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1 ***In situ* investigation of the Columnar-to-Equiaxed Transition during directional**
2 **solidification of Al–20wt.%Cu alloys on Earth and in microgravity**

3
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14
15 **Abstract**

16 Solidification experiments on Al–20wt.%Cu alloys with grain refiner were performed in a
17 Bridgman-type furnace to investigate the impact of gravity-driven phenomena on the
18 Columnar-to-Equiaxed Transition (CET) during directional solidification (*i*) in microgravity,
19 on board the sounding rocket MASER-14 and (*ii*) on Earth in three growth directions, namely
20 horizontal, vertical upward (counter-gravity direction) and vertical downward (in-gravity

1 direction). The CET was provoked by a step increase of the cooling rate and was visualised *in*
2 *situ* and in real-time using X-radiography. This paper reports direct observations of dendritic
3 columnar growth, CET and the subsequent equiaxed regime, and quantitative characterisation
4 of the microstructures. The increase in constitutional undercooling that induces the nucleation
5 of the first grains ahead of the columnar front can be related to the rapid thermal change
6 following the increase in cooling rate. The nucleation distance of equiaxed grains ahead of the
7 columnar front is larger in microgravity than on Earth and possible origins for this difference
8 are discussed. Both mechanical and solutal blocking mechanisms were observed at CET.
9 Mechanical blocking is attributed to the occurrence of [solidification-induced shrinkage flow](#).
10 The relative importance of each blocking mechanism is detailed for each growth configuration.
11 In particular, mechanical blocking is shown to be the principal type of impingement in the
12 microgravity experiment. The final equiaxed grain structure was characterised, and the results
13 confirm that gravity effects become less significant at high growth rates.

14

15 **Keywords:** Microgravity, directional solidification; Columnar-to-Equiaxed Transition, grain
16 characteristics, *in situ* X-radiography; Al-Cu alloy, gravity, convection

17

18 **1. Introduction**

19 The properties of metallic alloys are directly related to their solidification microstructures
20 so a detailed understanding of nucleation and growth processes is of tremendous importance in

1 engineering [1]. During industrial processes such as ingot casting, welding or additive
2 manufacturing, the formation of two types of microstructure is usually observed depending on
3 the solidification parameters (temperature gradient, cooling rate, alloy composition): the
4 columnar structure giving rise to directional properties, and the equiaxed structure with more
5 uniform and isotropic properties [2]. The columnar grain structure is required for instance for
6 aerospace turbine blades, while equiaxed grains are desired for automotive applications such as
7 car engines or suspensions. Equiaxed growth can be promoted by adding small particles known
8 as inoculants, which act as seeds to promote heterogeneous nucleation. During industrial
9 processes, a transition between those two types of grain structure is often observed: it is the
10 Columnar-to-Equiaxed Transition (CET). It is usually induced by an increase of the
11 solidification velocity or a decrease of the temperature gradient during the solidification process
12 [2]. This transition occurs when the columnar front is blocked by equiaxed grains, either by
13 hard blocking when equiaxed grains become in direct contact with the columnar front, or by
14 soft blocking when solutal or thermal interactions between equiaxed grains and the columnar
15 front cause the progressive deceleration of the columnar front and eventually its halt [3]. CET
16 can be sharp, leading to two distinct parts in the material with dramatically different properties,
17 or progressive, with the co-existence of both columnar and equiaxed structures [4–6]. It is worth
18 noting that the relative importance of each impingement mechanism for CET (hard vs. soft) is
19 still unclear, especially due to the lack of reference experiments using *in situ* observations to
20 investigate this topic.

1 On Earth, gravity is the cause of many phenomena that make the understanding of the
2 solidification process, and specifically the CET, difficult. Firstly, natural convection occurring
3 in the melt significantly affects the mechanism of heat and mass transport [7] and can thus
4 modify the position of the CET [6]. Another effect of gravity is buoyancy that gives rise to the
5 sedimentation or the flotation of solid grains [8,9], which can highly participate to or influence
6 the CET. Also, buoyancy leads to mechanical effects such as dendrites bending [10], which can
7 help to promote fragmentation and cause CET if the dendrite fragments are transported ahead
8 of the solidification front by fluid flow. Recently, variation of hydrostatic pressure by changing
9 the gravity level has also been highlighted as an effect that can modify the microstructure [11]
10 and cause CET [12]. To reduce such effects on the microstructure formation, thin alloy samples
11 and horizontal solidification can be used [13,14]. Nevertheless, gravity effects still exist even
12 in the latter configuration. Therefore, microgravity experimentation is the method of choice
13 since gravity effects are almost eliminated in such an environment and solidification
14 experiments can be carried out in nearly diffusive regime [15]. Benchmark data can thus be
15 obtained for the improvement of numerical studies [16] and comparative study between
16 experiments on Earth and in microgravity can be achieved to enlighten gravity effects [17,18].

17 Over the last couple of decades, X-radiography has become a relevant technique to
18 investigate microstructure formation dynamics in metal alloy solidification [19], and especially
19 gravity-induced effects [20–22]. For such studies, synchrotron sources are mainly used because
20 they provide a continuous range of photon energies and an intense X-ray beam [23]. However,

1 the synchrotron X-ray beam is horizontal, which constrains solidification experiments to a
2 vertical configuration (upward and downward growth), where gravity effects are expected to
3 be strong. Recent developments made compact microfocus X-ray sources, X-ray sensitive
4 cameras and optics more efficient, allowing home laboratory alternatives with substantial
5 brilliance to be used [13,24,25]. Solidification experiments could then be performed on ground,
6 in various directions (vertical, horizontal) [22,25–27], and also in microgravity either in
7 sounding rockets or on board of parabolic flights [12–14,28].

8 In the framework of the European Space Agency (ESA) project entitled XRMON (*In situ*
9 X-ray monitoring of advanced metallurgical processes under micro gravity and terrestrial
10 conditions), an *in situ* investigation of the columnar solidification in a non-refined Al–
11 20wt.%Cu alloy was conducted for the first time in microgravity, on board the MASER-12
12 sounding rocket [13]. This study provided direct comparison of terrestrial and microgravity
13 alloy solidification. In the subsequent stage of the project, the first *in situ* study in microgravity
14 of equiaxed solidification in isothermal conditions was performed in the MASER-13 sounding
15 rocket [29] with a refined Al–20wt.%Cu alloy [14]. In the continuation of those investigations,
16 the present work presents the first microgravity experiment dedicated to the *in situ* observation
17 of the Columnar-to-Equiaxed Transition in a refined Al–20wt.%Cu alloy. The experiment was
18 conducted in the XRMON-GF2 (Gradient Furnace) facility on board of the MASER-14
19 sounding rocket. To complement this microgravity experiment, three ground reference
20 experiments in horizontal, vertical upward and vertical downward configurations were

1 performed with nearly the same control parameters. A comparative analysis has been conducted
2 between the microgravity experiment in diffusive conditions and the ground-based experiments
3 to enlighten gravity effects during the different steps of the CET. Details of the experimental
4 configuration and the experiment timeline will be presented in the first part of this document.
5 In the second part, the columnar growth, CET and equiaxed regime will be depicted. In the last
6 part, discussions on the equiaxed grain nucleation step prior to CET, the blocking mechanisms
7 responsible for CET and the final equiaxed grain structures will be presented.

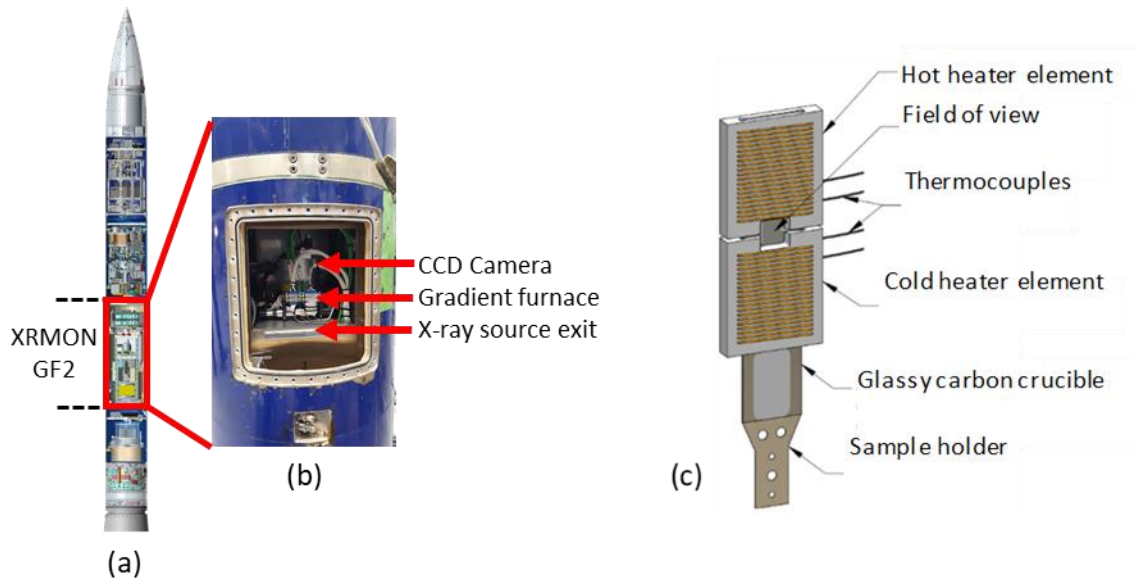
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9 **2. Experiment description**

10 **2.1. MASER-14 sounding rocket**

11 The MASER-14 (MAterials Science Experiment Rocket) sounding rocket is a sub-orbital
12 vehicle developed by the Swedish Space Corporation (SSC) to perform experiments during
13 short-duration microgravity flights. It was launched on June 25th, 2019 at the Esrange Space
14 Centre in northern Sweden. The 400 kg scientific payload was composed of 4 experimental
15 modules including the XRMON-GF2 experiment ([Fig. 1a](#)). The rocket followed a parabolic
16 trajectory from launch to landing with an apogee of 245 km, providing approximately 6 minutes
17 of microgravity.

