

1 **The role of ultrasonically induced acoustic streaming in developing fine equiaxed grains**
2 **during the solidification of an Al-2% Cu alloy**

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17 **Abstract**

18 Recent research and a simulation of heat transfer and solidification during acoustically
19 generated convection showed that the location of the coolest liquid, and thus the place where
20 the first grains are expected to form, is under the sonotrode. Further, the generated vigorous
21 convection produces a very flat temperature gradient in the bulk of the melt facilitating the
22 formation of a refined equiaxed structure throughout the casting. This study validates these
23 findings through a series of experiments on an Al – 2 wt.% Cu alloy, that evaluate grain
24 formation under the sonotrode over time and relates this to the formation of the
25 macrostructure of a cast ingot. Analysis of the results confirms the predictions of the
26 simulation and shows, for the conditions applied, that most grains nucleated in the cavitation
27 zone are swept into the melt by acoustically generated convection and over a period of 70
28 seconds the number of grains increase and they grow with spherical and globular morphology
29 gradually filling the casting with refined equiaxed grains. It was found that the
30 macrostructure of each casting is made up of three microstructural zones. A fine grained
31 equiaxed zone forms from the bottom of the casting below the sonotrode due to settling of
32 grains during and after termination of ultrasonic treatment (UST), which increases in size
33 with increasing duration of UST. Above this zone a coarse grained structure is formed due to
34 depletion of UST-generated grains on termination of UST. At the top of the casting a zone of
35 columnar grains growing from the top surface of the melt is formed. The latter two zones
36 decrease in size with increasing UST duration until 80 seconds where the macrostructure
37 consists entirely of the equiaxed zone.

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38 **Key words: aluminium alloy, grain refinement, acoustic streaming, ultrasonic treatment**

39 **1. Introduction**

40 Application of high intensity ultrasound to the processing of metallic melts has attracted
41 research interest for many years as summarized in monographs [1, 2]. In the last two decades
42 there has been a revival of interest in studying the fundamentals and to develop
43 technologically viable methods of implementing ultrasonic melt processing in industrial
44 casting processes. To date, ultrasound has been well demonstrated at the laboratory scale to
45 refine a broad range of metals and alloys including Mg alloys [3, 4], Al alloys [1, 3, 5-7],
46 steel [2, 8, 9], Zn [10], and a TiAl alloy [11].

47 The influence of ultrasonication on the refinement of microstructure is based on the
48 physical phenomena caused by high intensity ultrasound propagation in the melt, in particular
49 acoustic cavitation and acoustic streaming [1–3]. Although it has been recognized that
50 acoustic streaming plays an important role in many ultrasound-assisted industrial processes
51 including degassing, melt cleaning, homogenization, filtration and waste treatment [12,13], it
52 has not received as much attention as cavitation. Previous studies have dealt with the
53 immediate effect of the collapse of the cavities or bubbles but overlooked the effect of
54 acoustic streaming on solidification and grain formation. The lack of focus on acoustic
55 streaming may explain why the exact mechanisms of UST refinement are still being debated
56 [7].

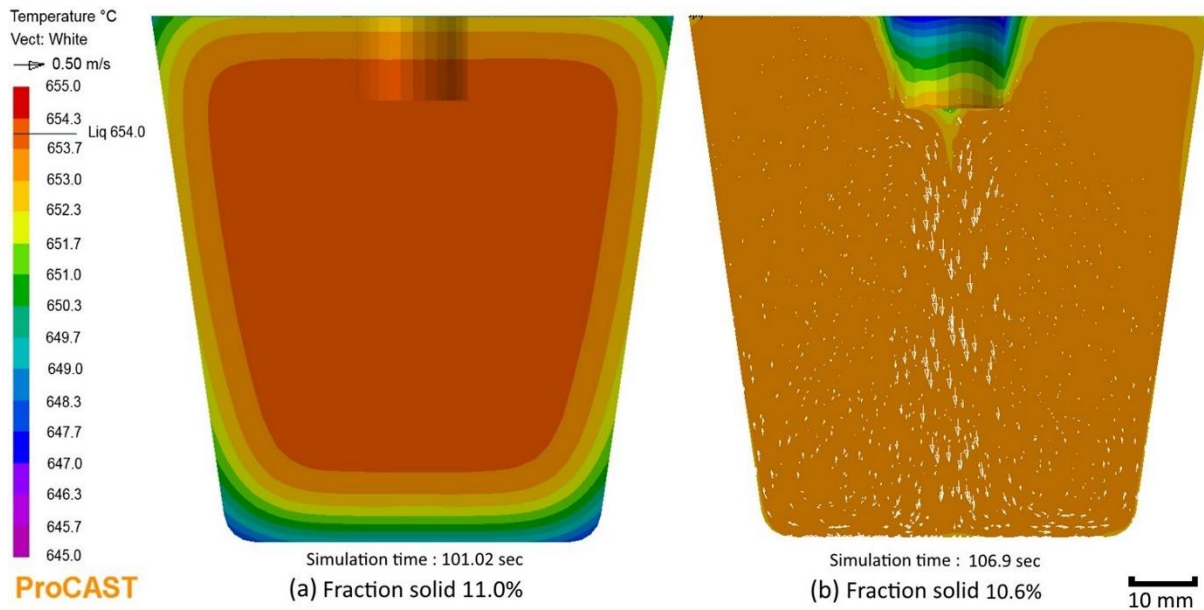
57 It has been reported that acoustic streaming generates convection with typical velocities in
58 the range 0.2–0.8 m/s with rapid attenuation as the distance from the ultrasonic source
59 increases [14-19]. Acoustic streaming assists in equilibration of the temperature field in the
60 liquid phase and interacts with the solidification front when the freezing range is wide [15].
61 Recent research highlighted the effect of acoustic streaming in generating convection patterns
62 that facilitate the formation of refined equiaxed zones [20]. It also showed that, for ingots the
63 same size as those used in the current study, when UST is applied the cooling curves from
64 near the wall and towards the centre of an ingot converge to within 1 °C of each other.

65 To gain better understanding of the relationship between acoustic streaming, thermal
66 equilibration and solidification, a simulation model was developed that simulates the effect of
67 ultrasonic treatment (UST) on acoustic streaming [15, 21] by assuming that the volume under
68 the sonotrode tip is a source of momentum that accelerates the surrounding fluid downwards
69 forming a jet based on the equations developed by Lighthill [22]. The model is able to predict

