Experimental Observation of Convective Breakdown during Directional Solidification

T. D. McCay, M. H. McCay, and P. A. Gray

Center for Laser Applications, University of Tennessee Space Institute, Tullahoma, Tennessee 37388

(Received 4 November 1988; revised manuscript received 3 February 1989)

Using confocal laser optical signal processing and neutrally buoyant particle laser tracking techniques, we have investigated the formation and breakdown of the diffusion layer which develops at the dendritic interface of a metal-alloy-model material (28 wt.% NH_4Cl in H_2O) undergoing directional solidification. The character of the breakdown for the system is shown to be a solutal Rayleigh instability with a Bénard-cell-type structure which is internal to the fluid, i.e., not terminated by physical or free boundaries.

PACS numbers: 81.30.Fb

The application of fluid analysis techniques to investigate the convective motions within the liquid region surrounding a solidifying metallic alloy has received extensive attention within the last decade. Several reviews on the subject have been written, one of the more recent being a thorough discussion of the phenomena and related problems by Glicksman, Coriell, and McFadden.[1] In his review, Glicksman refers to several types of instabilities, both morphological and convective, that concern the materials scientist. One of those, the thermosolutally driven salt-finger convective instability, is indicated as the culprit involved in localized macrosegregation (freckling) in many superalloys. Glicksman indicates that it has not been definitively established whether the onset phenomena associated with these instabilities involve both the diffusion-layer and interdendritic fluid or only the diffusion-layer fluid (Fig. 1); however, he does point out recent work by Sample and Hellawell[2] that indicates the onset of fluid motion is dictated by diffusion-layer rather than interdendritic fluid properties. Our goal was to obtain a more specific determination of the nature of the onset of convection.

We investigated the onset of fluid instability in a solidifying system by employing a solution of 28 wt.% NH_4Cl in H_2O sealed in a (25 mm high × 19 mm wide × 12 mm deep) quartz cuvette and directionally solidified in a constant vertical linear temperature gradient. This metal-alloy analog[3] solidifies dendritically, producing almost pure ammonium chloride dendrites.[4] The interdendritic liquid thus becomes water rich and, depending upon the magnitude of the opposing temperature gradient, buoyant. It should be noted that it is possible for the liquid to be nonbuoyant but still unstable due to double diffusive effects[5] but we chose to investigate the more prevalent case with a true density inversion, as indicated in Fig. 1.

A classical confocal optical processing (COP) technique, the central dark ground method,[6,7] was used in the experiments. The strong index-of-refraction variation with temperature and concentration for ammonium chloride and water produces well defined COP interference fringes which, when used in conjunction with external thermocouple measurements, permit one to monitor the development of the diffusion layer in front of the

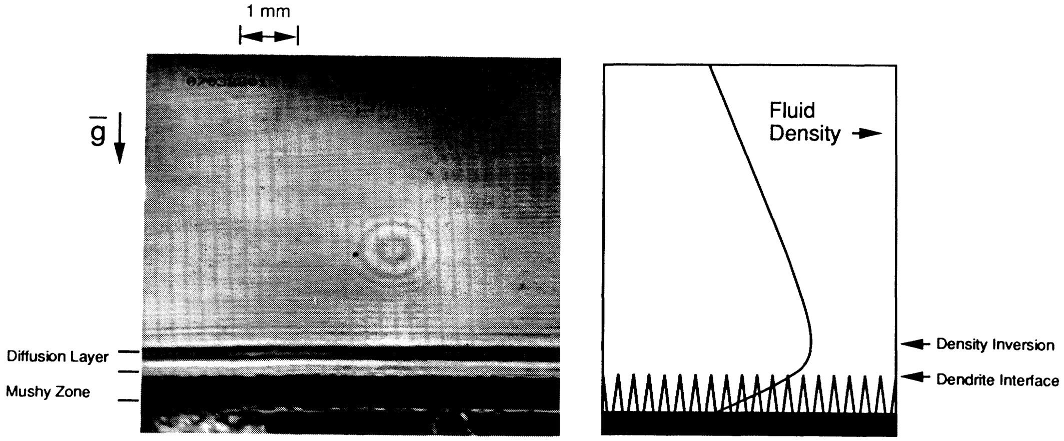

FIG. 1. Confocal optical signal processing view with schematic of directionally solidified 28 wt.% NH_4Cl in H_2O (cooling rate = 1.0 ± 0.02 °C/min, temperature gradient = 8.0 ± 0.5 °C/cm).

