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Title: Proton Radiography During Alloy Melting and Solidification Experiments

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Proton Radiography during Alloy Melting and Solidification Experiments

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T.J. Ott, M.R. Barker, F. O'Neill, J.J. Hill, M.G. Emigh[2]

Materials Science & Technology Division[1], MST-6[2], MST-7[3]
Physics Division, P-23[4], P-25[5]
Materials Design Institute[6]

MST-6 and the Proton Radiography Team completed the first in-situ examinations of melting and solidification in Al-In, Ga-Bi, Sn-Bi, and Al-Cu alloys at LANL's 800MeV Proton Radiography (pRad) Facility. A furnace was designed and built by MST-6 and inserted into pRad for these experiments, and metal alloy sections raging from 2 to 6 mm in thickness were imaged. Example results are presented. Advantages of using proton radiography for solidification experiments and future opportunities are also discussed.

Proton Radiography During Alloy Melting and Solidification Experiments

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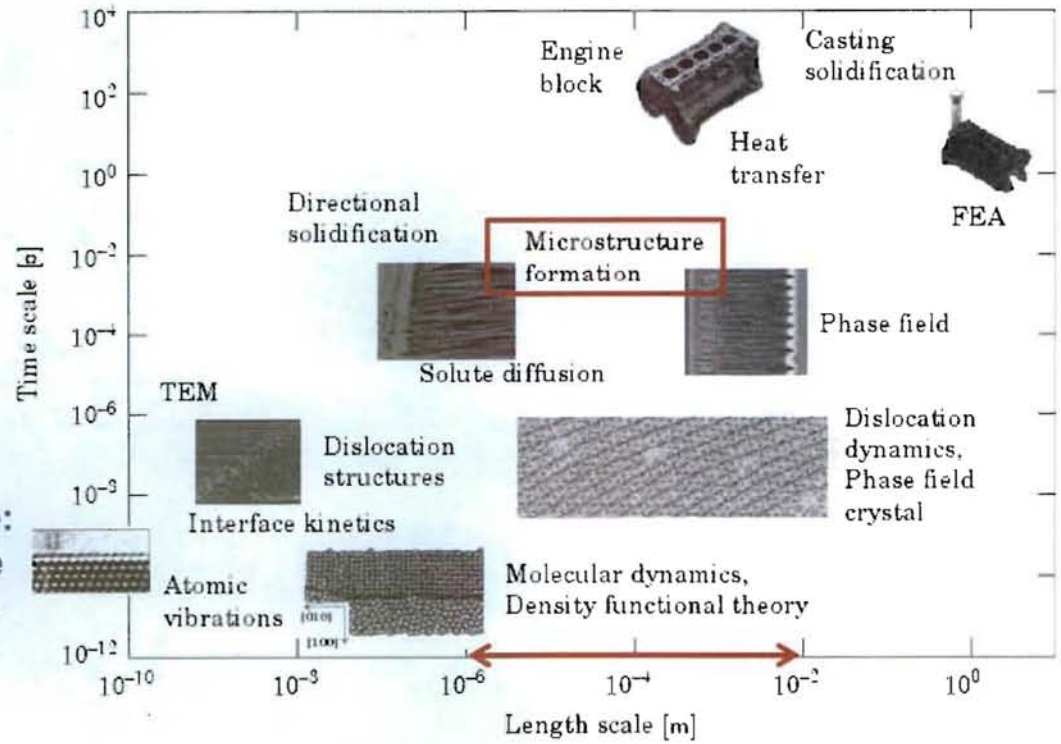
Overview

- Background and advantages of using proton radiography for solidification experiments
- Recent results obtained from in-situ melting and solidification experiments using 800 MeV Proton Radiography (pRad) at LANL
- Summary
- Future opportunities

Solidification Phenomena at Various Length and Time Scales

Micron to millimeter scale: Microstructure formation and chemical segregation occurs…



Macroscopic scale: Geometry and processing conditions determine solidification progress; fluid flow during solidification results in macrosegregation…

Atomic to nano-scale: Solid-liquid interface properties influence growth patterns…

Adapted from J.A. Dantzig and M. Rappaz, *Solidification*, EPFL Press: CRC Press, Taylor and Francis Group, 2009

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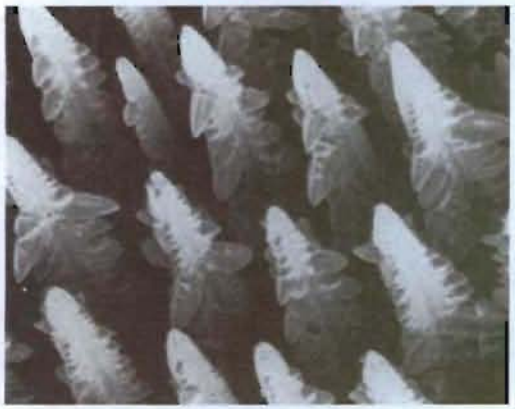
Direct Observations of Microstructure Evolution in Metal Alloys is Lacking

Decanting

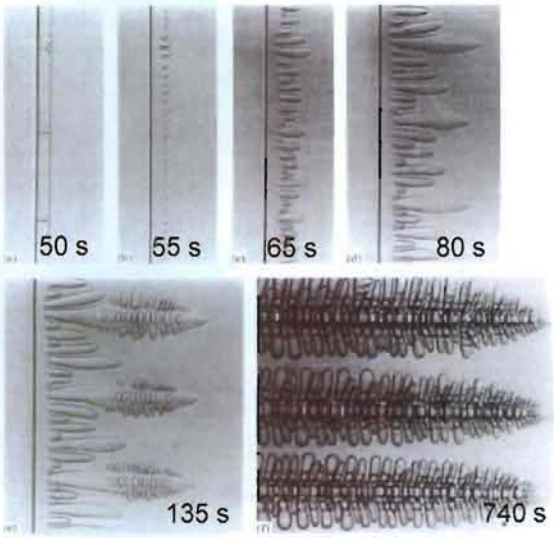


Scanning electron microscopy image showing the dendrite structure network in Ni-Ta-Cr-Mn after decanting (70 x)