18



1
 2 *Fig.1: (a) Scheme of the payload of the rocket with the XRMON-GF2 module located near the*
 3 *bottom of the payload, (b) photograph showing the XRMON-GF2 facility installed inside the*
 4 *module and (c) schematic layout of the XRMON-GF2 furnace (courtesy of A.G. Murphy,*
 5 *UCD)*

6
 7 **2.2. XRMON-GF2 facility**

8 The XRMON-GF2 facility is a duplicate of the XRMON-GF (Gradient Furnace) facility
 9 developed for the MASER-12 sounding rocket experiment [13] in the framework of ESA
 10 XRMON project dedicated to the application of X-radiography during microgravity
 11 experiments. It is also similar to XRMON-PFF (Parabolic Flight Facility), which was used
 12 during parabolic flights experiments [12] and the SFINX (Solidification Furnace with IN Situ
 13 X-radiography) laboratory device [29]. A complete description of those facilities has been
 14 presented in these papers. Only a short summary of the main features will be given below.

15 The XRMON-GF2 facility consists mainly of a Bridgman-type gradient furnace and an X-
 16 radiography system. The solidification furnace consists of two heaters separated by an adiabatic

1 gap. The adiabatic gap separating the heating elements contains a 5 mm × 5 mm window in its
2 centre that constitutes the Field of View (FoV) through which the X-rays pass, thus allowing an
3 *in situ* and real-time analysis of the solidification experiments. The two heaters impose a
4 longitudinal temperature gradient G_{app} controlled by two K-type thermocouples spaced 15 mm
5 apart: one placed in the hot zone 5 mm from the top of the field of view, the other one in the
6 cold zone 5 mm from the bottom of field of view. The gradient furnace enables directional
7 solidification with applied temperature gradient within the range of 5–15 °C/mm and controlled
8 cooling rates within the range of 0.01–1.5 °C/s.

9 The X-radiography system consists of a micro-focus X-ray source with a molybdenum
10 target (3 μm focal spot) delivering a photon flux with two peaks of energy 17.4 keV and 19.6
11 keV, ensuring a good contrast to study Al–Cu based alloys. The detector system comprises a
12 scintillator plate and a CCD digital camera with a pixel size of 20 μm. The microfocus source
13 delivers a conical beam leading to a magnification of the image depending on the respective
14 distance between the source, the sample, and the camera. For the current geometry, the
15 magnification is about 5 and the effective pixel size at the level of the sample is 4 μm.

16 The four foil-like samples investigated were Al–20wt.%Cu alloys with grain refiner (0.1
17 wt% Al–Ti–B) that were 50 mm in length, 5 mm in width, ~210 μm in thickness and 3 μm
18 finish-polished. The thickness value is a good compromise between having a good radiographic
19 contrast, reducing convection effects, and avoiding the superimposing of several layers of
20 dendrites, which would be detrimental to the picture legibility. The samples were sandwiched

1 between two flexible glassy carbon sheets sewn together with a silica thread, whilst held in
2 place in the middle of stainless-steel spacers. The holder/crucible/sample was then slid into the
3 furnace and straddled both heater elements to achieve the expected thermal profile.

4 Al–20wt.%Cu alloy with grain refiner was chosen to be consistent with the MASER-12
5 experiment where the same alloy, in non-refined form, was used to study columnar
6 solidification, and the MASER-13 experiment where the same alloy, also grain-refined, was
7 used to study equiaxed solidification. The contrast in radiography between the alpha-Al grains
8 and the Cu rich-solute is high for this alloy composition, ensuring a better visualisation of the
9 microstructure. Moreover, solid aluminium is lighter than liquid Al–20wt.%Cu, which enhances
10 buoyancy effects during vertical solidification experiments by pushing the aluminium grains
11 upward. Also, for downward solidification experiments, both thermal and solutal gradients are
12 destabilizing and strong convective flows are expected.

13

14 **2.3. Microgravity experiment parameters**

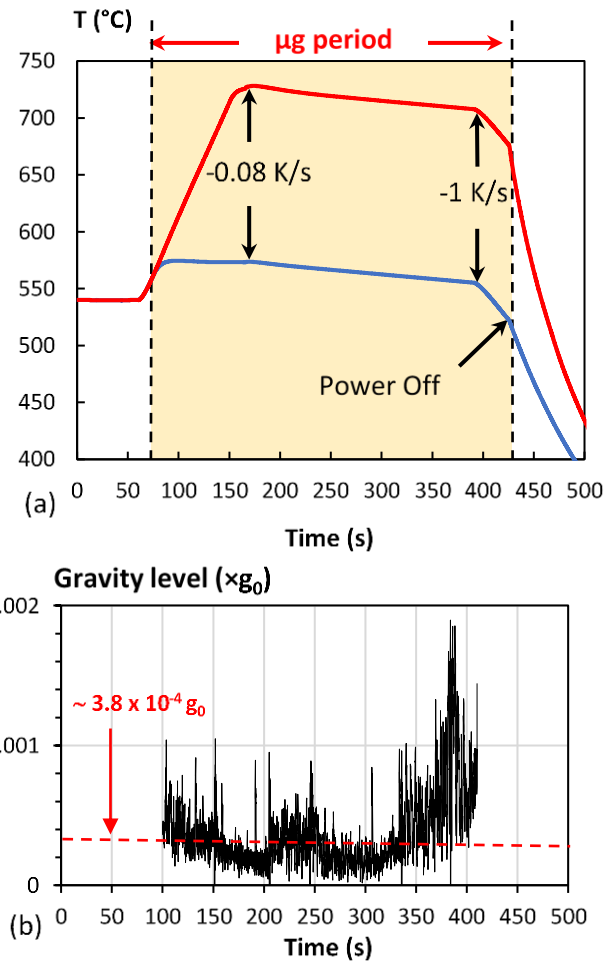
15 A well-adapted experimental timeline has been defined based on ground-based tests to
16 ensure that both the melting and the cooling stages would be achieved within the short
17 microgravity period of roughly six minutes. The chosen timeline is shown in Fig. 2 where $t_0 =$
18 0 s corresponds to the sounding rocket lift-off time. The sample was pre-heated before lift-off
19 at a rate of 2 °C/s from 20 °C to 540 °C, which is below the eutectic temperature $T_E = 548.2$ °C
20 to keep the sample fully solid. The sample melting phase started 14 s before the beginning of

1 the microgravity period by increasing the temperature of both heaters at a heating rate of 2 °C/s
2 until reaching the target temperatures. The melting phase ended at $t = 150$ s, when the “cold”
3 heater reached 572 °C and the “hot” heater reached 722 °C, which is above the alloy liquidus
4 temperature $T_L = 605$ °C and corresponds to an applied temperature gradient $G_{app} = 10$ °C/mm.
5 At the end of the melting phase, the sample was partially melted, and some solid phase remained
6 in the relatively cold part of the field of view, constituting the initial solid-liquid interface.

7 The solidification was triggered at $t_{RI} = 165$ s by applying the first cooling rate $R_1 =$
8 0.08 °C/s to both heaters. This value was chosen from ground-based tests to obtain a columnar
9 solidification front. A second cooling rate $R_2 = 1$ °C/s was applied on both heaters at $t_{R2} = 390$ s
10 to provoke the columnar-to-equiaxed transition. Finally, the two heaters were switched off at
11 the end of the microgravity period and the sample cooled down naturally.

12 During the whole experiment, uncompressed images were recorded and stored on board at
13 a frame rate of 3 images per second, which was sufficient to follow the microstructure
14 development. Image processing was enabled on board to improve the radiograph quality by a
15 flat-field correction. In this method, the solidification images were divided by a reference image
16 recorded at the end of the melting phase [30].

17



1
2 *Fig. 2. (a) MASER-14 flight timeline (Time $t_0=0$ s corresponds to the lift-off time) and (b)*
3 *gravity level with time during the μg period (the red dashed line corresponds to the average*
4 *gravity level value)*

7 **2.4. Ground-based experiment parameters**

8 For a sake of comparison, three experiments were performed on Earth using the same
9 XRMON-GF2 furnace, the same cooling rates R_1 and R_2 and applied temperature gradient G_{app}
10 but with different solidification configurations (horizontal, upward, and downward
11 solidifications). A fresh refined Al-20wt.%Cu sample was used for each experiment.