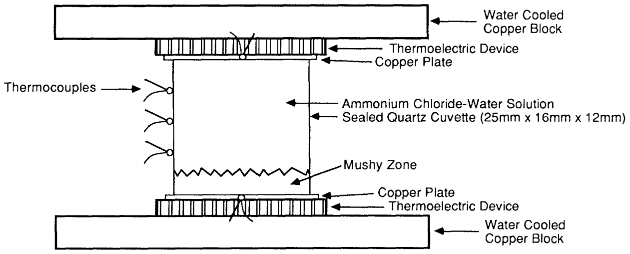

FIG. 2. Schematic of cuvette solidification assembly.

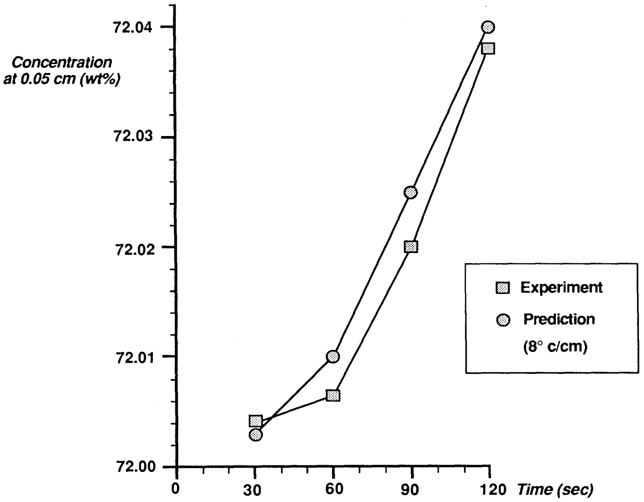

FIG. 3. Predicted and experimental values of the concentration of water 0.05 cm ahead of the mushy zone and bulk fluid interface as a function of time (cooling rate $=2.0\pm0.02\,^{\circ}\mathrm{C/min}$, temperature gradient $=8.0\pm0.5\,^{\circ}\mathrm{C/cm}$).

dendritic interface. This was accomplished for a matrix of cooling rates, temperature gradients, and initial mushy-zone (liquid plus dendrite region) heights. A computer-based solidification assembly (Fig. 2) was used to control the temperature gradient and cooling rate independently and simultaneously. The COP system and its associated optics permitted average diffusion-layer properties and mushy-zone heights to be accurately measured as functions of time. The diffusion-layer growth as measured by COP interference-fringe shifts matched one-dimensional theory[8] quite well (Fig. 3) up until the onset of convection.

The COP system was sufficient to establish onset[9] but not the nature of the convection. Based upon the COP data, the motion appeared to be periodic and vortical in nature but this was not conclusive.

A neutrally buoyant particle tracking system[10] which could be used in conjunction with the confocal optical processor (Fig. 4) was built to establish the convection patterns. Butane-filled plexiglass spheres approximately 10–25 μm in diameter were added to the solution and an argon-ion (488 nm) cylindrically lensed laser beam was used to illuminate the particles during solidification. By photographing 90° scattered light using an 8-sec time exposure, the fluid motion in the beam plane could be determined using the particle tracks. The three-dimensional character of the fluid motion was ascertained by translating (and rotating) the solidification assembly across the sheet of laser light and observing the 90° scattered light for several different parallel planes.

Figure 5 shows a sequence of photographs depicting the behavior of the fluid layer above dendritic interface immediately following the onset of convection, i.e., at breakdown of the diffusion layer. These photographs were taken for the sheet beam aligned with the center of the fluid container, a quartz cuvette. The cells shown were determined to be three deep by the movement of the assembly. We found the cellular convection motion to be vortical in nature, apparently bounded by the dendritic interface on the bottom and the thermally lightened fluid on top. The "eyes" of the vortices shown by the particle tracks were found to correspond to the center of the null (dark band) in the COP data. The null, a dark fringe associated with nearly constant refractive index, indicates the vertical density inversion point in the fluid.[9] An examination of the three-dimensional information provided by cuvette movement proved the vortices not to be paired rolls but rather axisymmetric vortical rings (doughnuts) resembling Bénard cells. The resolution of the current optical system is insufficient to establish whether the cells (rings) are actually circular or hexagonal in shape, but they are definitely three dimensional and have a characteristic wavelength.[11]

The cells grow with time and eventually move upward into initially undisturbed fluid while introducing undulations in the dendritic interface (which was flat prior to onset). Fluid from the mushy zone, containing dendrite fragments, begins to flow up through the center of the