Limited to the examination of coarse structures

J. Dejace, R. Matera, and G. Piatti, Journal of Materials Science, 1973, 8:754

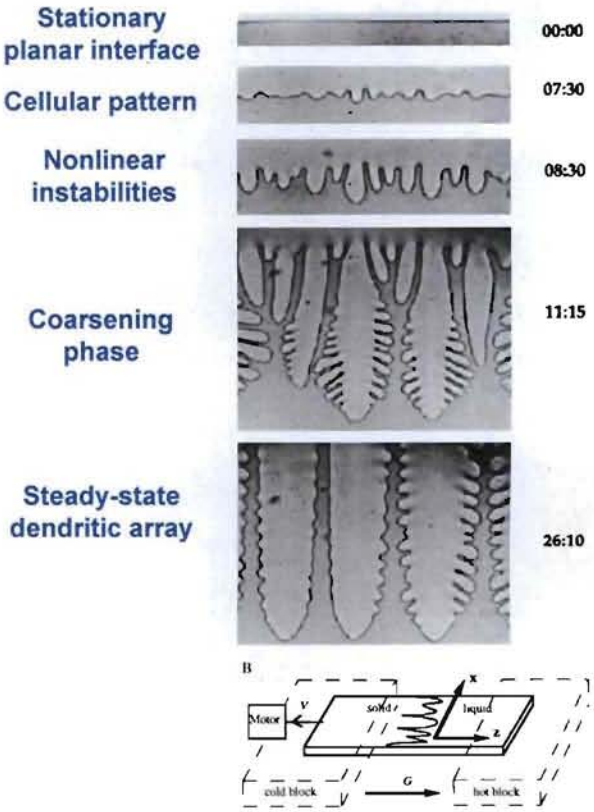
Transparent organic analogs of metallic systems



Development of solid-liquid interface shapes with change in growth rate from zero to 3.4 mm s^{-1} at a temperature gradient of 6.7 K mm^{-1}

Study pattern-forming instabilities and test theory; cannot study segregation

Adapted from R. Trivedi and K. Somboonsuk, Materials Science and Engineering, 1984, 65:65



Time evolution of the interface morphology in succinonitrile/rhodamine 6G at a constant pulling speed V (V=3.11 μm/s, G=2.8 K/cm , C_∞=0.325 wt.%)

Adapted from W. Losert, B.Q. Shi, and H.Z. Cummins, Proc Natl Acad Sci USA, 1998, 95:431

X-ray Radiography during Solidification and Tomography to Study Microstructure Evolution in Metal Alloys

Al-15.4at.%Cu

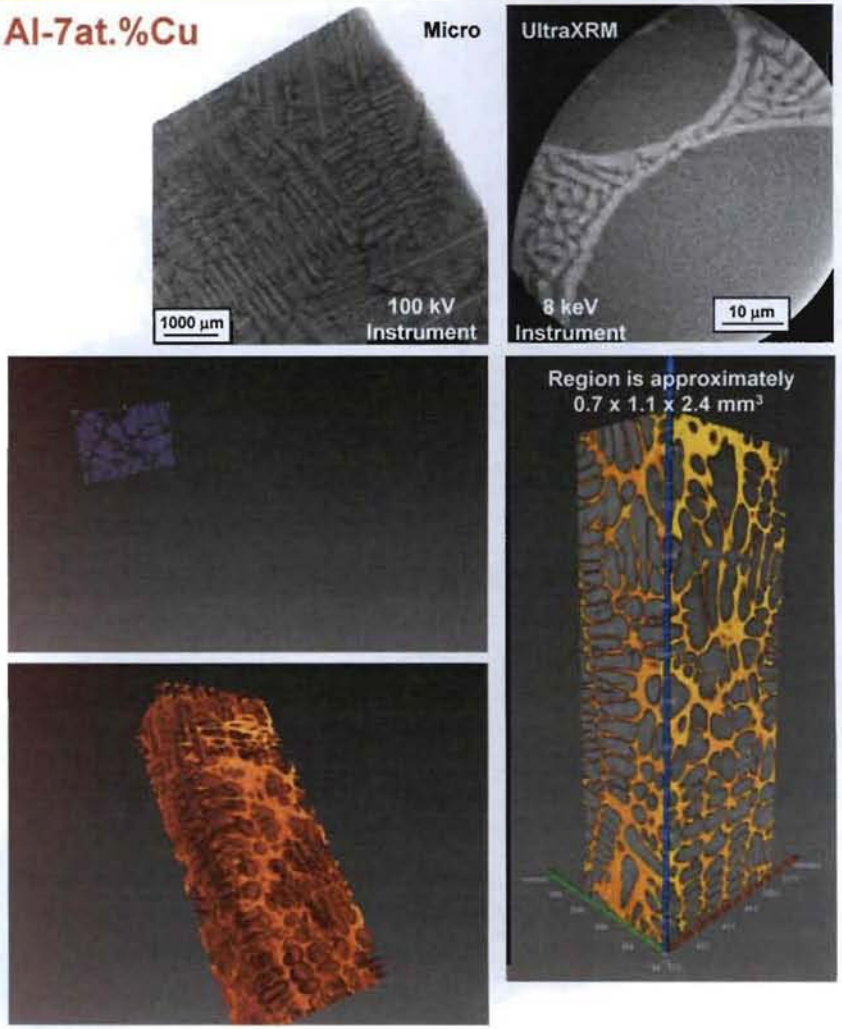
Time-resolved synchrotron x-ray radiography image after 35.55 s. The primary dendrite widths are approximately 200 μm, suggesting that artifacts, such as capillarity or surface nucleation, may be important in these 2D experiments.

