Growth of equiaxed dendritic crystals settling in an undercooled melt

A. Badillo and C. Beckermann
Dept. Mechanical & Industrial Engineering, University of Iowa, 2402 SC, Iowa City, IA 52242, USA

Abstract
Experiments were conducted to measure the dendrite tip growth velocities of equiaxed dendritic crystals settling in an undercooled succinonitrile (SCN)-acetone melt. The tip velocities were measured as a function of the settling speed of the crystal and the Eulerian angle between the dendrite arms and the flow direction relative to the crystal. The average of the measured tip growth velocities of all six dendrite arms of an equiaxed crystal was found to be in excellent agreement with the standard diffusion theory result. The individual tip velocities were correlated using a boundary layer model of free dendritic growth in the presence of melt flow that was modified to account for the flow angle dependence. The model is found to predict well the variations in the tip velocity that occur during settling due to crystal rotation and settling speed changes.

Keywords: Equiaxed dendritic growth, undercooled melt, settlement of crystals, SCN-ACE alloys.

1. Introduction
Equiaxed dendritic crystals are frequently observed in metal alloy castings. They freely grow from small nuclei or solid fragments that are suspended in thermally and/or constitutionally undercooled melt. The term ‘equiaxed’ stems from the fact that, for a cubic crystal structure, a seed develops six primary dendrite arms that grow at right angles to each other and at approximately the same rate; higher order dendrite arms are usually found behind the primary dendrite tips. During growth, latent heat of fusion and solute are rejected into the undercooled liquid surrounding the crystal. Understanding the growth of such equiaxed dendritic crystals is crucial for modeling evolution of the grain structure and the development of defects in castings [1–4]. The growth of equiaxed crystals in castings usually takes place in the presence of considerable melt flow and buoyant movement (i.e., settling or flotation) of the crystals themselves. Such flow can influence the growth of an equiaxed crystal and, in turn, its movement relative to the melt. The objective of the present experimental study was to perform detailed measurements of the growth of equiaxed dendritic crystals that are settling in an undercooled melt.

Previous experimental studies involving equiaxed dendritic crystals have primarily focused on the free growth of single, isolated dendrite tips into an undercooled melt [5–13]. Dendrite tip growth during settling of equiaxed crystals has been investigated by Ramani and Beckermann [14] and by Appolaire *et al.* [15,16] using transparent ammonium chloride-water (NH_4Cl-H_2O) solutions. In those experiments, a single NH_4Cl crystal was dropped inside a tall cylindrical glass tube containing an otherwise quiescent undercooled solution. These previous studies showed that dendritic growth is strongly affected by the flow relative to the settling crystal: the measured tip velocities, averaged over all six primary dendrite arms, were found to be between 15 and 68 times larger than those calculated from a theory that assumes purely diffusive solute transport in the melt. It should be noted that comparisons with the theories were hampered by the fact that the dendrite tip selection parameter, σ^*, and other properties (e.g., the Gibbs-Thomson coefficient) are not well established for NH_4Cl dendrites [17]. Also, these studies did not analyze in detail the dependence of the tip growth velocity on the angle between the dendrite axis and the flow [9,18,19].

The present settling experiments were conducted in a setup similar to that employed previously [14], but the transparent model alloy used was SCN-acetone. This alloy is well suited for such experiments because measurements of dendritic growth of pure SCN and SCN-acetone alloys have been performed under a variety of conditions in the past, and because all relevant thermophysical properties are known accurately [5–7,20]. Measurements of the growth velocities of each of the six primary dendrite arms of an equiaxed crystal are presented and analyzed as a function of the settling speed and the orientation of the arms with respect to the flow.

