, 1 and 2 mm s<sup>-1</sup>.  
294 We discovered that the maximum diameter a bubble can reach is about 180 μm at a  
295 traverse speed of 1 mm s<sup>-1</sup>, which is larger than the condition at a traverse speed of 2  
296 mm s<sup>-1</sup>, where a maximum bubble diameter is measured to be about 120 μm. It is  
297 speculated that this is due to the changes in the Marangoni flow and buoyancy force in  
298 the larger and deeper melt pool at a traverse speed of 1 mm s<sup>-1</sup>. At a traverse speed of 2  
299 mm s<sup>-1</sup> the large bubbles travel approximately the same periodic distance (in about half  
300 the time before escaping) as compared to 1 mm s<sup>-1</sup>. By counting the bubble number for  
301 coalescence, it is found that the average number of initial bubbles to coalesce the largest  
302 bubbles is over 30 at 2 mm s<sup>-1</sup> and over 70 at 1 mm s<sup>-1</sup>, indicating that the bubble  
303 coalescence consumes a large number of bubbles. Since the largest bubbles volumes at  
304 1 mm s<sup>-1</sup> were over double the largest bubbles volumes at 2 mm s<sup>-1</sup>, they were probably  
305 formed by approximately double the number of smaller bubbles. The growth of the  
306 large pore provides convincing evidence of bubble coalescence, and although there may



307 be some overlap of the bubble through the thickness, the high frame rate data shows  
308 small bubbles touch the larger pore and disappear, also providing strong evidence of  
309 coalescence, as shown in Supplementary Movie 3 at about  $t = 41$  and  $48$  ms.

310 From the observation, we noted that melt pool size is an important factor for the bubble  
311 growth dynamics. The maximum bubble diameter is larger at  $1 \text{ mm s}^{-1}$  than that at  $2$   
312  $\text{mm s}^{-1}$ . As shown in Fig. 3b, both the depth and width of the melt pool are larger at  $1$   
313  $\text{mm s}^{-1}$ , and the volume for bubble growth is larger, so the maximum bubble diameter  
314 is larger before it escapes.

315 The corresponding tomography results indicate that the maximum bubble diameter  
316 remains larger at  $1 \text{ mm s}^{-1}$  when the laser is off (see Fig. 3b and the full build track in  
317 Supplementary Fig. 4). The large bubbles are kept in the back of the melt pool rather  
318 than other positions, this could be attributed to the different melt flow in various  
319 locations, as the melt flow can push the bubbles down in the back of the melt pool.

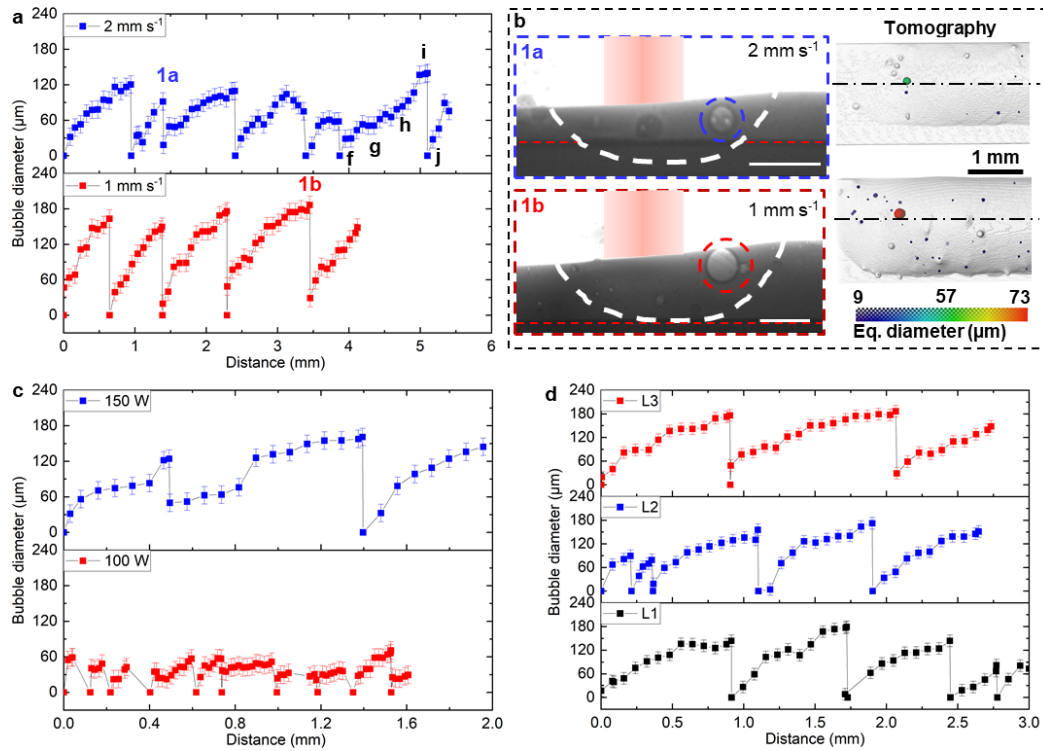
320 Fig. 3c plots the bubble diameter with moving distance at a laser power of  $150 \text{ W}$  and  
321  $100 \text{ W}$ . The maximum bubble diameter at  $150 \text{ W}$  is about  $160 \mu\text{m}$ , which is more than  
322 two times of the bubble diameter of about  $70 \mu\text{m}$  at  $100 \text{ W}$ . The larger maximum bubble  
323 diameter at a higher laser power could also be associated with the larger melt pool size  
324 for bubble growth. We also investigate the correlation between bubble behaviour and  
325 the different layers of build, as shown in Fig. 3d. From the results, we can confirm that  
326 the bubble behaviour, including the lifespan of the cycle and the maximum bubble

327 diameter, is not affected by the differences of build layers. It was also found that the  
328 diameter of a large bubble is larger at a higher powder flow rate (Supplementary Fig.  
329 5), this can be attributed to more argon pores entering the melt pool with a higher  
330 powder flow rate.

331 The melt pool size at different traverse speeds, laser powers and layers are plotted in  
332 Supplementary Fig. 6. The melt pool length and depth are both larger at a lower speed,  
333 higher laser power and greater powder feed rate, while the layer effect is insignificant.  
334 This is related to the bubble growth behaviour as shown in Fig. 3 and Supplementary  
335 Fig. 5, *i.e.*, the larger melt pool allows the larger maximum bubble size reached.