~1.4 x 1.4 mm^2 field of view, ~200 μm thick foil

Highlights the importance of in-situ solidification studies for advancing our understanding of dendrite formation, growth and fragmentation, solid-liquid interface morphology, and solute segregation

R.H. Mathiesen and L. Arnberg, Acta Mater, 2005, 53:947-956

Al-7at.%Cu



3D imaging is needed to fully understand the microstructure evolution → future tomography experiments at APS are planned

A.J. Clarke, B.M. Patterson, J.C. Cooley, and T.J. Ott

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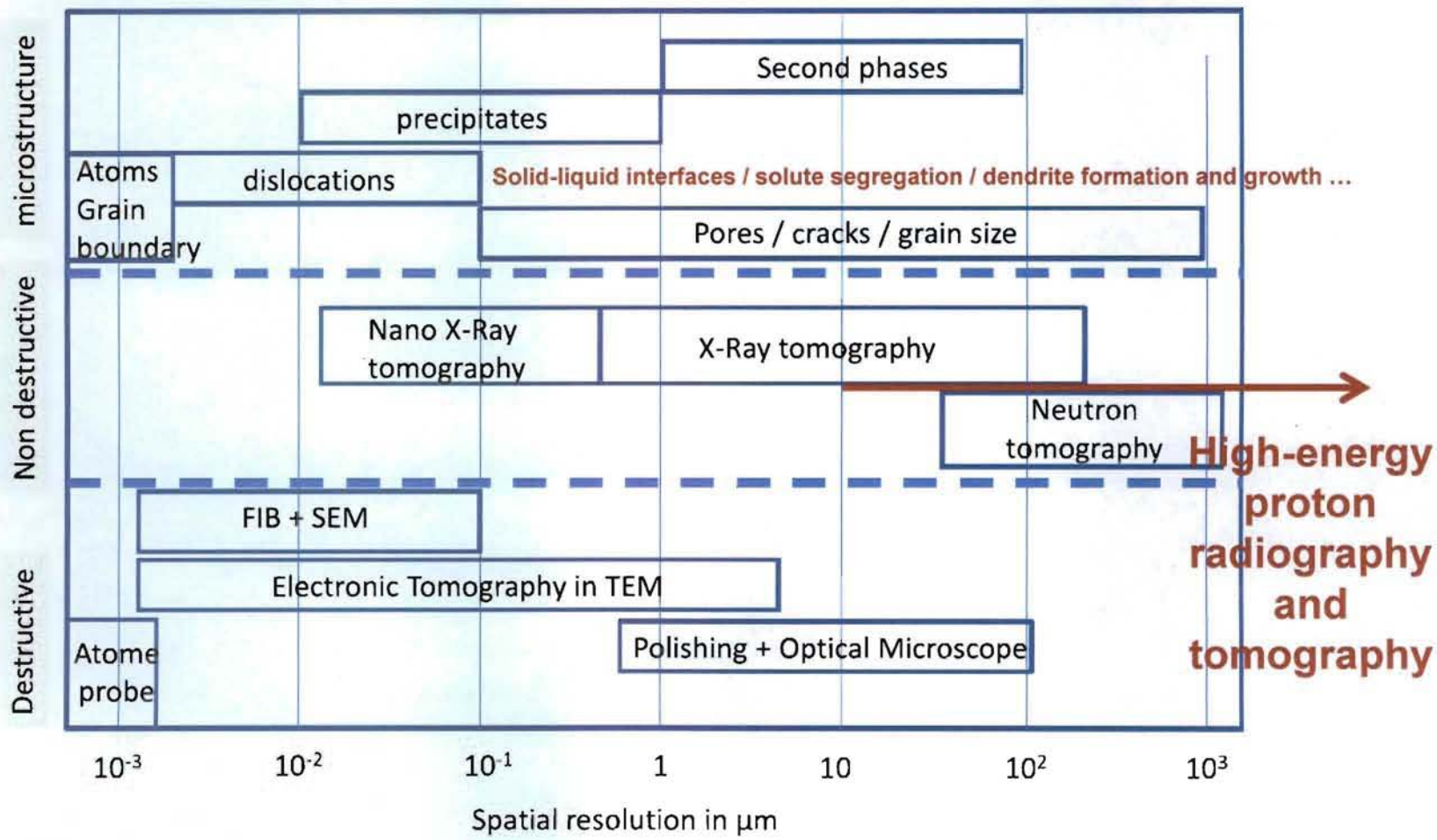
Example Tools for 3D Imaging in Materials - Opportunities for Proton Radiography