70 changes in convection patterns, velocity and temperature profiles generated throughout an
71 ingot during acoustic streaming and solidification. Very good correspondence between
72 experimentally measured and simulated cooling curves was obtained [15, 21]. Figure 1 shows
73 simulated temperature profiles when UST is not applied (Figure 1a) and while UST is being
74 applied (Figure 1b). When UST is applied the temperature gradient is very low compared to
75 the case without UST, and after UST is terminated a normal casting temperature profile with
76 a steeper temperature gradient is established. This quasi steady state created by UST during
77 cooling and nucleation of grains shows that the melt directly under the sonotrode is slightly
78 cooler than the bulk of the melt where the temperature gradient is very flat [15]. Given that
79 the melt is rapidly circulating under the sonotrode it would be expected that the temperature
80 under the sonotrode would be very close to the melt temperature. It has been suggested that
81 this condition where the ultrasound waves are at their most intense, is perfect for producing a
82 fine grain size as this is where cavitation occurs and acoustic streaming is generated [15, 23].
83 Once the grains are formed, acoustically generated convection transports them into melt of a
84 similar temperature and amount of undercooling which allows the grains to move without the
85 risk of remelting [15, 20] thus favoring the formation of an equiaxed grain structure.
86 Therefore, it was concluded that the convective flow induced by acoustic streaming plays a
87 critical role in promoting nucleation, growth, grain survival and their transport. However, it
88 should be noted that convection is dampened as the solid fraction increases, and could not be
89 maintained in the simulation once the solid fraction exceeded approximately 21%, the
90 coherency point [21, 24]. There are several types of coherency during solidification:
91 dendritic, globular, maximum packing fraction and tensile coherency [25]. For this work we
92 are referring to the morphological coherency: dendritic (where coherency occurs at a low
93 solid fraction) or globular (at higher solid fraction) which represents the point when the
94 grains begin to touch each other but are still not bonded together (i.e. the grains can still
95 move relative to each other). The smaller and rounder the grain size the greater the value of
96 coherency solid fraction. The value of 21% is the solid fraction after which computer
97 modelling showed that convection could no longer be maintained. Research has shown that
98 for large dendritic grains the morphological coherency point would be at 10% and for small
99 globular grains it could be as high as 45%. Thus, 21% indicates the situation for small
100 dendrites and/or medium sized globular grains.

101 The current work seeks to validate the model and to better understand the role of UST
102 during and after the nucleation stage on grain formation and microstructural refinement and

103 to identify the physical phenomena involved during solidification of an Al–2 wt% Cu alloy.
 104 The role of acoustic streaming and the development of solid fraction in the formation of the
 105 final grain structure are also discussed.



106

107 **Figure 1.** Simulated chill effect of the cold sonotrode on the melt temperature distribution at
 108 a simulation time of (a) 101 sec without UST and (b) 107 sec with UST, adapted from ref
 109 [15].

110 2. Materials, experimental methods and experimental design

111 An aluminium alloy with 2% Cu (compositions are in wt%) was produced in an electric
 112 melting furnace from 99.7% commercially pure aluminium and 99.9% pure copper. The
 113 liquidus and solidus temperatures measured using differential thermal analysis (DTA-TG)
 114 and the chemical composition measured by spectral analysis of the alloy are presented in
 115 Table 1. The temperatures were measured with an accuracy of 0.5 °C and the composition is
 116 accurate to 5 rel.%.

117 **Table 1.** Liquidus and solidus temperatures, and chemical composition of the Al–2% Cu
 118 alloy.

Liquidus (°C)	Solidus (°C)	Chemical composition (wt%)				
		Al	Cu	Si	Fe	Ti
655	620	Bal	2.02	0.03	0.09	<0.01

119

120 The ultrasonic device used in this work consists of a 2 kW commercial ultrasound
121 generator, an air cooled 20 kHz transducer, and a sonotrode made of molybdenum based
122 alloy with a tip 18 mm in diameter. About 1 kg of the alloy was melted inside a graphite-clay
123 crucible with dimensions 90 mm top diameter, 60 mm bottom diameter and 120 mm in
124 height. When the melt temperature reached $720\pm 5^{\circ}\text{C}$ the crucible was removed from the
125 electric furnace and transferred to the experimental platform, [which consists of the ultrasonic
126 device with an air cooling unit, a refractory brick for seating the hot crucible, a sonotrode
127 lifting unit, and a data acquisition system as illustrated in \[26\]](#). When the melt temperature
128 reached 695°C which is 40°C above the liquidus temperature, the powered sonotrode without
129 preheating was immersed into the melt to about 15 mm below the top surface of the melt. The
130 UST experiments were conducted with a fixed 1 kW input power. UST was terminated by
131 removing the powered sonotrode and then turning it off. Two K-type thermocouples were
132 inserted into the melt to one side of the ultrasonic probe. One was close to the edge of the
133 melt volume adjacent to the wall of the crucible and the other was 12.5 mm from the wall,
134 with both thermocouples placed 45 mm above the bottom of the crucible as indicated in
135 Figure 2a. The temperature data was collected by a data-acquisition system with a sampling
136 rate of 4 readings per second.

137 Three experiments were undertaken to provide information about the source of the
138 equiaxed grains (i.e. the location of the nucleation events), their transport into the bulk of the
139 melt, evolution of the micro and macro structures, and the morphology of the grains.

140 The first set of experiments was designed to confirm the location of the source of
141 nucleation of equiaxed grains and the convective flow generated by acoustic streaming for
142 the conditions used in this study. A gauze barrier has been used to isolate solidifying volumes
143 for many decades to determine the origin of the equiaxed zone [\[27-30\]](#) and the location where
144 new grains are nucleated [\[29-31\]](#). Yin et al used this technique to confirm that nuclei
145 dissociate from the mould wall with and without the application of electric current pulses [\[30\]](#),
146 and Li et al used the same method to investigate the origin of grains of an AZ80 Mg alloy
147 under a low voltage pulsed magnetic field [\[33\]](#). Gauze sheets were constructed to isolate the
148 liquid under the sonotrode as illustrated in Figure 2b. The melt was then cooled with the
149 sonotrode turned on [for 4 min after which it was removed from the solidifying melt](#) as
150 described above. The macrostructure was then evaluated. In addition, a glycerine-sand
151 analogue was subjected to similar UST conditions to that applied in the following
152 experiments to highlight the formation of convection due to acoustic streaming (Figure 2c).