- 1 • Horizontal solidification, with the main surface of the sample set perpendicular to the gravity
2 vector. In this configuration, gravity effects are expected to be minimized significantly [31].
- 3 • Vertical upward solidification (counter-gravity direction). In this configuration, both thermal
4 and solutal gradient are stabilising but buoyancy will induce the flotation of unconstrained
5 solid grains [21].
- 6 • Vertical downward solidification (in-gravity direction). In this configuration, both thermal
7 and solutal gradients are destabilising and strong convective flow is expected [22].

8 It is worth noting that for both vertical solidification experiments, grain flotation and
9 convective flow dramatically affected the melting of the sample. Consequently, the timelines
10 of melting phase of both vertical experiments were slightly adapted to better control the initial
11 position of the solid-liquid interface. More precisely, the temperature gradient G_{app} was applied
12 after the two heaters reached a temperature of 300 °C and a slower heating rate of 0.5 °C/s was
13 imposed to the heating elements during the melting phase.

14

15 **2.5. Grain structure characterisation**

16 The solidification process was observed *in situ* by using X-radiography. The solidification
17 sequence consisted of the growth of a columnar front followed by an equiaxed front propagating
18 from the cold zone towards the hot zone of the FoV. A quantitative analysis was made to
19 determine the grain size and grain elongation distribution of the equiaxed microstructure. The
20 procedure was described in detail in [22,29] and only the main steps are summarised hereafter.

1 Based on the radiographs recorded during the solidification process, the contouring of the
2 grains was performed manually using the free software ImageJ [32]. Then, using a semi-
3 automatic macro script, it was possible to determine two characteristic grain features: the grain
4 size, defined as the diameter d of a disk having the same surface area, also called the equivalent
5 disk. This parameter gives no information about the grain morphology and was complemented
6 by the grain elongation factor ϕ , defined as $\phi = L_1/L_2$ with L_1 the length of the longest straight
7 segment inscribed within the grain (main axis), and L_2 the length of the longest straight segment
8 inscribed within the grain and orthogonal to the segment that gives L_1 . This factor varies from
9 unity for a fully equiaxed grain to larger values for elongated grains.

10

11 **3. Experimental observations**

12 **3.1. Microstructure formation during columnar growth**

13 The objective of the present study was to experimentally investigate the Columnar-to-
14 Equiaxed Transition in a refined Al–20wt.%Cu alloy in microgravity conditions, and on ground
15 with different orientations of the sample with respect to the gravity vector. A series of
16 radiographs recorded during the solidification of the samples is shown in Fig.3 for these four
17 experiments: solidification in microgravity, horizontal solidification, vertical upward
18 solidification, and vertical downward solidification.

19 Four radiographs were selected for each configuration to show the formation of the
20 microstructure in the FoV. Complete solidification sequences are available online

1 (supplementary movies 1, 2, 3 and 4). As mentioned in section 2.3, the samples were first
2 partially melted. An initial solid-liquid interface was required to start the columnar
3 solidification from already present solid parts and thus prevent the occurrence of a high
4 undercooling in the liquid that would have triggered the nucleation of many grains on the
5 refining particles. The columnar growth started with the application of the low cooling rate R_l
6 = 0.08 °C/s and dendrites progressively developed from the solid parts of the initial solid-liquid
7 interface (Fig.3a and Fig.3b). A dark layer is visible ahead of the solidification front due to the
8 rejection of Cu during solidification as previously reported in synchrotron experiments [30],
9 and during the MASER-12 sounding rocket campaign [13]. It is worth mentioning that Cu-
10 rejection during the downward solidification experiment led to the development of solute
11 plumes, corresponding to a downward flow of Cu-rich liquid (Fig.3b3 and supplementary
12 movie 4) because the rejected solute (Cu) is denser than the solvent (Al). The solute plumes
13 impacted the growth of some columnar dendrites by locally increasing the Cu concentration
14 ahead of the dendrite tip and thus retarding their growth. Similar observations have been
15 reported in detail in previous studies [22,26,33,34].

16 Vertical lines and white areas are visible on the right-hand and left-hand sides of the
17 microgravity and horizontal samples. These features are attributed to sample deformation due
18 to solidification-induced shrinkage flow as well as bubble formation and collapse, which
19 induces a local change of the thickness and therefore of the X-ray transmission. The fact that
20 the changes in thickness appear with a high contrast is due to the contrast enhancement of the

1 images that was applied to better distinguish the solid and the liquid phases. Post-mortem
2 measurements showed that the sample was about 10 μm thicker in the centre after solidification.
3 It is assumed that these deformations did not significantly disturb the propagation of the
4 solidification front. Such deformations were not observed in the two vertical configurations
5 because the hydrostatic pressure of the liquid on the crucible wall maintained the sample shape
6 after the melting step. The use of a more rigid crucible might help reduce sample deformation
7 in future experiments.

8 In addition to the development of columnar dendrites, some elongated grains are also
9 visible (white circles in Fig.3a and Fig.3b). The observations show that these grains nucleated
10 ahead of the columnar front for the microgravity, horizontal and downward solidification
11 experiments, probably on refiners, impurities, or heterogeneities of the oxide layer. Those grains
12 slightly moved towards the cold part of the sample during their development. In the case of the
13 microgravity and horizontal experiments, this motion can be attributed to advective flow
14 induced by solidification shrinkage as already reported for experiments carried out in similar
15 configurations [21,22,31]. In the case of the downward solidification experiment, the grain
16 motion is due to buoyancy since the Al-rich solid parts are less dense than the surrounding
17 liquid. The origin of the grains was different for the upward solidification experiment. Close
18 inspection of the radiograph sequence revealed that no grain nucleation occurred, but copious
19 columnar dendrite fragmentation happened. The fragments floated due to buoyancy and moved
20 toward the hot part of the sample where they gradually melted. The grains visible in Fig. 3a2

1 and Fig. 3b2 (dashed white circles) are in fact fragments that remained stuck in the sample
2 thickness while floating. Such differences between the various experiments could not have been
3 deduced from post-mortem analyses, which highlight the interest of *in situ* observations [35].
4 It is worth mentioning that all fragments would float and melt for experiments carried out in
5 bulk samples, leading to a different final grain structure. This might not always be the case as
6 shown by Zimmermann *et al.* [35] on non-refined Al-10wt.%Cu alloys for which the solid and
7 liquid density are close. In the latter study, experiments in 3D bulk samples and in 2D sheet-
8 like samples produced the same results with respect to the fragmentation process and the
9 resulting grain structure.

10 As previously mentioned, the formation of gas bubbles due to the presence of residual
11 hydrogen gas was noticed. Bubbles that nucleated in the liquid melt grew and eventually
12 collapsed, leaving circular-shape deformations. Bubbles that nucleated in the mushy zone
13 caused small displacement of the dendrites while developing and left spherical liquid areas after
14 collapsing. Similar behaviors have been recently reported and studied in details for instance by
15 Murphy *et al.* [36] and Werner *et al.* [37]. In vertical configuration, the bubbles floated due to
16 buoyancy and/or thermal Marangoni effect [38]. In microgravity conditions, the bubbles moved
17 toward the hot part of the sample only due to thermal Marangoni convection because the applied
18 temperature gradient was sufficient to cause a significant variation of the surface tension at the
19 gas-liquid interface. Surface flow occurred in the direction of increasing tension, causing the
20 gas bubbles to move in the direction of increasing temperature. A similar behavior was not

1 observed in the horizontal configuration although the same temperature gradient was applied,
2 because the bubbles were stuck on the upper wall of the sample thickness due to buoyancy.
3 Here, Marangoni convection was not strong enough to induce bubble motion. For the upward
4 and downward experiments, only few bubbles occurred during the solidification step. This is
5 likely due to the longer time that was taken to position the solid-liquid interface. Bubbles
6 formed and collapsed during this adjustment period and most of the gas was released when the
7 solidification experiments started. A thorough investigation of the bubble behaviour is out of
8 the scope of the present study and is expected to be the subject of future studies. Besides, the
9 impact of gas bubbles on columnar growth and CET was poor for the present experiments.

10

11 **3.2. CET and equiaxed front propagation**

12 After the formation of columnar structures, the cooling rate was sharply increased from R_1
13 $= -0.08$ °C/s to $R_2 = -1$ °C/s to trigger a Columnar-to-Equiaxed Transition (CET). The CET
14 occurred in three steps. Firstly, the nucleation of many equiaxed grains occurred ahead of the
15 columnar front due to an increase of the constitutional undercooling in front of the dendrite tips.
16 Refiners present in the alloy were activated and a great number of nucleation events occurred,
17 in accordance with the free growth model proposed by Greer et al. [39]. The new grains
18 appeared as a layer of small equiaxed dendrites ahead of the columnar front (Fig. 3c). The
19 images in Fig 3.c and 3.d were divided by the last image recorded before application of the
20 second cooling rate to remove contrasts due to sample deformation, which was helpful to

1 distinguish more clearly the new grains and reveal the amount of solidified columnar dendrites
2 after initiating the second cooling rate. The first layer of grains appeared in approximately 6 to
3 7 seconds after application of the second cooling rate for the microgravity experiment, slightly
4 earlier than in the upward (8 s) and horizontal (9 s) configurations. Almost twice the time was
5 necessary to form the first layer of grains for the downward configuration (11 s to 12 s). In
6 addition, the new grains seemed to nucleate slightly farther from the columnar front in the case
7 of the microgravity experiment. This observation will be discussed in more detail with the help
8 of quantitative measurements in [section 4.1](#).