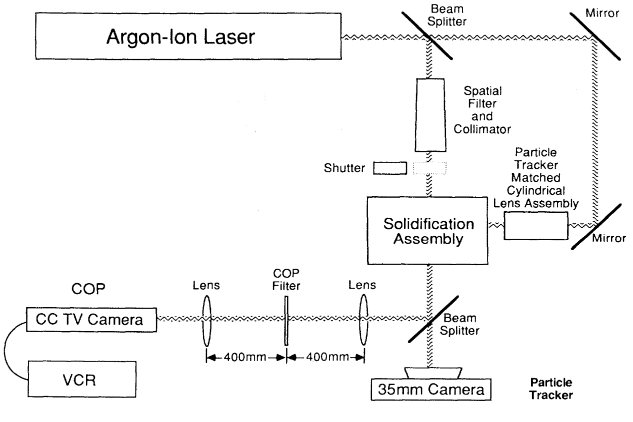

FIG. 4. Schematic of optical system.

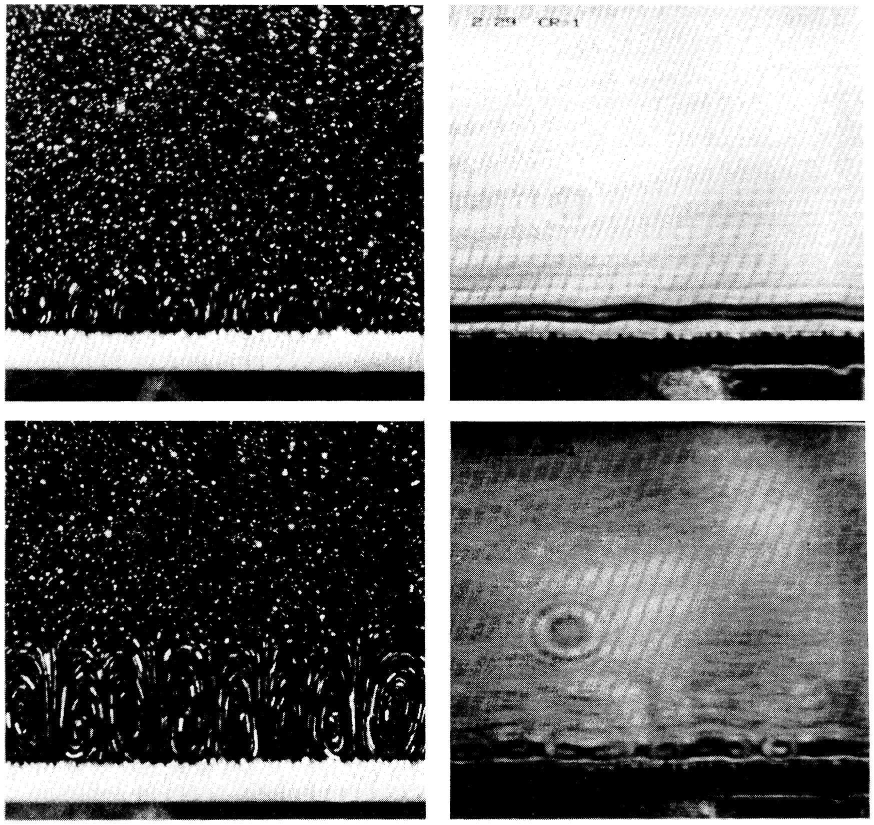

FIG. 5. Particle tracking and COP views during solidification sequence (cooling rate = 1 °C/min, temperature gradient = 8 °C/cm): (a) particle tracking at 2 min, 29 sec; (b) COP at 2 min, 29 sec; (c) particle tracking at 2 min, 59 sec; (d) COP at 2 min, 59 sec.

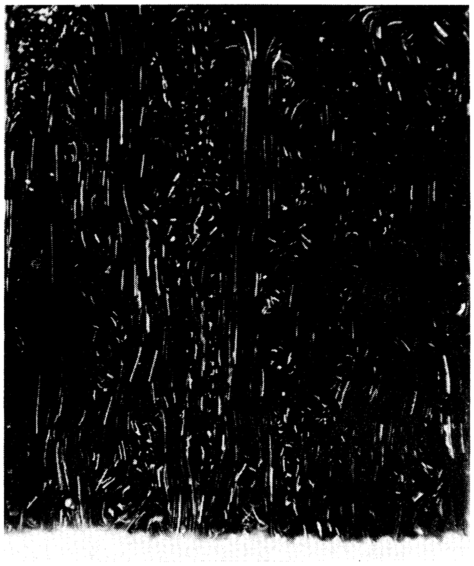

FIG. 6. Particle tracking view of elongated cells which occur subsequent to onset.