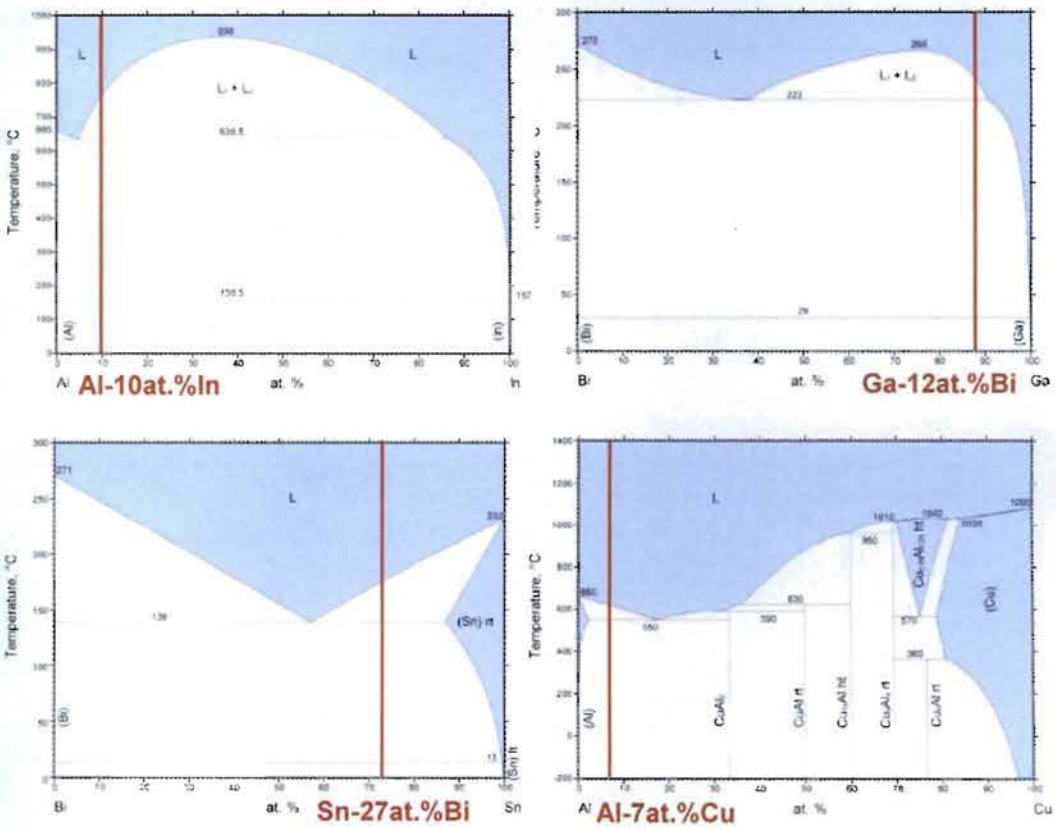
Adapted from L. Salvo, M. Suéry, A. Marmottant, N. Limodin, D. Bernard, C.R. Physique, 2010, 11:641

Advantages of Proton Radiography for Solidification Studies

- Time-resolved imaging to study dynamic processes (e.g., localized fluid flow and solute segregation) and microstructure evolution during solidification
- Large field of view (tens of millimeters) compared to that afforded by synchrotron x-ray radiography (typically less than 2 x 2 mm^2)
- Millimeter thick samples (relevant to casting) that allow for bulk material analysis
- Ability to examine high density materials
- Potential to perform tomography

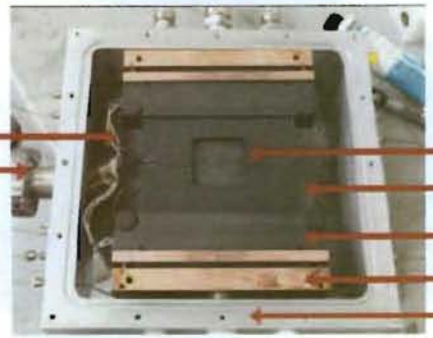
FY11 Laboratory Directed Research and Development - Goals

- Design and build a furnace for in-situ solidification experiments
- Perform the first in-situ melting and solidification experiments using 800 MeV Proton Radiography (pRad) at LANL (Al-In, Ga-Bi, Sn-Bi, Al-Cu)
  - Solid-liquid interfaces
  - Solute segregation
  - Alloy melt fluid flow
- Demonstrate the feasibility of using pRad to examine dynamic processes and microstructure evolution during solidification
- Examine a large field of view (~44 x 44 mm^2) and thick (2-6 mm) samples

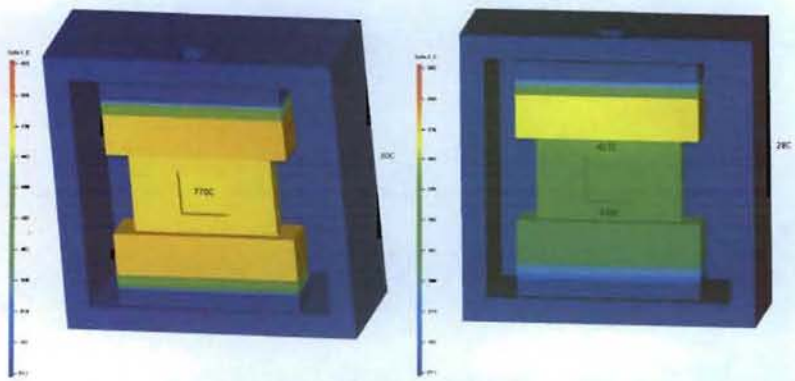


ASM Alloy Phase Diagrams Center, P. Villars, editor-in-chief; H. Okamoto and K. Cenzual, section editors; http://www1.asminternational.org/AsmEnterprise/APD, ASM International, Materials Park, OH, 2006