9 Secondly, the growth of the columnar grains was blocked by the development of the new
10 equiaxed grains. [The number of new grains was sufficient in each experimental configuration](#)
11 [to block the growth of the columnar dendrites and induce a sharp CET](#). Two blocking
12 mechanisms were observed: mechanical (hard) blocking when the equiaxed grains and
13 columnar dendrites came into direct contact, and solutal (soft) blocking when the accumulation
14 of solute in the interdendritic region led to the arrest of the columnar growth. The relative
15 importance of those two mechanisms on the blocking of the columnar front will also be
16 discussed in more detail in [section 4.2](#).

17 Finally, the growth of the columnar front is replaced by the propagation of an equiaxed
18 [effective front](#). [The newly nucleated grains develop in the temperature gradient direction and a](#)
19 [constitutionally undercooled liquid zone progressively forms ahead of the dendrite tips due to](#)
20 [solute rejection](#). Under these circumstances, a new layer of grains can only nucleate once the

1 liquid undercooling reaches the nucleation undercooling of the refining particles. Thus, the
2 nucleation phases are separated by no-nucleation periods during which grain growth occurs and
3 the equiaxed effective front propagates in a wave-like type as reported by many authors [40–
4 43]. The last images recorded during the experiments are shown in Fig.3d. They show that both
5 the equiaxed effective front and the grain structure are similar for all the solidification
6 configurations. This suggests that gravity did not impact much the solidification process at this
7 cooling rate ($R_2 = -1$ °C/s). This point will be investigated in more detail in section 4.3.

8

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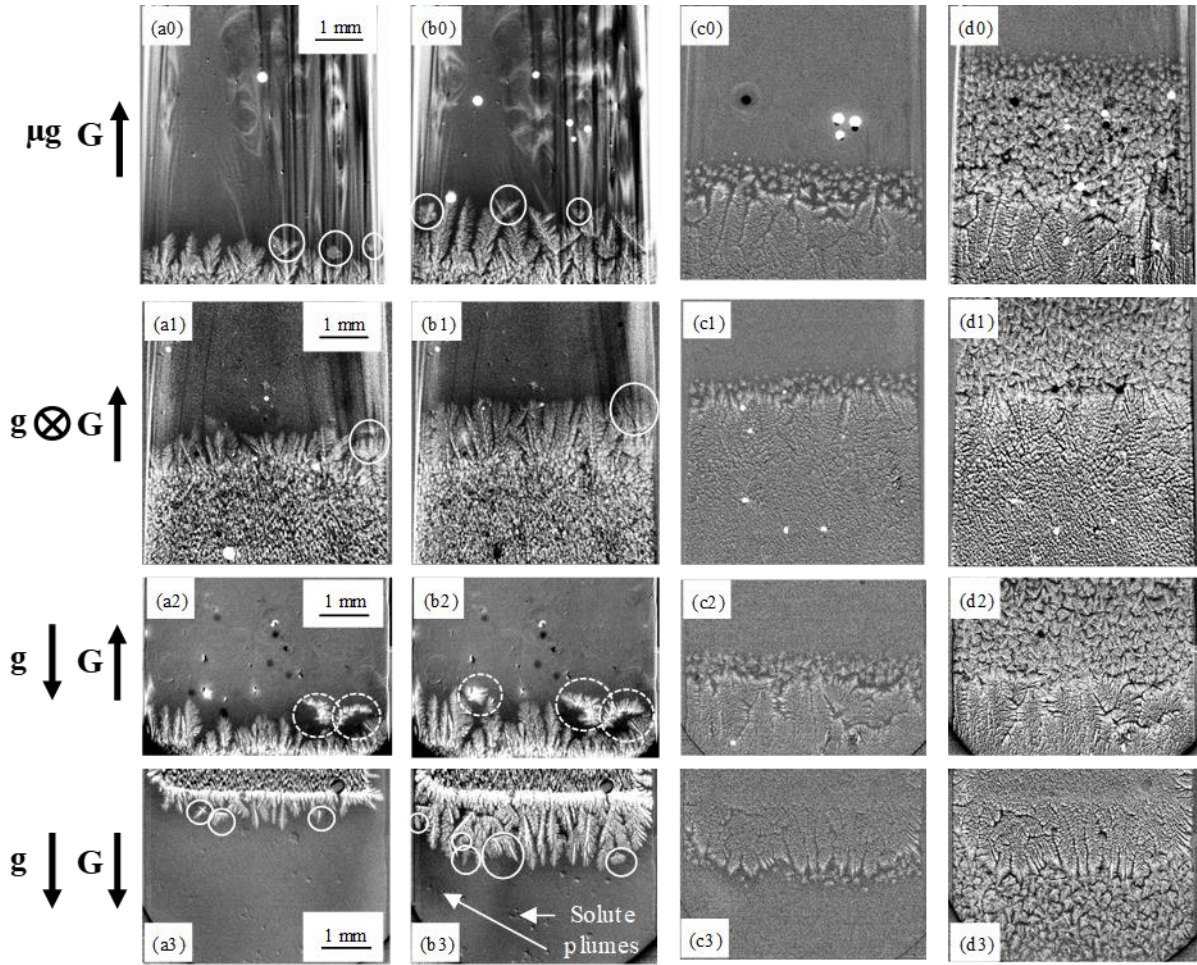
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1

2 *Fig.3: Radiographs of the whole solidification sequence. From top to bottom: microgravity*
 3 *experiment, horizontal configuration, upward configuration, and downward configuration. (a)*
 4 *Beginning of the columnar growth for the cooling rate $R_1 = 0.08 \text{ }^\circ\text{C/s}$, (b) solidification front*
 5 *before application of the second cooling rate $R_2 = 1 \text{ }^\circ\text{C/s}$, (c) nucleation of new grains*
 6 *(images were taken 11 s after application of R_2) and (d) final grain structure. Images in (c)*
 7 *and (d) were divided by the last image recorded before applying the cooling rate R_2 to better*
 8 *visualise the newly nucleated grains.*

9

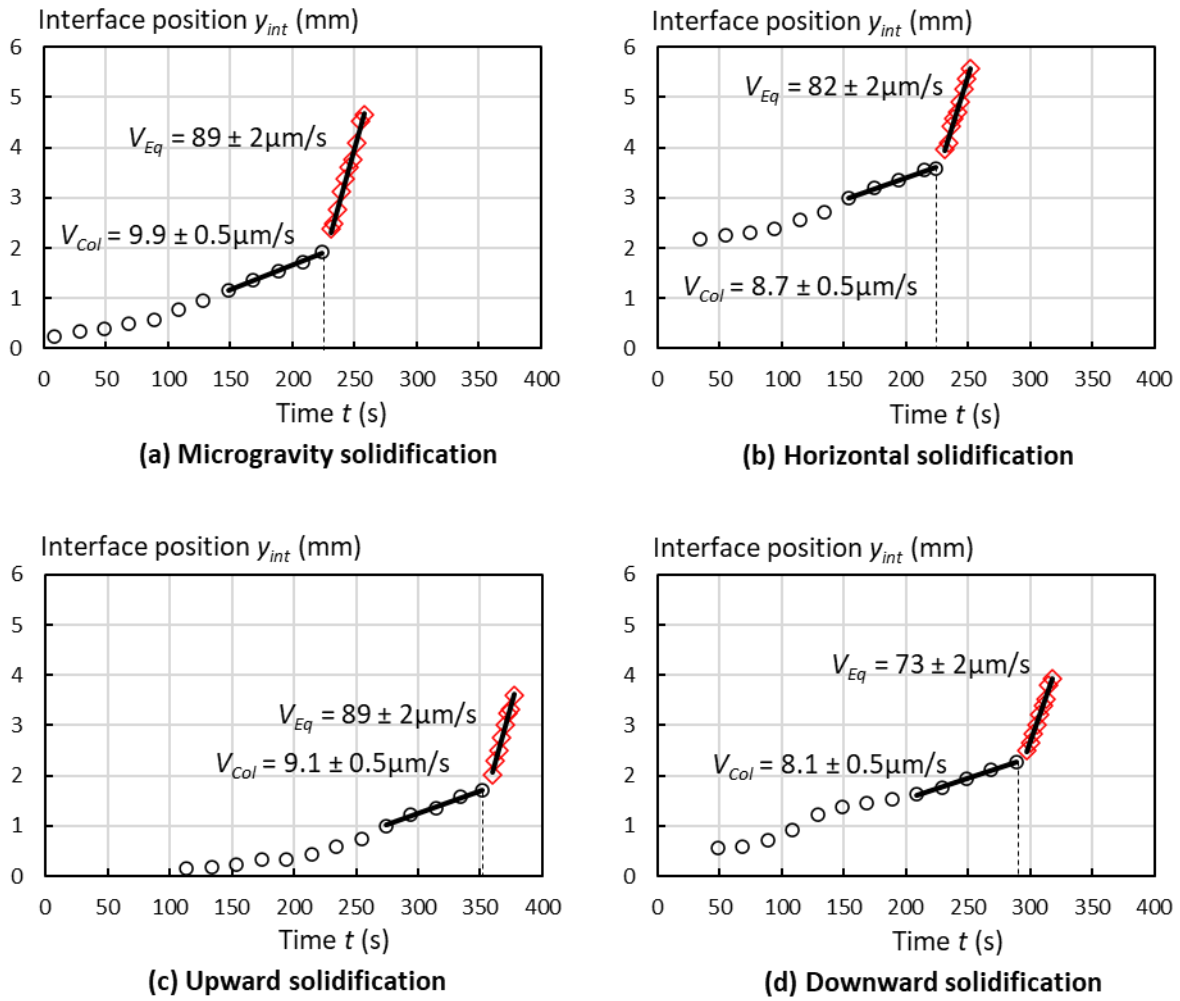
10 3.3. Growth rate measurements

11 The time evolution of the solidification interface position in the field of view y_{int} can be
 12 determined based on the radiographs. The results of these measurements are shown in **Fig. 4**
 13 for the four experimental configurations. During the columnar regime, the measured positions

