RM-645

FLIGHT IV TECHNICAL REPORT FOR EXPERIMENT 74-37 CONTAINED POLYCRYSTALLINE SOLIDIFICATION IN LOW-G

December 1977

(NASA-CR-150518) FLIGHT IV TECHNICAL REPORT FOR EXPERIMENT 74-37 CONTAINED POLYCRYSTALLINE SOLIDIFICATION IN LOW-G (Grumman Aerospace Corp.) 25 p HC A02/MF A01 CSCL 11F G3/26
N78-15232
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RESEARCH DEPARTMENT

GRUMMAN AEROSPACE CORPORATION
BETHPAGE NEW YORK

Grumman Research Department Memorandum RM-645

FLIGHT IV TECHNICAL REPORT FOR EXPERIMENT 74-37
CONTAINED POLYCRYSTALLINE SOLIDIFICATION IN LOW-G

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Postflight Technical Report, Contract NAS 8-31530

December 1977

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ABSTRACT

Experiments were performed to study the effect of a low-gravity environment on the columnar-to-equiaxed transition (CET) during polycrystalline solidification. Solutions of H_2O-30 wt % NH_4Cl and H_2O-37 wt % NH_4Cl were solidified in semicylindrical molds with radial heat extraction. In the ground base tests the general heat flow direction was parallel or anti-parallel to gravity. Both solutions were quenched from the same soak temperature (90^oC); the respective superheat temperatures were, therefore, approximately 57 and 23^oC. The lower superheat resulted in a completely columnar structure, and the higher superheat resulted in a 1/3 columnar - 2/3 equiaxed microstructure; these results were independent of the relationship between heat flow direction and gravity. Grain multiplication mechanisms observed were showering, thermal inversion driven convection cells, and compositionally induced density inversion driven convection cells.

Results obtained during the SPAR IV flight established the viability of the novel freon quenching system designed for this apparatus, but a partial blockage in the needle valve of the quenching system prevented solidification of the samples. Thus no data on the effect of gravity on the CET were obtained. Incomplete preflight melting of the NH_4Cl was also observed. It was established that slow solidification, a long waiting time at room temperature, and the presence of agitation during this time coarsened the room temperature structure of the solid and lengthened the time required for remelting. These observations have led to apparatus modification and revision of prelaunch procedures to prevent a future occurrence of the same problem.

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1. INTRODUCTION

The primary objective of experiment 74-37 is to study the effect of reduced gravity on the columnar-to-equiaxed transition (CET) in small castings (Ref. 1). The results obtained to date from SPAR I show the occurrence of an equiaxed microstructure in low gravity, instead of the expected columnar microstructure (Ref. 2). Similary, fine grained equiaxed, rather than columnar, microstructures were observed in the Skylab metals melting experiment (Ref. 3). On the other hand, Johnston and Griner (Refs. 4 and 5) were able to show that no dendrite arm break-off occurred during low gravity solidification of NH_4Cl and obtained a completely columnar structure. These two results are not necessarily contradictory because CET is a complex phenomenon and gravity driven convection leading to dendrite arm breakoff is only one of the mechanisms that is responsible for the formation of the equiaxed zone. Crystal nuclei may already pre-exist in the liquid, especially following inoculation (Ref. 6) r "big bang" nucleation (Ref. 7). Also, other gravity driven mechanisms may be responsible for transport of small crystals toward the central region of a casting (Ref. 8). A gravity independent mechanism for nucleation of the equiaxed zone has also been proposed (Ref. 9). Regardless of nucleation, it has been recently emphasized (Refs. 10 and 11) that the presence of nuclei is a necessary but not sufficient condition for the occurrence of CET. The growth of equiaxed grains must halt the advance of the columnar interface. This has been suggested to occur by attachment of equiaxed crystals to the columnar growth front (Ref. 10) or by the existence of thermal conditions favoring the growth of equiaxed rather than columnar grains (Ref. 12). This last suggestion, which has received some experimental confirmation (Ref. 11), may be particularly germane to the low gravity environment since rough estimates indicate that the predominant effect of reduced gravity on dendritic growth may be through modification of the thermal rather than solute boundary layer (Ref. 13).

It has been recently emphasized (Refs. 11 and 14) and it also follows from the above discussion that CET is not well understood. It depends on the geometry and thermal characteristics of the mold, heat transfer in the liquid and solid, superheat of the liquid, constitutional supercooling parameter, the presence of nucleation centers, and gravity driven fluid flow.

The SPAR IV flight of experiment 74-37 was performed in a simple apparatus using semicylindrical copper molds with transparent sides. Solutions of NH_4Cl in H_2O were selected as sample materials, because this system has been used successfully by others in previous experiments. The mold geometry was chosen to simulate weld bead solidification and to provide the simplification of radial heat extraction. Four separate cells were available, but all of the cells shared the same thermal environment, i.e., soak temperature and cooling rate. The adjustable parameters were solute concentration, inoculant concentration, soak temperature, and cooling rate. Values of these parameters selected for SPAR IV were such that the CET occurred at different locations in two of the cells, while the other two cells were redundant, i.e., identical to the first two. Thus, this flight experiment was expected to provide data on the effect of gravity on the CET for two conditions.