336 The bubble pushing behaviour over different build conditions was investigated. We  
337 noted that bubbles were being pushed in the melt pool while growing for a certain  
338 distance before they escaped. We have discussed previously that the pushing behaviour  
339 is related to the combination of Marangoni flow and buoyancy force. And the time of  
340 bubbles being pushed equals to the lifespan of the bubbles discussed in this section and  
341 is closely related to the growth rate we measured. We hypothesise that bubble pushing  
342 is also related to the melt flow around the large bubble. As shown in Supplementary  
343 Movies 1 and 4, small bubbles circulate around the large bubble due to Marangoni flow,  
344 indicating that the large bubble can be pushed in the melt pool with a downward force  
345 vector against the buoyancy force due to the high shear flow between the bubble upper  
346 surface and the melt pool surface.

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348

349 **Fig. 3 Quantification of the bubble growth mechanisms.** **a** Bubble growth over different  
 350 building parameters. Bubble diameter changes were tracked with moving distance over the  
 351 building process in the third layer of the build for a substrate traverse speed of 2 and 1 mm s<sup>-1</sup>,  
 352 respectively. The laser power is 160 W. The bubble diameter error bars are calculated as ±2  
 353 pixels, equivalent to the segmentation uncertainty. **b** Radiograph examples at 2 and 1 mm s<sup>-1</sup>  
 354 are shown in 1a and 1b (scale bars are 300 μm), with the corresponding tomographic rendered  
 355 images overlaid with the pore equivalent diameter. See the videos corresponding to **b** in  
 356 Supplementary Movies 1 and 4. **c** Bubble growth over the building process in the first layer of  
 357 build for a laser power of 150 and 100 W, respectively. The traverse speed is 1 mm s<sup>-1</sup>. **d** Bubble  
 358 growth over different layers. Bubble diameter changes were tracked over different layers of the  
 359 build, namely, layers 1-3, the laser power is 160 W, and the traverse speed is 1 mm s<sup>-1</sup>.

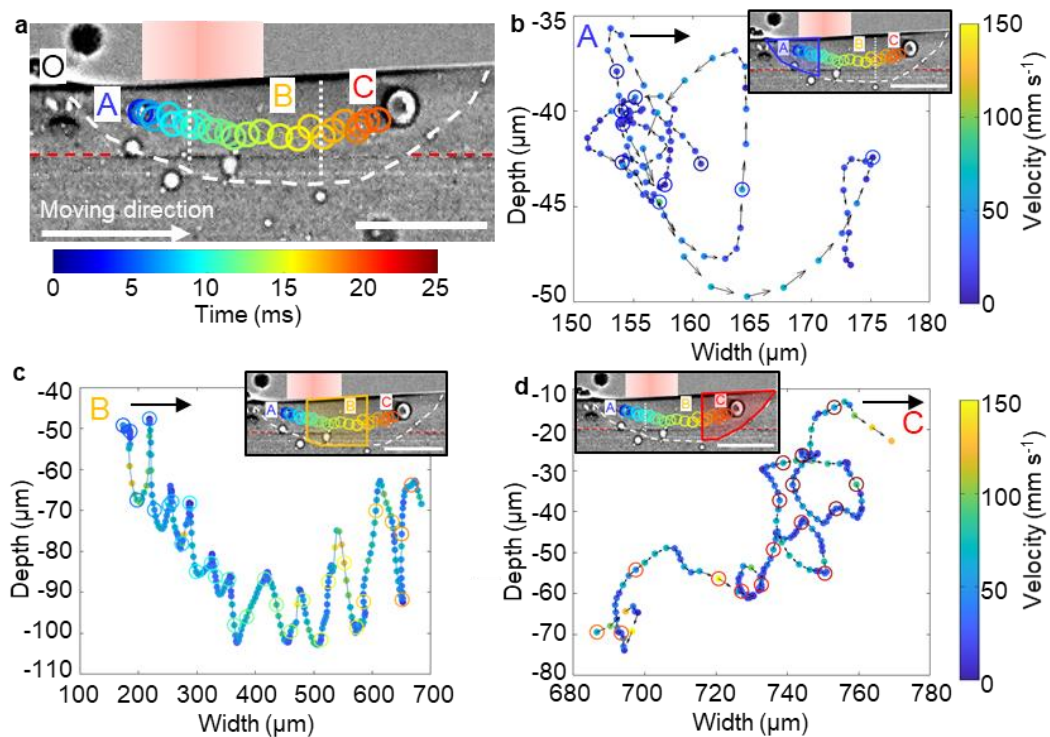
360 **Bubble migration mechanisms.** We tracked the 2D projections of the bubble  
 361 movements during the DED process from the radiographs, as shown in Fig. 4. As  
 362 mentioned in the pore behaviour section, depending on the regions of the melt pool,

363 bubbles are observed to migrate laterally or circularly. Based on this observation, we  
364 divided the melt pool into three regions, namely regions A (front), B (middle) and C  
365 (back), as shown in Fig. 4a. We then tracked the movements of a bubble which passed  
366 through these three regions as shown in Fig. 4b-d. In region A (Fig. 4b), which is the  
367 front of the melt pool, the bubble is observed to circulate counter clockwise, and the  
368 maximum velocity is measured to be  $\sim 88 \text{ mm s}^{-1}$ , driven by Marangoni flow in the front  
369 of the melt pool.

370 The bubble then moves into region B (Fig. 4c), which is the middle of the melt pool;  
371 the bubble is observed to move up and down. We hypothesise that there are 4 flow cells:  
372 front, back, and one at each side. In the middle region, the bubble is in one of the side  
373 flow cells, and is going up/down and in/out of the page. In this region, the pore appears  
374 stationary in the laser frame of reference, which means it is pushed forward by the rear  
375 recirculation at the speed the substrate is moving ( $2 \text{ mm s}^{-1}$ ). At some stage, the drag at  
376 the bottom moves the pore back into the rear recirculation flow. This backward  
377 migration will be a balance of the recirculation flow ( $>10 \text{ mm s}^{-1}$ ), substrate motion  
378 ( $2 \text{ mm s}^{-1}$ ), and the capillary force originating from the thermal gradient.

379 When the bubble finally moves into region C, which is the back of the melt pool, its  
380 circular motion is observed to be clockwise, and its maximum velocity is  $\sim 196 \text{ mm s}^{-1}$ ,  
381 driven by the Marangoni flow in the rear of the melt pool, as shown in Fig. 4d. In  
382 Supplementary Movie 5, this small bubble coalesces with the large bubble, and the large  
383 bubble is formed by the coalescence of small bubbles.

384 We measured the instantaneous velocity of the bubble. We observed that the velocity of  
 385 the bubble oscillates and accelerates between the highest and lowest points in each cycle,  
 386 and decelerates when the bubble approaches these peaks. The mean and maximum  
 387 velocity of the bubble in region B are measured to be  $51 \text{ mm s}^{-1}$  and  $145 \text{ mm s}^{-1}$ ,  
 388 respectively. These values are higher than the corresponding velocities of  $28 \text{ mm s}^{-1}$   
 389 and  $88 \text{ mm s}^{-1}$  in region A.