2. Experimental setup and procedures
The present experiments on growth of equiaxed dendritic crystals settling in an undercooled melt were performed using dilute SCN-acetone alloys. The SCN-acetone alloy was filled into a 40 cm tall cylindrical glass tube having an inner diameter of approximately 8 cm. The glass tube was inserted into a large rectangular bath with transparent walls through which a temperature controlled ~ 20% glycol-water

mixture was circulated. Using carefully calibrated thermocouples, the temperature of the SCN-acetone alloy was verified to be within about 0.05°C of the desired set point everywhere inside the glass tube. Small seeds, from which the equiaxed crystals were grown, were produced in-situ using a so-called 'crystal generator' located at the top of the 40 cm long glass tube. The location, size, shape, and orientation of the equiaxed crystals were measured as a function of time during the experiments using two orthogonal cameras. Before each experiment, the liquidus temperature of the SCN-acetone melt was measured, and then the temperature of the bath was lowered until the desired melt undercooling was reached. Figure 1 shows a typical image of a settling equiaxed dendrite. The two orthogonal images were used to measure, as a function of time, the settling speed, U, the length, L, of each of the six primary dendrite arms (from the center of the crystal to the tip), and the Eulerian angle, θ, of each arm. An arm pointing exactly downward has an Eulerian angle of zero degrees, and an arm pointing exactly upward has $\theta = 180°$. Tip growth velocities were obtained from the slopes of relatively short straight line segments that were fitted to the arm length versus time data.

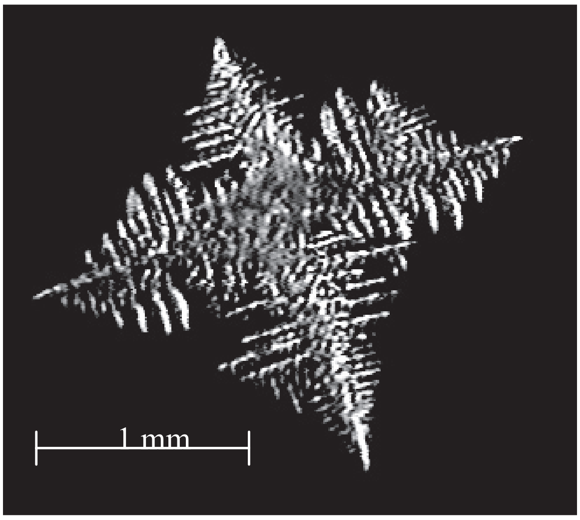

Figure 1: Typical image of an equiaxed SCN-acetone dendrite settling in an undercooled melt.