2. APPARATUS AND EXPERIMENTAL PROCEDURE

A dedicated apparatus was designed and constructed for this experiment. Figure 1 shows an overall view. The apparatus consists of five major components: 1) sample chamber, 2) electronics box that automatically operates the experiment and conditions the telemetry signals, 3) a 250 exposure motor-driven camera, 4) a freon reservoir designed to deliver liquid freon, and 5) is the supporting structure. A more detailed description of the apparatus is given in Ref. 15. Figure 2 shows a close-up view of the sample chamber assembly. Four independent, semi-cylindrical pockets are contained between plexiglass faces. All of these chambers share the same thermal environment; that is, soak temperature and cooling rate. The cylindrical walls of the pockets and the central portion of the metallic block are machined from one piece of copper. The flat "top" walls of the sample pockets are made of stainless steel that is brazed to the copper. This is done in order to provide a slower cooling rate on the "top" of the sample which would better simulate weld bead solidification. Lighting for photographic purposes is provided by small lamps at the top of each cell, and slots are machined in the stainless steel to admit the light. One of the sample pockets is instrumented with four thermistors (numbered 1-4) placed at equal intervals along the radius, and another pocket has a thermistor (no. 5) at the apex. Two additional thermistors (no. 6 and 7) are attached to the copper block on the "outside" of the cylindrical chill wall. Two heaters are attached to the copper block and two small pieces of stainless steel tubing direct liquid freon onto the web of the copper block at central locations. Not shown in Fig. 2 are fine ruled grids which were placed on the inside rear

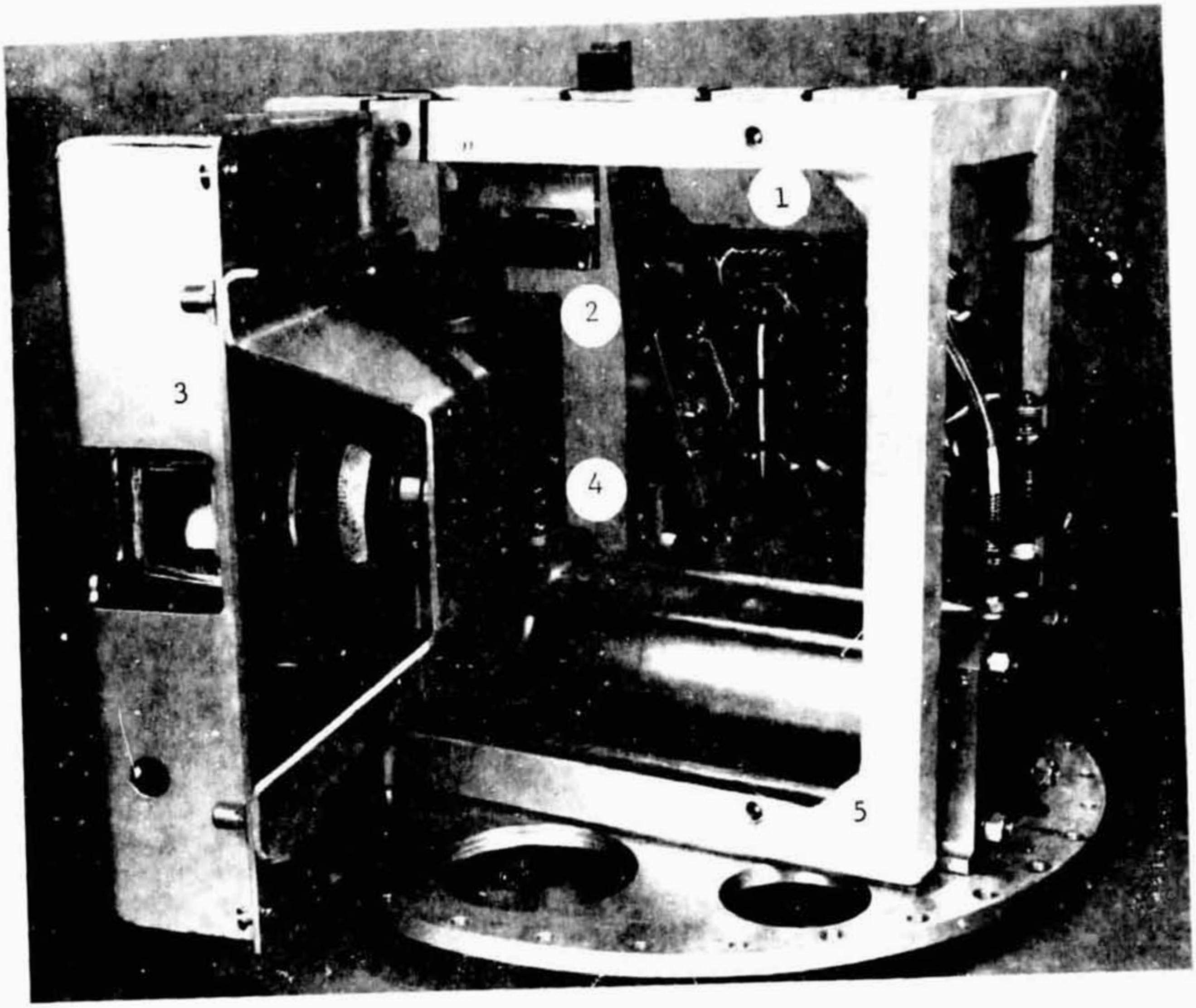

Fig. 1 Overall View of Apparatus 1) sample chamber, 2) electronics box, 3) motor-driven camera, 4) freon reservoir, 5) supporting structure

