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# **Influence of growth velocity on fragmentation during directional solidification of Al – 14 wt.% Sn alloy studied by in-situ synchrotron X-radiography**

**L. Abou-Khalil<sup>a\*</sup>, K. Sabat da Cruz<sup>b</sup>, G. Reinhart<sup>a</sup>, N. Mangelinck-Noël<sup>a</sup>, H. Nguyen-Thi<sup>a</sup>**

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## **Abstract:**

Directional solidification of Al – 14 wt.% Sn alloy was investigated by means of synchrotron X-radiography at the European Synchrotron Radiation Facility (ESRF). The dynamics of the solidification microstructure formation was studied in-situ and in real-time. In the present manuscript, a special attention is given to the influence of increasing growth velocity on the fragmentation phenomenon of dendrite arms, which has an important role and impact on the final grain structure. Fragmentations of both secondary dendrite arms and more strikingly part of primary trunks have been observed. The experiments show that the number of fragments increases concomitantly with the solidification growth rate until it reaches a limit value after which fewer or no more fragments are observed. The possible origins of the fragmentation are discussed in detail and the likelihood of each of these potential mechanisms is estimated to understand the reason of both primary and secondary arm detachment. It is shown that the fragmentation in a transient regime is correlated to the variations of the solute accumulation, latent heat rejection and the curvature of the attachment neck during solidification. Moreover, during these experiments, Columnar-to-Equiaxed Transition (CET) induced by the fragmentation phenomenon is observed. The velocity of the dendrite fragments due to the buoyancy force

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1 is a major parameter to control the CET and our results show that a reverse Equiaxed-to-  
2 Columnar Transition can even be observed. Once the growth velocity is of the same order  
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4 or higher than the fragment flotation velocity, the fragments are trapped within the  
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6 columnar dendritic microstructure, and CET cannot take place or the equiaxed regime  
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8 cannot be sustained.  
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12 **Keywords:** Solidification, X-ray imaging, Al-Sn, Dendrites, Fragmentation, CET.  
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## 16 17 **1. Introduction** 18

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20 Solidification of metal alloys generally leads to the formation of microstructures at the  
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22 solid-liquid interface, like cells and dendrites, which determine to a great extent the final  
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24 mechanical properties of the elaborated material. It is well established that those  
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26 microstructures are at the origin of severe micro-segregation, as well as of several defects in  
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28 castings, ingots and welding like hot tears or freckles.  
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32 To obtain a regular and stationary dendritic array, it is mandatory to maintain the growth  
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34 conditions constant during the whole process, as far as possible. However, even in this case,  
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36 dendrite fragmentation phenomenon can take place during the solidification process, which can  
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38 generate segregation, microstructure inhomogeneity and defects. Fragmentation is the process  
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40 of detachment of secondary or tertiary dendrite arms from the primary trunk, or possibly of the  
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42 primary trunk itself. This mechanism occurs for example during the casting of steel ingots for  
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44 which massive fragmentation is observed during solidification in the centre of the ingot where  
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46 the temperature gradient is approaching zero [1,2]. In non-refined alloy casting, the detachment  
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48 of dendrite fragments is the mechanism that can explain the Columnar-to-Equiaxed Transition  
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50 (CET). It is generally thought that CET can happen if the detached fragments are transported  
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52 by convection/buoyancy force not too far ahead of the columnar front and can grow to block  
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54 the progress of the columnar front [3,4]. Another industrial example for which dendrite  
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1 fragmentation plays a detrimental role in the manufacturing of turbine blades made of Nickel-  
2 based superalloys [5]. Indeed, the relationship between dendrite fragmentation and freckle  
3 occurrence remains an open question for the specific processes of turbine blade fabrication.  
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7 To explain why secondary/tertiary dendrite arms detach from primary trunk during  
8 solidification, several mechanisms based on the curvature difference at different parts of  
9 dendrite arms have been proposed. One of them is the remelting of the dendrite arm neck  
10 because of recalescence during solidification from an undercooled melt [6]. This phenomenon  
11 is associated to rapid solidification and it can be at the origin of grain refinement [7,8]. Another  
12 mechanism is related to solute effects that cause a curvature-driven remelting. The dendrite  
13 arms can remelt during coarsening process due to the Gibbs-Thomson effect [9] or by solute  
14 enrichment caused by natural convection [10,11] or forced melt flow [12–14], which modify  
15 the equilibrium temperature, thus promoting fragmentation.  
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29 Clarifying the underlying mechanisms that induce solutal-driven detachment requires  
30 experimental methods that allow in-situ studies of both morphological and solutal aspects.  
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32 Ruvalcaba *et al.* [10] presented results obtained from directional solidification experiments  
33 observed by synchrotron X-radiation microscopy. According to their in-situ observations, the  
34 solute enrichment at the neck of the tertiary dendrite branches is produced by the solute-rich  
35 melt pushed by buoyancy force that flows through the mushy zone (interdendritic liquid). Liotti  
36 *et al.* [13] analysed the dendrite fragmentation in the mushy zone by synchrotron X-ray  
37 radiography during the directional solidification of Al-Cu alloys with the application of a pulsed  
38 electromagnetic field. The authors confirmed that the main mechanism of the fragmentation  
39 and detachment of the dendrite arms is the solute pile-up at the bottom of the dendrite branches  
40 causing the re-melting. Zimmermann *et al.* [15] and Shevchenko *et al.* [16] studied the effect  
41 of pulling rates combined with different fluid flow on the dendritic microstructure. It was shown  
42 that the fragmentation is at the origin of the grain refinement at low solidification velocities  
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1 independently of the flow intensity in the melt in a non-refined alloy. When increasing the  
2 pulling speed, columnar growth dominates, the number of grains decreases, and the mean grain  
3 size concomitantly increases in the case of natural convection or of weak intensity forced  
4 convection. Oppositely, many equiaxed grains are observed under intense forced flow. These  
5 observations are attributed to the competition between the solidification front growth rate and  
6 the fluid flow velocity and their effect on the fragment transportation.  
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14         Apart from the solutal mechanism, Ananiev *et al.* [17] studied the effect of mechanical  
15 loading due to buoyant forces on the solid-liquid interface in thermodynamic equilibrium. They  
16 showed that elastic energy can lead to what they called catastrophic elastic remelting of the  
17 neck that attaches the secondary arms to the primary trunk and can lead to fragmentation. Indeed,  
18 buoyant forces can be at the origin of cumulative bending moments due to the lower moment  
19 of inertia there. Secondary arms grow and become heavier and after a certain time the stress is  
20 exceeded at the necks, which causes their sudden and irreversible rotation or bending [18].  
21 Frequently, remelting/dissolution of thin solid neck follows arm bending, which results in  
22 dendrite fragmentation. The rotation and bending effects have also been analysed by Reinhart  
23 *et al.* [19] and Jung *et al.* [20] during the solidification of Al – 7 wt.% Si. Furthermore, Salloum-  
24 Abou-Jaoude *et al.* [21] underlined the major impact of gravity on the fragmentation  
25 phenomenon, as well as on the movement of fragments in Al – 20 wt.% Cu by performing a  
26 comparison between solidification experiments carried out on Earth and in microgravity. In a  
27 review paper, Akamatsu *et al.* [22] reported dramatic difference of fragmentation density  
28 between experiments carried out in microgravity conditions, without buoyancy force and fluid  
29 flow, and under normal conditions. They concluded that the fragmentation is most likely not a  
30 purely diffusive process. Cai *et al.* [23] emphasized the impact of buoyancy forces on dendrite  
31 fragmentation during vertical upward solidifications of Al – 15wt.% Cu samples via X-ray  
32 tomography. The authors concluded that mechanical deformation from various sources may  
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1 play a much greater role in grain fragmentation during solidification than previously thought.  
2 In a very recent paper, Nelson *et al.* [24] found a marked difference in fragmentation density  
3 between vertical upward and downward solidification for the same alloys. They attributed this  
4 to the change of buoyancy force that modulates the solute flow convection and that stresses the  
5 dendrite arms.  
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11 The fragmentation during directional solidification of Al – 14wt.% Sn alloys is the subject  
12 of the present work. Al-Sn alloys are well known for having excellent tribological [25] and  
13 mechanical properties, making this kind of system suitable for the application in combustion  
14 engine pistons and cylinder liners [26]. The origin of their outstanding properties relies on the  
15 Sn particles spread over a continuous Al-rich matrix. The tough Al-rich matrix, which is more  
16 resistant to high mechanical loads, works in combination with the Sn particles that act as solid  
17 lubricants. The experiments presented in this paper were carried out at the European  
18 Synchrotron Radiation Facility (ESRF) where in-situ X-radiography technique was used to  
19 reveal the dynamics of grain structure formation with a particular focus on fragmentation and  
20 on CET induced by fragmentation. The solidification was achieved in a transient condition that  
21 is similar to industrial casting processes such as direct-chill casting [27], with the concomitant  
22 increase of growth velocity and decrease of temperature gradient. This allows a better  
23 understanding of the fragmentation phenomenon during the elaboration of the industrial  
24 material and its effect on the final structure [4,20]. Several mechanisms at the origin of  
25 fragmentation are discussed in details as well as the impact on the resulting microstructure and  
26 grain structure.  
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## 2. Experiments