390

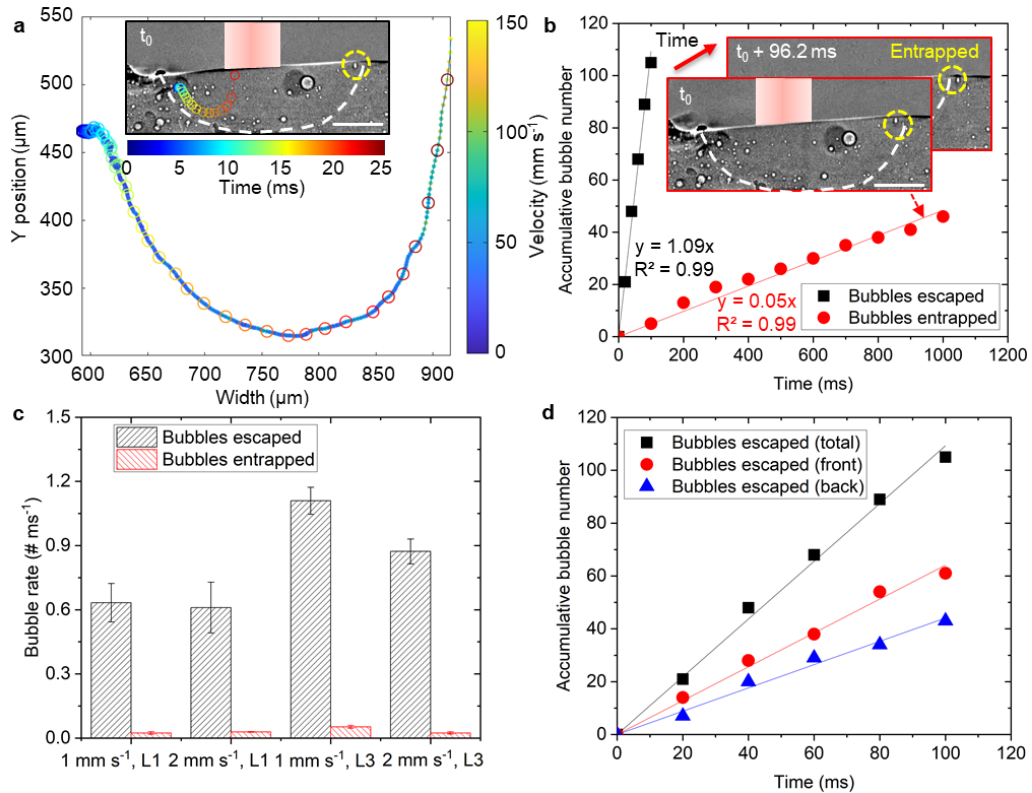
391 **Fig. 4 Bubble migration from front to back of the melt pool.** **a** The melt pool is divided into  
 392 regions A (front), B (middle) and C (back). The location of point O in the left intersection of  
 393 the solid/liquid/air boundary was regarded as the starting position (depth = 0, Width = 0). Laser  
 394 power is 160 W and traverse speed is  $2 \text{ mm s}^{-1}$ , layer 3. See the video in Supplementary Movie  
 395 5. **b** Motion track and velocity of the bubble circulation in region A, the velocity value is shown  
 396 in the colour bar, and the arrow shows the moving direction. **c** Motion track and velocity of the  
 397 bubble in region B. **d** Motion track and velocity of the bubble circulation in region C. The scale  
 398 bars in **a** and inset figures in **b-d** are  $300 \mu\text{m}$ .

399 **Bubble escape and entrapment mechanisms.** Some bubbles follow the Marangoni-  
400 driven recirculating flow in the melt pool up to the surface and escape (Fig. 5a, see the  
401 video in Supplementary Movie 6). Some bubbles coalesce into large bubbles as  
402 discussed above, and some are entrapped into the solidification front.

403 Bubbles will escape if the buoyancy force is greater than the downward component of  
404 the recirculating cell. Another important factor, we hypothesise, is the location and  
405 velocity of the bubble inside the recirculation cell, as this also affects the upward  
406 component of the bubble, which ranges from  $88 \text{ mm s}^{-1}$  (Fig. 4b) to  $247 \text{ mm s}^{-1}$  (Fig.  
407 5a) when the bubble changes from recirculation mode to escape. This indicates that the  
408 maximum bubble velocity in the vertical direction will affect bubble motion and hence  
409 escape.

410 In Fig. 5b, we compared the number of small bubbles that escape, coalesce, and are  
411 entrapped versus time, and the bubble versus time was defined as bubble rate. The  
412 number of bubbles for escaping and coalescing was observed to increase linearly with  
413 time. More bubbles escaped than were entrapped, as the bubble rate of  $1.09 \text{ # ms}^{-1}$  in  
414 escaped bubbles is higher than the bubble rate of  $0.05 \text{ # ms}^{-1}$  in entrapped bubbles. In  
415 Supplementary Fig. 7, the bubble rate by counting is  $0.07 \text{ # ms}^{-1}$  in coalesced bubbles,  
416 indicating that more bubbles coalesced than were entrapped but less than escaped. The  
417 bubble number for coalescence by counting is in the range of the bubble number  
418 calculated using the large coalesced bubble volume divided by the initial bubble volume  
419 with the minimum, mean and maximum diameters of 17, 31 and  $55 \text{ }\mu\text{m}$ , and is close to

420 the bubble number calculated by the mean diameter (Supplementary Fig. 7). This  
421 indicates that the counting results capture the average bubble coalescence behaviour  
422 under the conditions studied. We also compared the number of escaped bubbles per unit  
423 time against processing parameters, as shown in Fig. 5c. In layer 1, the rate of bubble  
424 escape and entrapment is shown to be constant despite the differences in traverse speed.  
425 The rate of both bubble escape and entrapment in the 3<sup>rd</sup> layer (L3) is higher than in the  
426 1<sup>st</sup> layer (L1). We hypothesise that this is due to more bubbles being present in the tracks  
427 laid during the experiment than in the industrial machine-built substrate for layer 1.  
428 There is no significant difference in the bubble behaviour as a function of traverse speed.  
429 We also investigated where the bubbles escape from the molten pool (Fig. 5d). More  
430 bubbles are observed to escape from the front of the melt pool. This could be due to the  
431 different velocities of the Marangoni flow in these two regions, and bubbles could stay  
432 longer at the back of the melt pool.  
433



434

435 **Fig. 5 Bubble escape from the melt pool and entrapment by the solidification front. a**

436 Motion track and velocity of a bubble escape following Marangoni flow. The velocity value is

437 shown in the parula colourmap. The time is shown in the jet colourmap. See the video in

438 Supplementary Movie 6. **b** Accumulative number of bubble escape, coalesce and are entrapped

439 with increasing time, and it is fitted linearly. Entrapped bubbles are shown in the inset figure. **c**

440 The rates of bubble escaped and entrapped in a traverse speed of 1 mm s<sup>-1</sup>, layer 1; 2 mm s<sup>-1</sup>,

441 layer 1; 1 mm s<sup>-1</sup>, layer 3; 2 mm s<sup>-1</sup>, layer 3, respectively. Error bars represent standard deviation.