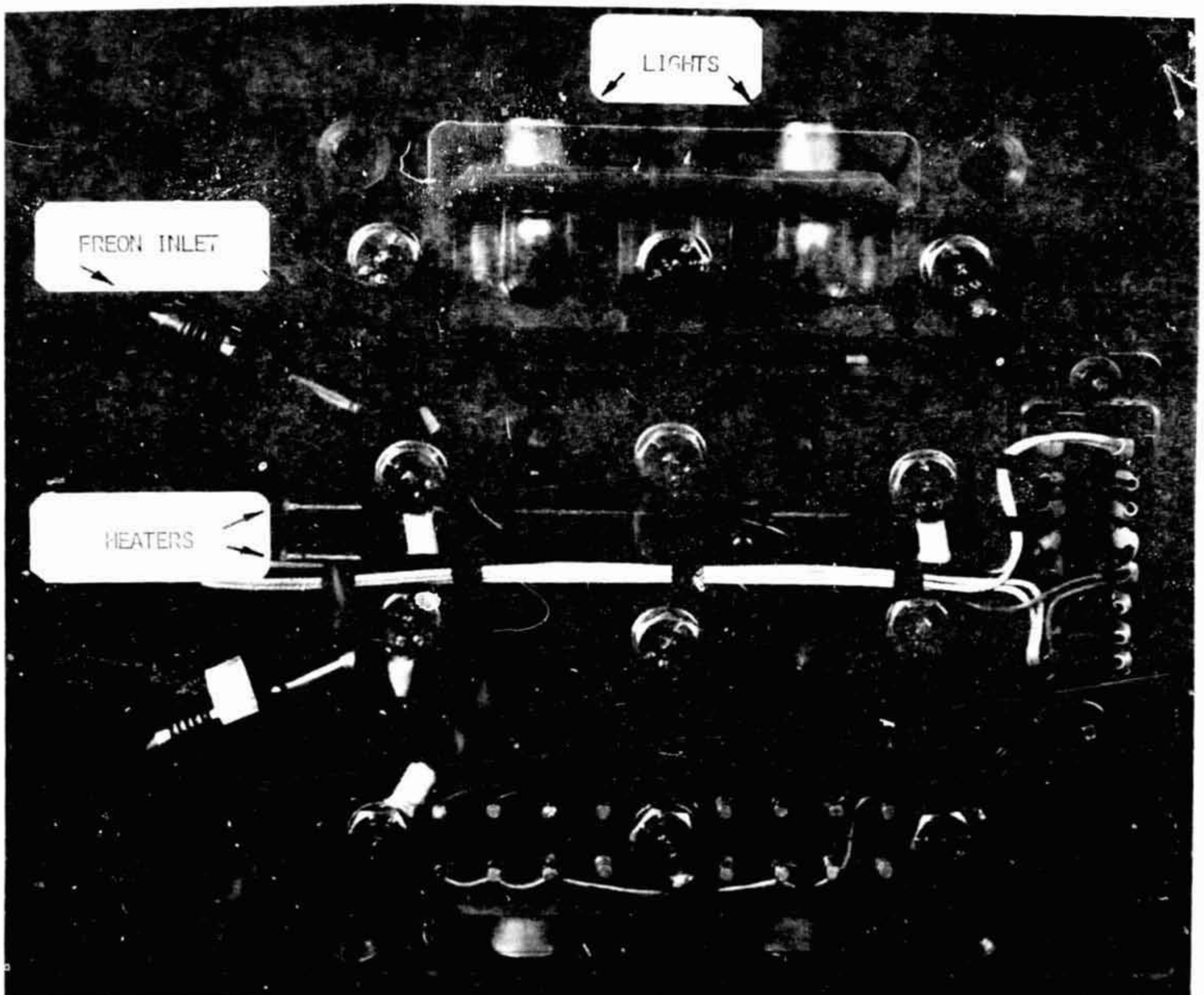

Fig. 2 Close-up View of Sample Chamber

wall of each pocket to allow simple Schlieren observation of convection.

In operation, the heaters were activated 45 minutes before lift-off and the entire sample chamber assembly was heated and controlled at 90°C. Upon lift-off the heater connection was severed. Approximately 30 seconds after attainment of low-g the freon quench was initiated and the camera started. The freon delivery rate was adjusted to give complete solidification of all four samples before the end of the film (220 exposures at approximately 1 frame/sec).

The samples were solutions of NH_4Cl in water. Compositions of 30 wt% and 37 wt% NH_4Cl - H_2O were chosen; their liquidus temperatures were 33 and 67°C, respectively. The solutions were made from laboratory purity NH_4Cl and filtered before use.

Laboratory simulations of the flight experiment were performed to generate one-gravity baseline data. The apparatus was installed in a vacuum bell jar which was equipped with electrical and fluid feed throughs, and the prelaunch and flight sequence was followed. The telemetered data were recorded on strip-chart recorders and the flight camera was allowed to operate automatically. Some tests were also performed using a 16 mm motion picture camera to record the progress of solidification.

3. GROUND BASE RESULTS AND DISCUSSION

The major results of the ground base experiments are included in a 16 mm motion picture film that has been submitted to the SPAR Program Office at NASA/MSFC. A copy of the film is available from the authors for loan to interested parties. The film illustrates visually the important effects of solute concentration and heat flow direction on cast microstructure. Figures 3, 4, 5, and 6 show selected frames taken during a typical ground base simulation; they show the progress of solidification in this experiment. The main results arranged in a chronological sequence from the beginning to end of the experiment, are summarized as follows:

1. Before nucleation occurs, thermal inversion, i.e., cold liquid above hot liquid, causes vigorous convection in the lower cells, Fig. 3, cell IV. This is due to a decrease in density of the solution with increasing temperature. This convection is damped soon after the solid starts growing along the chill, presumably because the release of latent heat of fusion decreases substantially the temperature difference between cell top and bottom.

2. Thermal inversion-induced convection is responsible for some grain multiplication, as illustrated in Fig. 4, cell III.