Directional solidification experiments were carried out on beamline ID19 at ESRF (Grenoble, France) in a Bridgman furnace set-up previously described in detail in [28,29]. The nominal alloy composition was Al – 14 wt.% Sn. The sample dimensions were 40 mm in length, 5 mm in width and about 300  $\mu\text{m}$  in thickness. It was sandwiched between two graphite foils and two molybdenum diaphragms, held together with two clips and fixed to a sample holder. The whole assembly was then inserted vertically into the Bridgman furnace placed in a high vacuum chamber (Fig.1).

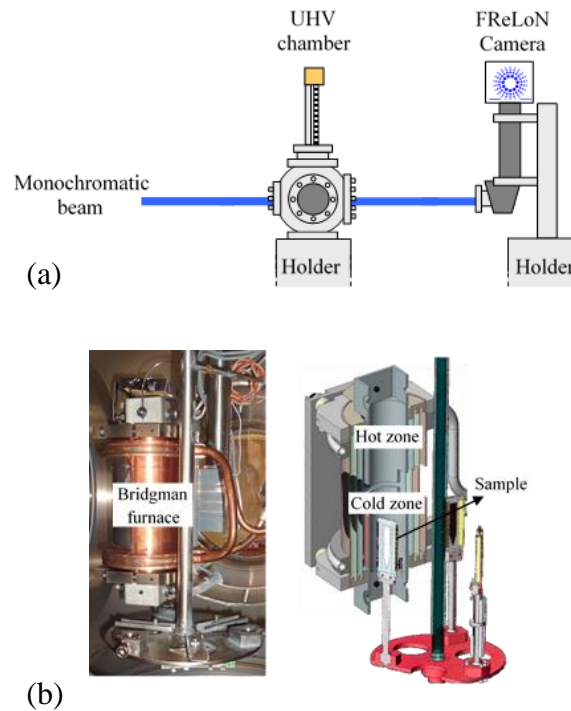


Figure 1: (a) Sketch of in-situ synchrotron X-ray imaging setup. (b) Picture and 3D drawing of Bridgman furnace.

The temperatures of the top and bottom zones of the furnace were first adjusted to achieve the desired initial temperature gradient  $G_i$  (Table 1). The temperature gradient was applied between the two heater elements regulated by two embedded K-type thermocouples 4.5 cm

1 apart. Then, solidification was triggered by cooling only the top heater of the furnace ( $R_T$ ) while  
 2 maintaining the bottom heater temperature constant, with displacement of neither the sample  
 3 nor the furnace. The application of a cooling rate only on the top heater induced a progressive  
 4 decrease of the temperature gradient in the sample, similar to industrial casting conditions, for  
 5 example direct-chill casting [27]. For all experiments, an initial transient period was observed.  
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 7 After a while, the decrease of the temperature gradient became linear. The values of the  
 8 temperature gradient variation with respect to time ( $\dot{G}$ ) are given in Table 1. The observation  
 9 and analysis of experiments were performed during the period of linear decrease of the  
 10 temperature gradient.  
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24 Table 1: Experimental parameters;  $R_T$  is the cooling rate applied to the top heater element. The  
 25 temperature of the bottom heater element is maintained constant.  $G_i$  and  $G_f$  are the initial and  
 26 final temperature gradients, respectively.  $\dot{G}$  is the temperature gradient variations with respect  
 27 to time.

| 28 Experiments | 29 $R_T$ (K/min) | 30 $G_i$ (K/cm) | 31 $G_f$ (K/cm) | 32 $\dot{G}$ (K /cm/min) |
|----------------|------------------|-----------------|-----------------|--------------------------|
| 33 (A)         | 0.2              | 22.4            | 10.6            | -0.04                    |
| 34 (B)         | 0.5              | 26.0            | 14.1            | -0.11                    |
| 35 (C)         | 8.0              | 23.7            | 15.3            | -1.70                    |
| 36 (D)         | 16               | 22.6            | 17.0            | -2.44                    |

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 41 The availability of high energy X-rays in synchrotron accelerators allows the emergence  
 42 of characterization methods than can be applied to the study of solidification microstructure  
 43 formation of metallic alloys [30,31]. In this paper, synchrotron X-radiography was used to  
 44 investigate the dynamics of the microstructure formation during solidification and solute  
 45 distribution in the melt. This technique is based on the local changes in the amplitude of a  
 46 transmitted X-ray beam due to differential absorption by an inhomogeneous sample [32,33]. In  
 47 Al-based alloys, contrast firstly results from segregation of the chemical species and secondly  
 48 from the difference of density between the solid and liquid phases. X-radiography enables us  
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1 to follow the dynamics of the solid-liquid interface [33], the grain motion [21,34,35] and under  
2 certain conditions to determine quantitatively the solute concentration in the liquid phase  
3 [36,37]. In our experiments, the main surface of the sample (40 x 5 mm) was perpendicular to  
4 the horizontal incident monochromatic X-ray beam with an energy of 14 keV. The Fast  
5 Readout–Low Noise (FReLoN) CCD camera developed at ESRF was used as detector and the  
6 camera optics were chosen to obtain a good compromise between a large field of view (14 mm  
7 x 6 mm) and a suitable spatial resolution (pixel size 7.46 x 7.46  $\mu\text{m}^2$ ). The high intensity of  
8 synchrotron radiation makes it possible to record images with enough contrast. Images were  
9 recorded with different exposure time (0.5 to 4 s) and time intervals (1 to 14 s) depending on  
10 the solidification velocity and on the phenomena of interest.

11 For the experiments presented in this paper, successive images of the solid-liquid zone  
12 were recorded during the whole solidification step, and it was thus possible to display videos  
13 of the time evolution of the solid-liquid interface. Image processing was applied to improve the  
14 quality of the radiographs, which consisted in dividing the raw images of the sample taken at  
15 time  $t$  during solidification by a reference image. This reference image was recorded just before  
16 cooling down started, when the sample was mostly liquid and homogeneous with a constant  
17 initial solute concentration  $C_0$  [36]. The result was an enhanced contrast and a defect-free image.  
18 In these images, the Al-dendrites that have a lower density relatively to the Sn-enriched liquid  
19 appear in white or light grey and Sn-enriched liquid in dark grey.