Experimental Approach



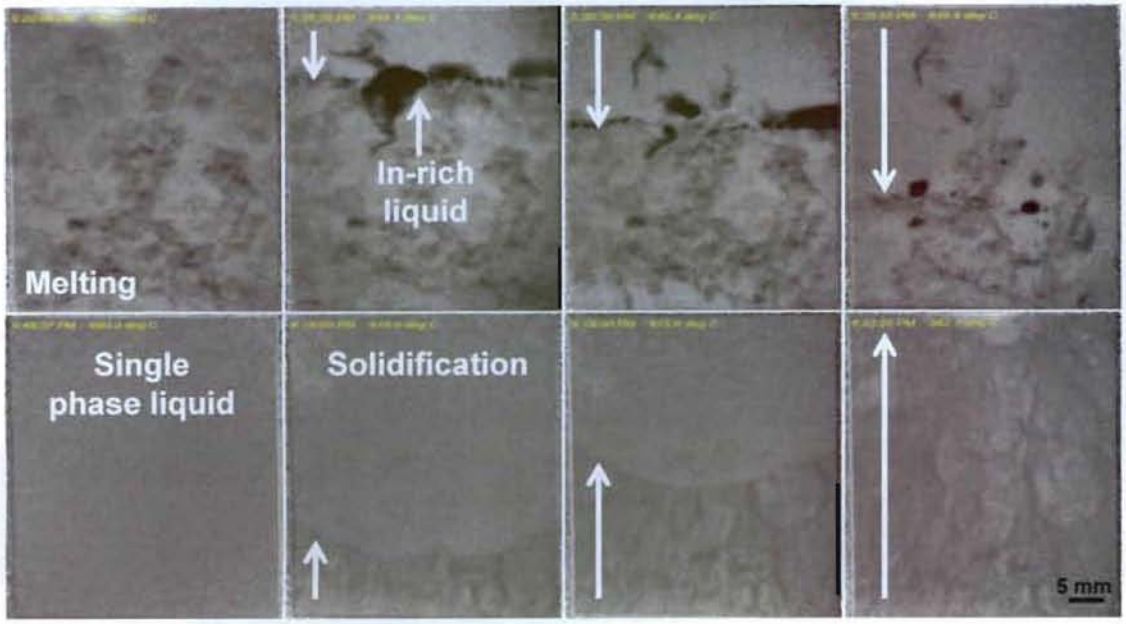
A furnace was designed and built for insertion into pRad (August, 2011). (Downstream view; the outer Al box is 305 mm x 305 mm (12 in x 12 in). The back Al plate is removed.)



Early furnace heat-up simulations were performed using TRUCHAS, a LANL developed casting simulation code (D.A. Korzekwa).

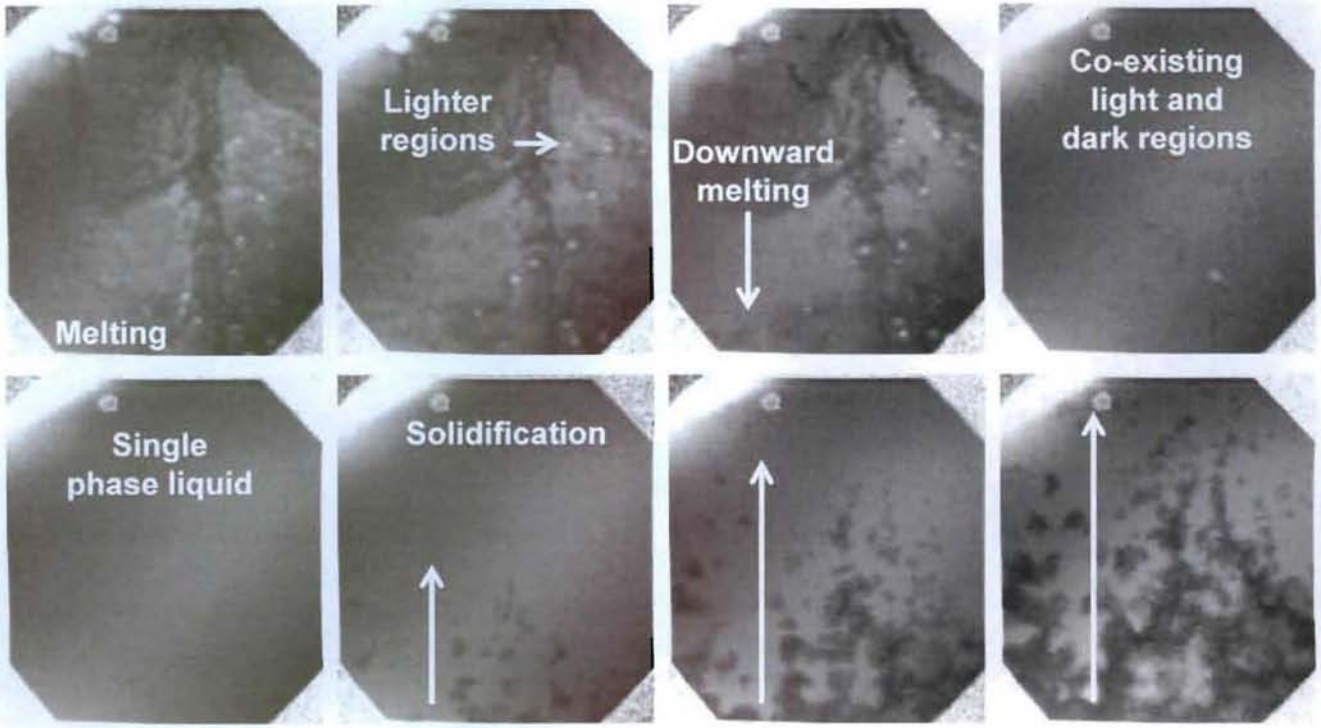
The X3 magnifier set-up at pRad was used with 65 μm resolution and a ~44 x 44 mm^2 field of view.