442 **d** Accumulative number of bubble escape in total, front and back of melt pool. In **a**, **b**, and **d**,

443 the laser power is 160 W and the traverse speed is 1 mm s<sup>-1</sup>, layer 3. Scale bars in **a-b** are 300

444 μm.

445 **Melt flow and bubble behaviour revealed by multiphysics modelling.** The

446 multiphysics model developed (based on ref. <sup>50</sup>) uses a control volume solution of the

447 mass, momentum and temperature transfer in the DED process, including phase change,

448 bubble migration and coalescence, and powder particle impact on the surface of the

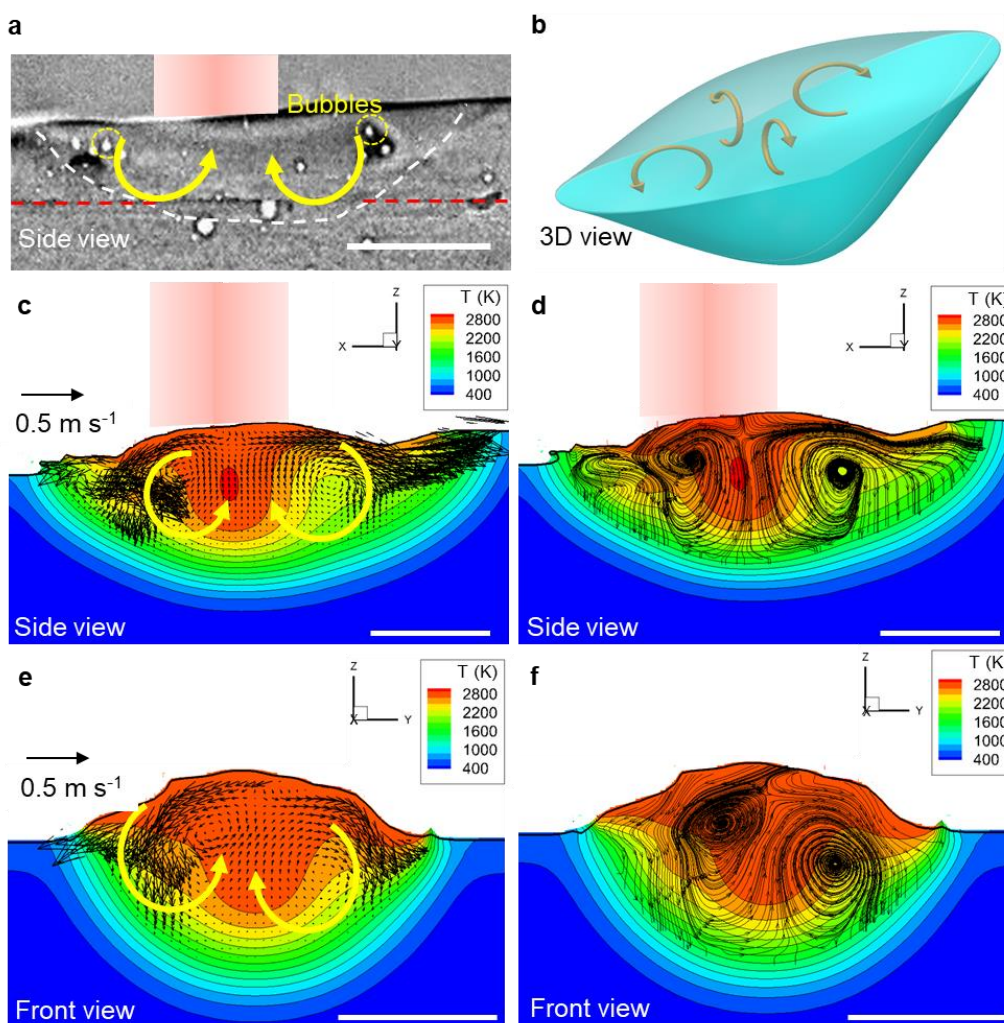


449 molten pool. Full details of the model are in Methods and Supplementary Information.  
450 We used this high-fidelity Multiphysics model of DED<sup>50</sup> to validate the hypotheses we  
451 have formulated from the *in situ* X-ray imaging experiments on melt pool flow and  
452 bubble formation mechanisms.

453 **Melt pool recirculating flow cells.** Fig. 6a shows an X-ray radiograph of the melt pool,  
454 together with schematic arrows showing proposed Marangoni-driven recirculating  
455 flows at the front and back of the melt pool. Fig. 6b shows a schematic illustration of  
456 our hypothesis above that there are four main recirculation flow cells in the melt pool,  
457 with two cells at the centre in and out of the page of the radiograph in Fig. 6a. The  
458 model predicted flows are shown in Fig. 6c and d, predicting recirculating flow cells at  
459 the front and back of the pool. These predictions match the X-ray results shown in Fig.  
460 1 and Fig. 6a (also see videos in Supplementary Movies 1 and 2), where the pores  
461 recirculate in the front and back of the melt pool. The model also predicts two more  
462 flow cells, shown in a front view cut at the centre of the melt pool (Fig. 6e and f). This  
463 matches our hypothesis that there are two into and out of the page flow circulations,  
464 and explains the pores oscillation up and down in the middle zone in Fig. 4, as bubbles  
465 migrate from the front to the back of the melt pool.

466 As shown in Supplementary Fig. 8, the melt pool region was divided into the surface  
467 region and the inner periphery region. The temperature is higher in the surface region  
468 than in the inner periphery region. The flow velocity is higher in the region near the  
469 surface and generally increases with increasing temperature. The magnitude of the

470 predictions of the flow also nicely matches the measured ones, as shown in  
 471 Supplementary Fig. 8b, for a bubble with a diameter of 160  $\mu\text{m}$ , where the average  
 472 velocity is 100  $\text{mm s}^{-1}$  (20~400  $\text{mm s}^{-1}$ ), which is consistent with the velocity that we  
 473 measured by X-ray imaging (Fig. 4 and Fig. 5).



474

475 **Fig. 6 Modelling results showing the melt pool flow without bubbles during DED.** **a** A X-  
 476 ray image showing the melt pool. See the video in Supplementary Movie 2. **b** 3D view  
 477 schematic showing the melt pool flow. **c** Side view and **d** corresponding 2D projected  
 478 streamlines by modelling. **e** front view and **f** corresponding 2D projected streamlines by  
 479 modelling. T in **c-f** represents temperature in K. The traverse speed is 2  $\text{mm s}^{-1}$ , and the laser  
 480 power is 160 W. Scale bars in **a** and **c-f** are 300  $\mu\text{m}$ .