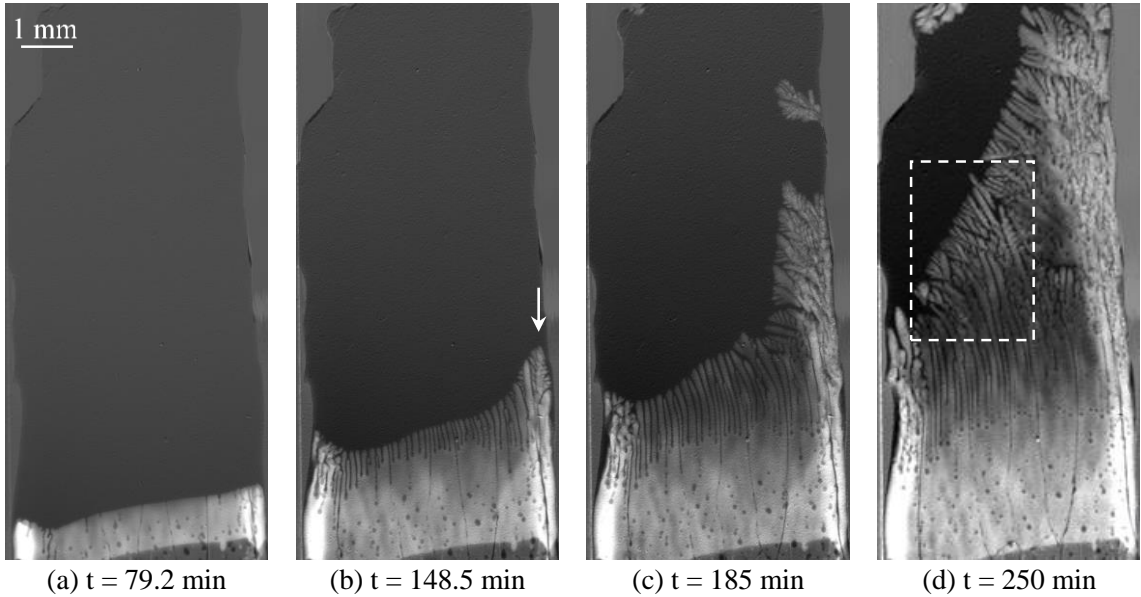
## 20 **3. Results**

### 21 **3.1. In-situ investigation of the microstructure evolution/interface destabilization**

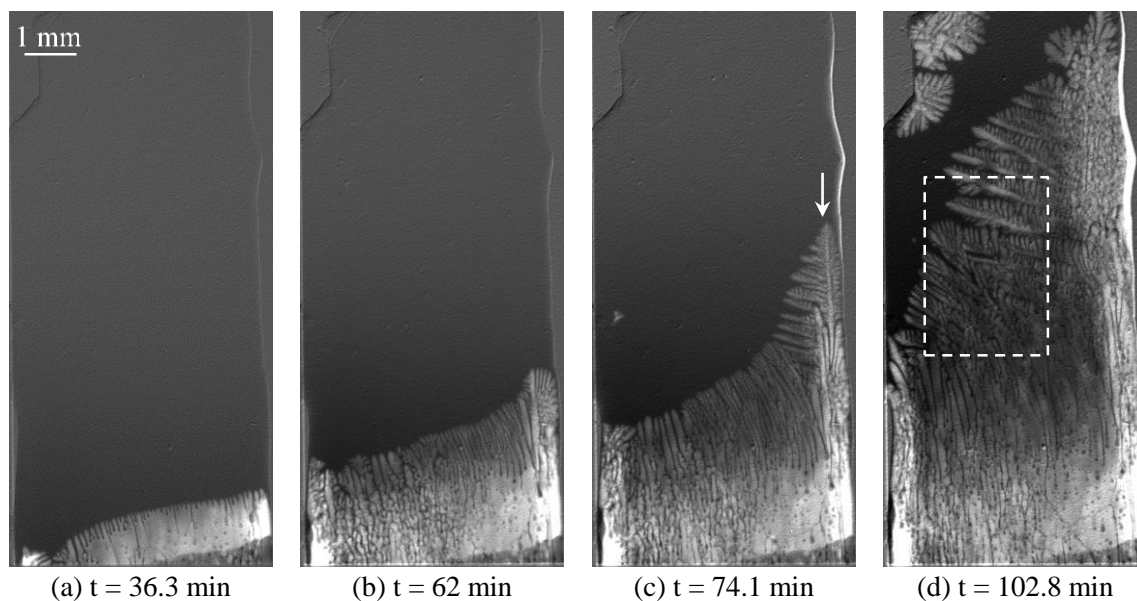
22 Figures 2 and 3 display two sequences of radiographs showing the evolution of  
23 solidification microstructures of Al – 14 wt.% Sn alloy, at a very slow cooling rate of 0.2 K/min  
24 and 0.5 K/min on the top heater element, respectively. Radiographs show that Al-Sn samples

1 did not keep their initial rectangular shape and were deformed during the melting phase.

2 At the early stages of the cooling process, the initial solid-liquid interface is smooth and inclined  
3 (Fig.2a), most likely due to a residual transverse temperature or solutal gradients. As  
4 solidification proceeds, the gradual decrease of the longitudinal temperature gradient augments  
5 the instability level of the solid-liquid interface, so that a transition from a planar to cellular  
6 front occurs after few minutes of the cooling process, and a dendritic microstructure is formed  
7 on both sides of the sample (Fig.2b and Fig.3a,b). The dendritic pattern then spreads towards  
8 the centre of the solid-liquid interface until the whole interface become dendritic. As the sample  
9 cooling continues, the formation of developed microstructures with more branches that become  
10 more pronounced is observed (Fig.4).

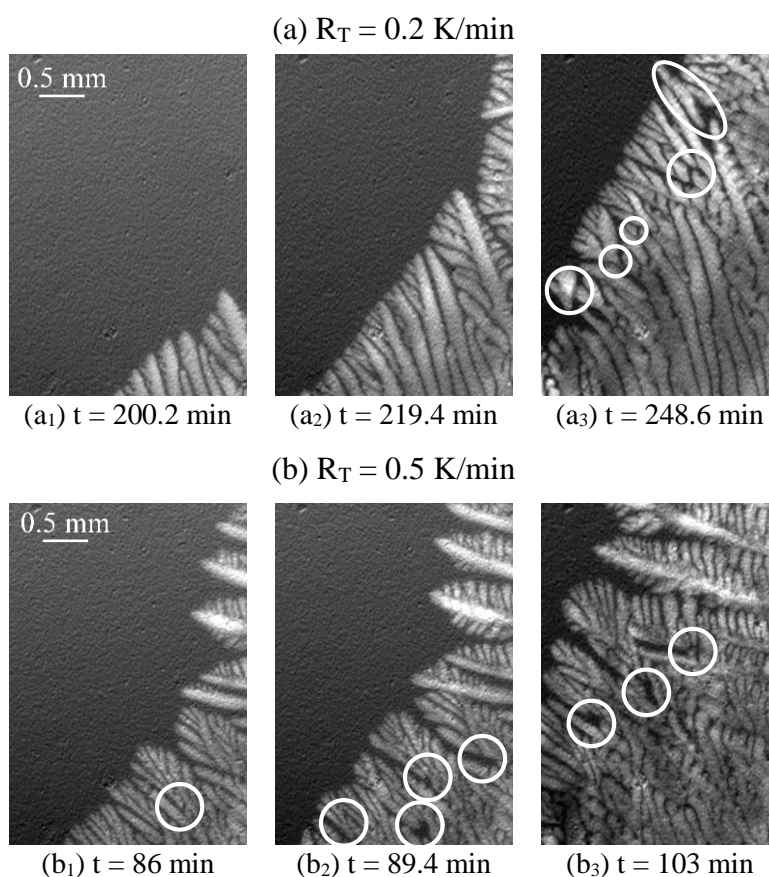


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Figure 2: Sequence of radiographs showing the evolution of the solidification microstructure of Al – 14 wt.% Sn by applying a cooling rate of  $R_T = 0.2$  K/min on the top heater element only. The white arrow points to the dendrite whose growth rate evolution is plotted in Figure 5a. A close-up of the growth of the microstructure growth inside the dashed white zone is shown in Figure 4a (top). (See supplementary material for the corresponding movie).