In-situ Monitoring of Alloy Melt Fluid Flow and Solidification Using pRad at LANL (August 2011); Al-In, 6 mm thick



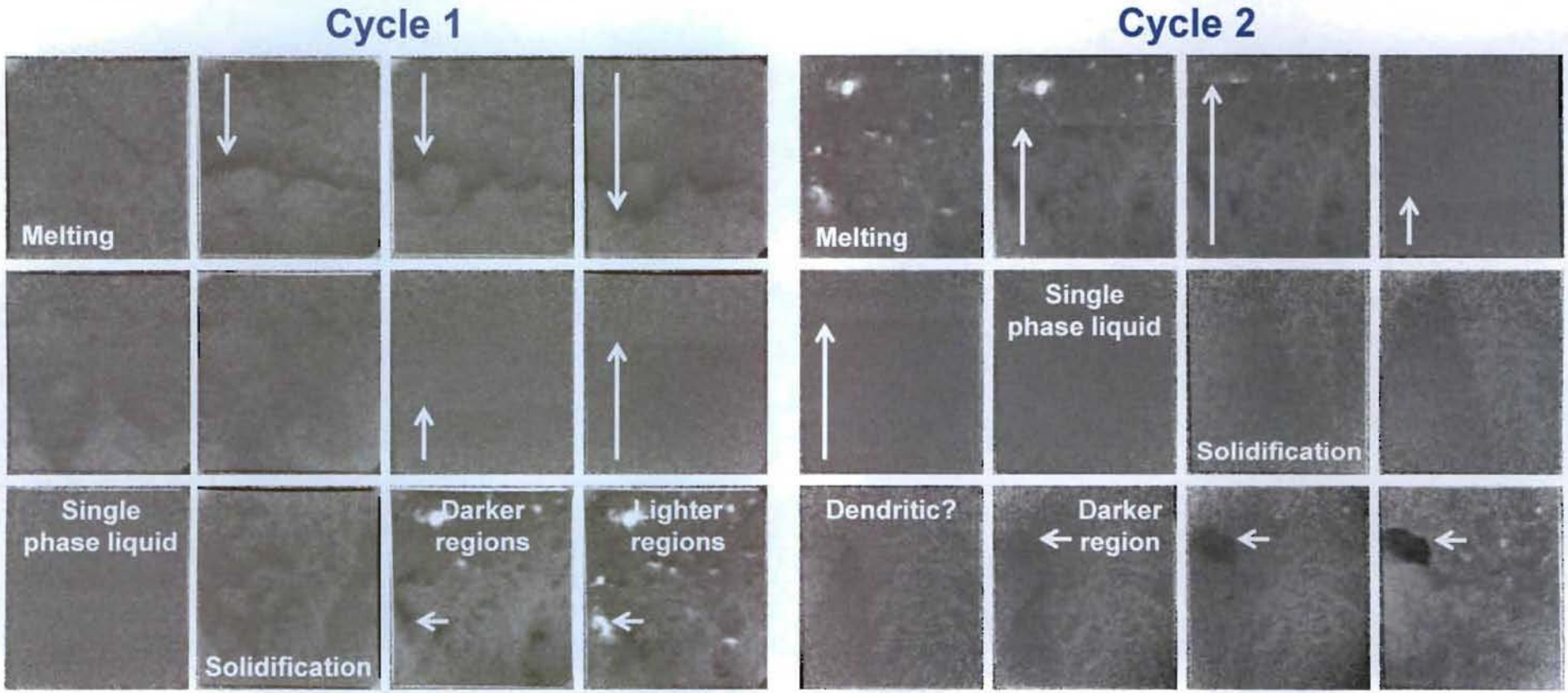
Evidence of liquid-liquid phase separation during melting…

In-situ Monitoring of Alloy Melt Fluid Flow and Solidification Using pRad at LANL (August 2011); Ga-Bi, 4 mm thick



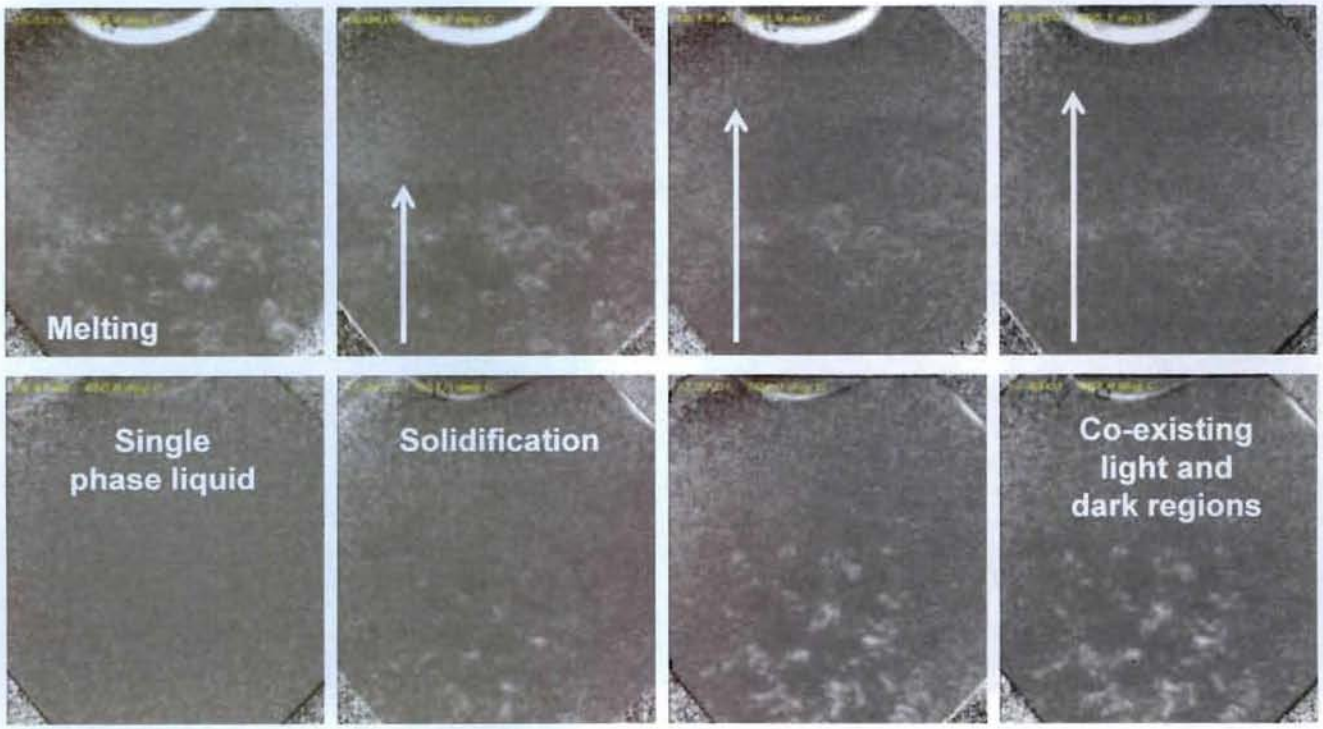
Movement of lighter regions and evidence of dendritic solidification…