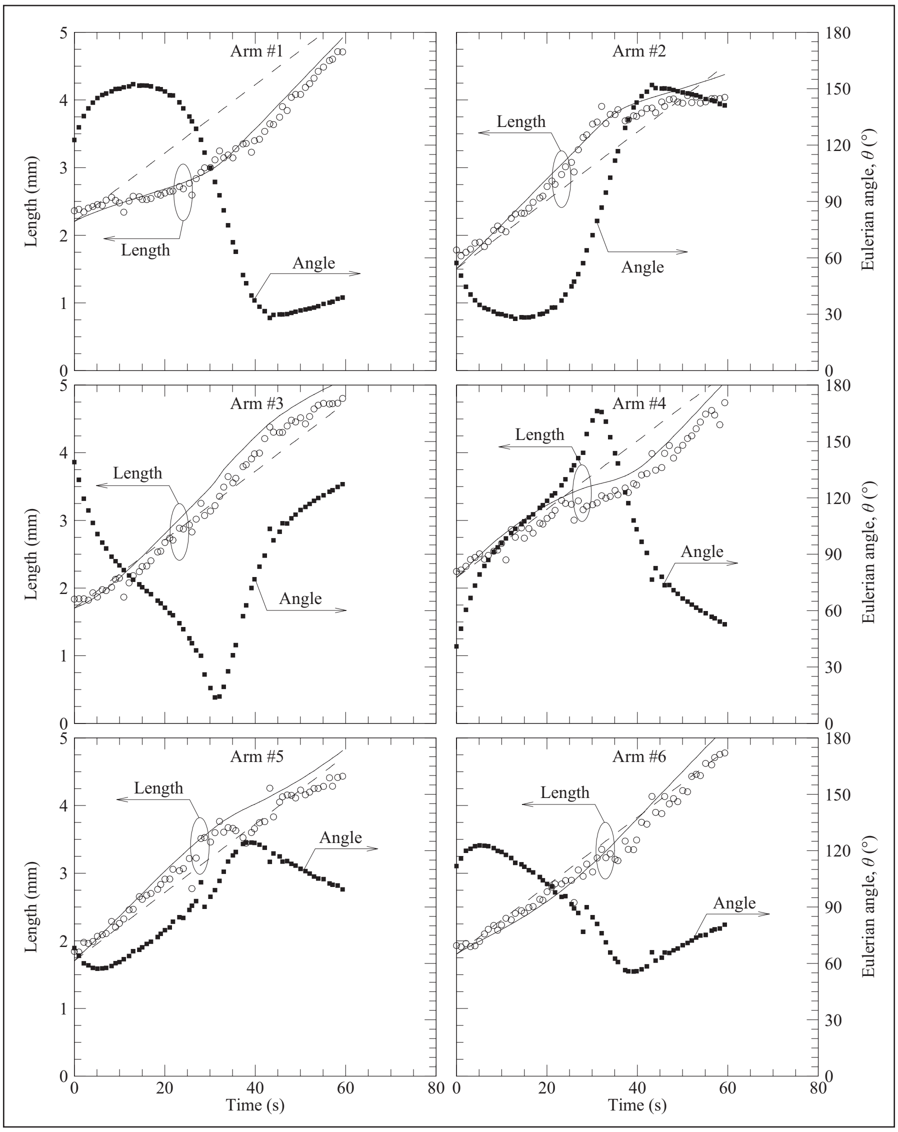

Figure 2: Measured dendrite arm lengths (open circles) and Eulerian angles (solid squares) as a function of time; and predicted dendrite arms lengths from diffusion theory (dashed line) and from the convection model (solid line).

3. Results and Discussion

Eight settling experiments were performed covering a range of acetone concentrations, C_0, and melt undercoolings, ΔT. In all experiments, the settling speed was found to increase in a linear fashion. The ratio of the settling speed to the average tip growth velocity, $U/\bar{v}_t$, ranged from 62 to 572. The measured lengths and Eulerian angles of the six primary dendrite arms for an experiment where C_0 = 1.08 wt% and ΔT = 0.84 K are shown in Figure 2. It can be seen that the six dendrite arms of a settling equiaxed crystal generally grow at different rates, even though the melt is uniformly undercooled. Furthermore, non-constant tip growth velocities can be observed. Considerable crystal rotation can be inferred from the angle variations.

The measured data are first compared to the standard diffusion theory for free dendritic growth of an alloy into an undercooled melt [21–24]. The selection parameter σ^* was taken equal to the well-established diffusion value of 0.02 for SCN. Since, for purely diffusive transport, the predicted tip velocity is constant, the length of a dendrite arm as a function of time can be calculated from $L = L^i + v_t t$ where t is the time from the beginning of the measurements and L^i is the measured initial length of a dendrite arm (at t = 0). The dendrite arm length variations predicted in this manner are included in Figure 2 as dashed lines. As expected, the arm length variations from the diffusion theory generally do not agree with the measurements.

In Figure 3, the measured dendrite tip growth velocities, normalized by the diffusive growth velocity predicted for each experiment, v_{meas}/v_{diff}, are shown as a function of the measured Eulerian angle in a polar plot. It can be seen that the tip growth velocities decrease with the Eulerian angle increasing from 0° to 180°. The dendrite arms with an Eulerian angle greater than about 90° have a growth velocity that is generally smaller than the diffusion value, i.e. the ratio v_{meas}/v_{diff} is less than unity. The arms in the wake of the equiaxed dendrite grow in a less undercooled melt, since heat and solute rejected by upstream arms is advected around the crystal. Hence, their tip velocity can be reduced to a value below the diffusion value corresponding to the original undercooling, despite the presence of convection. This finding is in contradiction to the convection models of Sekerka *et al.* [9] and Gandin *et al.* [18], which give dendrite tip growth velocities as a function of the Eulerian angle that are always greater than the diffusion value. For Eulerian angles between 0° and 90°, the growth velocity ratio, v_{meas}/v_{diff}, generally exceeds unity. This can be explained by the enhancement of heat and solute transport at the dendrite tips by the flow, since for these angles the flow is more or less impinging on the dendrite tips and the melt undercooling is not reduced by heat and solute rejection from upstream dendrite arms.