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Figure 3: Sequence of radiographs showing the dynamics of the solid-liquid interface during the solidification of Al – 14 wt.% Sn by applying a cooling rate of  $R_T = 0.5$  K/min on the top heater element only. The white arrow points to the dendrite whose growth rate is plotted in Figure 5b. A close-up of the growth of the microstructure inside the dashed white zone is shown in Figure 4b (bottom). (See supplementary material for the corresponding movie).



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Figure 4: Sequence of radiographs showing the evolution of the microstructure and the dendrite branching inside the white dashed zones in Figures 2 and 3 solidified by applying a cooling rate of  $R_T = 0.2$  K/min and 0.5 K/min on the top heater element only, respectively. The white circles represent the location of some of the dendrite fragments.

1 The dendrite growth velocities of the two far right dendrites in Figures 2 and 3 are measured  
 2 from the radiographs and are plotted in the graphs in Figure 5. As expected, the growth velocity  
 3 increases during the experiment as solidification is achieved by cooling only the top heater of  
 4 the furnace while maintaining the bottom heater temperature constant. The growth rate  
 5 increases, from  $\sim 1.2 \mu\text{m/s}$  to  $\sim 3 \mu\text{m/s}$  for the experiment solidified at  $R_T = 0.2 \text{ K/min}$  and from  
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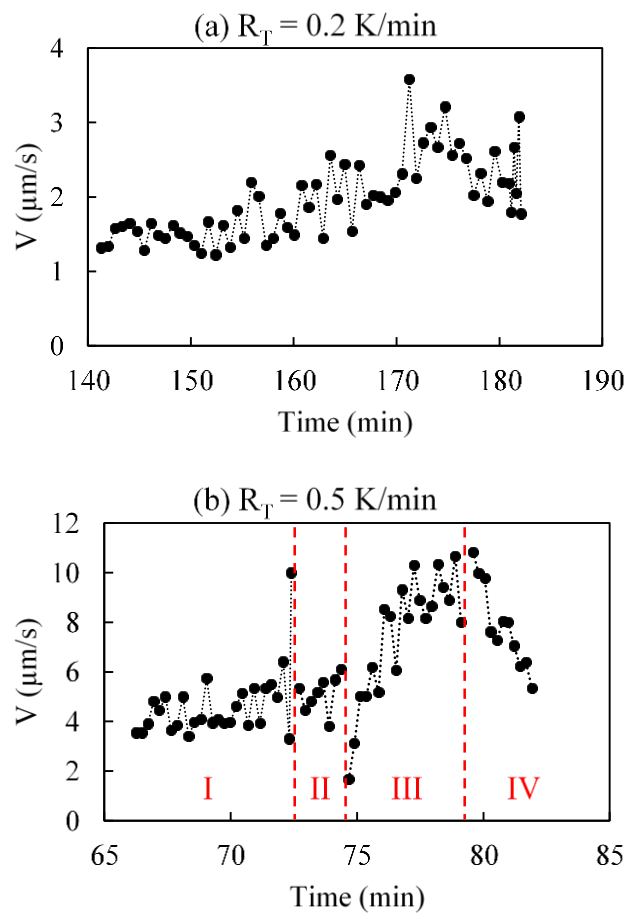


Figure 5: Tip growth rate as a function of time of the two far-right dendrites (pointed by white arrows) for the experiments solidified by applying (a)  $R_T = 0.2 \text{ K/min}$  (in Figure 2) and (b)  $R_T = 0.5 \text{ K/min}$  (in Figure 3). The red dashes indicate when the dendrite detachments occur.

### 3.2. In-situ investigation of fragmentation

In the experiments solidified at cooling rates  $R_T = 0.2$  K/min and  $R_T = 0.5$  K/min, the first fragmentation phenomena are surprisingly observed during the cellular growth for a growth velocity larger than  $\sim 1.7$   $\mu\text{m/s}$ ; below this value no fragment is detected. The detachment of few cells is observed directly by in situ X-radiography for the first time to the best of our knowledge (Fig.6). In Figure.6b, the cell to dendrite transition occurs with the development of some secondary arms in the sample thickness.

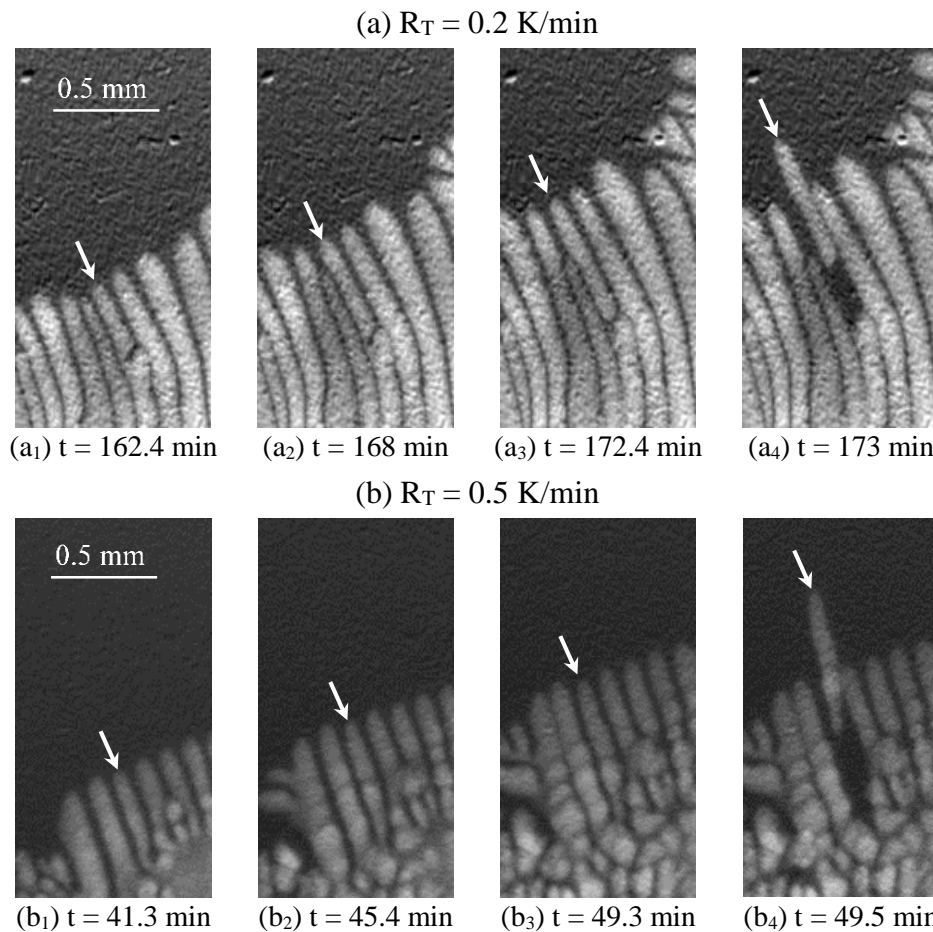


Figure 6: Sequence of radiographs, taken from the experiments solidified by applying a cooling rate of (a)  $R_T = 0.2$  K/min and (b) 0.5 K/min on the top heater element only, showing the evolution of cell neck shape and size before its detachment.