1 correspond to the highest dendrite tip of the columnar grain network (black circles in Fig. 4).
2 During the equiaxed regime, the measured positions correspond to the highest dendrite tip of
3 the equiaxed grain network forming the effective equiaxed front (red diamonds in Fig. 4).

4 After application of the first cooling rate ($R_1 = -0.08 \text{ }^\circ\text{C/s}$) the columnar interface moves
5 slowly toward the hot end of the field of view then gradually accelerates to eventually reach a
6 quasi-linear regime. The corresponding growth rate V_{Col} was deduced by fitting the data to a
7 linear law (black lines in Fig. 4). The effective equiaxed front growth rate V_{Eq} was also
8 measured by using the same procedure after initiating the second cooling rate ($R_2 = -1 \text{ }^\circ\text{C/s}$).
9 The measured velocities are summarised in Table 1. The systematic error on the velocity values
10 was deduced from the error on the position measurement ($\pm 4 \text{ } \mu\text{m}$) that depends on the pixel
11 size. For the columnar regime, growth velocities are in the same order of magnitude within the
12 error bars, which indicates that the four configurations are comparable. For the equiaxed regime,
13 the growth velocities are also in the same order of magnitude except in the case of the downward
14 configuration where the growth velocity is slower. This is likely because the whole
15 experimental system could not be rotated upside-down to obtain a temperature gradient parallel
16 to gravity. Instead, the system remained with the same orientation and the temperature values
17 of the heaters were swapped (the “hot” heating element became the “cold” heating element, and
18 the “cold” heating element became the “hot” heating element). Then, because of the asymmetry
19 of the sample holder design as visible in Fig.1c, a different heat extraction and thermal field can
20 have induced slower growth.

1 The actual temperature gradient in the sample could be estimated by using the relation $G =$
2 $R.V$, with V the velocity of the solidification front. The temperature gradient values are
3 summarised in Table 1. It is worth noting that the measurements show that the actual
4 temperature gradient inside the sample was slightly different from the applied temperature
5 gradient $G_{app} = 10$ °C/mm. The discrepancy can be attributed to the difference in thermal
6 diffusivity between the sample, the crucible, and the furnace. Further numerical simulations of
7 the experimental device need to be considered to get a better understanding of the temperature
8 field evolution and will be the subject of future investigations.



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Fig. 4: Measurement of the solidification interface position y_{int} in the field of view as a function of time (columnar regime in black circles, equiaxed regime in red diamonds, color available online): (a) microgravity, (b) horizontal, (c) upward and (d) downward experiment. The reference time ($t = 0$ s) is taken at the application of the first cooling rate. The vertical dashed line in each plot indicates the time of the cooling rate increase from $R_1 = -0.08$ °C/s to $R_2 = -1$ °C/s. The columnar (resp. equiaxed) interface growth rate V_{Col} (resp. V_{Eq}) was deduced by fitting the data to a linear law (thick black lines).

	Microgravity	Horizontal	Upward	Downward
V_{Col} ($\mu\text{m/s}$)	9.9 ± 0.5	8.7 ± 0.5	9.1 ± 0.5	8.1 ± 0.5
G_{Col} ($^{\circ}\text{C/mm}$)	8.1 ± 0.4	9.2 ± 0.5	8.8 ± 0.5	9.8 ± 0.6

V_{Eq} ($\mu\text{m/s}$)	89 ± 2	82 ± 2	89 ± 2	73 ± 2
G_{Eq} ($^{\circ}\text{C/mm}$)	11.2 ± 0.2	12.2 ± 0.3	11.2 ± 0.2	13.7 ± 0.4

1 *Table 1: Measured solid-liquid interface velocities and estimated temperature gradients for*
2 *the four experiments.*

4. Discussion

5 The comparison between ground and microgravity experiments is an effective approach to
6 reveal the impact of convection and buoyancy on solidification microstructure formation as
7 demonstrated for instance by Dupouy *et al.* [44], Nguyen-thi *et al.* [13] or Murphy *et al.* [31].
8 It is also an essential step to obtain benchmark data for comparisons with theoretical models
9 and numerical simulations. The impact of gravity on the CET for these different configurations
10 will be discussed in the following parts.

4.1. Nucleation step

12 As mentioned in [section 3.2](#), the first layer of equiaxed grains appeared later in the downward
13 case upon initiating the second cooling rate ([Fig. 3c](#)). This indicates that a longer time was
14 necessary to increase the constitutional undercooling ahead of the solidification front and
15 induce grain nucleation. To investigate the origin of this feature, the temperature data were
16 checked, which showed that this difference was not due to a heater regulation issue. A different
17 composition profile ahead of the columnar front can be a possible origin for the different
18 undercooling distribution in the liquid. In radiograph images, the gray level values in the liquid

1 phase are qualitatively related to the solute concentration: the lower the gray level, the higher
2 the solute concentration. Fig.5 shows gray level profiles measured for the four solidification
3 configurations in front of the highest dendrite tip of the columnar grain network and just before
4 initiating the second cooling rate. A moving average was calculated (red line in Fig.5) to smooth
5 out fluctuations due to noise in the images. A solute layer is clearly visible for the microgravity,
6 horizontal and upward experiments (Fig.5a to Fig.5c). It was also detected for other dendrites
7 of the columnar fronts. The extension of the solute layers are of the same order of magnitude
8 as the characteristic diffusive solutal length $l_s = D_s/V \approx 333 \mu\text{m}$, with $D_s = 3 \times 10^{-9} \text{ m}^2/\text{s}$ the
9 solute diffusion coefficient [45] and $V = 9 \mu\text{m}/\text{s}$ the average growth velocity. In stark contrast
10 to those experiments, no solute layer was visible ahead of the dendrite tip in the downward
11 experiment with the current spatial resolution. Instead, a constant gray value was obtained and
12 only very short solute layers, often difficult to distinguish, were observed for other dendrites.
13 This difference in solute boundary layer is ascribed to thermo-solutal convective flow that was
14 most likely strong enough to sweep away the solute rejected during solidification [22].
15 Consequently, additional time after initiating the second cooling rate was necessary to build up
16 a solutal layer with a sufficiently constitutionally undercooled zone where equiaxed grains
17 could nucleate like in the other experimental configurations.