In-situ Monitoring of Alloy Melting and Solidification Using pRad at LANL (August 2011); Sn-Bi, 2 mm thick



Repeated interface movement during melting and signatures of microstructure evolution during solidification…
Improved resolution is needed!

In-situ Monitoring of Alloy Melting and Solidification Using pRad at LANL (August 2011); Al-Cu, 2 mm thick



Interface movement during melting…
Improved temperature control is needed.
Improved resolution needed!

Summary

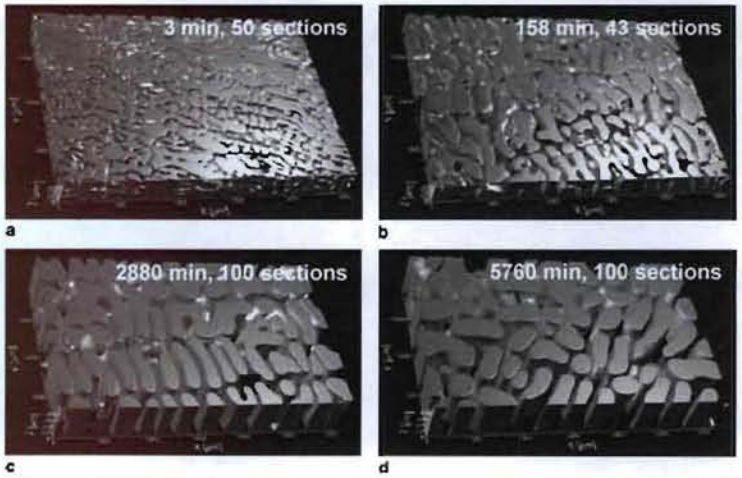
- A furnace for in-situ solidification experiments at pRad was designed and built
- The first in-situ melting and solidification experiments were performed at pRad in August 2011
  - Time-resoled images of dynamic processes and solid-liquid interfaces were obtained
  - Localized fluid flow was observed
  - Signatures of solute segregation and microstructure evolution were obtained, but improved resolution is needed
- A large field of view (~44 x 44 mm^2) and thick (2-6 mm) samples were examined, demonstrating the ability to examine high density alloys
- pRad is a promising tool for 3D in-situ solidification studies

Future Opportunities - Solidification

- FY12 LDRD funding for in-situ solidification experiments beyond these initial proof-of concept experiments:
  - Time-resolved imaging and tomography at Argonne National Laboratory's Advanced Photon Source (toward tomography at pRad)
  - Post-mortem examinations (e.g., serial sectioning and x-ray radiography at LANL) of FY11 pRad samples, demonstrating the importance of 3D information
  - Pairing of time/temperature information with the images from pRad

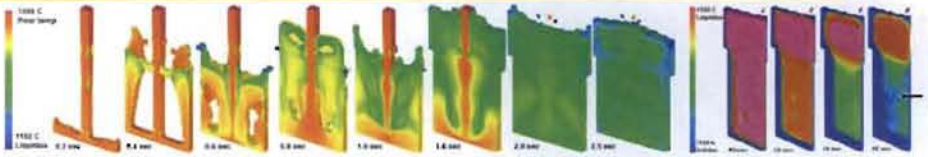
3D reconstructions from serial sectioning of Pb-Sn coarsened 2 °C above the eutectic

D. Kammer and P.W. Voorhees, Acta Materialia, 2006, 54:1549

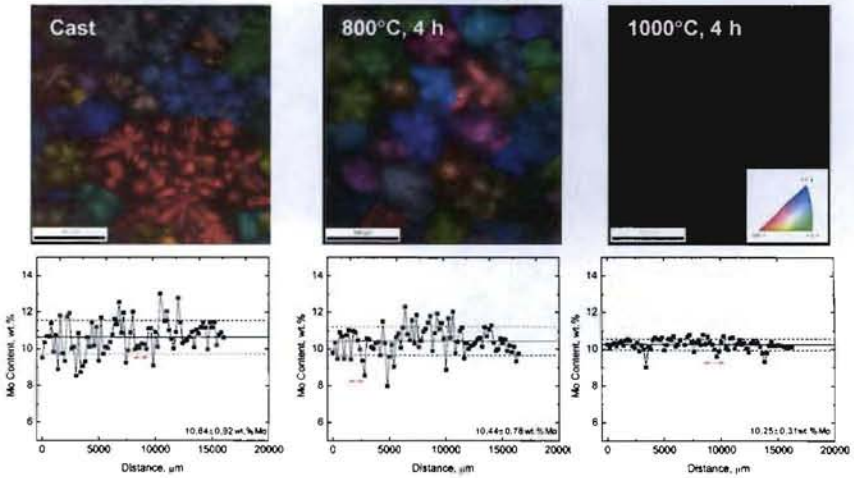


Additional Future Opportunities - Solidification

- Tomography at pRad
- Casting mold filling at pRad
- In-situ homogenization studies at pRad
- Application of pRad to study high density alloys
- Application of new experimental findings to validate and advance solidification theory and modeling (incorporation of solute segregation and microstructure evolution)
- Advancements in casting technology (LANL and industry relevance)