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2 At a later stage, following the development of the dendritic microstructure, several  
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4 detachments of dendritic branches occur during the two experiments. As the growth velocity  
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6 increases, the number of fragments also increases progressively. Additionally, during the  
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8 dendrite development, secondary dendrite arm bending is repeatedly observed and generally  
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10 followed by detachment. They rotate around their neck in the upward direction by 3° to 7° and  
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12 then detach (Fig.7). Moreover, a repetitive and somewhat unexpected phenomenon in these  
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14 experiments is the fragmentation of the primary dendrite trunks (Fig.2c, Fig.3c and Fig.7). By  
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16 using in-situ observation, the detached fragment behaviour is followed in detail. The free  
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18 fragments, i.e., not blocked inside the mushy zone, float towards the top region (i.e., towards  
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20 higher temperatures) and melt while others get stuck in the thickness of the sample and continue  
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22 to grow locally (see supplementary materials).  
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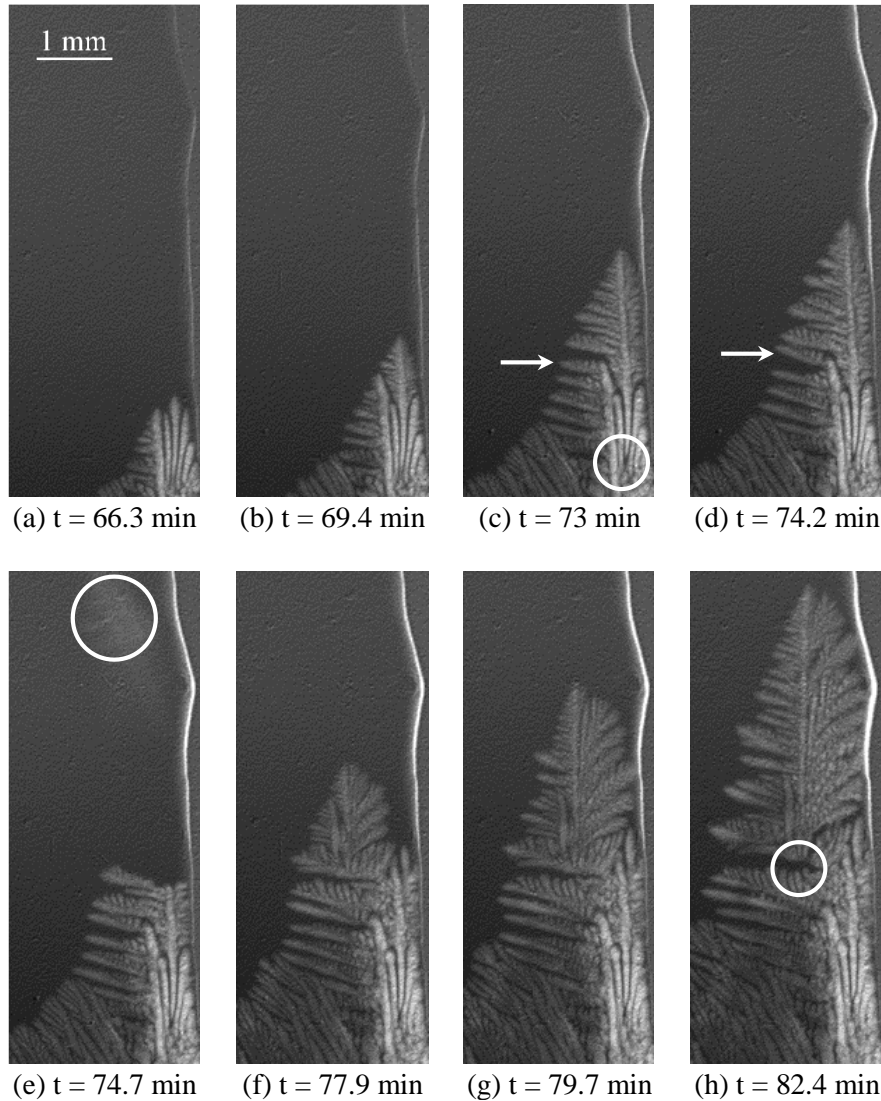


Figure 7: Sequence of radiographs showing the dynamics of the far-right dendrite pointed by a white arrow in Figure 2. The detachments of the primary trunk are highlighted by white circles. The white arrows showing the position of the secondary arm before and after bending.

A remarkable example of the phenomena observed in our experiments is the dynamics of the far-right dendrite in Figure 2, which is initially a tertiary arm. The behaviour of this dendrite is presented in Figure 7 and described in detail in the following. First, its growth velocity increases from  $\sim 4 \mu\text{m/s}$  to  $\sim 10 \mu\text{m/s}$  with a maximum of growth rate just before its first detachment (Fig.7a,b and region I in Fig.5b). The broken primary branch (white circle in Fig.7c) stands close to its initial position and starts growing again. The upward bending of some secondary arms is observed (Fig.7c,d). Several seconds later, a new maximum value of the growth velocity is reached ( $\sim 4.5 \mu\text{m/s}$  to  $\sim 6 \mu\text{m/s}$ ) for the tip of the far-right dendrite and a

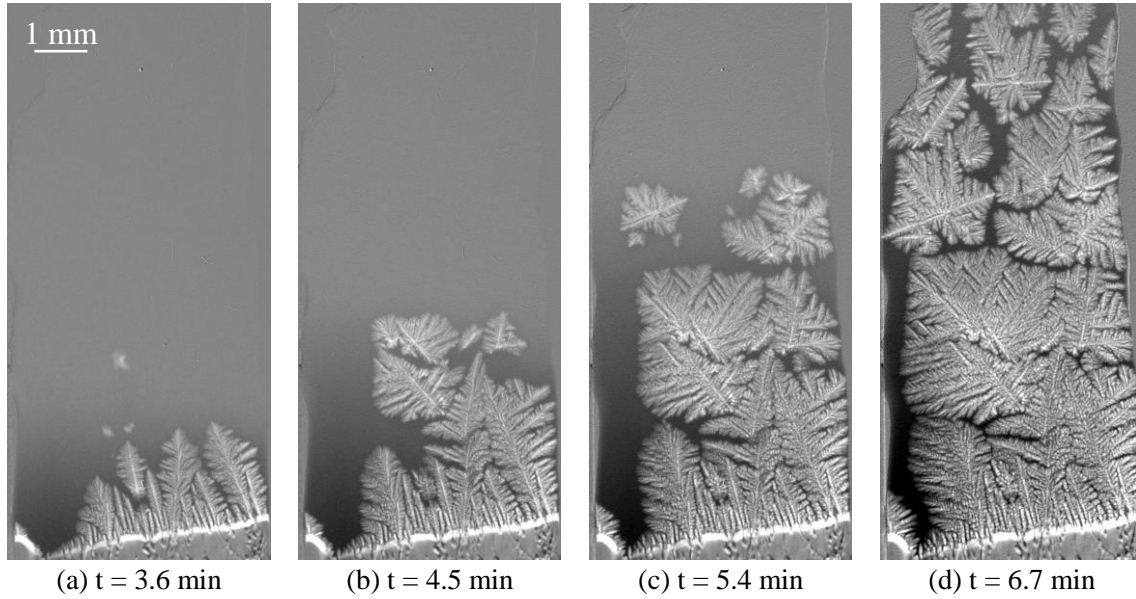
1 new detachment of the primary trunk occurs (region II in Fig.5b). This fragment floats and  
2 melts while reaching the hot region of the sample (Fig.7e). A tertiary arm takes over and grows  
3 (Fig.7f). Its growth rate first decelerates under the effect of the melted fragment, then it  
4 accelerates (region III in Fig.5b), and the tertiary arm continues its growth along the direction  
5 of the temperature gradient as a primary trunk ( $\sim 1.6 \mu\text{m/s}$  to  $\sim 10.6 \mu\text{m/s}$ ). Then, a bending of  
6 the secondary arm from which this tertiary arm started occurs (Fig.7g and region IV in Fig.5b).  
7 Consequently, the tip of the tertiary arm moves slightly upward in a liquid region with a lower  
8 solute concentration. A deceleration of the tertiary arm growth follows, which is probably due  
9 to the time necessary to recreate a solutal layer in front of the tip. Few minutes later, the  
10 secondary arm with its corresponding tertiary arm detaches, the fragment is blocked and  
11 continues its growth (Fig.7h).  
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26 It is worth noticing that the final structure of the sample solidified at 0.2 K/min and 0.5  
27 K/min appears columnar with no clear evidence of the fragmentation history except for the  
28 indirect effect of the fragmentation that induces dendrite disorientations from the initial growth  
29 direction (Fig.2d and Fig.3d).  
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### 39 **3.3. Columnar-to-Equiaxed Transition (CET) by fragmentation**

