Crystal Shape Evolution in Detached Bridgman Growth

M. P. Volz¹, K. Mazuruk²

¹NASA, Marshall Space Flight Center, EM31, Huntsville, Alabama, USA
²University of Alabama in Huntsville, Huntsville, Alabama, USA
Research Motivation

• What are the conditions for detachment in microgravity and how do they depend on the governing parameters?
  ➢ Growth angle
  ➢ Contact angle
  ➢ Pressure differential
  ➢ Bond number (ratio of gravity to capillarity)

• Which detached growth solutions are dynamically stable?

• How does an initial crystal radius evolve to one of the following states?
  ➢ Stable detached gap
  ➢ Attachment to the crucible wall
  ➢ Meniscus collapse

• What are the effects of angular dependence on the crystal shape (faceting effects)?
Schematic Diagram of Detached Solidification

\[ \alpha: \text{growth angle} \]
\[ \theta: \text{contact or wetting angle} \]
\[ \Delta P = P_H - P_C: \text{Dimensionless pressure differential across the meniscus} \]
\[ z(r): \text{meniscus shape} \]
\[ Z(r): \text{crystal shape} \]
Equilibrium solutions

\[ \frac{dr}{dZ} = 0 \]

Gap Width vs. Pressure Differential (Microgravity)

\( \theta + \alpha < 180^\circ \)
\( \theta + \alpha > 180^\circ \)

\( B = 0 \)
\( \alpha = 14.3^\circ \) (Ge)
\( \alpha = 25^\circ \) (InSb)

\( \theta = 110^\circ \); InSb
\( \theta = 135^\circ \); InSb
\( \theta = 115^\circ \); Ge
\( \theta = 140^\circ \); Ge
\( \theta = 172^\circ \); Ge
Equations in Zero Gravity

\[ \frac{\partial z}{\partial r} = \pm \frac{\Delta P(r^2 - 1) - 2 \cos \theta}{\sqrt{4r^2 - (\Delta P(r^2 - 1) - 2 \cos \theta)^2}} \]

Meniscus shape equation, \( z(r) \): 2 possible solutions for \( g = 0, B = 0 \)

\[ \frac{dZ}{dr} = \tan (\alpha + \beta) \]

Crystal shape equation, \( Z(r) \): 2 possible solutions in zero gravity

\[ \frac{dZ^\pm}{dr} = \frac{\sqrt{4r^2 - y^2} \tan \alpha \pm y}{\sqrt{4r^2 - y^2} \mp y \tan \alpha}, \quad y = \Delta P(r^2 - 1) - 2 \cos \theta \]
Two Solutions: Crystal Radius can Increase or Decrease

Material: Ge
Starting position, \( r_0 \): 0.6
Growth Angle: 14.3
Contact Angle: 140
\( \Delta P = 1 \)

\( Z^- \): meniscus collapses when \( r = 0.32 \)
\( Z^+ \): crystal eventually attaches to wall
Ge Crystal Evolution for $\theta = 140$ ; $Z^+$ solution

Ge

$\alpha = 14.3$

$\theta = 140$

$Z^+$ (crystal height)

$r_0$ (crystal radius)
Ge Crystal Evolution for $\theta = 140$; $Z^{-}$ solution

Ge
$\alpha = 14.3$
$\theta = 140$

$Z^{-}$ (crystal height)

$r_0$ (crystal radius)
Ge Crystal Evolution for $\theta = 172 \, ^\circ$; $Z^+$ solution

Ge

$\alpha = 14.3$

$\theta = 172$
Ge Crystal Evolution for $\theta = 172$ ; $Z^-$ solution

Ge
$\alpha = 14.3$
$\theta = 172$

$P = 2.5$
$P = 1.9$
$P = 1.4$
$P = 1.3$
$P = 1.2$
Dynamic Stability of Crystallization

Deflection of the growing crystal radius from the stationary condition is governed by the equation

$$\delta \dot{R} = A_{RR} \delta R + A_{Rh} \delta h$$

In microgravity, the coefficient $A_{Rh} = 0$

Solution: $\delta R(t) = \exp( A_{RR} t )$

Dynamic stability achieved when $A_{RR} < 0$

For vertical Bridgman crystal growth

$$A_{RR} = -V \frac{\partial \beta}{\partial R}$$

where $\beta$ is the angle between the tangential line to the meniscus and a horizontal line

$$A_{Rh} = -V \frac{\partial \beta}{\partial h}$$

$V$ is the growth velocity
Dynamic Stability Results

\[ A_{RR} = -V \frac{\partial \beta}{\partial R} \]

\[ \frac{\partial \beta}{\partial R} = \frac{(\Delta PR - \cos \alpha) \sin^2 \alpha}{R \sin^3 \alpha - B(1 - R)^2 (5/24 + R/8)} \]

In microgravity, only the coefficient \( A_{RR} \) is nonzero.

Therefore, dynamic stability only depends on capillarity and occurs when \( \Delta P \cdot R > \cos \alpha \)

Dynamic stability calculations are in agreement with crystal evolution calculations.

On Earth, dynamic stability will depend on the specific thermal conditions of the system.
“Influence of Containment on the Growth of Silicon-Germanium” (ICESAGE) is a collaborative investigation between NASA and the European Space Agency (ESA)

The ICESAGE experiments will be conducted in the Low Gradient Furnace (LGF) in the Materials Science Laboratory on the International Space Station (ISS)

ICESAGE will test the theories of crystal shape evolution in detached Bridgman growth

Dependence on the parameters $\Delta P$, $\theta$, and the crystal starting position $r_0$ will be examined

Launch is currently planned on a SpaceX flight in 2015
A theory describing the shape evolution of detached Bridgman crystals in microgravity has been developed.

A starting crystal of initial radius $r_0$ will evolve to one of the following states:
- Stable detached gap
- Attachment to the crucible wall
- Meniscus collapse

Only crystals where $\alpha + \theta > 180^\circ$ will achieve stable detached growth in microgravity.

Results of the crystal shape evolution theory are consistent with predictions of the dynamic stability of crystallization (Tatarchenko, *Shaped Crystal Growth*, Kluwer, 1993).

Tests of transient crystal evolution are planned for ICESAGE, a series of Ge and GeSi crystal growth experiments planned to be conducted on the ISS.