In Figure 4, the average of the six primary arm lengths (symbols) shown in Figure 2 is compared to the diffusion theory (dashed line). Good agreement can be observed; in fact, agreement for the other seven experiments is even better (not shown here due to space limitations). This good agreement with the diffusion theory is surprising, since it indicates that the relative flow between crystal and melt due to settling has no effect on the average dendrite tip growth velocity in the present experiments. Ramani and

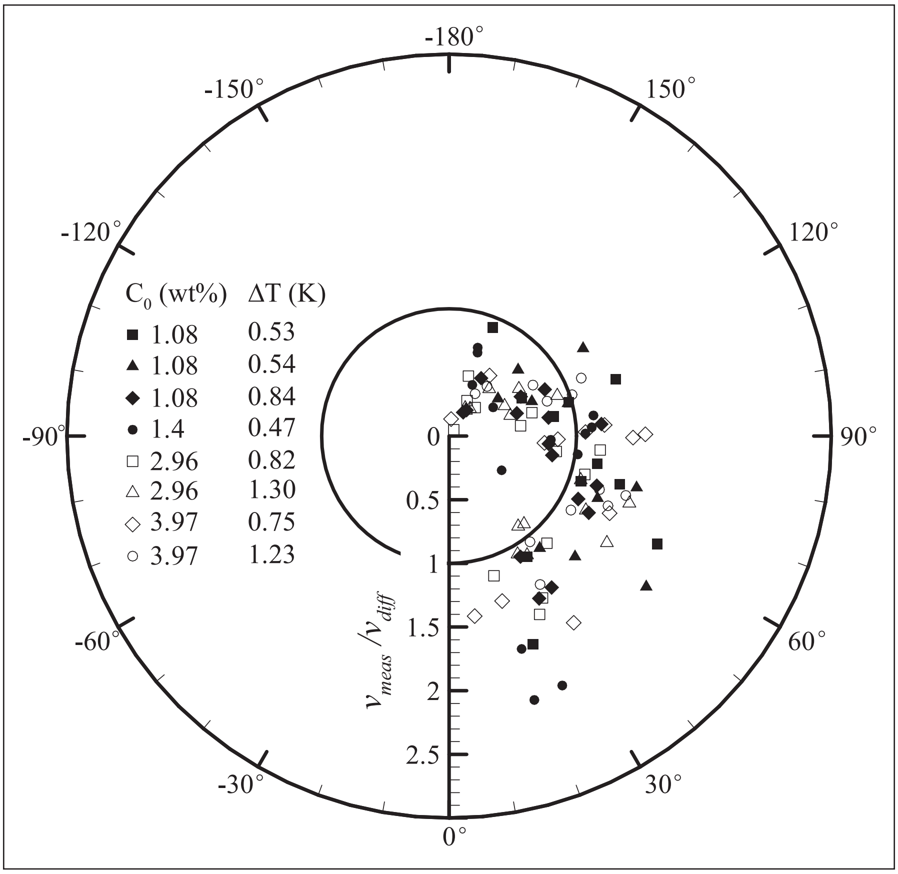

Figure 3: Ratio of measured dendrite tip velocities to those predicted from the diffusion theory as a function of the Eulerian angle.

Beckermann [14] and Appolaire *et al.* [15,16] found that the measured average growth velocities in their NH_4Cl-H_2O settling experiments were between 15 to 68 times larger than those predicted by diffusion theory. The settling speed to average growth velocity ratio in the NH_4Cl-H_2O experiments ranged from 330 to 1,550, while it ranged from 62 to 572 in the present SCN-acetone experiments. This difference is not significant enough to explain the much higher average tip growth velocities during settling in the NH_4Cl-H_2O experiments.

Boundary layer models have emerged as a relatively simple but accurate means of representing the effects of convection on free dendritic growth [9,11,18,25]. This approach is followed in the present study, but a modification is introduced to account for the flow angle dependence. The Ivantsov relation for the dimensionless (thermal or solutal) undercooling, Ω, as a function of the (thermal or solutal) growth Péclet number, Pe, is replaced in the diffusion theory by a modified stagnant film solution of Cantor and Vogel [26] (see Ref. [11] for details):

$$\Omega = Pe \exp(Pe)\{E_1(Pe) - f(\theta)E_1[Pe(1+2\delta/R)]\} \quad (1)$$

where E_1 is the exponential integral function, R is the dendrite tip radius, and δ is the (thermal or solutal) boundary layer thickness. For melt flow directly opposite to the dendrite tip growth direction (i.e. $\theta = 0°$), the boundary layer thickness is calculated from the following correlation developed by Gandin *et al.* [18],

$$\delta = \frac{2R}{0.5773\, Re^{0.6596}(Pr \text{ or } Sc)^{0.5249}} \quad (2)$$

where Re, Pr and Sc are the flow Reynolds, Prandtl and Schmidt numbers, respectively. This correlation was obtained by fitting Equation 1 to the exact Stokes flow solution of Ananth and Gill [10].