cells. This central flow leads to a pluming situation (Fig. 6). After a competitive period between the cells a limited number of these central flows dominate the movement of fluid and fully developed chimneys and plumes can be observed. This behavior is consistent with the predictions and long-time-scale observations of Chen.[12,13]

The observations we have shown here are for the case of a single temperature gradient (8 ± 0.5 °C) and cooling rate (1 ± 0.02 °C/min), but are representative of all cases studied. The bottom temperature was varied to produce various initial mushy-zone heights. Typically the starting bottom temperature was approximately 20 °C. The temperature of the top and bottom were ramped down at a controlled rate to maintain the linear thermal gradient. The temperature was always maintained above the eutectic (−15 °C) temperature during the experiments.

A comparison with theoretical predictions with regard to characteristic wave number and Rayleigh number at breakdown will appear in a future article.[14] The significance of these experiments is the direct (*in situ*, real time) observation of the onset of convection in an alloy undergoing directional solidification. The phenomena are consistent with solutally driven convective instabilities[15] and can provide experimental stability criteria[16] for light-solute-rejecting systems undergoing dendritic growth.

---

[1] M. E. Glicksman, S. R. Coriell, and G. B. McFadden, Annu. Rev. Fluid Mech. **18**, 307 (1986).

[2] A. K. Sample and A. Hellawell, Metall. Trans. A **15**, 2163 (1984).

[3] K. A. Jackson and J. D. Hunt, Acta Metall. **13**, 1212 (1965).

[4] We have measured the segregation coefficient, 0.004, for the system.

[5] S. R. Coriell, G. B. McFadden, R. F. Boisvert, M. E. Glicksman, and Q. T. Fang, J. Cryst. Growth **66**, 514 (1984).

[6] M. Born and E. Wolf, *Principles of Optics* (Pergamon, Oxford, 1980).

[7] J. W. Goodman, *Introduction to Fourier Optics* (McGraw-Hill, San Francisco, 1968).

[8] M. H. McCay, T. D. McCay, and L. M. Smith, "Solidification Studies Using a Confocal Optical Signal Processor" (to be published).

[9] T. D. McCay, M. H. McCay, S. A. Lowry, and L. M. Smith, "Convective Instabilities During Directional Solidification" (to be published).

[10] P. A. Gray, M.S. thesis, University of Tennessee Space Institute, Tullahoma, 1989 (unpublished).

[11] The size of the cuvette and the cooling rate dictate the wavelength. Reference 9 gives complete details.

[12] F. Chen and C. F. Chen, J. Heat Transfer **110**, 403 (1988).

[13] C. F. Chen (private communication).

[14] T. D. McCay, M. H. McCay, and P. A. Gray, "Solutal Breakdown of Diffusion Layers during Directional Solidification" (to be published).

[15] The absolute value of the solutal Raleigh number exceeds the thermal Raleigh number by over an order of magnitude. The occurrence of plumes is a long-time-scale observation.

[16] The criteria include the effect of mushy-zone porosity and are discussed in Ref. 9.

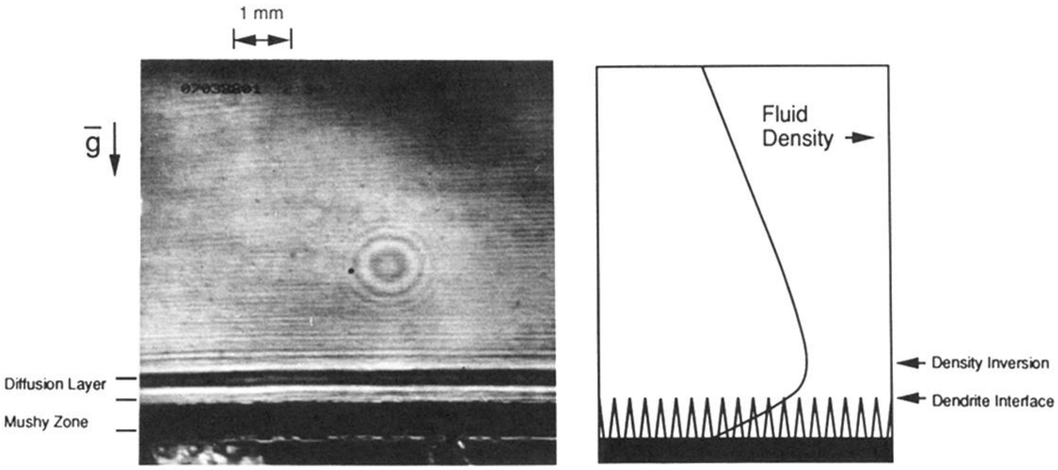

FIG. 1. Confocal optical signal processing view with schematic of directionally solidified 28 wt.% NH_4Cl in H_2O (cooling rate $= 1.0 \pm 0.02\,°C/min$, temperature gradient $= 8.0 \pm 0.5\,°C/cm$).