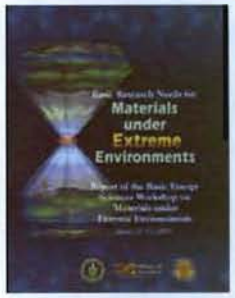


Casting filling and solidification sequences of a U-Mo alloy predicted using TRUCHAS (developed at LANL). The fluid flow can influence the quality of the casting, controlling factors such as macrosegregation and homogeneity, voids and shrinkage, and the as-cast microstructure. (D.R. Korzekwa)

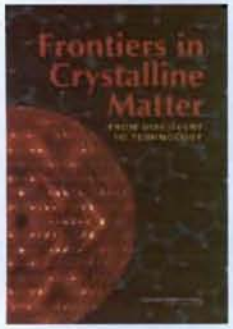


Electron backscatter diffraction (EBSD) results highlighting solute segregation in as-cast DU-10wt.%Mo. Monolithic low enriched uranium (LEU)-10wt.% Mo fuel is a potential replacement for highly enriched uranium (HEU) dispersion fuel in research and test reactors. (A.J. Clarke and P.A. Papin)

LDRD Project 20110512ER
- Relevance



Basic Research Needs for Materials under Extreme Environments; (2007 BES workshop report)
- Highlights the need to advantageously use extreme environments for materials design and synthesis, and for characterization on the scale of fundamental interactions

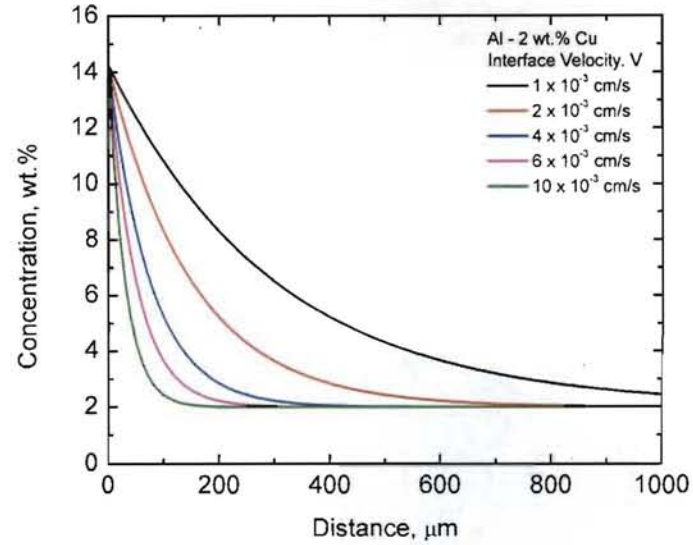


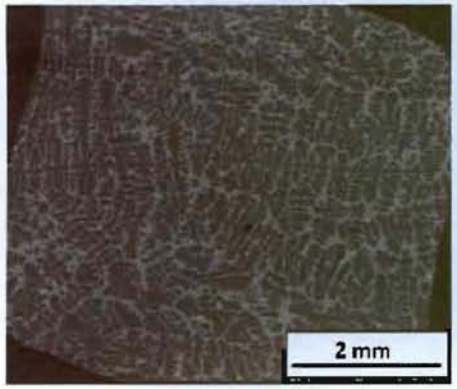
Frontiers in Crystalline Matter from Discovery to Technology; (2009 National Research Council of the National Academies report)
- Addresses the need to support the discovery and growth of crystalline materials within the United States (developing multiagency initiatives and centers of expertise, developing education and training opportunities, supporting collaborations, and promoting academic programs)

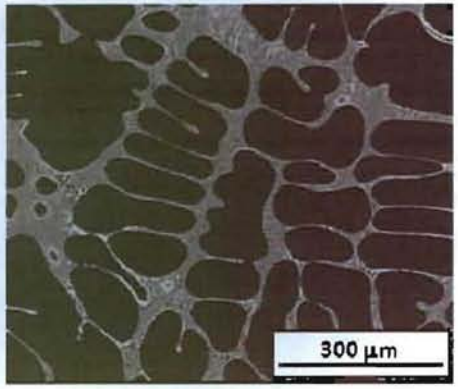
Decadal Challenges for Predicting and Controlling Materials Performance in Extremes; (2009 LANL MaRIE workshop report)
- Supports the need for a facility where *in-situ* characterization of materials during processing, specifically during casting, could be performed

LDRD Project 20110512ER
- Calculated Solute Segregation Curves in the Liquid and an Example Coarse Solidification Microstructure

Calculated copper concentration versus distance in the liquid of Al-2wt.% for a variety of interface growth rates. A steady-state diffusion field ahead of a moving planar interface at the solidus temperature was assumed. For these calculations, the initial alloy concentration C_0 is 2 wt.% Cu, the distribution coefficient k is 0.14, and the diffusion coefficient in liquid D_l is 3×10^{-9} m^2/s. The maximum composition at the interface is C_0/k, which decreases to C_0 away from the interface. Note that increasing growth rate results in a sharper gradient.







Al-15wt.%Cu (Al-7at.%Cu) microstructure consisting of dendrites (dark, branched structure) and interdendritic eutectic (two phase microconstituent) structure, *courtesy of Jason Cooley and Thomas Ott.*