481 **Bubble coalescence.** Our hypotheses on bubble behaviour were also investigated with  
482 the model. One typical phenomenon is bubble coalescence. The bubble coalescence  
483 behaviour was investigated by first simulating the flow without bubbles to establish the  
484 four recirculating flows (see Fig. 6), and then bubbles were inserted at varying positions  
485 into the melt pool. Our observations of bubble coalescence were replicated in the model,  
486 showing that when 3 separate pores are placed in the flow, they are all driven towards  
487 the centre of a recirculation cell and coalesce (Fig. 7a, Supplementary Fig. 9 and  
488 Supplementary Movie 7). The front view shows this most clearly, with two bubbles  
489 coalescing to form a dumbbell shape. Due to surface tension, this shape is transient,  
490 quickly converting to a near-spherical large bubble.

491 For bubbles in the mid-front but the deep location (Supplementary Fig. 10 and  
492 Supplementary Movie 8), the bubbles are pushed between the front and side  
493 recirculation cell, where the flow velocity is lower, with a high flow velocity above. In  
494 this front-deep location, two bubbles also coalesce into a larger bubble, indicating this  
495 is conducive to coalescence. For bubbles in the middle location (Supplementary Fig.  
496 11), bubbles are trapped in the centre of recirculation, and the local velocity is low, and  
497 bubble coalescence also occurs. These phenomena are similar to the bubble coalescence  
498 that occurred in the back of the melt pool. Therefore, bubble pushing occurs in back,  
499 front-deep and centre locations, as the Marangoni flow circulations can push bubbles  
500 down. Bubble coalescence is much more likely to occur in a larger melt pool of DED  
501 than in LPBF, as the residence time of bubbles is much greater, enabling them to

502 coalesce to form large bubbles. The strong recirculating flow in a large pool constrains  
503 both the small and large bubbles' flow, creating conditions appropriate for bubble  
504 collision, with coalescence occurring when the film of liquid between colliding bubbles  
505 ruptures <sup>60</sup>. Coalescence reduces the overall free energy as it minimises the total bubble  
506 surface area <sup>60</sup>.

507 **Bubble pushing at the surface.** One surprising experimental observation was that  
508 large, coalesced bubbles did not immediately rise to the surface (due to buoyancy force)  
509 and pop. We hypothesised that this was due to the constriction of high-shear Marangoni  
510 flow. To test this a large bubble was put close to the surface in the back region, as shown  
511 in Fig. 7b, Supplementary Fig. 12, and Supplementary Movie 9. The model predicts  
512 that the shear flow circulates over the large bubble and pushes it in the melt pool. This  
513 pushing behaviour is consistent with the experimental results shown in Fig. 1 and Fig.  
514 2, which confirms that the Marangoni flow contributes to pushing bubbles down in the  
515 melt pool, overcoming the buoyancy force, until the bubble reaches a critical size.  
516 Therefore, although bubble coalescence and growth can contribute to the larger  
517 buoyancy, the bubbles constrict the Marangoni flow, causing a downward force on the  
518 bubbles that delays their escape.

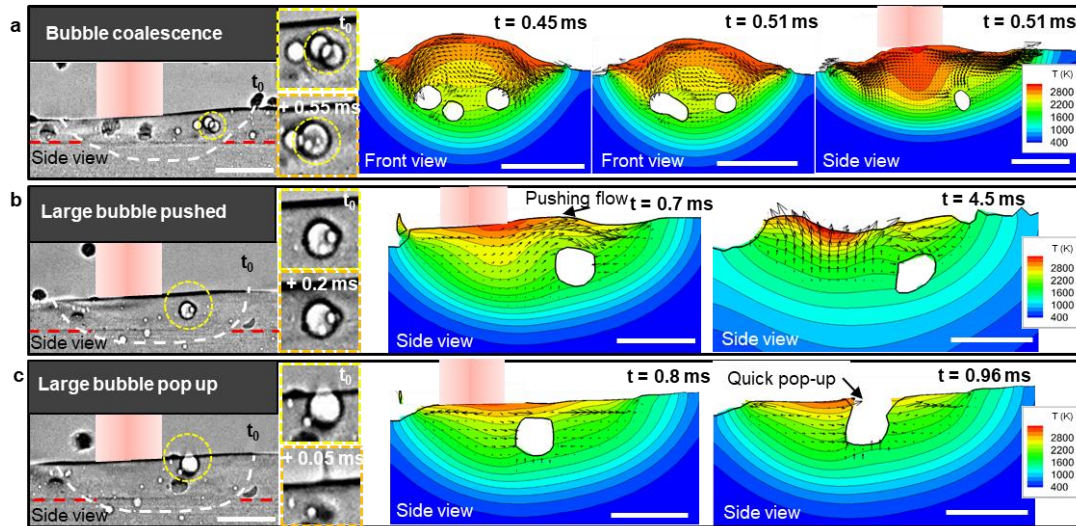
519 In Supplementary Discussion 2, the force balance onto the large bubble is calculated by  
520 comparing static buoyancy, shear and pressure forces induced by the molten metal flow.  
521 According to the corresponding simulation results of these forces in Supplementary  
522 Table 3, the large horizontal shear force can push the large bubble in the horizontally

523 backward direction. The strong transverse Marangoni flow above the bubble pushes the  
524 bubble downward, balancing the buoyancy and positive shear force in the vertical  
525 direction. Therefore, the bubble can be pushed downward and backward when this flow  
526 structure is formed.

527 **Large bubble escape.** Fig. 7c (and Supplementary Fig. 13) shows an example where a  
528 very large bubble can escape from the top of the melt pool. The large bubble touches  
529 the top liquid surface when the bubble grows into a critical size and moves by the flow  
530 disruption, and then the top liquid surface ruptures to release the large bubble. Here the  
531 bubble is both very large (and hence large buoyancy force) and is located in the middle  
532 of the melt pool, between the flow recirculation cells, breaking the balance of forces,  
533 so the bubble pops up, explaining the experimentally observed behaviour.  
534 Computational fluid dynamics simulation in the Supplementary Information (e.g., see  
535 Supplementary Fig. 13), show how changes in the Marangoni driven flow cells can  
536 create conditions entrapping bubbles within the flow cell, or pushing them to the melt  
537 pool surface, rupturing.