3. Grain "showering" is illustrated in Fig. 4, cells III and IV and in Fig. 5, cells I and III.

4. Convection cell formation and grain multiplication through compositionally induced density inversion is observed in Fig. 4, cell I. During columnar solidification there is rejection of water between the dendrites, making the interdendritic

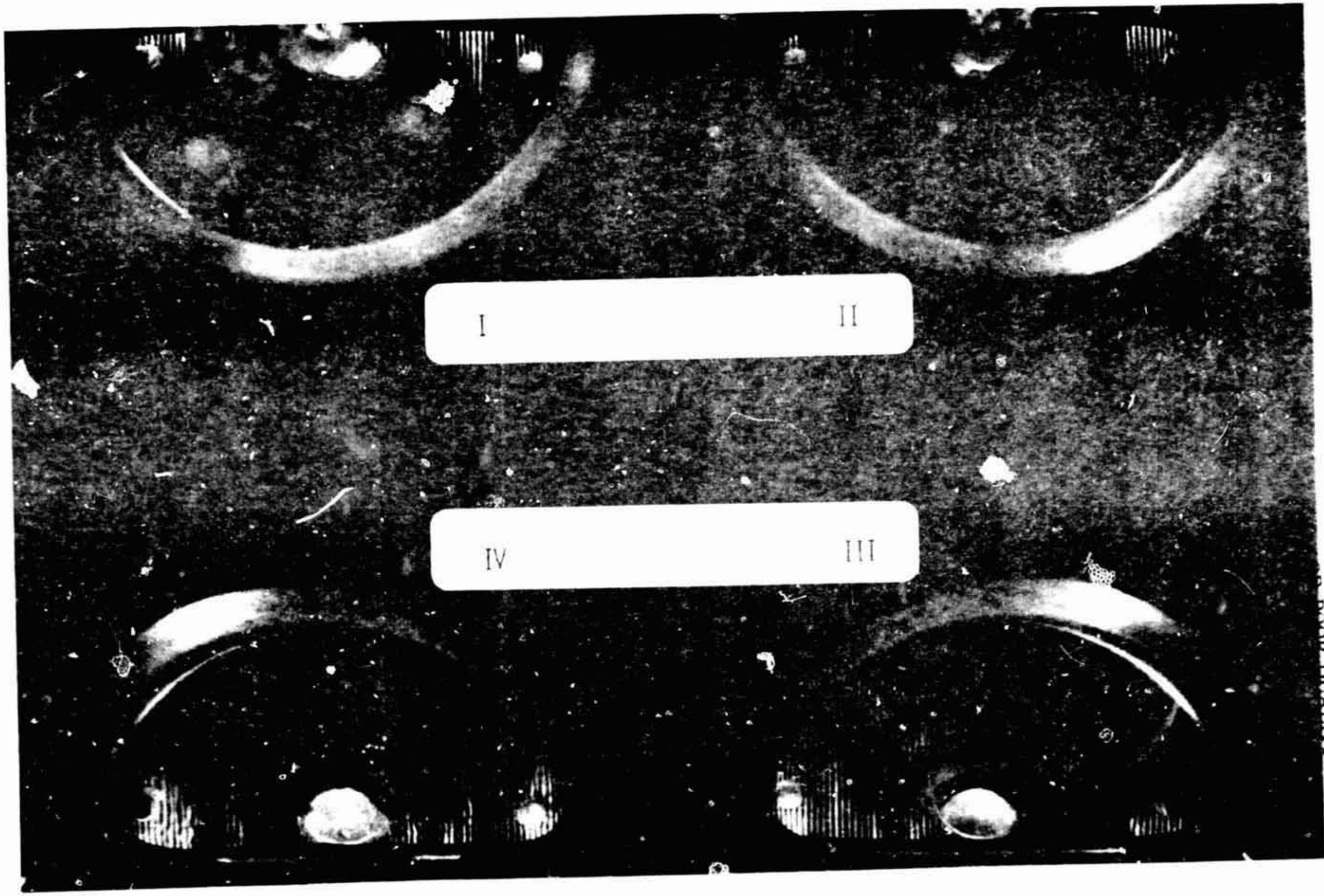

Fig. 3 Ground Base Simulation, 150s After Lift-Off

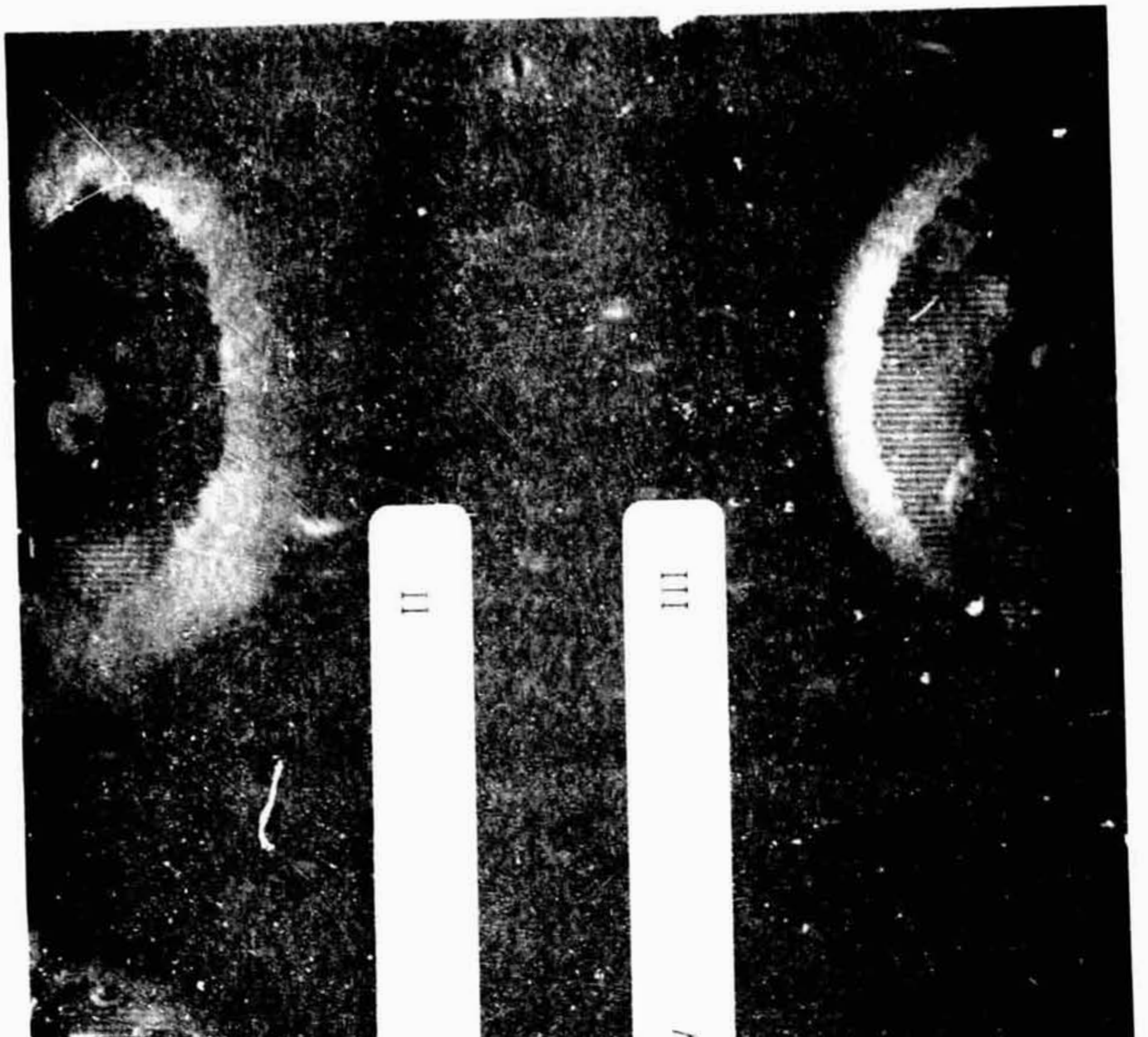

Fig. 4 Ground Base Simulation, 243s After Lift-Off

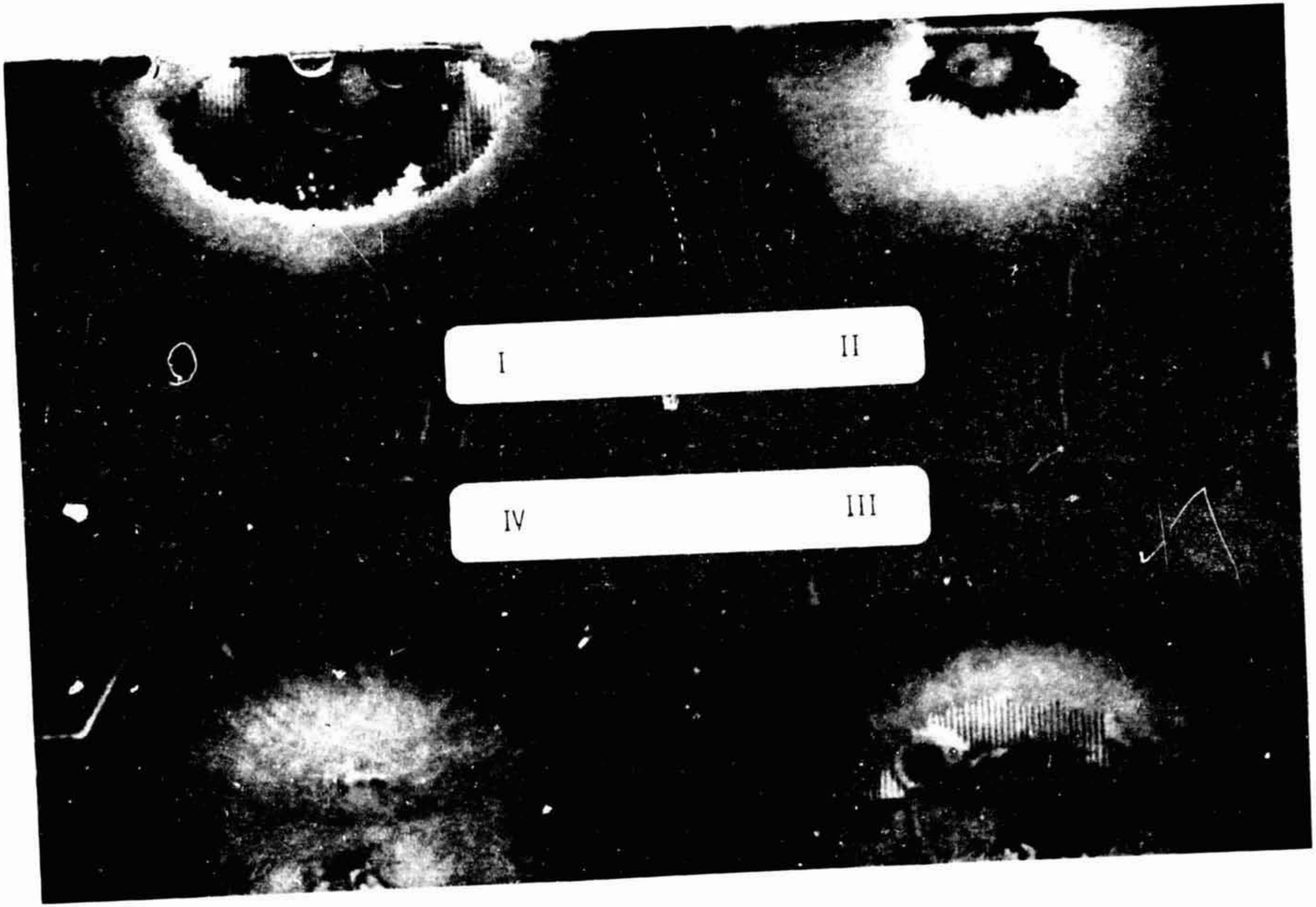

Fig. 5 Ground Base Simulation, 283s After Lift-Off

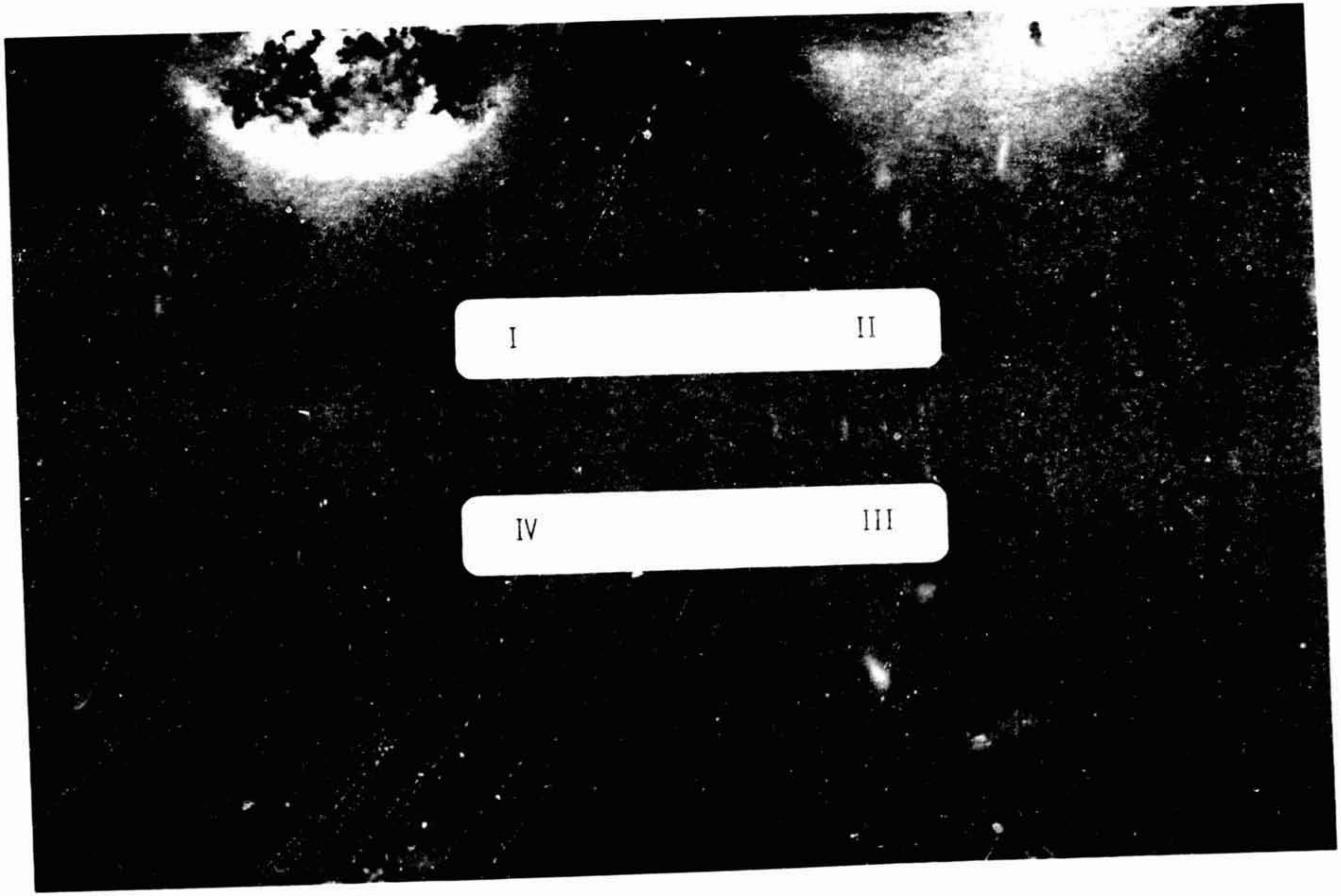

Fig. 6 Ground Base Simulation, 314s After Lift-Off

liquid less dense than the original solution. The rising interdendritic liquid accelerates dendrite arm remelting and coarsening and generates solid fragments which grow into a new generation of grains (Ref. 16).