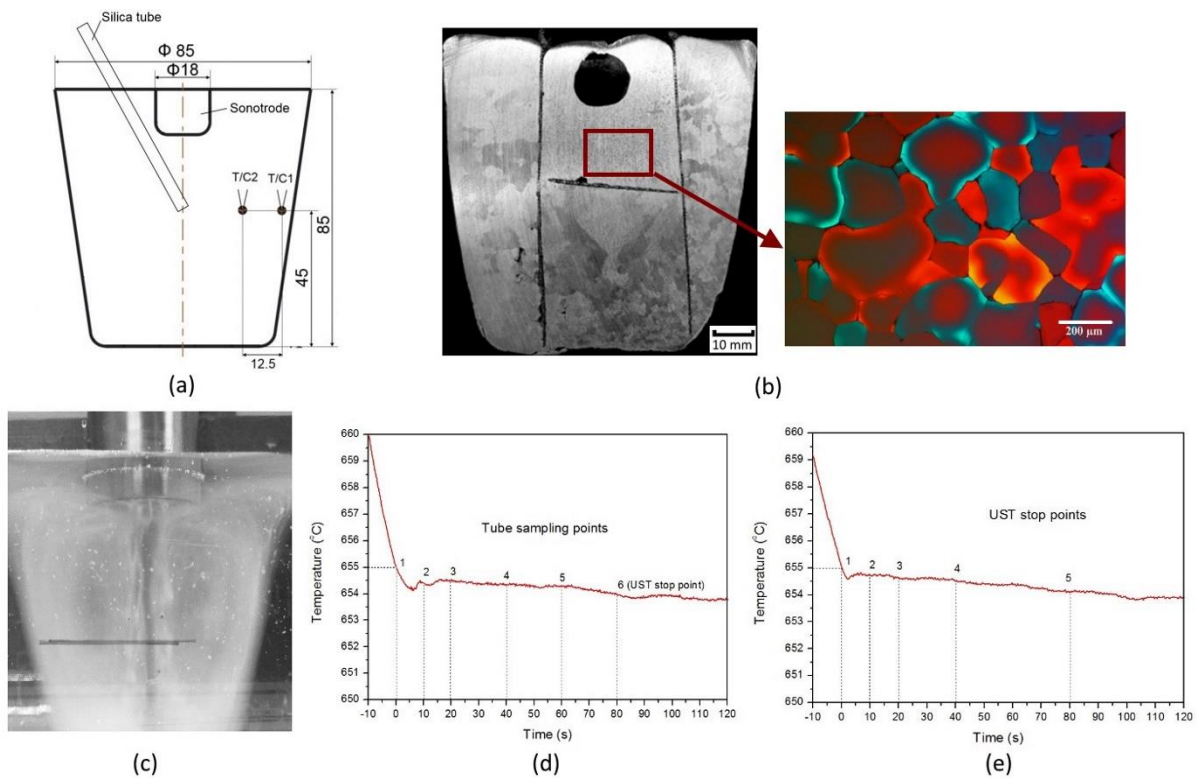
153 Although glycerine is more viscous than liquid aluminium and thus the degree of cavitation
154 and streaming will be different [34], it is likely a similar pattern of convection will be
155 induced as indicated by the simulation [15].

156 In the second series of experiments the evolution of grains under the sonotrode was
157 investigated by inserting a small 5 mm diameter silica tube of 1 mm wall thickness into the
158 melt to draw hot samples from under the sonotrode at a depth close to the position of the
159 thermocouple nearest to the sonotrode (Figure 2a), followed by quenching the sample into
160 cold water. Prior to inserting the tube into molten aluminium, it was preheated to 300°C to
161 keep the tube dry and moisture free to avoid a violent moisture reaction with molten
162 aluminium. It was expected that on insertion into the melt, the tube would quickly increase in
163 temperature due to heat transfer from the surrounding melt. Some nucleation may be
164 triggered by the relatively cold tube but this would be small compared to the total amount of
165 nucleation that contributes to the the final grain size, and similar for each sample such that
166 the measured trends would be unaffected. The time between filling and quenching was less
167 than 2 seconds so any drop in temperature would be considered to be part of the quenching
168 process. There was no observable reaction between the melt and the silica tube and the
169 sample was readily removed after quenching. The samples were taken during UST at 0, 10,
170 20, 40, 60 and 80 s after the measured melt temperature reached 655°C. The sampling points
171 are indicated on the cooling curve shown in Figure 2d to indicate the order in which the
172 samples were extracted by the silica tube. Recalescence would be occurring while the first
173 and second samples were extracted. Figures 2d and e show that despite the temperature
174 fluctuating during solidification the undercooling remains within 2°C below the liquidus
175 temperature. This condition facilitates further nucleation and the survival, movement and
176 growth of grains [23] while a large amount of liquid (> 75%, as determined by Thermocalc)
177 is present for the length of time that UST was applied.

178 In the third series of experiments the evolution of the macrostructure of the cast ingots was
179 studied. For each test approximately 1 kg of alloy was melted in the crucible and then the
180 activated sonotrode was applied without preheating, from 695°C and terminated after 10, 20,
181 40 and 80 s (as shown in Figure 2e) by removing the sonotrode and turning off the ultrasonic
182 power to allow the melt to solidify. After growth of the newly formed grains that occurs
183 during UST, solidification continues as the melt cooled in air until solidification was
184 complete below the solidus temperature. Previous work [15, 20] showed that a superheat of
185 40°C was sufficient to heat the sonotrode above the liquidus temperature and remelt any chill

186 crystals formed on immersion of the cold sonotrode before solidification began. The final as-
187 cast grain structure was then examined.

188 Metallographic samples for ingots were sectioned along the central symmetrical axis,
189 mechanically ground and polished using standard metallographic equipment for observation.
190 Macroetching was done using a solution of hydrofluoric, nitric and hydrochloric acids. In
191 order to measure the grain size, small samples were cut at 45 mm (the same height as the
192 thermocouples) from the bottom of the sectioned piece. Tube samples were cold mounted in
193 resin moulds, ground and polished along the axial direction axis. For both tube and ingot
194 samples, micrographs were obtained using a Leica Polyvar microscope with polarized light
195 after anodizing using a 0.5% HBF_4 water solution for about 20 seconds at 30 VDC. The
196 number of spherical/globular equiaxed grains in the tube's total longitudinal cross section
197 were manually counted. This number was converted to grain density per cm^{-3} . The grain size
198 was measured using the linear intercept method (ASTM E112-10). Statistical analysis of the
199 results was performed. In order to understand the relationship between temperature and solid
200 fraction during the solidification process, DTA-TG curves were obtained using a Netzsch
201 STA 449C at a cooling rate of 10–20 °C/min which is a slower cooling rate than that of the
202 cast sample cooled in the ingot crucible which had a cooling rate of 30-40°C/min above the
203 liquidus temperature.



206 **Figure 2.** (a) Schematic of a cast ingot indicating the internal dimensions of the casting
207 cavity, the location of the ultrasonic probe (sonotrode), thermocouples and the tube for
208 collecting samples of the semisolid alloy from under the sonotrode; (b) macro- and micro-
209 structure obtained from a gauze experiment to isolate the source of equiaxed grains ([the large](#)
210 [pore formed when the sonotrode was removed for the ingot](#)); (c) acoustic streaming observed
211 upon UST of glycerine; (d) a cooling curve highlighting the times when sampling occurred
212 during UST applied from 695°C for different durations measured from the time the melt
213 reached the liquidus (655°C) at 0 s (sample 1), 10 s (sample 2), 20 s (sample 3), 40 s (sample
214 4), 60 s (sample 5), and 80 s (sample 6); and (e) the UST termination points for five separate
215 ingot castings are shown after the melt reached 655°C at 1 (0 s), 2 (10 s), 3 (20 s), 4 (40 s)
216 and 5 (80 s). The cooling curve is from the thermocouple nearest the sonotrode.