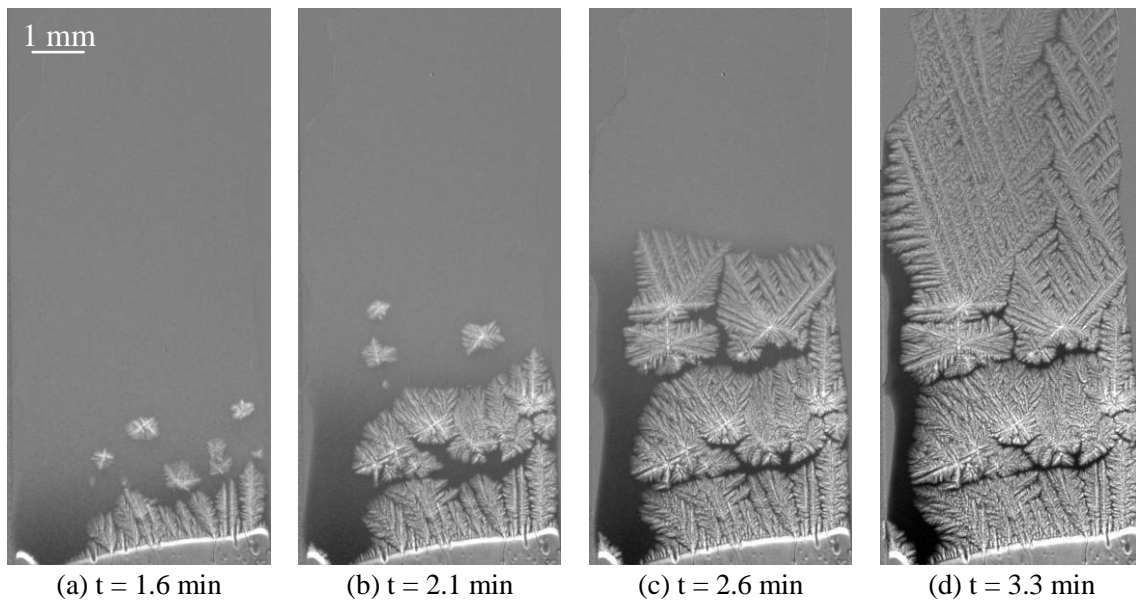
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41 Figures 8 and 9 show the microstructure evolution of Al – 14 wt.% Sn solidified at higher  
42 cooling rates of 8 K/min and 16 K/min on the top heating zone, respectively. For these higher  
43 cooling rate values, the solid-liquid interface becomes rapidly dendritic after the application of  
44 the cooling. Concomitantly, a lot of fragmentations of primary, secondary and tertiary branches  
45 are observed (Fig.8a, Fig.9a).  
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Figure 8: Sequence of radiographs showing the Columnar-to-Equiaxed Transition due to the fragmentation during the solidification of Al – 14 wt.% Sn by applying a cooling rate of  $R_T = 8$  K/min on the top heater element only. (See supplementary material for the corresponding movie).



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Figure 9: Sequence of radiographs showing the Equiaxed-to-Columnar Transition during the solidification of Al – 14 wt.% Sn by applying a cooling rate of  $R_T = 16$  K/min on the top heater element only. (See supplementary material for the corresponding movie).

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In the experiment solidified at  $R_T = 8$  K/min, some fragments float and melt once they reach the hot region while the others are blocked during their flotation and continue to grow locally forming a layer of equiaxed grains that blocks the initial columnar front (Fig.8a,b). The

1 equiaxed grains develop and become more and more elongated. Then, new fragmentations  
2 occur, and new grains are blocked in the thickness. This process is repeated causing successive  
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4 Columnar-to-Equiaxed Transitions (Fig.8c). The final grain structure is composed of several  
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6 layers of packed elongated grains either connected or separated by a Sn-rich segregated band  
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8 (Fig.8d).  
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11 For the experiment performed at  $R_T = 16$  K/min, the same microstructure behaviour is  
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13 observed in the beginning where one layer of elongated grains is formed that blocks the initial  
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15 columnar front, then two fragments from this first layer are blocked above the interface  
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17 (Fig.9a,b). Contrary to the previous experiment, at the end, a columnar front develops from the  
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19 solidification front without any further fragmentation (Fig.9c). Therefore, an Equiaxed-to-  
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21 Columnar Transition is observed (Fig.9d). It is worth noticing that fragmentation phenomena  
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23 are mainly observed inside the mushy zone in both samples during the coarsening step.  
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## 31 **4. Discussion**

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33 The most important observation in these experiments is the great number of fragmentation  
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35 events of secondary, tertiary and even primary arms during the sample cooling. As seen in the  
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37 introduction, several mechanisms can be at the origin of dendrite detachment such as  
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39 mechanical and solutal effects. These effects will be discussed in detail in this section, as well  
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41 as the consequence of fragmentation on the final grain structure.  
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### 47 **4.1. Effect of growth velocity on microstructure evolution**

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49 In the case of Al – 14 wt.% Sn alloy, most of solute is rejected during the liquid to solid  
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51 transformation because of the value of the partition coefficient  $k \approx 0.033$  which is well below  
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53 unity. As the solute (Sn) is denser than the solvent, it accumulates in the melt ahead of the solid-  
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55 liquid interface. This solute layer induces a constitutional undercooled region, which is at the  
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57 origin of the solid-liquid interface instability and the formation of cellular or dendritic patterns  
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1 [38,39]. Interface destabilisation depends on the temperature gradient ( $G$ ), growth velocity ( $V$ )  
2 or cooling rate and alloy concentration. The change in the interface morphology is controlled  
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4 by the ratio  $G/V$ , where the solid-liquid interface tends to be dendritic when the ratio is small  
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6 (small temperature gradient or high growth rate) [40]. In the experiments presented in this  
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8 article, the solidification occurs in transient conditions because of the gradual decrease of the  
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10 temperature gradient and the progressive increase of the growth velocity as shown in Figure 5.  
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12 Accordingly, the ratio  $G/V$  gradually slows down, which leads to the breakdown of the planar  
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14 solidification front to give cells and then, dendrites. In the case of the experiments with the  
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16 highest cooling rates applied on the top heater (8 and 16 K/min), the cellular pattern is skipped  
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18 and the solid-liquid interface is dendritic from the very early stages of solidification (see Fig.8  
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20 and Fig.9 and corresponding videos in the supplementary material).  
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26 The initial inclination of the solid-liquid interface observed for all experiments (Fig.2a,  
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28 Fig.3a, Fig.8a and Fig.9a) is caused by a transverse temperature gradient in the sample and  
29  
30 worsened due to the shape of the sample. This transverse temperature gradient induces a  
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32 transverse convective flow. Consequently, the solute concentration varies along the solid-liquid  
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34 interface from one side to the other. Solute accumulates in depressed regions, which modifies  
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36 locally the equilibrium temperature, and leads to the delay in the growth on the left side and to  
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38 the dendrite formation on the right side (Fig.2b, Fig.3b) [31]. Those two effects amplify  
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40 throughout the growth process.  
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46 The growth velocity also affects the dendrite dimensions. As the growth rate increases, the  
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48 microstructure becomes finer: a reduction in the diameter size of the dendrite arm is observed  
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50 and a decrease of the secondary arm spacing is detected [41]. As an example, in the experiment  
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52 where the applied cooling rate is  $R_T = 0.5$  K/min, the secondary arm spacing of the right dendrite  
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54 decreases from  $\sim 200$   $\mu\text{m}$  to  $\sim 100$   $\mu\text{m}$  and a lot of ramifications occurs as shown in Figure 4  
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56 due to the progressive increase of the growth rate.  
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## 4.2. Impact of mechanical effect