5. The amount of liquid superheat prior to solidification has a significant effect on ingot microstructure. At a superheat of 23°C, which corresponds to a H_2O - 37% NH_4Cl alloy, a columnar structure was obtained regardless of chill location (top or bottom), Fig. 6, cells II and IV. At a superheat of 57°C, which corresponds to a H_2O - 30% NH_4Cl alloy, the structure was columnar along the first 1/3 of the mold cavity radius and equiaxed along the remaining 2/3, again regardless of chill location, Fig. 6, cells I and III. These results are consistent with observations of CET transitions in large ingots, in which columnar microstructures are observed during the initial stages of solidification in the high temperature gradient zones and equiaxed microstructures are observed later in lower temperature gradient zones. Different results can be obtained when nucleation rather than growth is the controlling step.

4. FLIGHT RESULTS AND DISCUSSION

The experiment was performed on SPAR IV. Inspection of the telemetered thermal data showed that the chill block failed to cool adequately (Fig. 7). Examination of the flight film showed that no solidification occurred during the entire length of the film, and that unmelted solid was present throughout the experiment (Fig. 8). These two problems are discussed below.

ABSENCE OF SOLIDIFICATION

The telemetered data were carefully analyzed. We find that for an interval of approximately twenty seconds between 300 and 320s the sample holder was cooled at a rate of about 0.9^{o}C/s, which is the same cooling rate as our preflight ground base simulation (see Fig. 7). Thus, the freon system was capable of cooling at the desired rate even in the absence of gravity. Unfortunately, this cooling rate occurred only for a short interval, rather than for the full 200s. The fact that the proper cooling rate is achievable in zero gravity is, therefore, established and the cause of lack of cooling must be related to freon delivery, not low gravity effects.