The flow direction factor $f(\theta)$ is added in an ad-hoc manner to the term in the stagnant film solution, Equation 1, that accounts for the effect of convection. The flow direction factor has the properties that (i) $a_2 = 1.67 \times 10^{-3}$, such that the solution for flow opposite to the dendrite tip growth direction is recovered, (ii) $f(\theta) > 0$ ($f(\theta) < 0$) for angles that correspond to measured dendrite tip velocities that are greater (smaller) than the diffusion value, and (iii) $f(\theta) = 0$ for angles where $v_{meas}/v_{diff} = 1$. By allowing $f(\theta)$ to be negative, it is possible to predict tip velocities that are less than the diffusion value, as observed in the experiments. Discrete values for the flow direction factor were determined by matching the tip velocities predicted by the convection model with the measured tip velocities. These values were then fitted to a fifth order polynomial given by

$$f(\theta) = 1 + \sum_{i=1}^{5} a_i \theta^i \quad (3)$$

where θ is in degrees. The coefficients are given by: $a_1 = -6.11 \times 10^{-2}$, $a_2 = 1.67 \times 10^{-3}$, $a_3 = -2.02 \times 10^{-5}$, $a_4 = 1.065 \times 10^{-7}$, and $a_5 = -2.08 \times 10^{-10}$.

The above convection model was tested by comparing its predictions with the measured dendrite arm length variations. The instantaneous tip growth velocity of each of the six primary dendrite arms was calculated as a function of the measured instantaneous settling speed, U, and flow angle, θ, of a dendrite arm. Since U and θ vary continuously during an experiment, the predicted tip velocity is not constant and the dendrite arm length must be obtained by numerical integration of the equation $dL/dt = v_t$. The result is shown as a continuous solid line in Figure 2. Relatively good agreement can be observed between the measured and predicted arm length variations, even at those times when the tip growth velocity varies strongly and/or is very different from the diffusion value.

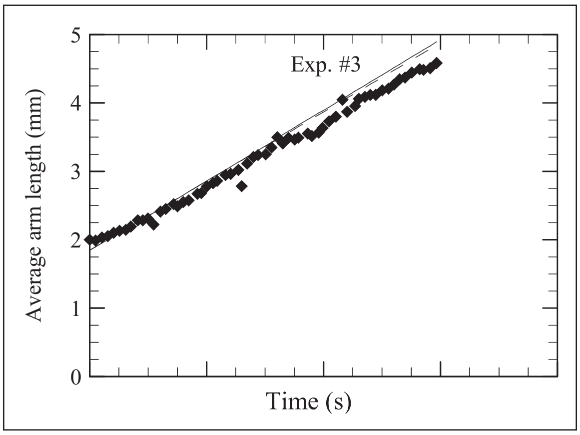

Figure 4: Measured dendrite arm lengths averaged over all six arms (symbols); and predicted average dendrite arm lengths from diffusion theory (dashed line) and the convection model (solid line).

Since the individual dendrite arm lengths are predicted well, it is not surprising that the measured and predicted *average* arm length variations for the entire equiaxed crystal agree too. Figure 4 shows that the differences between the convection and diffusion model predictions for the average arm length are negligibly small, and both agree with the measurements. This indicates that the $f(\theta)$ function given by Equation 3 is sufficiently symmetric about zero. Such symmetry is needed so that the increases in the tip growth velocities due to convection for the three arms with a flow angle less than 90° approximately cancel the reductions for the three arms with $\theta > 90°$, and the average corresponds to the diffusion limit ($f(\theta) = 0$).