### 4.2.1. Mechanical effect on primary trunks

During the solidification of Al – 14 wt.% Sn, the detachment of primary trunks growing parallel to the temperature gradient direction is observed. In Al-Sn alloy, the density of the formed solid ( $\rho_S = 2585 \text{ kg/m}^3$ ) is lower than the density of the surrounding liquid ( $\rho_L = 2617 \text{ kg/m}^3$ ) so that the buoyancy force exerted on the solid is upward like in Al-20wt%Cu alloys [42] and contrary to Al-Si and Al-Ni alloys where this force is exerted downwardly [19,43]. A model was proposed by R.H. Mathiesen *et al.* [11,44] to determine the limit thickness of the primary arm neck for which the deformation of the neck is plastic and thus can lead to fragmentation by a mechanical effect. In this model, the potential fragment is assimilated to a sphere with an equivalent radius  $R_e$  and a neck radius  $r$  (Fig.10). The detachment takes place when  $R_e$  exceeds a value given by the equation 2:

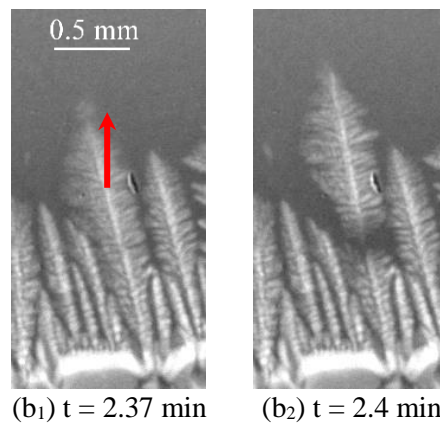
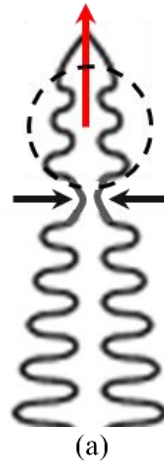
$$R_e > \left( \frac{2\sigma_B}{4\Delta\rho g} r^2 \right)^{1/3} \quad (2)$$

with  $\sigma_b$  the critical stress, and  $\Delta\rho$  the density difference between liquid and solid phases.

From equation (2), it is possible to determine the minimum value range of primary arm neck radii for which the deformation is plastic. We impose a value range of  $R_e$  from  $50 \text{ }\mu\text{m}$  to  $150 \text{ }\mu\text{m}$  (keeping in mind that the thickness of the sample is  $300 \text{ }\mu\text{m}$ ) and  $\sigma_B = 0.14 \text{ MPa}$  for aluminium [43]. According to this model, the calculated range of the neck radius is  $\sim 0.02 \text{ }\mu\text{m} < r < 0.12 \text{ }\mu\text{m}$ , which means that the neck radius must be as small as these values to obtain detachment due solely to mechanical effect.

However, in our experiments, the dendrite arm neck of the growing microstructures is much larger (micrometers to tens of micrometers). Therefore, it can be concluded that buoyancy force alone is not sufficient to provoke fragmentation of primary trunks. This result is in agreement with B. Cai *et al.* [23] work, where they proposed that buoyancy induced mechanical effect is

1 too small to cause the fragmentation without additional mechanisms. Mechanisms at the origin  
2 of the neck arm pinch-off and so of subsequent dendrite arm fragmentation will be discussed in  
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4 the following.  
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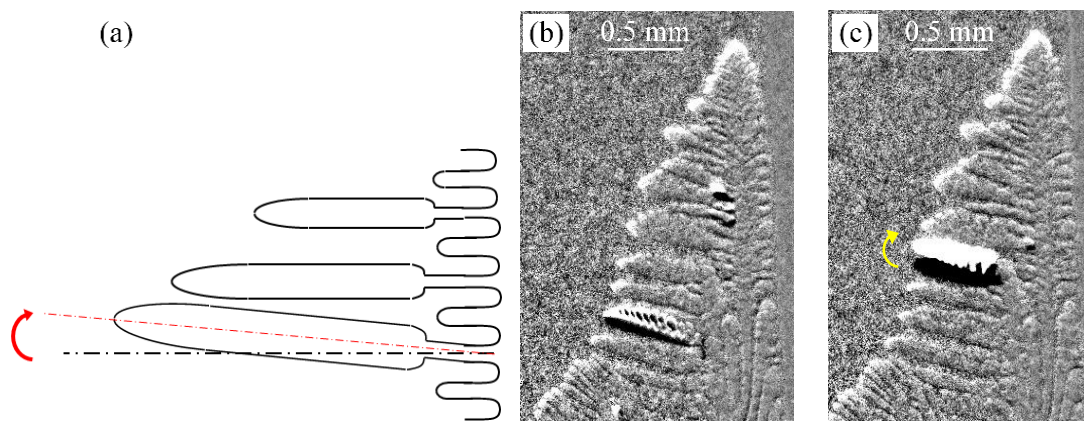


40 Figure 10: (a) Sketch illustrating the irreversible neck deformation of the primary trunk under  
41 the buoyancy force leading to the detachment of the top of the dendrite. The potential fragment  
42 is assimilated to a sphere with an equivalent radius  $R_e$  and a neck radius  $r$ . (b) Two consecutive  
43 radiographs from experiment (D) solidified at  $R_T = 16$  K/min, showing the detachment of a  
44 primary trunk.  
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#### 47 48 49 **4.2.2. Upward bending of secondary arms** 50

51 The radiographs in Figure 11b and 11c show the upward bending and detachment of two  
52 secondary arms. These pictures are obtained after an image processing named frame  
53 differencing, which consists in dividing a radiograph by a previous one. The positions of the  
54 secondary arms before bending are illustrated in black and their new positions in white. The  
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1 cause of the bending is linked again to density difference between liquid and solid phases, which  
2 is at the origin of an upward buoyancy force. Reinhart *et al.*[43] analysed the mechanical  
3 stresses that are localized at the thin solid necks that attach the secondary arms to the primary  
4 trunk. They showed that the mechanical stresses accumulate there as the arms grow.  
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7 trunk. They showed that the mechanical stresses accumulate there as the arms grow.  
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9 Subsequently, the yield stress can be exceeded at the necks, which causes their sudden and  
10 irreversible rotation as presented in Figure 11. Secondary arm detachment is observed all along  
11 the solidification experiments and sometimes follows dendrite bending. However, using the  
12 same argument as in section 4.2.1, it can be concluded that the buoyancy force is not sufficient  
13 to provoke fragmentation alone. In our previous work, detachment of secondary arms is also  
14 observed in Al – 7wt.% Si following dendrite bending [19] although due to a reverse solid and  
15 liquid ratio, the bending is observed in the downward direction.  
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42 Figure 11: (a) Sketch depicting the upward bending of a secondary arm due to buoyancy force.  
43 (b) and (c) two pictures showing the bending of two different secondary arms during experiment  
44 (B) solidified at  $R_T = 0.5$  K/min. These pictures are obtained by an image processing (frame  
45 differencing), the positions of the secondary arms before and after upward bending appear  
46 respectively in black and in white.  
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### 51 4.3. Origin of fragmentation