A ground based test was performed on the apparatus as received immediately after the flight. The freon supply cylinder was removed from the system, and a supply of freon 12 connected in its place. The complete warm-up and launch sequence was followed and the major temperatures monitored. The test revealed that the system was still blocked. The sample holder was then removed to eliminate it as the source of blockage. The test was performed again and the blockage remained. This procedure isolated the blockage in the solenoid valve-needle valve combination.

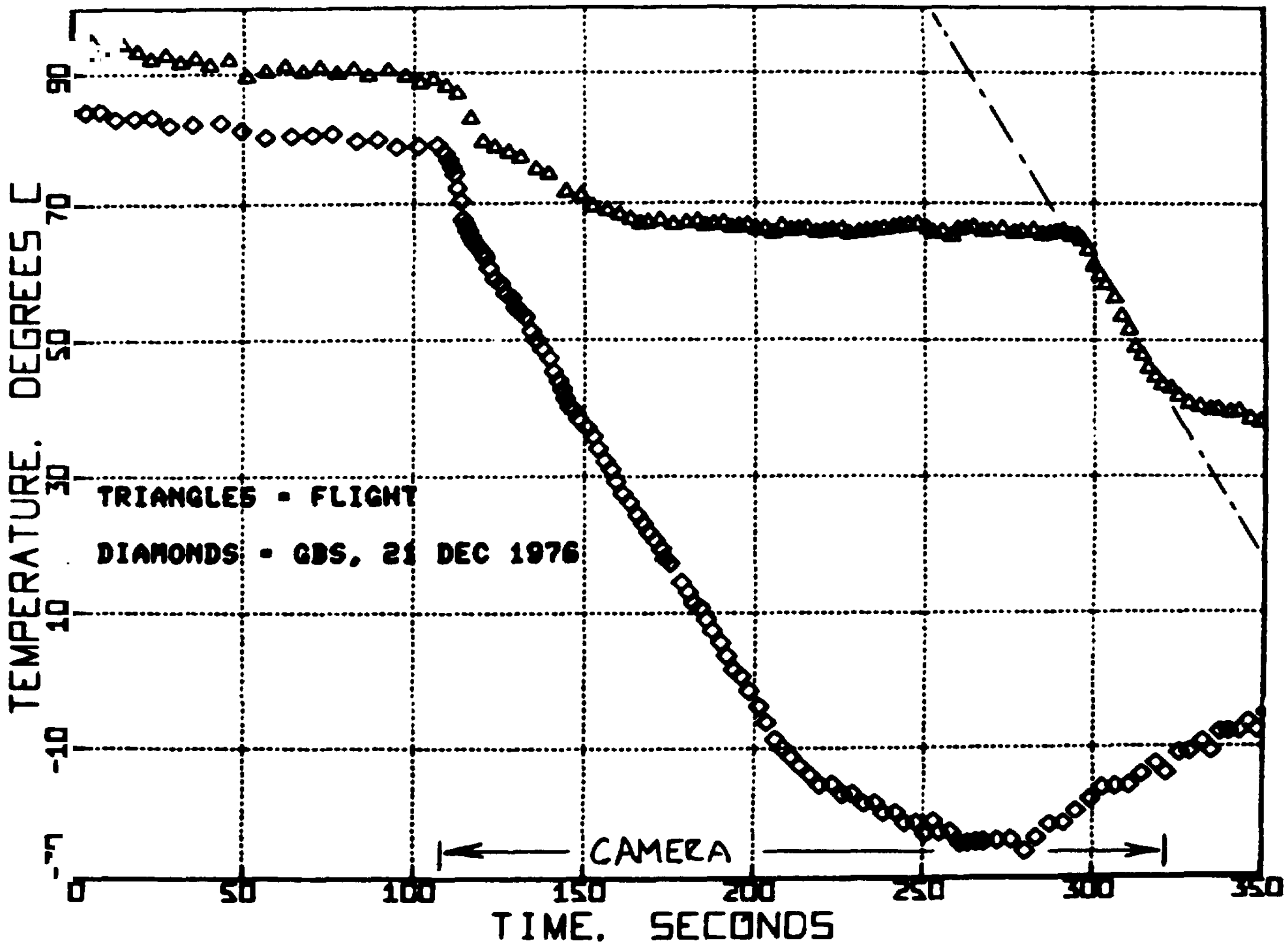

Fig. 7 Thermal History of Flight Experiment and Ground Base Simulation

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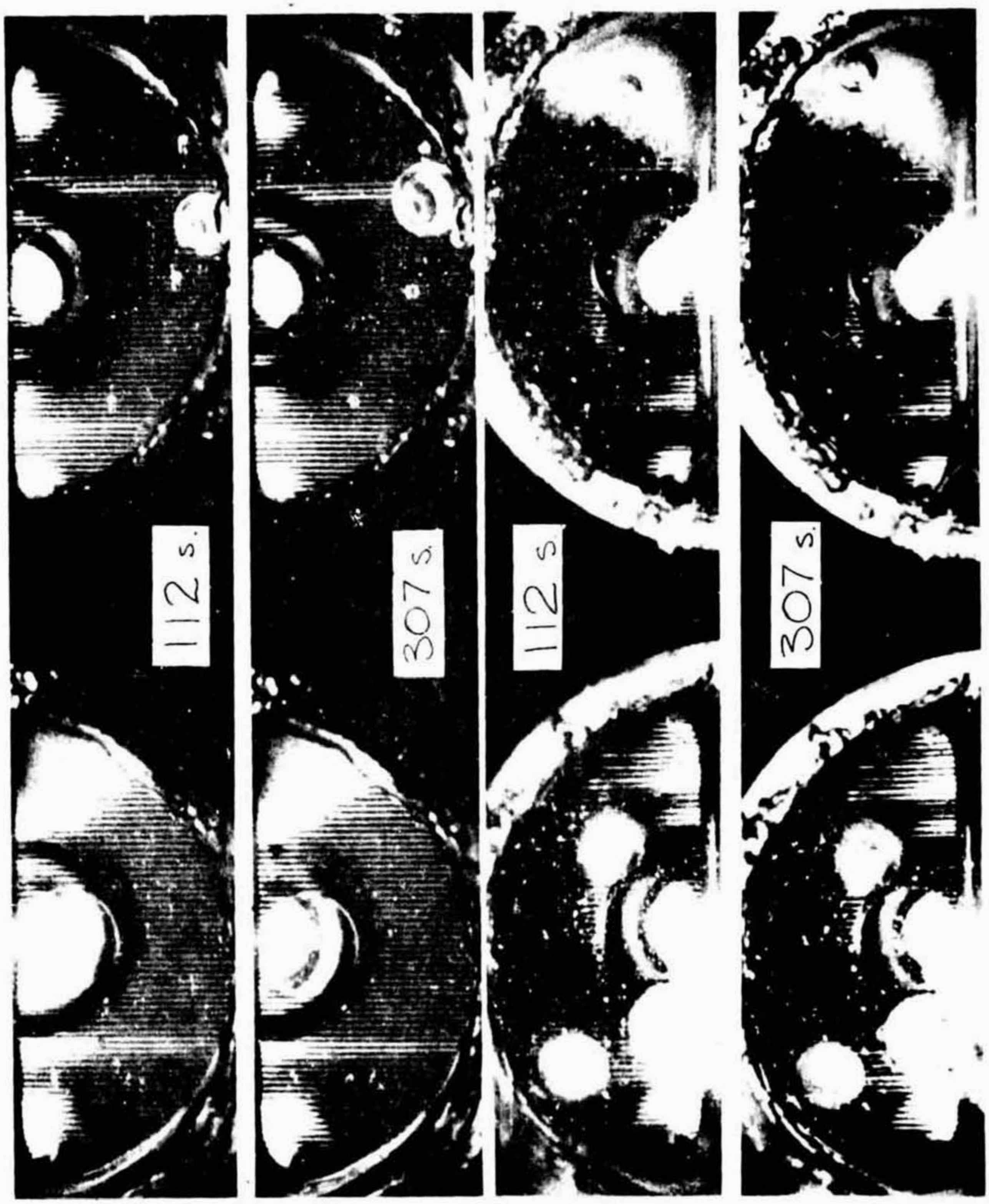

Fig. 8 Results of Flight Experiment at 112s and 307s After Lift-Off

Careful disassembly of each of these valves resulted in the following observations and conclusions:

There were no foreign objects in the solenoid valve, therefore, this unit did not clog

The following foreign objects were found in the needle valve:

- A small black particle about 2 mils in diameter, from the O-ring in the freon bottle
- Shredded material, thread-like, either teflon sealing tape or from the Kel-F seat seal
- A grease, completely surrounding the tip of the needle, suspected to be silicone O-ring lubricant used in the freon bottle

Either the grease or the thread-like material could have clogged the needle valve and reduced the flow rate to an inadequate level, but the combination of the two is thought to provide a potent source of blockage.

Based on these findings, it is concluded that the absence of solidification during the flight experiment was due to a blockage in the freon delivery system. Subsequently, modifications have been made to the apparatus to prevent a future occurrence of this problem.

INCOMPLETE MELTING

To elucidate the reasons for incomplete melting (dissolution) a series of experiments were conducted using the flight apparatus specimen holder. These experiments and the results obtained are summarized in the table on the following page. The three process variables investigated were: original solidification rate, waiting time at room temperature prior to remelting, and agitation during this time. All experiments were performed in the absence of agitation during the remelting stage.

