MUSHY ZONE EVOLUTION: EXPERIMENTAL AND THEORETICAL APPROACH

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Summary Kinetics of melting pivalic acid dendrites was observed under convection-free conditions on the space shuttle *Columbia* launched in 1997 as STS-87. At low melting Stefan numbers, $St \ll 1$, dendrites melt and shrink steadily toward extinction. Individual fragments follow a characteristic time-dependence derived using quasi-static theory based on conduction-limited melting under shape-preserving conditions. Fragmentation of the dendrites is observed at higher initial supercoolings where the dendritic crystals are initially finer. Capillarity effects are also observed toward the end of each melting cycle. Simulations using three-dimensional adaptive boundary integral methods are performed to quantify the effect of capillarity in melting and fragmentation and to test the shape-constraint assumption (i.e. prolate spheroids) that is made in the quasi-static theory. In addition, crystal/crystal interactions during melting are analyzed numerically.

INTRODUCTION

Melting and freezing phenomena play key roles in materials and biological sciences, and might also influence geophysical and nebular processes. The Isothermal Dendritic Growth Experiment (IDGE), a series of space flight experiments launched by NASA tested existing mathematical models that predict how melt supercooling affects dendrite growth velocity and tip radius. The IDGE instruments, on board the STS-87, provided CCD images (telemetered to Earth during each flight), NASA-processed 35-mm film negatives (available postflight), and, real-time streaming of 30 fps video data telemetered via the K-band (high-frequency) antenna. For the last minute of melting, where the thermal fields approach the stable melt temperature, approximately 1.8K above T_m, individual .tiff files were exported for every video frame.

MICROGRAVITY EXPERIMENTS

Quasi-Static Theory

To interpret microgravity melting experiments two of the authors (MEG/AL) developed a quasi-static model for the conduction-limited melting of prolate spheroids by solving the quasi-static moving boundary problem in the absence of capillarity, assuming that the ratio of major to minor axes, $\mathcal{C}/\mathcal{A}$, is constant [1]. The kinetics are expressed in terms of major axis changes $\mathcal{C}(t)$ (rescaled with respect to its initial value, $\mathcal{C}_0$), namely

$$\mathcal{C}(t)/\mathcal{C}_0 = \sqrt{1 - K_{prol} \cdot St \cdot Fo}. \tag{1}$$

Here K_{prol} is the kinetic coefficient, Fo is the Fourier number, and St is the Stefan number. As melting begins, the tertiary dendritic side-arms begin to shorten, and the larger secondary branches detach from the primary stem. During the final stages, the remaining dendritic fragments present a large variety of shapes, from nearly spherical blobs to elongated needle-like crystals, all of which eventually melt away. In microgravity, it is important to note that the individual crystalline elements of the mushy zone remain motionless, and the melting proceeds by pure thermal conduction. The mushy-zone melting events recorded every $1/30^{\mathrm{th}}$ second as digital frames provides, to our knowledge, the first data compilation of convection-free melting over a significant range of length scales, from about 10^{-2}m down to about 5×10^{-5}m. The large dynamic range of these length scales allows a detailed kinetic analysis of the convection-free melting process.

Capillarity Effects

Capillarity plays an increasingly important role in the late stages of melting, as the curvatures diverge when a melting crystal nears extinction. In Figure 1, left, the $\mathcal{C}/\mathcal{A}$ ratio versus time is shown for an isolated melting prolate spheroid (i.e. the mush zone is almost completely destroyed. The $\mathcal{C}/\mathcal{A}$ ratio gradually increases and falls precipitously until extinction occurs. Until the rapid decrease, our quasi-static theory correctly predicts value of $\mathcal{C}(t)/\mathcal{C}_0$, assuming that the $\mathcal{C}/\mathcal{A}$ ratio is constant (i.e. self-similar evolution). It is thought that capillary effects accelerate melting at the poles relative to melting at the equator, thereby decreasing $\mathcal{C}/\mathcal{A}$. These experiments provide the first evidence for the onset of capillary-enhanced melting—an effect that must eventually occur during melting at sufficiently small length scales—but which is not included in our model of diffusion-controlled melting.