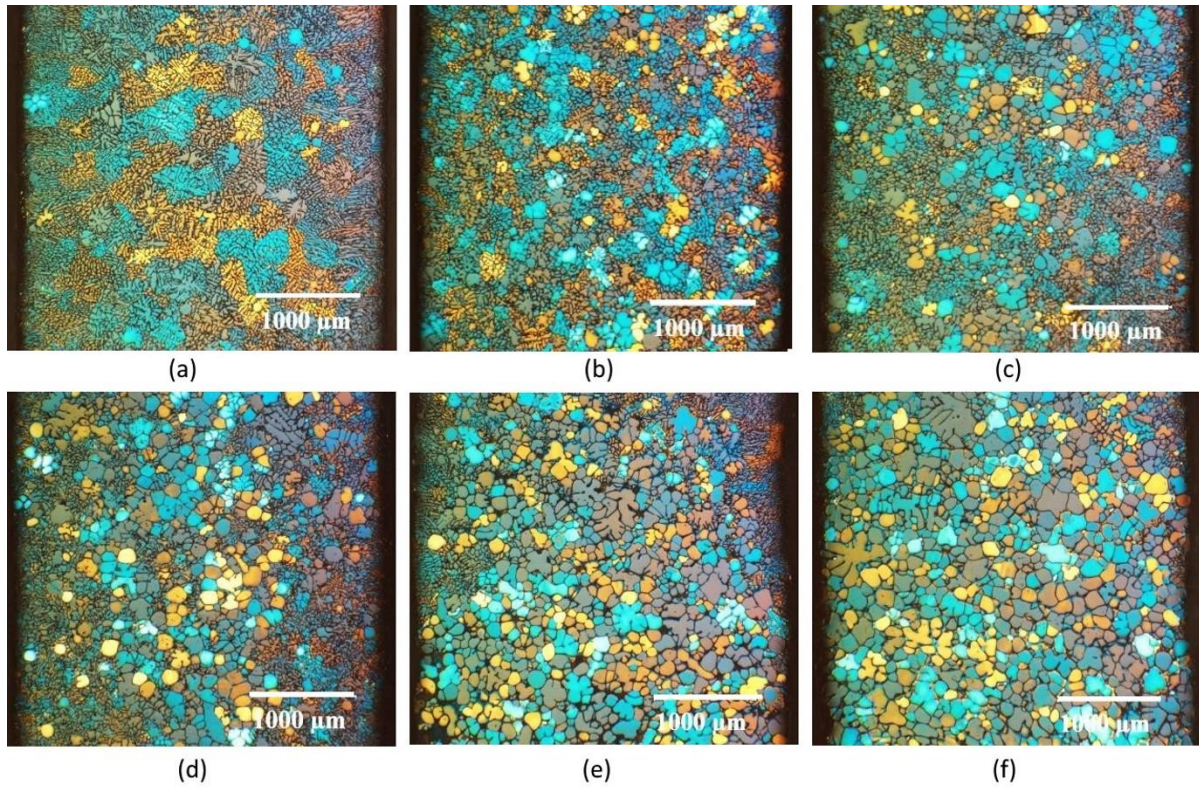
217 **3. Results and discussion**

218 **Source of grains and their transport:** As mentioned in the Introduction, the focus of this
219 study is on the effects of acoustic streaming rather than cavitation which is assumed to be
220 occurring under the sonotrode. This means the actual nucleation events have not been studied
221 but their location, the number forming over time, how they are transported throughout the
222 melt and the degree of grain growth that occurs. Figure 2b confirms that the main source of
223 equiaxed grains is from the region under the sonotrode (i.e. the cavitation zone). The grain
224 size in this zone is refined (see adjacent microstructure) while outside this zone on the other
225 side of the gauze barrier, large and columnar-like grains are observed. The glycerine-sand
226 analogue of a similar size to the ingot crucible, Figure 2c, shows that the sonotrode generates
227 streaming that causes the liquid to flow vertically downwards. Upon reaching the bottom of
228 the container the melt flows across the bottom and then up the walls of the container
229 generating a circulating flow pattern as this liquid is then sucked into the region under the
230 sonotrode. This observation confirms the simulation results presented in [15]. The sand
231 particles are carried by convection although some particles become lodged in the bottom
232 corners of the container. Figure 2c and simulations in Figure 1 [15] indicate that the newly
233 nucleated grains of the Al-2%Cu melt will also be forced to move vertically downwards
234 along with any fragmented grains eventually circulating back to the region under the
235 sonotrode and then circulated repeatedly until they become mechanically lodge in the
236 corners of the casting cavity. It would be expected that as the number and size of grains
237 increases the size of the mechanically locked regions would become deeper and spread across
238 the bottom and up the walls of the ingot.

239 It should be noted that the observations made from these experiments apply to a relatively
240 small ingot. For much larger ingots the velocity of acoustic streaming will gradually decrease
241 with distance from the sonotrode due to attenuation weakening convection. Because the melt
242 is pushed towards the bottom of the ingot, some melt will flow upwards into the cavitation
243 zone maintaining a circulating flow probably in the upper part of the ingot due to the
244 resistance of the melt below. However, it would be expected that as the depth of the ingot
245 increases the decreasing strength of convection may assist many grains of higher density than
246 the liquid to continue sinking to the bottom of the ingot. Depending on the temperature
247 gradient, the settling grains will impede the growth of adjacent grains forming a refined
248 equiaxed microstructure. If a temperature gradient is maintained by heat being extracted
249 through the container walls then a columnar zone could form until stopped by the settling
250 grains as observed for UST of Zn [10].

251 **Evolution of grains:** Figures 3a-f show the evolution of grains in the liquid during UST in
252 samples extracted from the melt volume directly under the sonotrode. As the samples were
253 quenched in water immediately after extraction, their microstructure reflects grain formation
254 as UST progresses. The abrupt increase in the cooling rate at the cessation of UST causes
255 large dendrites with fine branches to form during quenching of the remaining liquid as
256 observed in [33]. The small spherical and globular grains (i.e. nondendritic grains) are formed
257 by nucleation and growth during the application of UST. Figure 4 shows the corresponding
258 quantitative relationships between (a) the number of nondendritic grains and (b) their average
259 size versus UST duration after the temperature of the melt reached just below the liquidus
260 temperature (655°C). Figure 4a shows that a small number of grains formed in the extracted
261 volume of melt. Note that zero time on the cooling curve (i.e. when the temperature reaches
262 the liquidus temperature) is when the temperature of the thermocouple nearest the sonotrode
263 reaches the liquidus temperature. The actual zero directly under the sonotrode should be
264 when the melt that is extracted reaches the liquidus temperature which will be a few seconds
265 earlier. An estimate of zero time can be made by extending the curves in Figure 4b to the x-
266 axis to the left of the y-axis. Depending on the actual steepness of these curves, the start of
267 solidification under the sonotrode would begin at about -6 s in relation to the temperature at
268 the thermocouple closest to the cavitation zone of the casting. For clarity during the
269 discussion, the grain size and grain density at 0 s on the cooling curve corresponds to
270 approximately 6 s after nucleations begins under the sonotrode. Therefore, reconsidering
271 Figure 4a the small number of grains observed were formed during the first 6 s of UTS after

272 the melt reached the liquidus temperature. Also note that Figure 4a is not representative of the
273 whole casting but only the area under the sonotrode from where the sample is extracted.
274 Acoustic streaming will quickly sweep these grains into the bulk melt. Each extracted sample
275 is a combination of the grains newly nucleated by the action of the sonotrode and grains that
276 have returned with the circulating melt to a position under the sonotrode one or more times.
277 Thus, a size distribution develops as illustrated by Figure 4b.