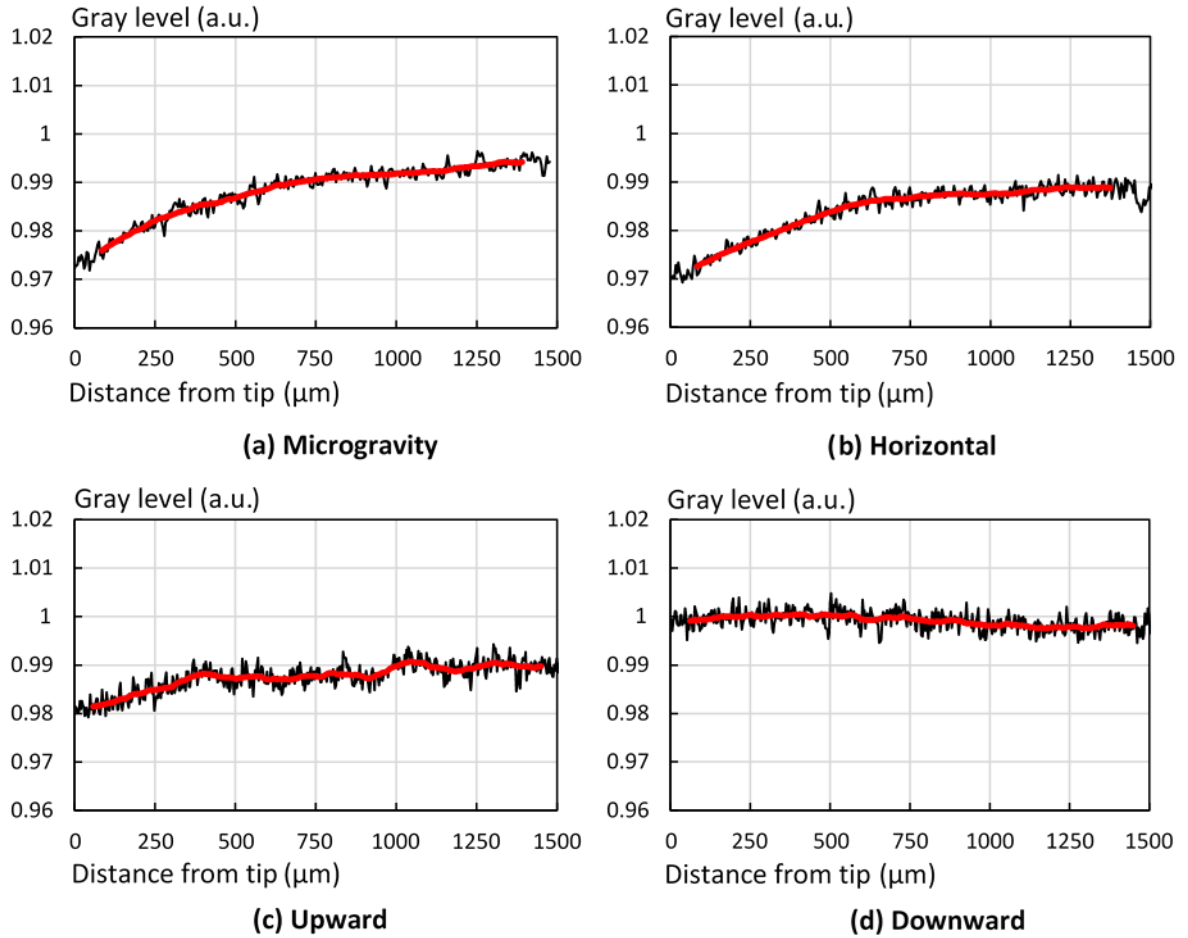
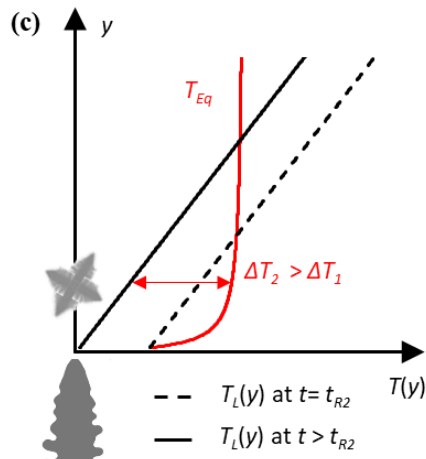
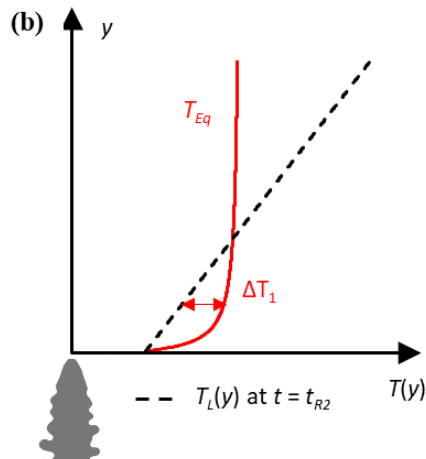
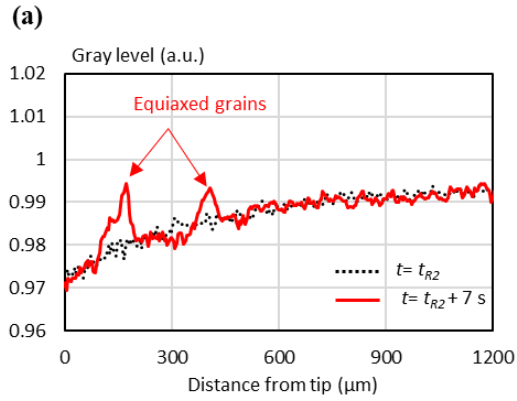


Fig. 5: Gray level variation ahead of the highest dendrite tip for the four experimental configurations. The red curves correspond to the moving average.

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5 Interestingly, the solute profile ahead of the columnar front only slightly changed when the first
6 equiaxed grains appeared as illustrated in Fig. 6a for the microgravity experiment. The two
7 curves show the gray level variation in front of the highest columnar dendrite when the second
8 cooling rate was applied at $t = t_{R2}$ (black curve), and later at $t = t_{R2} + 7s$ (red curve). The two
9 peaks in the red curve correspond to two equiaxed grains that nucleated in front of the dendrite
10 tip. Indeed, the X-ray absorption of the Al-rich solid grains is lower than the Cu-rich liquid,

1 therefore they appear with a higher gray level. The equiaxed grains nucleated even though the
2 concentration profile did not substantially change. This suggests that a change in concentration
3 profile was not the main cause of the constitutional undercooling increase in the liquid, which
4 is necessary to activate the refiners. Given that the Lewis number, defined as the ratio between
5 the thermal diffusivity D_T and the solutal diffusivity D_S is large ($Le = D_T / D_S = 2.17 \times 10^4$ with
6 $D_T = 6.5 \times 10^{-5} \text{ m}^2/\text{s}$ [46] and $D_S = 3.5 \times 10^{-9} \text{ m}^2/\text{s}$), the sample temperature rapidly decreased
7 after the abrupt change of the cooling rate. Meanwhile, the concentration profile did not have
8 time to significantly evolve (Fig.6b and Fig.6c). Consequently, the increase of the constitutional
9 undercooling $\Delta T = T_{eq} - T_L$ (with T_{eq} the local liquidus temperature deduced from the phase
10 diagram and T_L the temperature of the liquid imposed by the temperature gradient) can be
11 attributed to the rapid decrease of the temperature as illustrated in Fig.6c.

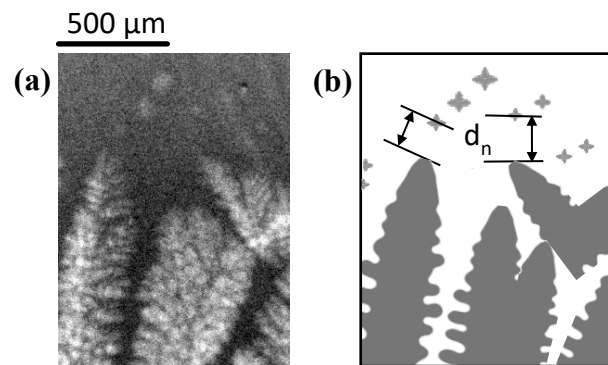


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Fig.6: (a) Gray level variation ahead of the highest columnar dendrite tip (microgravity case). The black curve is the gray level measured at $t = t_{R2}$ and the red curve is the gray level measured at $t = t_{R2} + 7\text{s}$. The two peaks in the red curve corresponds to equiaxed grains that nucleated in front of the dendrite. (b) Graph illustrating the constitutional undercooling intensity ahead of a columnar dendrite at $t = t_{R2}$ and (c) the increase of the constitutional undercooling intensity after the rapid change in cooling rate at $t > t_{R2}$

1 Another difference related to grain nucleation was also observed between the experiments.
2 As mentioned in [section 3.2](#), it seems that the grains nucleated farther from the columnar front
3 in the microgravity case. Complementary measurements were made to quantitatively analyse
4 the distance at which the equiaxed grains nucleated from the columnar dendrites. This
5 nucleation distance d_n was defined as the distance between the centre of the newly nucleated
6 grains and the nearest columnar dendrite tip as illustrated in [Fig.7](#). This measurement was
7 performed for the first layer of equiaxed grains, which corresponds to approximately 30 grains,
8 and for all solidification configurations (it is worth noting that the number of grains of the first
9 layer are very close for the four experiments). The results are shown in [Fig.8](#).