538 Most bubbles escape through the top liquid surface of the melt pool, it requires the high-  
539 speed X-ray imaging with a frame rate of 20 kHz to capture these phenomena (see  
540 videos in Supplementary Movies 2, 3, 5 and 6), as the X-ray imaging at a low frame  
541 rate of 1 kHz may miss a short escaping period due to the fast bubble escaping speed  
542 of  $247 \text{ mm s}^{-1}$  in Fig. 5. A large bubble also escapes in the rear of the melt pool (see  
543 Supplementary Movie 2). The large bubble in the rear of the melt pool grows close to

544 the top liquid surface of the melt pool, and the powder particle hits the melt pool and  
 545 disrupts the Marangoni flow near the large bubble to break the force balance, so the  
 546 large bubble can escape.



547

548 **Fig. 7 Comparison between experimental data and modelling results of bubble**  
 549 **coalescence, push-down and pop-up in the back of the melt pool. a** X-ray images and  
 550 corresponding simulation images showing bubble coalescence at  $t = 0.45$  ms and  $t = 0.51$  ms  
 551 (shown in the front and side view images) (see simulation images in Supplementary Fig. 9 and  
 552 video in Supplementary Movie 7). **b** X-ray images and corresponding simulation images  
 553 showing a large bubble pushed by Marangoni shear flow at  $t = 0.7$  ms and  $t = 4.5$  ms from  
 554 bubble insertion  $t = 0$  ms (shown in the side view image) (see simulation images in  
 555 Supplementary Fig. 12 and video in Supplementary Movie 9). **c** X-ray images and  
 556 corresponding simulation images showing the large bubble pop-up at  $t = 0.8$  ms and  $t = 0.96$   
 557 ms from bubble insertion  $t = 0$  ms (shown in the side view image) (see simulation images in  
 558 Supplementary Fig. 13). T in the colour bar represents temperature in K. Scale bars in **a-c** are  
 559 300  $\mu\text{m}$ .

560 **Influence of powder particles hitting the melt pool surface.** One possible  
 561 explanation for the cyclic bubble migration in Fig. 4c and the circulating motion in Fig.

562 4d could be the disruption of the Marangoni flow when feedstock powder particles hit  
563 the surface, locally quenching the pool and altering the thermal, and hence surface  
564 tension gradient. Fig. 8 shows the modelling results of direct particle bombardment on  
565 the surface, causing surface oscillation and local flow disruption. As the melt pool flow  
566 is disturbed by the bombardment, the bubble migrates similarly to the experimental  
567 observations.

568 In Fig. 8, to consider powder-hitting effects in our modelling, two approaches including  
569 forced and direct bombardment cases were applied. Based on the forced case (see the  
570 details in Methods), Fig. 8b plots the temperature field, and smaller flow cells were  
571 observed in the melt pool. The corresponding velocity and trace of a bubble are shown  
572 in Fig. 8c. The up-down migration of a bubble under forced oscillation on the surface  
573 and migration from the front to the back of the melt pool, caused by the formation of  
574 circulation cells, which is consistent with experimental flow result that is shown in Fig.  
575 8a and c. This indicated that the phenomena can be attributed to the velocity and  
576 temperature perturbations induced by powder particles hitting.

577 For the direct bombardment case, as shown in Fig. 8d and e, the temperature field and  
578 flow direction near the powder change significantly. This can disrupt the normal  
579 Marangoni flow instantly and locally. As a result, the bubbles oscillate up and down  
580 and do not follow the normal circulating path. In addition, in the modelling results  
581 shown in Fig. 8e, an outward flow cell forms near hitting particles. In Fig. 8e and f,  
582 these flow cells can drive bubbles to migrate from the front to the rear of that melt pool

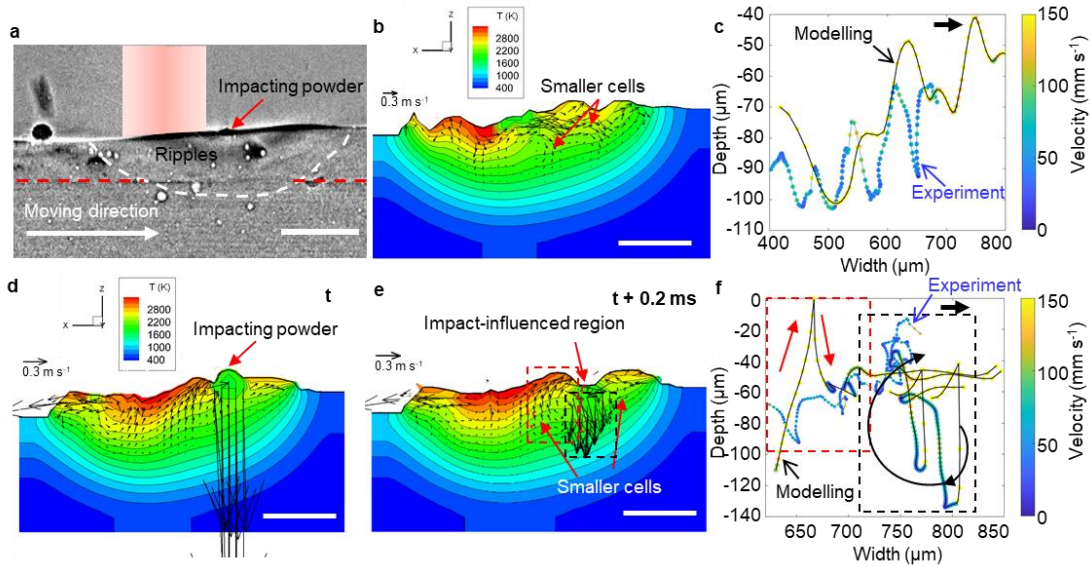
583 (in the region indicated with a red dashed box) and then circulate outward (in the region  
584 indicated with a black dashed box). These phenomena are consistent with the  
585 experimental results of bubble migration in Fig. 8f and Fig. 4d. These results indicate  
586 that the flow cells generated by the particle impact can promote the bubble migration.

587 When the powder particle hits the melt pool, it can mainly generate two effects, namely,  
588 i. the impact ripple waves of the particle when the powder particle just touches the melt  
589 pool surface and subsequent standing waves, which can affect the flow and bubble  
590 migration near the particle; ii. after that, the powder particle gradually melts and  
591 quenches the melt pool, which can change the local temperature and flow pattern and  
592 bubble migrations near the particle. As shown in Fig. 8a-c and Fig. 4, the modelling  
593 results considering velocity and temperature perturbations for the powder effects are  
594 consistent with experimental results, in which the impact wave of powder particle  
595 causes the initial flow disruption and small flow cells (in accordance with the standing  
596 wave generation) are formed (Fig. 8d-f).

597 The motion trajectory in Fig. 4c and Fig. 8c is supposed to be mainly related to the  
598 simultaneous effects of Marangoni flow cells and powder impact effects. Although the  
599 powder particles can hit different locations of the melt pool at different times, the  
600 powder flow rate is controlled to be constant and high, which can produce a relatively  
601 consistent powder hitting, thus to change the flow pattern in the melt pool. It is also  
602 speculated from the experimental results that the later standing wave formation is nearly  
603 similar although the initial ripple formation and the temperature effect occur in random



604 places. Therefore, the bubble motion trajectory exhibits an organised pattern.