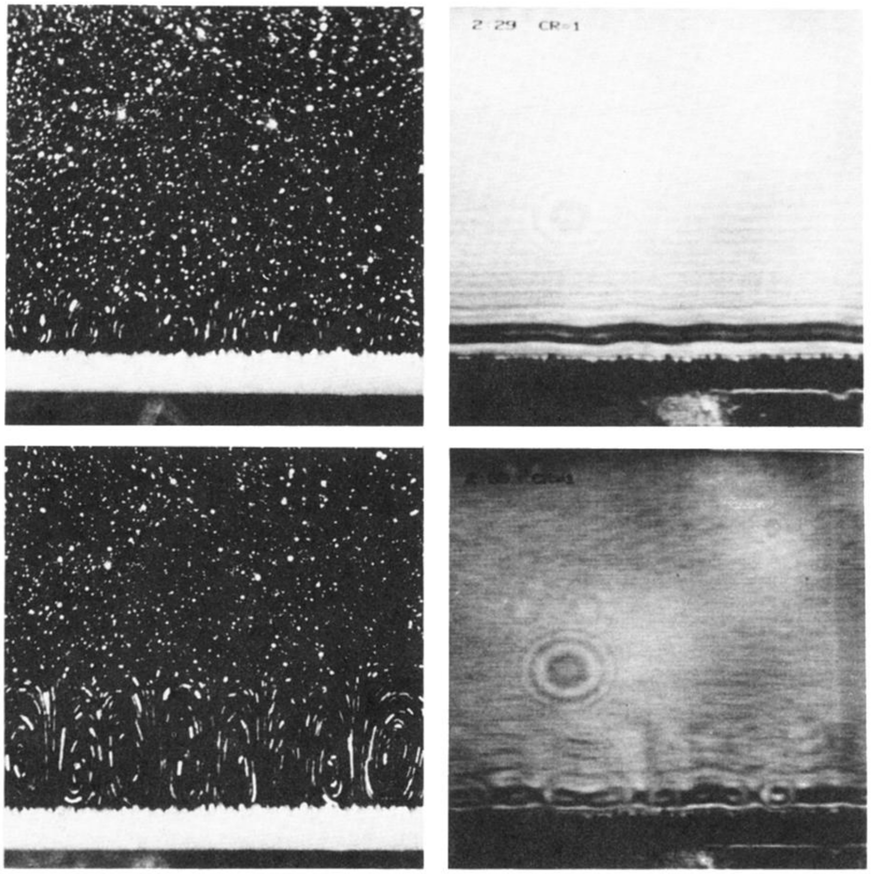

FIG. 5. Particle tracking and COP views during solidification sequence (cooling rate = 1 °C/min, temperature gradient = 8 °C/cm): (a) particle tracking at 2 min, 29 sec; (b) COP at 2 min, 29 sec; (c) particle tracking at 2 min, 59 sec; (d) COP at 2 min, 59 sec.

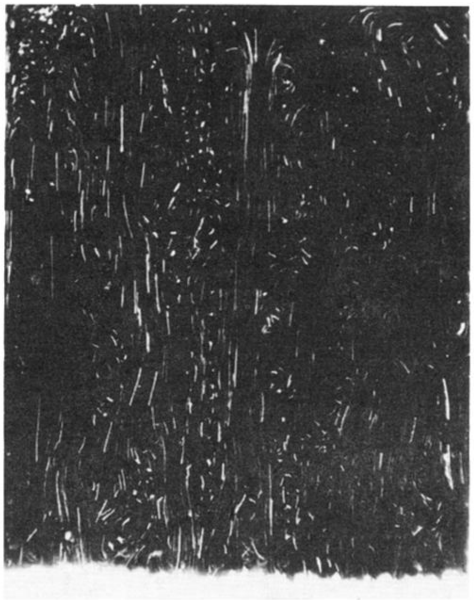

FIG. 6. Particle tracking view of elongated cells which occur subsequent to onset.