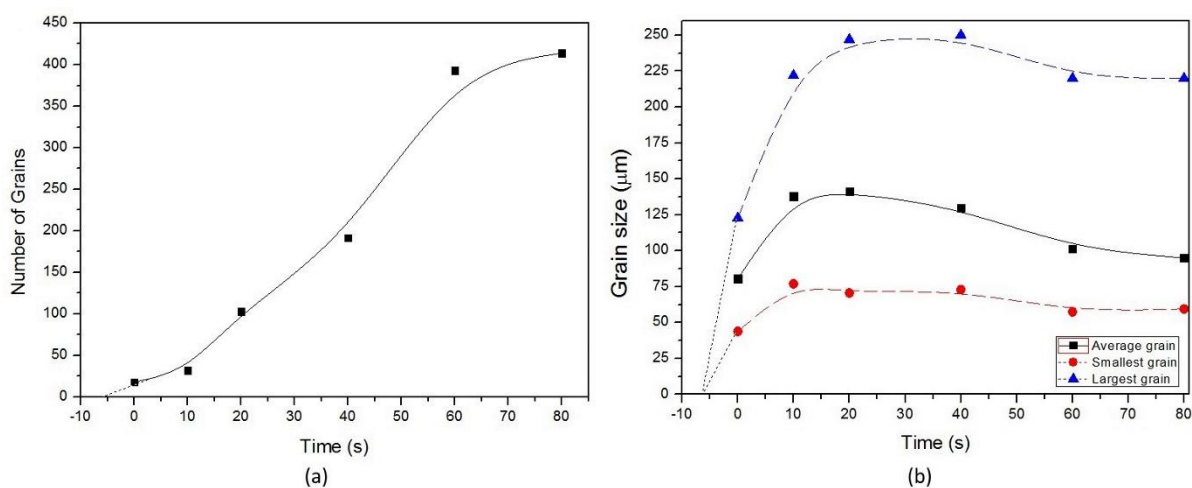
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279 **Figure 3.** The grain structure of the tube samples taken from the melt during UST after the
280 melt temperature at the thermocouple reached 655 °C at (a) 0 sec, (b) 10 sec, (c) 20 sec, (d)
281 40 sec, (e) 60 sec, and (f) 80 sec (see Figure 2d for reference). The larger fine dendritic
282 structure observed in (a) to (e) is formed on quenching of the preheated tube sample extracted
283 from below the sonotrode. The area fraction of these fine dendrites decreases as the number
284 and size of spherical and globular grains increases.

285 As noted above, the microstructure corresponding to just below the liquidus temperature
286 (Figure 3a) shows only a few small spherical grains. This observation suggests that UST prior
287 to reaching the liquidus temperature does not have a significant impact on the activation of
288 substrates for the nucleation of the primary grains, which is in good agreement with our
289 previous work [16]; and/or that some of the nucleated grains are too small to be observed.
290 Figure 3b and Figure 4a show that the number of spherical nondendritic grains in the

291 quenched dendritic matrix approximately doubles during the first 10 s, indicating that
 292 continued nucleation occurs during 10 s of UST. According to Figure 2d this period is during
 293 recalescence. However, the region under the sonotrode would be beyond the time of
 294 recalescence which occurs for about 10s (Figure 2d). With the time increasing from 20 to 40,
 295 60 and 80 s, the number of nondendritic grains increases significantly (Figure 4a) while the
 296 grain size begins to stabilize in size after 20 s as shown in Figure 4b. After 20 s the
 297 distribution of nondendritic grains becomes more and more uniform and after 60 s a **uniform**
 298 solid network appears to be established, evidenced by interconnected nondendritic grains and
 299 porosity formed due to the lack of feed liquid (Figure 3e). Upon continuous application of
 300 UST for 80 s after reaching the liquidus temperature nondendritic grains dominate the
 301 microstructure (Figure 3(f)). It is possible that small non-dendritic grains formed during
 302 quenching. The maximum number of these grains would be less than 10 grains measured at
 303 zero (i.e. 6 s after the liquidus temperature is reached). Compared to the number of grains
 304 measured at subsequent times the quenched grains would have little affect on any conclusions
 305 drawn from the data.

306 Figure 2b and the simulation in Figure 1b highlight that the new grains are produced in the
 307 region under the sonotrode. However, we cannot determine whether some of these grains
 308 nucleated on the bottom surface of the sonotrode or all grains were formed by cavitation-
 309 enhanced heterogeneous nucleation in melt adjacent to this surface. Either mechanism is
 310 possible [15, 20].

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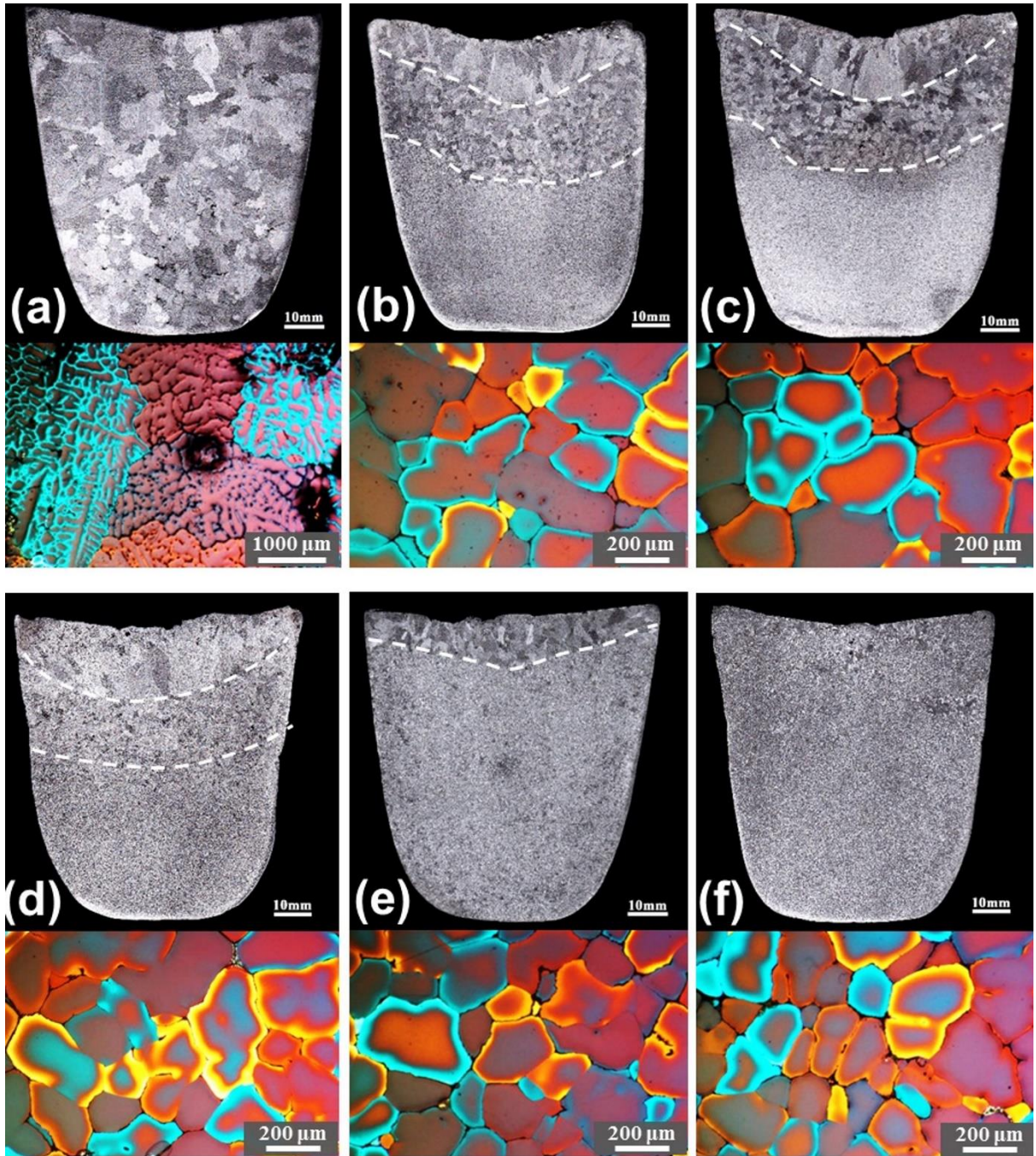