605

606 **Fig. 8 Comparison between experimental data and modelling results of bubble migration**

607 **in the melt pool. a** A radiograph showing melt pool with impacting powder. **b** For the forced

608 case, the temperature field obtained by modelling with the same parameters as the X-ray  
609 imaging experiments, velocity and temperature perturbations given to the surface to simulate

610 powder hitting effects, and **c** corresponding velocity and trace of a bubble inside the melt pool.

611 The up-down migration of a bubble under forced oscillation on the surface, caused by the  
612 formation of circulation cells compared with the large Marangoni circulation shown in Fig. 6c

613 and **d**. Modelling and experiment results are shown in blue and black lines, respectively. **d**

614 Temperature field considering impacting powder at  $t$ , **e** formation of smaller cells at  $t + 0.2$  ms.

615 And **f** corresponding velocity and trace of a bubble inside the melt pool. The modelling and  
616 experimental curves are connected in black and blue lines, respectively. Direct simulation of

617 random powder bombardment where sudden velocity increase is induced in the impact region,  
618 which causes irregular bubble migration such as the up-down migration and local circulation.

619 Modelling and experiment results are shown in blue and black lines, respectively. For the forced

620 case, the (circular) surface wave period is set as 0.6 ms, surface wave number is 5 in the pool

621 lateral direction. For the direct bombardment case, the impacting velocity is  $4 \text{ m s}^{-1}$ , the powder

622 diameter is  $90 \text{ }\mu\text{m}$ , the impacting interval is 0.5 ms and the powder temperature is 1800 K for

623 simplicity. These values for modelling are determined by the X-ray imaging experimental video.  
624 T in the colour bar in **b** and **d** represents temperature in K. The velocity unit in **b**, **d** and **e** is m  
625 s<sup>-1</sup>. Scale bars in **a**, **b**, **d**, and **e** are 300 μm.

626 In summary, we have applied *in situ* high-speed synchrotron X-ray imaging and multi-  
627 physics modelling to reveal pore behaviours in the DED process, including pore  
628 formation, bubble coalescence and growth, pushing, migration, escape and entrapment.

629 We found that the majority of bubbles in the melt pool originated from argon pores in  
630 the feedstock powder. Although many of these small bubbles escaped from the melt  
631 pool surface, some were entrapped by the solidification front and some coalesced into  
632 larger bubbles; those entrapped in the solid are often entrained in the pool in the next  
633 layer of track. The large bubbles are formed by up to one hundred small bubbles  
634 coalescing, and are pushed ahead of the solidification front until they reach a critical  
635 size. High-fidelity multi-physics modelling demonstrates that the constriction of the  
636 Marangoni shear flow between the melt pool surface and the top of the large bubbles  
637 provides sufficient downward force to overcome the upward buoyancy force, keeping  
638 the bubble entrained in the pool. After the bubble reaches a critical size, it interacts with  
639 the recirculating flow along the bottom of the melt pool, and is pushed to the pool  
640 surface and then pops out. We demonstrate the growth of large bubbles through  
641 coalescence and their subsequent periodic escape is a function of pool size and hence  
642 build conditions, including laser power and traverse speed. Although some prior studies  
643 of DED mention feedstock pores might be entrained, it is only through the *in situ*  
644 observations shown here that the key phenomena of bubble coalescence to form large

645 pores have been revealed. This coalescence of up to 70 pores with a diameter of 20 - 50  
646  $\mu\text{m}$  to form a single 180  $\mu\text{m}$  pore may control final component properties.

647 The bubble dynamics also includes their interaction with the fluid flow causing their  
648 entrainment or escape from the surface, and their interactions with solid/liquid interface,  
649 causing entrapment or pushing. To the best of the authors' knowledge, no bubble  
650 coalescence and growth in a large melt pool of AM was reported in previous studies.  
651 The solid/liquid interface entrapment or pushing of bubbles was reported in directional  
652 solidification<sup>13,25</sup>, but direct observation has not been reported in DED. Bubble  
653 entrainment, escape and entrapment in the solid were seen for keyhole pores in LPBF  
654<sup>33</sup>, but not in DED.

655 The bubble behaviour should be related to the Marangoni flow in the melt pool. The  
656 Marangoni flow was observed by Mills et al.<sup>58</sup> and Lee et al.<sup>61</sup> using *ex situ*  
657 observations, and modelled by Paul & Debroy<sup>62</sup>, and more recently *in situ* observations  
658 by Aucott et al.<sup>63</sup> for welding and Guo et al.<sup>64</sup> in LPBF. However, our observations in  
659 DED also elucidate that some small bubbles follow the flow, some float out, some are  
660 entrapped, and some coalesce; whilst the large bubbles stay in the melt pool. This  
661 study contributes to a greater fundamental understanding of pore evolution and  
662 dynamics mechanisms during additive manufacturing processes, providing a potential  
663 pathway for developing a pore minimisation strategy for the DED process.

## 664 **Methods**

665 **Material characterization.** The gas atomised nickel-based superalloy RR1000 powder

666 was characterized with scanning electron microscopy (SEM) JEOL JSM-6610V. The  
667 SEM image of the powders and corresponding powder size distribution was plotted in  
668 Supplementary Fig. 2. The powders were segmented using Otsu's method and then  
669 separated using a watershed algorithm in MATLAB to measure the powder size.

670 **Blown Powder Additive Manufacturing Process Replicator II (BAMPR II) system**  
671 **and processing conditions.** *In situ* synchrotron X-ray imaging was performed on the  
672 ID19 beamline at the European Synchrotron Radiation Facility (ESRF) to capture the  
673 pore dynamics and formation during DED. BAMPR II was a custom-designed system  
674 to replicate the commercial DED process that can be integrated into synchrotron  
675 beamtime. It includes an environmental chamber (Saffron, Scientific Equipment Ltd),  
676 a high-precision 3-axis platform (Aerotech, US), a coaxial DED nozzle, and a  
677 Ytterbium-doped laser (SPI lasers Ltd, UK) in continuous wave mode with a  
678 wavelength of 1070 nm and a maximum power of 200 W. The beam reducer (Optogama,  
679 Lithuania) was equipped to focus the beam size down to 400  $\mu\text{m}$  with a symmetric  
680 Gaussian shape. The laser beam spot size is defined with  $1/e^2$ , and the profiled laser  
681 beam is plotted in Supplementary Fig. 14. The measured beam spot size is about 390  
682  $\mu\text{m}$  near the focal point. The environmental chamber was filled with argon gas to reduce  
683 oxidation, and the oxygen level was generally controlled to be below 10 ppm during  
684 the experiments. The powder was delivered to the nozzle in a stream of argon gas by  
685 the industrial powder feeder system (Oerlikon Metco TWIN-10-C) and then blown to  
686 normal to the substrate plate. The powder feed rate in this work is 1.8 - 2.7  $\text{g min}^{-1}$ . The

687 substrate with dimensions of 60 mm × 20 mm × 1.5 mm was mounted in a moving  
688 platform with a maximum traverse speed of 50 mm s<sup>-1</sup>. The high-speed imaging for the  
689 melt pool and pores was captured at spatial (4 μm) and temporal resolutions (20 kHz)  
690 using a CMOS camera (type: SAZ, Photron, Japan) lens-coupled to a LuAG:Ce single-  
691 crystal scintillator. The low-speed imaging was captured at spatial (3.7 μm) and  
692 temporal resolutions (1 kHz) using a CMOS camera (type: Dimax, PCO AG, Germany)  
693 lens-coupled to a LuAG:Ce single-crystal scintillator as well to observe a longer  
694 duration period.