Experiment Number	Solidification Rate	Waiting Time (days)	Agitation	Time Required for Complete Melting (min)
3	Quench	0	No	11
7	Quench	0	Yes	35
1	Quench	3	Yes	>90
10	Quench	1	No	>90
5	Slow Cool	1	No	>90
11	Slow Cool	0	No	22

From these results it appears that slow solidification, a long waiting time, and the presence of agitation contribute to a longer time being required for complete remelting. Comparison of experiments nos. 3 and 11 shows that a low solidification rate can double the remelting time. Comparison of experiments nos. 3 and 7 shows that the presence of simple agitation at room temperature has an even greater effect on remelt time. Furthermore, comparison of experiments nos. 3, 7, 11, and 1, 10, 5 shows that the effect of waiting time at room temperature is the most important of the three process variables.

To explain these results it is necessary to observe that at room temperature the system is above the eutectic temperature and is part liquid, part solid. It consists of primary dendrites of pure NH_4Cl surrounded by a liquid containing about 27 wt% NH_4Cl. During reheating, a dissolution of NH_4Cl occurs at the surface of dendrite arms or solid particles, enriching the surrounding liquid in NH_4Cl. The dissolution process depends on the surface to volume ratio of the solid, therefore, the time required for complete dissolution depends on the dendrite arm or solid particle radius. A rapid initial solidification rate would refine the primary NH_4Cl dendrites and hence would reduce the time required for complete dissolution or remelting. Also, because solid is in contact with liquid at room temperature, Ostwald ripening or diffusional coarsening of the solid would occur during long waiting times. Thus, dendrite arms or solid particles would become coarser, hence a longer time would be required for their remelting. Finally, agitation of a solid-liquid mixture at room temperature accelerates diffusional coarsening by enhancing transport of NH_4Cl through the liquid and causes collisional coalescence of various dendritic or nondendritic particles. Again, this would contribute to coarsening dendrite arms or solid particles and would, therefore, lengthen the time required for complete remelting. As a result of this analysis a revised prelaunch procedure can be developed to assure complete dissolution of NH_4Cl particles before lift-off.

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