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313 **Figure 4.** (a) The total number of spherical/globular nondendritic grains measured in the
 314 tube's longitudinal cross section (9 mm^2) of the extracted volume and (b) the size of

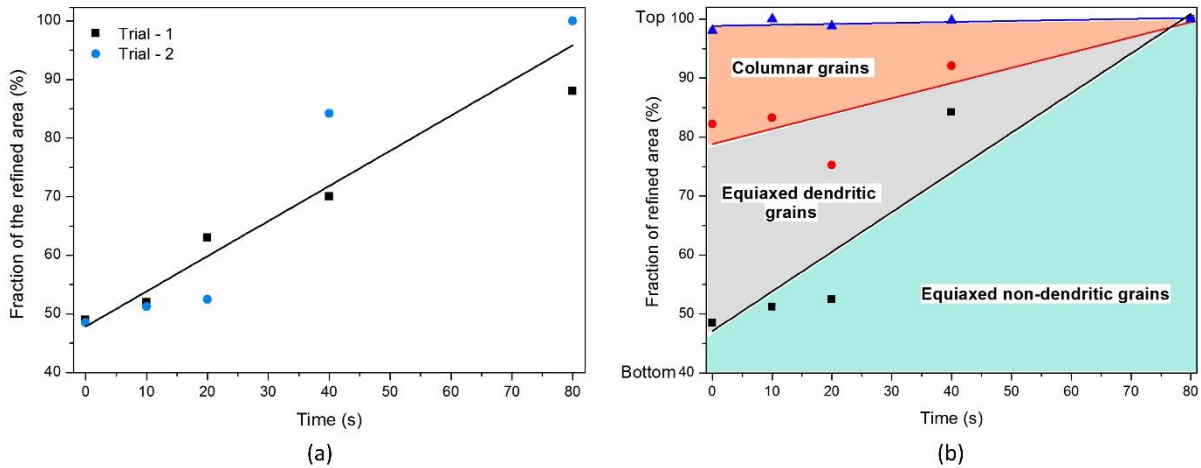
315 spherical/globular grains versus UST duration after the liquidus temperature is reached (see
316 Figure 2d for reference).

317 **Evolution of macrostructure:** Figure 5 shows the macrostructures of the ingots cast without
318 (Figure 5a) and with (Figures 5b-f) UST corresponding to the range of UST termination times
319 marked in Figure 2e. Without UST a fully dendritic grain structure of large grains with rather
320 thick secondary dendrite arms are observed in Figure 5a. In the case of the ingots produced
321 with UST (Figures 5b–f), the vertical cross section of each casting is clearly divided into
322 three regions as indicated by the superimposed boundary lines, i.e. a fine grain region at the
323 bottom and a coarse grain region at the top with a structure similar to that in Figure 5a, and a
324 large columnar grained area at the top of the ingot. The lack of a columnar zone next to the
325 walls and bottom of the ingot and the similar grain size obtained throughout the equiaxed
326 zone indicate that the temperature gradient measured between the two thermocouples extends
327 throughout the melt as predicted by the simulation (Figure 1, [15]). The size of the region of
328 fine equiaxed grains progressively increases with increasing ultrasonication time at the
329 expense of the coarse grained region (Figure 6a). Figure 6b highlights the corresponding
330 regions of grain structure delineated by dashed lines mapped across the cross section area of
331 Figures 5b-f. The grain size in the fine equiaxed region decreases sharply in the first 20 s of
332 UST with possible slight coarsening occurring afterwards, as illustrated in Figure 7a. Figure
333 7b compares the grain density of the extracted tube samples and the equiaxed zone of the cast
334 ingots showing that the grain densities of both the extracted and ingot samples tend towards
335 the same approximate value. Therefore, UST duration is important for generating a large
336 region of fine equiaxed grains but not necessarily for controlling the grain size in the fine
337 grain region at the bottom.



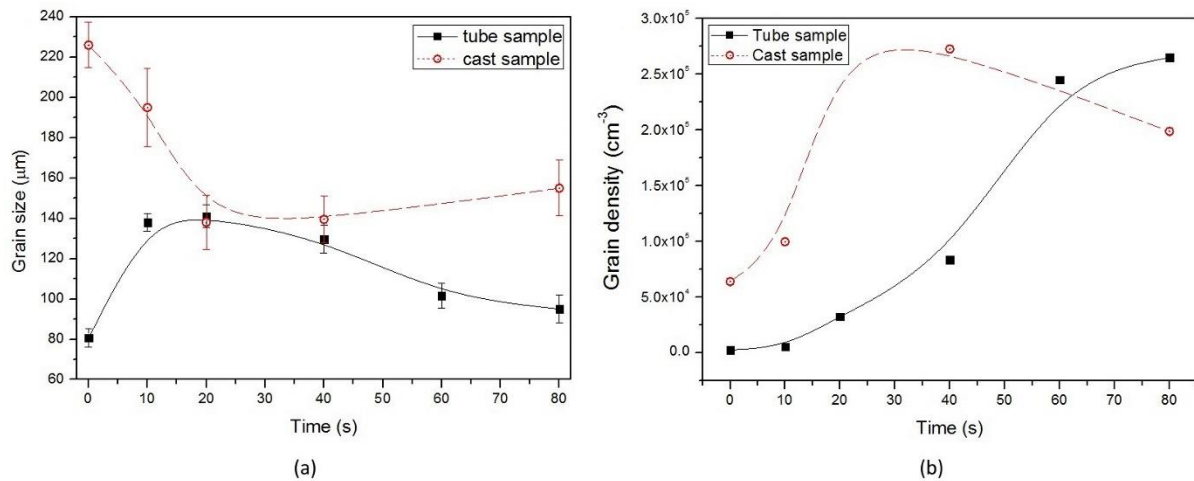
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339 **Figure 5.** Grain structure of the cast ingots (a) without UST, and with UST stopped at (b) 0 s
 340 (c) 10 s (d) 20 s (e) 40 s and (f) 80 s (see Figure 2 e for reference) after reaching the liquidus
 341 temperature of 655°C. Note the different scale for the microstructure in 5 a.