52 The microstructure formation in the experiments presented in this work takes place in a  
53 transient regime. Decreasing the temperature gradient progressively induces a gradual increase  
54 of the dendrite growth velocity (Fig.5), which is accompanied by a continuous increase of the  
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1 solute and latent heat rejection. The solute is transported from the cell/dendrite tips toward their  
2 roots as it is denser than the solvent. The solute accumulation around the microstructure,  
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4 together with the heat rejection, modify locally the temperature and restrict the neck growth  
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6 which leads to its size reduction and can ultimately contribute to its detachment as pointed out  
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8 by Ruvalcaba *et al.* [10]. The solute concentration was measured locally at some detachment  
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10 points by applying the image processing presented in A. Buffet *et al.* [36], where the grey scale  
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12 of the radiographs was converted to solute concentration. The fragmentation occurs for an  
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14 enrichment of 20 – 25 wt.% Sn in the beginning and then later during solidification the  
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16 concentration increases further to reach 30 wt.% Sn. As shown in Figure 6, the detached cells  
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18 already represent small radii that reduce respectively during the growth due to the solute  
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20 accumulation which leads to their detachment.  
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26 Moreover, an important feature to mention is that several fragmentation phenomena are  
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28 preceded by a sudden increase of the growth rate (Fig.5), which goes with increased local solute  
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30 and heat rejection. However, during the directional solidification of transparent alloys, Liu *et*  
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32 *al.* [45] showed that under sharp decreasing growth rate, the arm detachment is accelerated due  
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34 to the rising of interdendritic solute concentration that induces the dissolution of narrow  
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36 attachment necks of the primary stems. This observation confirms that fragmentation is  
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38 correlated to the variations during solidification of the local columnar tip growth rates, to the  
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40 curvature of the attachment necks and to the geometrical arrangement of neighbour dendrites.  
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42 So, it seems that the transient conditions applied in the present work enhance the fragmentation  
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66 Some detached fragments can also contribute to further fragmentation as can be seen in  
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68 experiments (C) and (D) solidified at  $R_T = 8$  K/min and 16 K/min, respectively (see  
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70 supplementary materials). As the fragments float, they can melt or grow and thus reject solute  
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72 that is transported by the flow toward the mushy zone and provokes the melting of other necks  
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that can induce new fragmentation events [10,11].

Moreover, as discussed in section 4.1, the dendritic microstructure evolution strongly depends on the growth velocity and this evolution can make the microstructure more prone to fragmentation. Indeed, because of the progressive increase of the growth rate, the microstructure becomes finer and shows several successive ramifications (Fig.4). The attachment necks of these new branches are already thin, which makes them easy to detach. In most cases, fragmentation occurs after several successive branching and thus in the case of finer dendrites (Fig.4). In fact, some detached primary trunks are initially tertiary branches (Fig.7). The fact that the structure becomes finer [41] during the growth process can facilitate the fragmentation. H. Neumann-Heyme *et al.* [46] showed that if the secondary arm spacing ( $\lambda_2$ ) is large, the solidification effect is predominant compared to the capillarity effect (coarsening). Oppositely, the tendency for pinch-off (detachment) is enhanced for smaller  $\lambda_2$  (finer structure).

Adding to the above effects, the mechanical effect can also contribute to the detachment processing in Al-Sn alloy when the buoyancy force is acting upwardly; either by the stretching of the arms or of parts of the primary trunks which can reduce the neck radius and facilitate the fragmentation due to the mechanisms described above.

#### **4.4. Effect of the fragmentation on the final grain structure and CET**

The final microstructure is strongly related to the growth velocity and to the fragmentation phenomenon that occur during the solidification process. In the experiments solidified at the lowest cooling rates  $R_T = 0.2$  K/min and  $R_T = 0.5$  K/min (Fig.2d, Fig.3d), the final microstructure is columnar with a segregated channel due to the convective flow with no clear traces of fragmentation when looking at the final grain structure. Without the in-situ investigation, the fragment dynamics could not be detected in detail from the final structure. No direct impact of fragmentation on the final structure is observed but it does affect the dendrite crystallographic orientation, especially when the fragments rotate while floating or due



to the small fragments that are detached inside the mushy zone during the coarsening.

The final grain structure of the sample solidified at  $R_T = 8$  K/min is composed of successive layers of elongated equiaxed grains (Fig.8d) either connected due to the coalescence and coarsening phenomena or separated by a Sn-rich segregated band. These equiaxed grains are initially fragments that are blocked above the solid-liquid interface and continue their growth whereas the ones that go in the upper part of the sample are remelted. In our experiments, the fragment floatation is hindered due to the thin samples. However, it can also occur in bulk ingots when a high amount of fragments are competing with each other and stop the growth of the columnar front [4].

Each new layer is due to fragmentations from the grains below. So, the fragmentation can produce a Columnar-to-Equiaxed Transition grain structure that is repeatedly observed during solidification even in this non-refined alloy and at a relatively low cooling rate.

As seen in the result section 3.3, CET is also observed (Fig.9a,b) at the beginning of the experiment solidified at the highest cooling rate ( $R_T = 16$  K/min). As a second step, columnar growth starting from the second layer of equiaxed grains is characterised. The growth rate of the solidification front is compared to the floating velocity of fragments to understand these transitions. From the beginning of the solidification, detachment of dendritic branches is observed, they float toward the hot zone with a velocity of  $\sim 100$   $\mu\text{m/s}$  to  $\sim 200$   $\mu\text{m/s}$  while the growth rate of the interface is increasing from  $\sim 35$   $\mu\text{m/s}$  to  $\sim 60$   $\mu\text{m/s}$ . As long as the solidification velocity is lower than the fragment floating velocity, the fragments have enough time to leave the interface and to move up due to the buoyancy force. As a first step and similarly to experiment (C), a layer of elongated equiaxed grains issued from fragmentation develops above the columnar front so that a Columnar-to-Equiaxed Transition is observed. A second layer is formed by the same mechanism. However, as soon as the growth velocity reaches the floating fragment velocity, the fragments are trapped because the buoyancy force is

1 not sufficient to bring them out from the solidification interface. As a result, a columnar front  
2 dominates and an Equiaxed-to-Columnar Transition is observed (Fig.9d). Thus, there is an  
3 optimum of solidification parameters to obtain and maintain CET by fragmentation.  
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## 9 **5. Conclusion**

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12 Directional solidification of Al – 14 wt.% Sn alloy was investigated by means of  
13 synchrotron X-ray radiography imaging techniques. In order to be close to industrial conditions  
14 such as direct-chill casting [27], solidification was triggered by simultaneously decreasing the  
15 temperature gradient, and increasing the solidification rate. A strong correlation between the  
16 growth velocity and the fragmentation phenomenon was demonstrated under transient regime.  
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24 The results show that the number of detached secondary, tertiary arms and even primary  
25 trunk sections raises gradually by increasing the growth velocity while reducing the temperature  
26 gradient. This phenomenon takes place until the growth velocity reaches the fragment floatation  
27 velocity so that the fragments cannot anymore escape from the mushy zone.  
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34 The mechanical effect induced by the buoyancy force is the cause of the irreversible  
35 secondary arms bending and of the deformation of the primary arms size, which can contribute  
36 to the fragmentation. However, we show that it cannot be the only mechanism at stake. In fact,  
37 increasing the growth velocity is at the origin of the increase of solute and latent heat rejection  
38 and of finer and ramified dendrites which make the microstructure more prone to the  
39 fragmentation phenomenon by remelting and capillarity effect.  
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49 It is also shown that the fragmentation can have a major impact on the final grain structure.  
50 It can be at the origin of CET and of an equiaxed grain structure in a non-refined alloy solidified  
51 at relative low growth rate for optimum solidification conditions and can create local  
52 disorientations within the final grain structure. The CET can take place and the equiaxed regime  
53 can be maintained by the fragmentation phenomenon only if the fragment floatation velocity is  
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higher than the front growth velocity.

## 6. Supplementary data

Supplementary data related to this article can be found at (link to supplementary material).

## 7. Acknowledgments

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