4. Conclusions

The present measurements of the tip growth velocities of equiaxed dendritic SCN-acetone crystals settling in an undercooled melt reveal a number of interesting effects that apparently contradict previous measurements and models. The average of the measured tip growth velocities of the six primary dendrite arms of an equiaxed crystal is found to be constant, despite significant settling speed increases and crystal rotation during an experiment. Furthermore, this average tip growth velocity is found to be in almost perfect agreement with the prediction from the standard free dendritic growth theory for purely diffusive heat and solute transport (with $\sigma^* = 0.02$). The agreement is remarkable because the individual dendrite arms grow at velocities that are not only very different from the diffusion value, but that are also not constant during settling. This finding is different from the results of previous experiments with

NH_4Cl-H_2O solutions [14–16]. The present experimental results are also used to develop a model for the effect of convection on dendrite tip growth that takes the flow angle effect into account. Unlike previous models [9,18], it can predict growth velocities that are smaller than the diffusion value. However, this model should not be interpreted as a rigorous theory, because the present measurements were used to calibrate it. Hence, it is not known whether it applies to conditions other than those of the present experiments. By using the measured settling velocity and flow angle variations as input, the model is shown to predict well the tip growth velocity variations that occur during the experiments. It is recommended that a more rigorous theory of equiaxed dendritic growth with melt flow be developed.

Acknowledgements

This work was supported by NASA under contracts NCC8-199 and NNM04AA18G.

References

1. M. Rappaz, Internat. Mater. Rev., **34** (1989) 93.
2. C. Beckermann, C.Y. Wang, Ann. Rev. Heat Transfer, **6** (1995) 115.
3. C. Beckermann, Internat. Mater. Rev., **47** (2002) 243.
4. C.Y. Wang, C. Beckermann, Metall. Mater. Trans. A, **27A** (1996) 2754.
5. M.B. Koss, J.C. LaCombe, L.A. Tennenhouse, M.E. Glicksman, E.A. Winsa, Metall. Mater. Trans. A, **30A** (1999) 3177.
6. S.C. Huang, M.E. Glicksman, Acta Metall., **29** (1981) 701.
7. M.A. Chopra, M.E. Glicksman, N.B. Singh, Metall. Trans. A, **19A** (1988) 3087.
8. Y.W. Lee, R. Ananth, W.N. Gill, J. Crystal Growth, **132** (1993) 226.
9. R.F. Sekerka, S.R. Coriell, G.B. McFadden, J. Crystal Growth, **154** (1995) 370.
10. R. Ananth, W.N. Gill, J. Crystal Growth, **108** (1991) 173.
11. Q. Li, C. Beckermann, J. Crystal Growth, **236** (2002) 482.
12. Y.W. Lee, R.N. Smith, M. Glicksman, M.B. Koss, Ann. Rev. Heat Transfer, **7** (1996) 59.
13. R. Ananth, W.N. Gill, J. Crystal Growth, **179** (1997) 263.
14. A. Ramani, C. Beckermann, Scripta Metall., **36** (1997) 633.
15. B. Appolaire, V. Albert, H. Combeau, G. Lesoult, Acta Mater., **46** (1998) 5851.
16. B. Appolaire, V. Albert, H. Combeau, G. Lesoult, ISIJ Internat., **39** (1999) 263.
17. J.M. Liu, Z.G. Liu, G.C. Wu, Scripta Metall., **32** (1995) 445.
18. Ch.A. Gandin, G. Guillemont, B. Appolaire, N.T. Niane, Mater. Sci. Eng. A, A **342** (2003) 44.
19. X. Tong, C. Beckermann, A. Karma, Q. Li, Phys. Rev. E, 63 (2001) 061601.
20. D.L. Ceynar, C. Beckermann, J. Crystal Growth, **222** (2001) 380.
21. J. Lipton, M.E. Glicksman, W. Kurz, Mater. Sci. Eng., **65** (1984) 57.
22. J. Lipton, M.E. Glicksman, W. Kurz, Metall. Trans. A, **18A** (1987) 341.
23. J. Lipton, W. Kurz, R. Trivedi, Acta Metall., **35** (1987) 957.
24. R. Trivedi, W. Kurz, Internat. Mater. Rev., **39** (1994) 49.
25. D.S. Schrage, J. Crystal Growth, **205** (1999) 410.
26. B. Cantor, A. Vogel, J. Crystal Growth, **41** (1977) 109. ■