342
 343 **Figure 6.** (a) The area fraction of the refined region versus UST duration after the liquidus
 344 temperature is reached where each point corresponds to one casting and (b) the corresponding
 345 regions of grain structure delineated by dashed lines mapped across the cross section area of
 346 Figures 5 b-f.

347
 348 In the current study, terminating UST just below the liquidus (655°C) (i.e. after 0 s based
 349 on the cooling curve but actually after 6 s under the sonotrode) resulted in grain refinement of
 350 about 50% of the volume, and the grains were refined to about 220 μm, as shown in Figure 8a.
 351 At the same time, only a few small nondendritic grains were observed in the sample taken in
 352 situ from under the sonotrode (Figure 3a). The continuously increasing UST duration resulted
 353 in a gradual increase in the proportion of refined structure. After 20 to 40 s of UST the
 354 minimum grain size of 140 μm is reached. After that time a coherent network of solid grains
 355 would begin to form and penetration of the acoustic flow into this region will become
 356 hindered. At the same time, the ultrasonic energy is still being supplied to the system,
 357 resulting in slower cooling (see the relatively flat cooling curves in Figures 2d and e) and
 358 possibly grain coarsening as has been reported elsewhere [35].



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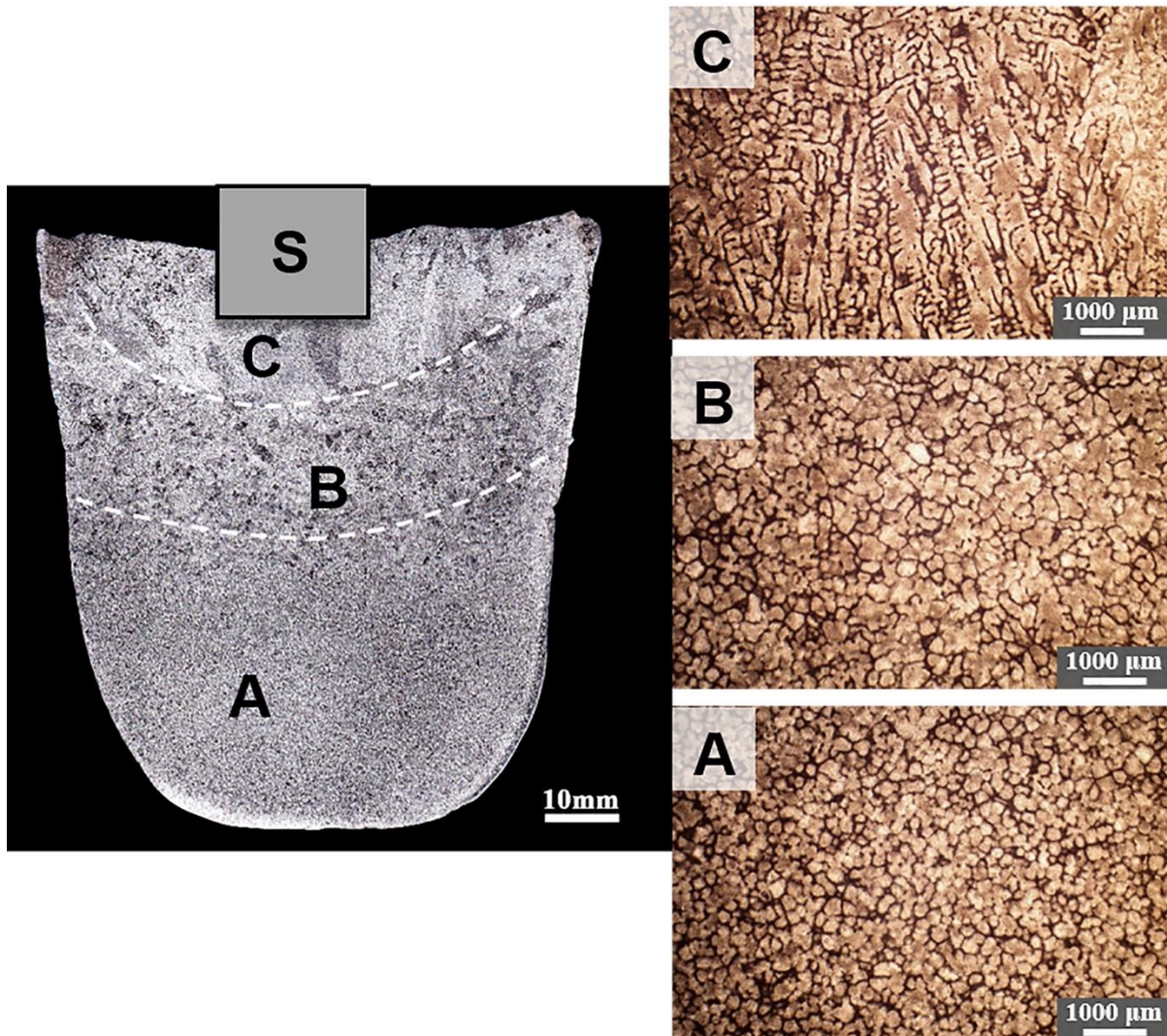
361 **Figure 7.** (a) Grain size comparison between tube samples and the refined equiaxed region of
362 the cast ingots and (b) comparison of grain density between tube samples and ingots versus
363 time.

364 The combination of refined nondendritic equiaxed grains in the bottom region and coarse
365 dendrites in the upper region of the sample (Figures 4b–e) can be explained as follows: (1)
366 during the nucleation stage the initially formed and refined grains are generated continuously
367 by UST up to a solid fraction of less than 25%; (2) below 25% solid fraction and as long as
368 UST continues, the nucleated grains are distributed into the bulk of the melt by acoustic
369 streaming and are kept in suspension and moving, therefore a uniform refined grain structure
370 can be achieved in the refined equiaxed region due to the bulk of the melt being below the
371 liquidus temperature; (3) terminating UST after establishment of the coherent solidified
372 network leads to a fully refined equiaxed grain structure throughout the ingot casting; (4) the
373 unrefined region is formed after termination of UST because the vigorous convection stops
374 and solidification reverts to the case when UST is not applied and large grains are formed in
375 the remaining melt (Figure 5a). Many of the grains present in the melt after termination of UST
376 sink due to the density differential between the liquid and grains depleting the liquid of grains
377 [35] while very few new crystals are generated at this stage of solidification. The grain size in
378 the equiaxed zone is controlled by the amount of growth that occurs while in suspension and
379 by the point during settling when they mechanically contact each other.

380 The samples taken from below the sonotrode include new grains and grains that are re-
381 circulating due to convection. Figure 4a shows that the number of grains below the sonotrode
382 gradually increase. However, the size range of these particles become more or less constant
383 after about 10 seconds (Figure 4b) despite the number of grains continuing to increase. In the

384 extracted samples the rate of increase is about 7 grains per second in the quasi-steady-state
385 regime from 10 to 60 seconds.