695 **Image processing.** The acquired radiographs were processed using ImageJ and  
696 MATLAB. A flat field correction was conducted via the equation:  $FFC = (I_0 - Flat_{avg}) /$   
697  $(Flat_{avg} - Dark_{avg})$ , where FFC is the flat field corrected image,  $I_0$  is the raw image,  
698  $Flat_{avg}$  is the average of 100 flat field images (imping beam profile without sample) and  
699  $Dark_{avg}$  is the average of 100 dark field images (sensor noise without any impinging  
700 radiation).

701 **Multi-physics modelling.** The temperature, velocity and bubbles in the melt pool were  
702 simulated using multi-physics modelling which is validated with experimental  
703 parameters<sup>50</sup>. The fluid flow equations of mass, momentum and temperature are solved  
704 along with interface capturing by the Coupled Level-Set/Volume-Of-Fluid (CLSVOF)  
705 method.

706  $(\text{mass}) \frac{\partial \rho}{\partial t} + (\mathbf{u} \cdot \nabla) \rho = -\rho \nabla \cdot \mathbf{u} \quad (1)$

707 (momentum)  $\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} = -\frac{\nabla p}{\rho} + \mathbf{Q}_u + \mathbf{g} + \mathbf{F}_{u,surf}$  (2)

708 (temperature)  $\frac{\partial T}{\partial t} + (\mathbf{u} \cdot \nabla) T = -\frac{p \nabla \cdot \mathbf{u}}{\rho c_p} + Q_T$  (3)

709 where  $\rho$  is the density,  $\mathbf{u}$  is the velocity,  $T$  is the temperature,  $p$  is the pressure and  $c_p$  is  
710 the constant-pressure heat capacity.  $\mathbf{Q}_u$  represents the Newtonian viscous force and  
711 Darcy's force in the mushy zone,  $\mathbf{g}$  is the gravitational acceleration and  $\mathbf{F}_{u,surf}$  represents  
712 the interfacial surface tension force including the Marangoni effect.  $Q_T$  represents the  
713 heat transport, including heat conduction by Fourier's law, viscous work, latent heat for  
714 phase change and radiation. The laser power is given to the melt pool surface by the ray  
715 tracing method. Material accumulation on the surface is calculated by the conservation  
716 of mass. Details of the numerical method can be found in Supplementary Information  
717 and the reference<sup>50</sup>. The physical properties such as viscosity and thermal conductivity  
718 are derived as in the reference<sup>65</sup>. The fine grid resolution is 16  $\mu\text{m}$ . The resolution for  
719 long-time simulation is 32  $\mu\text{m}$  in cases of Fig. 7b-c, 8b and 8d-e. Still, we justify the  
720 use of this grid since we have confirmed that the same Marangoni flow structure can be  
721 reproduced. For the small bubble tracking cases in Fig. 8b and 8d-e, these bubbles are  
722 assumed to be sufficiently small that they can be treated as Lagrangian point particles  
723 (see Supplementary Method 1 and Supplementary Fig. 15 for justification). For the  
724 large bubbles (e.g., those in Fig. 7), the bubbles are explicitly modelled using the level-  
725 set method to capture the liquid gas interface, simulating the surface shape and bubble  
726 coalescence (see Supplementary Method 1).

727 In the forced case in Fig. 8, velocity and temperature perturbations were directly applied  
728 to the melt pool surface. From the experimental observation, standing waves are seen  
729 after particle bombardment. For simplicity, the perturbations to give on the surface are  
730 set as follows; the wavelength  $\lambda$  is one-fifth of the longitudinal melt pool length (5  
731 standing waves in the melt pool), the period  $T$  is 0.6 ms, the displacement amplitude  $A$   
732 is 30  $\mu\text{m}$ , and the velocity amplitude is  $A\omega$ , where  $\omega = 2\pi/T$ . In the assumed region of  
733 particle bombardment, the surface temperature is set at 1800 K, but this temperature  
734 effect is minor.

#### 735 **Data availability**

736 The authors declare that the data supporting the findings of this study are available  
737 within this article and its Supplementary Information file, or from the corresponding  
738 authors upon request.

739

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909

## 910 **Acknowledgements**

911 This research is financially supported by the Engineering and Physical Sciences  
912 Research Council (EPSRC) via MAPP: Future Manufacturing Hub in Manufacture  
913 using Advanced Powder Processes (EP/P006566/1), Rolls-Royce plc. via the Aerospace  
914 Technology Institute program REINSTATE (contract 51689), and the Royal Academy  
915 of Engineering (CiET1819/10). We also acknowledge the use of facilities and support  
916 provided by the Research Complex at Harwell and thank the ESRF for providing the  
917 beamtime proposal (MA-4857) and the staff at ID19 beamline for technical assistance.

918 This work is partially supported by Next Generation TATARA Project sponsored by the  
919 Government of Japan and Shimane Prefecture. C.P. would like to acknowledge the  
920 funding from Innovation Fellowship funded by Engineering and Physical Science  
921 Research Council (EPSRC), UK Research and Innovation, under the grant number:  
922 EP/S000828/2.

## 923 **Author contributions**

924 K.Z., M.A.J. and P.D.L. conceived the research. K.Z. wrote the manuscript. K.Z. and  
925 P.D.L. finalised the manuscript, with all authors contributing. K.Z. led and performed

926 data analysis and image processing (with help from P.D.L., C.L.A.L., Y.C., X.F. and  
927 S.B.). The experiments were performed remotely during the lockdown, with all authors  
928 virtually participating, but S.M., Y.C., M.F., M.M., B.L., K.J. and A.R. who were based  
929 at ESRF physically present. J.S. and C.P. performed modelling.

930

931 **Additional information**

932 **Supplementary Information** accompanies this paper.

933 **Competing interests:** The authors declare no competing interests.