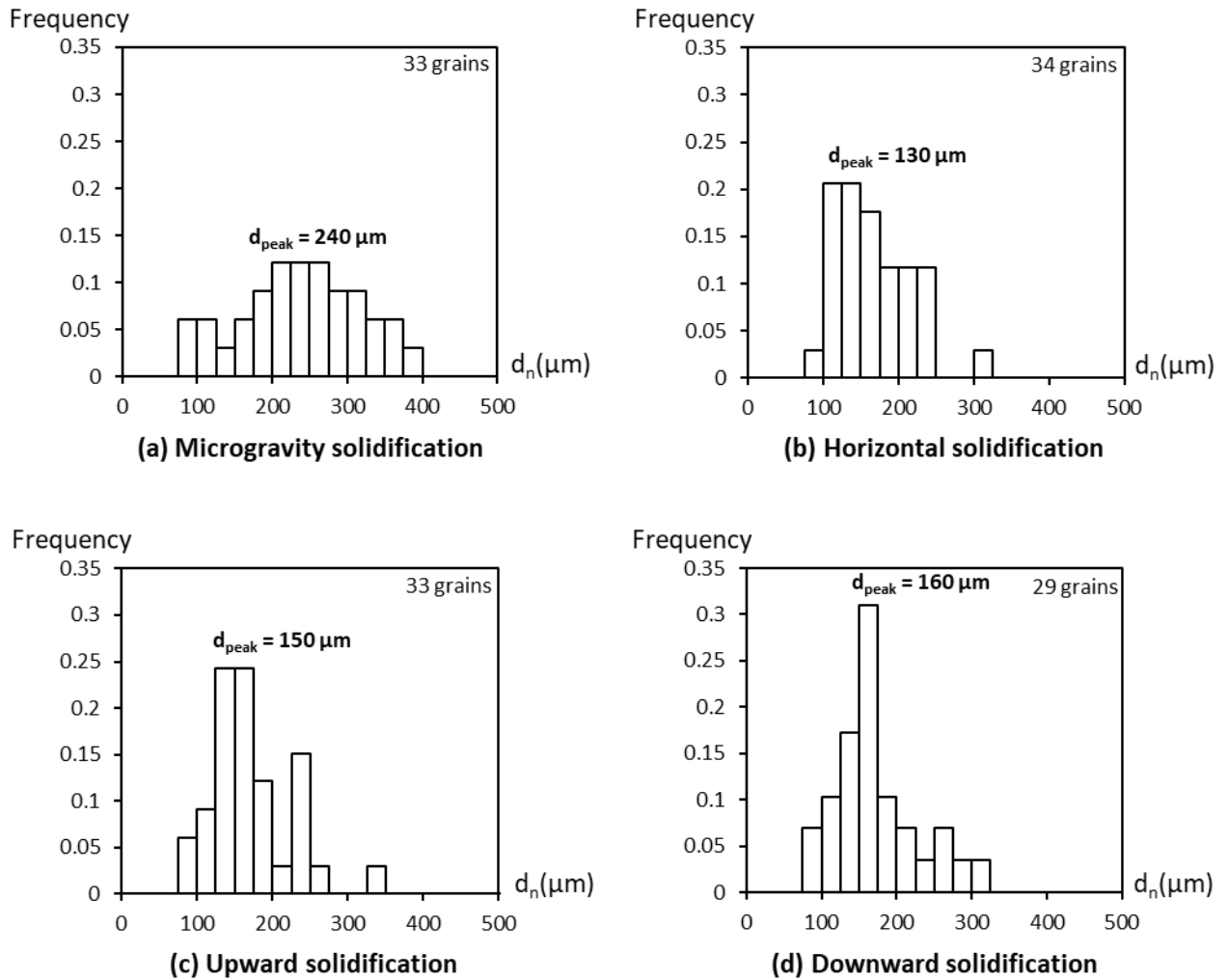
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12 *Fig.7: (a) Radiograph showing columnar dendrites with newly nucleated equiaxed grains*
13 *ahead and (b) sketch showing the nucleation distance d_n between the centre of an equiaxed*
14 *grain that nucleated ahead of the columnar front and the nearest columnar dendrite tip.*

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3 *Fig.8: Distribution of the nucleation distance. (a) Microgravity experiment, (b) horizontal*
4 *configuration, (c) upward configuration and (d) downward configuration.*

5
6 For every experiment, a distribution of the nucleation distance was obtained within a range
7 of 75 μm to 400 μm. This can be attributed to the fact that there exists a distribution of the
8 refining particle size. According to the free growth model proposed by Greer [39], particles
9 with small diameters require a higher undercooling intensity than particles with a large diameter
10 for grain nucleation to occur. Thus, grains that nucleate on particles with a large diameter will

1 appear far from the columnar dendrites, where the constitutional undercooling is lower. In the
2 same way, grains that nucleate on particles with a small diameter will appear closer to the
3 columnar dendrites, where the constitutional undercooling is higher.

4 In addition, clear differences can be noticed between the four graphs. The measurements
5 confirm that the equiaxed grains appeared farther from the columnar front for the microgravity
6 experiment, at around 240 μm from the columnar dendrite tips, which was much larger than the
7 values of around 150 μm for the ground experiments. These discrepancies suggest that gravity
8 and convective flow had a significant impact on the grain nucleation process. It is well known
9 that convective flows on Earth can induce a narrower solute layer compared to the purely
10 diffusive case in microgravity. Models based on the concept that solute transport is assumed
11 diffusive inside a boundary layer (also referred as stagnant film) and that the solute is
12 completely homogenised due to convection outside of the boundary layer have been successful
13 to mimic the effect of convective flow on solidification [47,48]. Thus, the closer distance at
14 which equiaxed grains appeared for the ground experiments can be attributed to convective
15 flow that shortens the solute layer ahead of the columnar front and reduces the extent of the
16 undercooled zone where grains can nucleate. The gray-level profiles reported Fig.5 show that
17 the solute layer before increase of the cooling rate is shorter for the upward and downward
18 solidification experiments. This is less obvious for the horizontal solidification that has a solute
19 profile comparable to the microgravity experiment but for which the equiaxed grains nucleated
20 the closest to the columnar front. The origin of the latter discrepancy is not clear. As for

1 downward solidification, the horizontal configuration is both thermally and solutally unstable
2 with respect to convective flow [47]. Thus, the concentration pile-up, and therefore the
3 concentration profile, could be modified in a way that the maximum of constitutional
4 undercooling would be closer to the solidification front than in microgravity. Experiments with
5 better spatial and temporal resolution would be necessary to carry out a more accurate analysis
6 of the modification of the solute profile during the transient stage following the application of
7 the second cooling rate. Another phenomenon that cannot be characterised currently with the
8 present experimental observation technique is the impact of buoyancy and convective flow on
9 the refining particle distribution, which could also modify the position at which the first grains
10 would nucleate. Advanced numerical simulation that would account for both dendrite growth
11 and convective flow would be helpful to improve the understanding of the present experimental
12 observations.

13

14 **4.2. Blocking of the columnar dendrites**

15 The CET was triggered by initiating the second cooling rate $R_2 = -1 \text{ }^\circ\text{C/s}$ which was
16 significantly faster than $R_1 = -0.08 \text{ }^\circ\text{C/s}$. As discussed in the previous section, this rapid change
17 of the cooling rate induced an increase of the constitutional undercooling in the liquid ahead of
18 the columnar dendrites, which led to the refiner activation and numerous equiaxed grain-
19 nucleation events. The high number of new grains was sufficient in each experimental
20 configuration to eventually lead to the blocking of the columnar dendrites and induce a sharp

1 CET.

2 The two blocking mechanisms reported in the literature were observed. During mechanical
3 (hard) blocking [2], the equiaxed grains came in direct contact with the columnar dendrites as
4 illustrated in Fig. 9a. During solutal (soft) blocking [3], there was no contact and the growth of
5 the equiaxed and columnar grains was progressively stopped due to the accumulation of solute
6 in the interdendritic region as illustrated in Fig. 9b. Solute accumulation between the dendrites
7 induced a smooth increase in the far-field concentration with concomitant decrease in the solute
8 gradient that drives the growth rate, as reported by Bogno *et al.* [45] and Becker *et al.* [49] in
9 the case of isothermal experiments.

10 Additional measurements were made to quantitatively determine the relative importance
11 of each mechanism on the blocking of the solidification front. The measurements were made
12 on the most advanced dendrites of the columnar front and the results are reported in Fig. 9c. It
13 shows that the main blocking mechanism for the microgravity and horizontal experiments was
14 mechanical blocking. This may seem surprising since equiaxed grains could be expected to be
15 fixed in the absence of gravity, as well as in horizontal configuration when grain motion due to
16 buoyancy is limited to the small thickness of the sample. However, the drag flow induced by
17 solidification shrinkage can be significant during solidification experiments as studied by
18 Vernède *et al.* [50] and reported by Salloum-Abou-Jaoude *et al.* [21]. Drag flow induced by
19 solidification shrinkage is a non-gravity-related effect and was at the origin of equiaxed grain
20 motion toward the columnar front. [The motion of 15 representative equiaxed grains has been](#)

1 investigated. The average velocity was 20 $\mu\text{m/s}$ and varied between 10 and 30 $\mu\text{m/s}$ depending
2 on the interaction with the neighbouring grains. Grain motion was observed as soon as the
3 grains were visible, and most dendrites were consequently mechanically blocked.

4 For the two vertical experiments, both mechanical and solutal blocking mechanisms were
5 observed in more similar proportions. On one hand, like in the microgravity and horizontal
6 experiments, solidification shrinkage was at the origin of grain motion toward the cold part of
7 the sample, which induced mechanical blocking. On the other hand, grain motion was also due
8 to buoyancy force in the case of the vertical configuration. The origin of solutal blocking is
9 different for the upward and downward solidification experiments. The image sequence shows
10 that during upward growth, the equiaxed grains did not move toward the cold part as soon as
11 visible as in the microgravity and horizontal experiments. This delay is probably due to a
12 competition between shrinkage flow and buoyancy. For some grains, the shrinkage flow is
13 strong enough to overcome the buoyancy force and mechanical blocking occurs. For other
14 grains, the balance between shrinkage flow and buoyancy leaves time to grains to form a
15 coherent network preventing them from moving and thus promoting columnar dendrite
16 blocking through solutal interaction. In the downward case, the grains moved toward the cold
17 top part of the sample under the sum of both shrinkage flow and buoyancy. In some regions,
18 the grains piled up to form bridges ahead of columnar dendrites, which lead to the solutal
19 blocking of the latter.