386 Figure 4b suggests that after 20 s the size range of grains becomes approximately constant.
387 When the grains mechanically contact each other the rate of settling would cease or
388 dramatically decrease. It would be expected that grains will first collect at the corners and
389 edges of the ingot much like the sand particles in Figure 2c. This implies that there are three
390 zones responsible for the formation of the macrostructure which are illustrated schematically
391 in Figure 8. Zone A is where the grains settle during and after UST is terminated forming a
392 refined equiaxed microstructure. In the melt above Zone A grains continue to move and grow
393 while acoustically-assisted convection continues to be generated. Zone B forms when UST is
394 terminated and the recirculating grains sink into Zone A depleting this Zone of grains
395 allowing the remaining grains to grow larger. Zone C forms in response to the radiant heat
396 cooling the top of the melt such that grains nucleate and grow with a columnar morphology
397 into the melt when the few grains remaining in this region sink into Zone B such that a region
398 of unrefined columnar grains form.



399

400

401 **Figure 8.** Three distinct zones formed in the macrostructure are identified as A, B and C
 402 under a specified duration of ultrasonication after reaching the liquidus (655°C) of the Al-
 403 2wt.%Cu alloy. Zone A represents fine equiaxed grains formed during UST that sink to the
 404 bottom of the crucible until UST is terminated after 40 seconds. Zone B is depleted of many
 405 of the last to form grains when they sink into Zone A providing more room for growth into
 406 larger dendritic or rosette type grains. Zone C refers to the columnar grains that grow under
 407 the influence of the radiant heat transfer from the top surface of the ingot in a region that is
 408 unaffected by UST.

409 **The relationship between grain formation and macrostructure development**

410 Figure 7 allows comparison of the development of, and the relationship between, grain
 411 size and grain density between under the sonotrode and in the bulk melt. Figure 7a shows the
 412 difference between grain size in the tube samples and the final grain size in the ingots. During
 413 the first 20s the grain size in the tube samples increases from 80 to 140 mm while in the

414 ingots it decreases from 225 to 140 μm . This reflects the space available in the melt for grain
415 growth. By referring to Figure 7b the grain density in the tube sample increases slowly while
416 it increases faster in the ingot. Thus, at short times the lower grain density of grains produced
417 can grow over a larger distance until impeded by other grains. As time increases the grain
418 density increases with grains accumulating in the melt reducing the distance between grains
419 such that the average grain size in the ingot decreases. After 20 s the number density under
420 the sonotrode continues to increase as new grains are formed until the grain density becomes
421 similar to that in the cast ingot. Growth is now severely restricted thus the grain size under
422 the sonotrode decreases indicating that new grains are still being generated. The grain size in
423 the refined equiaxed zone begins to increase slightly possibly due to grain coarsening.
424 However, the size of the error bars suggest there may be little change in grain size after 20 s.
425 The difference in grain density between the equiaxed zone in the ingot and under the
426 sonotrode represents the proportion of grains circulating in the melt that are not recirculated
427 under the sonotrode. Figure 6a shows that the area fraction of the equiaxed zone in the ingots
428 increases steadily from 50 to about 85%. Figure 7b shows that the grain density stays about
429 the same after 20 to 30 s implying that the increase in area fraction of fine equiaxed grains
430 from then on is due to an increase in the number of grains being produced as evidenced by
431 the curve for the tube samples.

432 **Morphology of grains:** Due to the imposed agitation the grains mostly remain spherical or
433 globular in morphology with few dendritic grains being observed from their first appearance
434 until 80 s when UST is terminated. Also, the average grain size remains relatively constant
435 and possibly decreases after 20 s of UST (Figure 4b). These observations imply that a
436 dendritic morphology does not readily form and, therefore, fragmentation is unlikely to
437 significantly contribute to grain density for this alloy. This conclusion is consistent with the
438 analysis by Kotadia et al. [7] who concluded that fragmentation does not occur in CP Al and
439 Al-10%Cu alloy.

440 **4. Concluding remarks**

441 An Al-2Cu alloy was subjected to UST over a range of durations while cooling from 40°C
442 above the liquidus temperature to below the liquidus for up to 80s. The grains that form an
443 equiaxed zone are initially nucleated under the sonotrode. The microstructure of samples
444 extracted from below the sonotrode were compared with the macrostructure on the as-cast
445 ingots. The results are consistent with the predictions of the simulation study [15]: nucleation

446 occurs under the cooler sonotrode, a relatively flat temperature gradient forms in the warmer
447 but undercooled bulk melt, and the importance of acoustically-generated convection in
448 forming an equiaxed zone of refined grains.

449 A number of insights were revealed:

- 450 • The cooling curves show that although the temperature fluctuates under UST, the degree
451 of undercooling remains within less than 2 °C below the liquidus temperature such that
452 the grains can grow in a constitutionally supercooled liquid ensuring the survival of
453 grains while considerable liquid (> 75%) remains present. The heat generated by the
454 ultrasonic energy would contribute to maintaining a relatively stable temperature range.
- 455 • A quasi-steady-state is created under the sonotrode where, within the field of view, seven
456 grains were formed every second from about 20 to 60 s. This process continues as the size
457 of the equiaxed zone increases from about 20 to 85% of the ingot's cross-sectional area.
458 40 s is the optimum time of UST to ensure the finest grain size over the largest area while
459 after 80 s the largest area of equiaxed grains of a slightly larger size are formed.
- 460 • The development of macrostructure over time shows three microstructural zones form
461 with an increasing zone of fine equiaxed grains, a diminishing zone of large globular and
462 dendritic grains, and a zone of columnar grains at the top of the casting. Many of the
463 grains appear to remain in the convective field until UST is terminated after which time
464 they settle towards the bottom of the ingot forming an equiaxed zone. The grain size in
465 the equiaxed zone is set by the density of settling grains. Initially, an increasing grain
466 density contributes to an increased size of the equiaxed zone. After about 20s, continued
467 application of UST does not increase the grain density but the number of grains generated
468 over time continues to increase which in turn increases the area fraction of the refined
469 equiaxed zone up to 85%. After 80 s the zones of larger globular, dendritic and columnar
470 grains are suppressed while the fine equiaxed zone fills the macrostructure.
- 471 • Grain size is controlled by the number of grains present in the melt and the time available
472 for settling. Thus, few grains produced at shorter times grow to a larger size than when,
473 after a longer time, many grains have formed with comparatively limited space left to
474 grow.
- 475 • The new grains initially form with a spherical morphology. The morphology becomes
476 globular with some dendritic grains present after 80 s of UST. The globular morphology
477 is maintained by the vigorous convection circulating in the ingot.

- 478 • The observation of mostly spherical and globular grains throughout the application of
479 UST indicates that fragmentation is not a significant source of additional grains during
480 UST of an Al-2Cu alloy. The fully dendritic grains observed in the tube samples and the
481 large grains above the equiaxed zone are formed when UST is terminated either by
482 quenching or cooling in air.

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