Crystal/crystal interactions

Very little is understood at present of how the presence of neighboring crystals influences the melting and freezing kinetics. When crystals were in close proximity, the melting of an individual crystal was found to be not self-similar, i.e., the $\mathcal{C}/\mathcal{A}$ ratio changed with time, in contrast to the case of isolated crystals. Figure 1, right, shows a representative example where the data and the theoretical predictions are plotted. The theoretical predictions use a "sectorized" approach in which the $\mathcal{C}/\mathcal{A}$ ratio is taken to be piecewise constant. Interestingly, using this approach, the melting kinetics predicted over the entire lifetime of this dendrite fragment is in good agreement with experiment.

Figure 1. ***Left:*** *$C/\mathcal{A}$ ratio versus time for a melting prolate spheroid. The $C/\mathcal{A}$ ratio gradually increases and falls precipitously for the last 10s, until extinction occurs. It is thought that capillary effects accelerate melting at the poles relative to melting at the equator, thereby decreasing $C/\mathcal{A}$.* ***Right:*** *Experimental melting kinetics (data points) for the non-self-similar melting of a mushy zone crystallite, and the theoretical prediction from quasi-static potential theory (solid curve) using rolling averages of the $C/\mathcal{A}$ ratio. Error bars are 5%.*

Fragmentation of Mushy Zones

When dendrites of PVA form at higher initial supercoolings (0.46K) we observe more complex melting phenomena, including fragmentation, capillary effects, and the melting of a primary stem and secondary arm. Despite the appearance of capillarity and fragmentation during melting, the kinetic data are again well predicted by the sectorizing analysis described above.

3D BOUNDARY INTEGRAL METHODS

The other authors (VC/JL) recently developed adaptive 3D boundary integral methods for quasi-steady diffusional phase transformations [2, 3, 4]. These algorithms combine a novel time and space rescaling together with an efficient and accurate numerical descretization of the boundary equations that allows accurate simulations of evolving crystals to be performed for much longer times than has been possible previously. A crucial component of this algorithm is the use of a 3D adaptive surface triangulation developed by Cristini et al. [5]. Simulations using these adaptive algorithms are performed to quantify the effect of capillarity in melting and fragmentation and to test the shape-constraint assumption (i.e. prolate spheroids) that is made in the quasi-static theory. In addition, fragmentation and crystal/crystal interactions during melting are analyzed numerically. An example simulation of a growing anisotropic crystal is shown in Figure 2.

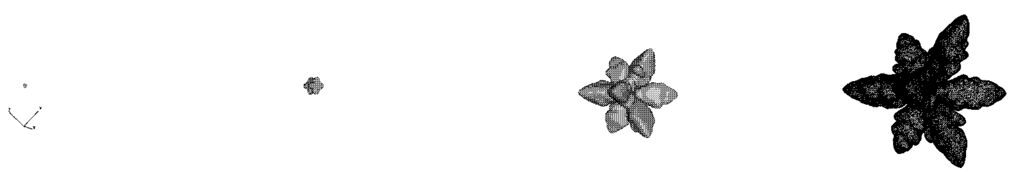

Figure 2. *A 3D adaptive simulation of the growth morphologies of an anisotropic crystal [4]; the computational mesh is shown in the last frame (there are approximately 23,200 computational nodes).*

SUMMARY

Microgravity melting experiments allow observations of the crystal⟶melt phase transformation without any accompanying convective motion. Melting occurs by quasi-static heat conduction down to length scales where capillary effects become important. Capillary effects cause rapid changes in the $C/\mathcal{A}$ ratio of fragments that are approximated as prolate spheroids. Fragmentation also occurs, especially where the crystals grew originally at higher supercooling. Simulations using three-dimensional adaptive boundary integral methods are performed to quantify these effects without any constraint on the shape.

References

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