20 The present observations and measurements show that both mechanical and solutal

1 blocking mechanisms come into play to induce CET. This highlights the fact that models
2 describing both grain packing, as recently investigated both experimentally and numerically by
3 Olmedilla *et al.* [51], and solutal interactions, as studied for instance by Boukellal *et al.* [52]
4 using a phase-field model, are necessary to improve the understanding of CET. Such a study
5 was recently reported by Sakane *et al.* [53] for two-dimensional simulations and required the
6 use of GPU (Graphics Processing Units) supercomputers.

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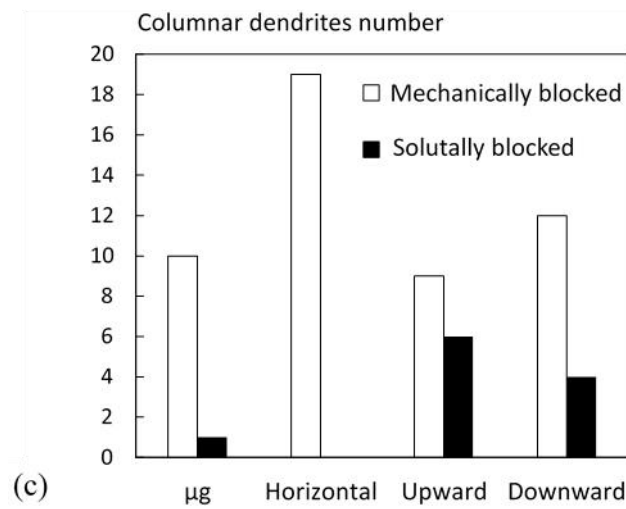
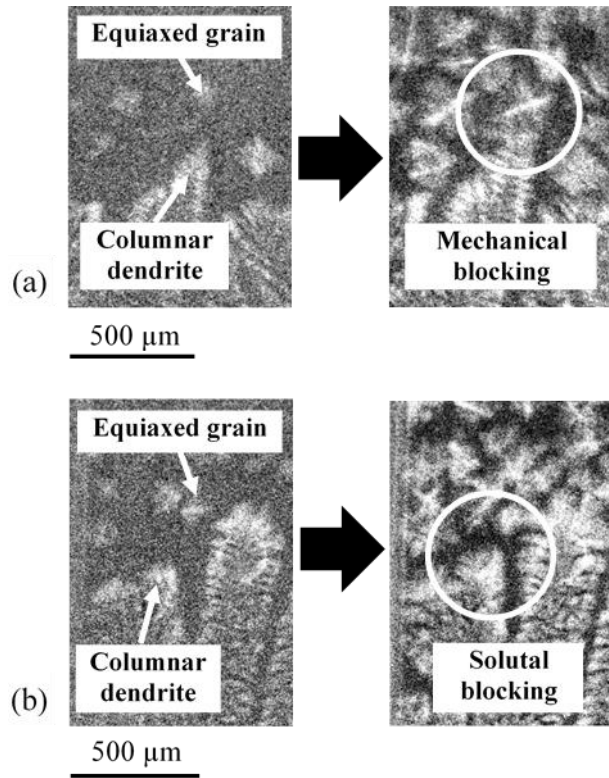


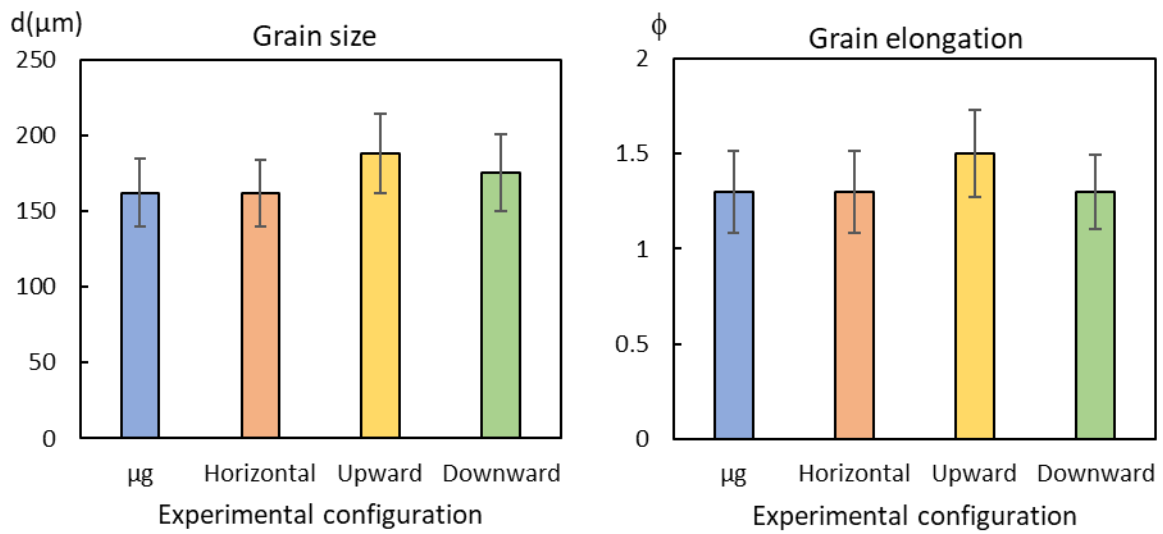
Fig.9: Radiographs illustrating (a) the mechanical blocking mechanism and (b) the solutal blocking mechanism. (c) Distribution of mechanical and solutal blocking for each solidification configuration.

4.3. Equiaxed grain structure after CET

1 The CET was followed by the propagation of an equiaxed front, as described in [section 3.2](#)
2 and in Soltani et al. [29]. The equiaxed grain structure was quantitatively characterised by
3 measuring the grain size and the grain elongation factor defined in [section 2.5](#). Distribution
4 plots were obtained for these two parameters to determine the average value and the standard
5 deviation. The results are summarised in [Fig. 10](#) where the error bar corresponds to the standard
6 deviation. Both the equiaxed grain size and elongation factor are very similar for all
7 experiments. This was expected for the microgravity and horizontal experiments since gravity
8 effects are removed or significantly reduced, respectively, in those cases. In the upward case,
9 the shrinkage flow and the rapidly propagating equiaxed front prevent the grain motion after
10 nucleation, leading to the development of a grain structure like the one obtained in microgravity
11 and horizontal configuration. In a similar way for the downward case, the effective front
12 propagation was much faster than solute sedimentation. Consequently, no solute plume
13 developed during the equiaxed regime, and the equiaxed grain structure formation was not
14 impacted. These observations confirm that gravity-related effects are reduced when increasing
15 the solidification velocity [44].

16 Finally, it is worth noting that the grain elongation factor ϕ was about 1.3 for all
17 experiments. According to Hunt's criterion, the grains can be considered as equiaxed ($\phi < 2$)
18 [2], but the elongation factor did not reach unity even for the presently very high cooling rate,
19 which means that the temperature gradient still had an influence on the grain development.

20



1
2 *Fig.10: (a) Grain size distribution and (b) elongation distribution for the four investigated*
3 *experiments.*

5. Conclusion

6 The purpose of this study was to assess the impact of gravity-related effects on the
7 Columnar-to-Equiaxed Transition in microgravity condition on board of the MASER-14
8 sounding rocket, and on Earth for three different experimental configurations: horizontal,
9 vertical upward and vertical downward solidification. For each case, the different steps leading
10 to a CET were observed *in situ* by using X-radiography. Analysis of the radiographs shows that
11 the increase of the constitutional undercooling necessary to activate the refiners and induce the
12 nucleation of the first grains could be attributed to the rapid decrease of the temperature after
13 application of the rapid cooling rate. In the microgravity case, the nucleation distance is higher
14 than in the three other experiments *i.e.*, equiaxed grains nucleated farther from the columnar
15 dendrites. The low value of nucleation distance for ground experiments is attributed to

1 convective flow on Earth that reduces the solutal layer extent and leads to a shorter nucleation
2 distance.

3 CET due to soft impingement by solutal interaction is prevalent in both vertically aligned
4 solidification experiments. However, mechanical blocking, or hard impingement, is the
5 dominant blocking mechanism in all experiments, due to the shrinkage flow that induces
6 equiaxed grain motion toward the columnar dendrites. This observation highlights a significant
7 importance of solidification-induced shrinkage flow, which is often overlooked. Further
8 investigations on this subject would be worthy of interest, both experimentally for samples with
9 similar geometry or bulk samples, and theoretically.

10 The analyses show that gravity did not impact the grain morphology and size in the
11 equiaxed regime at very high cooling rate. The data collected during this campaign will
12 thereafter be used as benchmark inputs for comparison with numerical simulations and
13 theoretical models to improve the understanding the various processes occurring during CET
14 in metal alloys.

15

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18 the European Space Agency (ESA) and by the French National Space Agency (CNES). The
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20 the XRMON-GF2 facility and the technical support. We would also like to thank Antonio Verga

1 from ESA for his support and the fruitful discussions throughout the sounding rocket campaign.

2

3 **Appendix A. Supplementary data**

4 Supplementary data related to this article can be found at ([link to supplementary material](#))

5

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