In situ diagnostics and optimization of single crystal compound scintillator and semiconductor materials through energy-resolved neutron imaging

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Goals and Objectives

Optimize crystal growth process: fast development cycle, good quality and reproducibility

- Enable feedback control of crystal growth process in real-time, with quantification on composition homogeneity and dopant distribution. Prevent crystal cracking.
- Use in-situ diagnostics to validate model, observe outcomes, optimize new growth strategies
- In combination with computational models of heat transfer and fluid dynamics enable real-time control of crystal growth conditions
- Optimize:
  - ✓ Solid/liquid interface shape
  - ✓ Uniform growth velocity
  - ✓ Uniform temperature gradients at growth interface
  - ✓ More uniform composition (axial and/or radial)

Methods

- Energy resolved neutron imaging with quantification data analysis for absorption resonances and diffraction imaging.
- Finite-element computational models for high-temperature heat transfer and crystal growth.

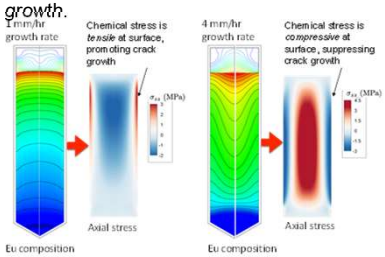

Fig. 1. Eu segregation from melt flow and solidification is predicted. High-temperature neutron diffraction parameters are used in calculations of chemical stress in crystal. The growth experiments are consistent with model predictions, namely cracking was more likely at slower growth rates.

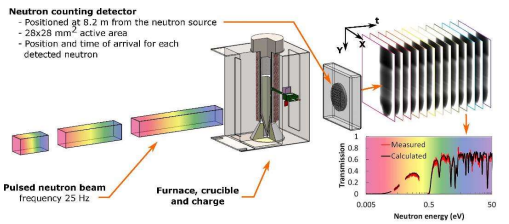

Fig. 2. Schematic diagram of the experimental setup. A pulsed neutron beam travels towards the neutron counting detector. Both position (~ 55 μm) and time (0.1-1 μs) are measured by the detector for each registered neutron. A furnace with a material charge is installed a few centimeters from the detector. A set of images, each corresponding to a particular neutron energy, is acquired in each experiment, spanning neutron energies from epithermal range (1 - 100 eV) to cold neutrons of meV energies. 262,144 spectra are acquired simultaneously (within each of the 512x512 pixels of 55x55 μm² area).

5-zone furnace used in all latest runs

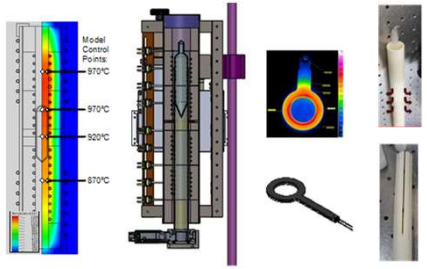

Fig. 3. (*A*) Schematic diagram of the 5-zone furnace. The sample can be translated vertically and rotated for tomographic imagining, if needed. The booster heater is used for the control of interface shape.

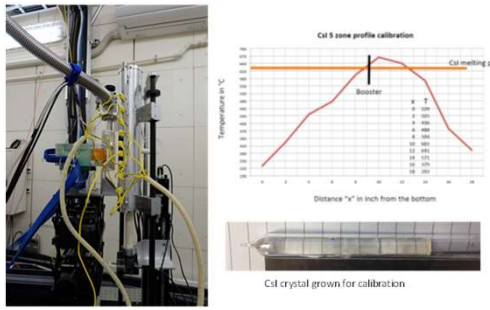

Fig. 4. (Left) Photograph of the furnace installed in a neutron beam. 10 thermocouples are used to measure and control the temperature profile within the furnace. Neutron counting detector is installed ~1 cm behind the furnace. (Right). Temperature profile calibration of the furnace. The booster location is indicated in the graph.

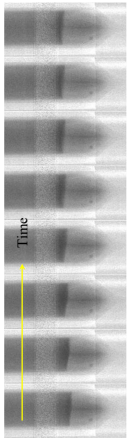

Fig. 5. Image sequence acquired during growth of CsI:0.5%Eu crystal used to test the booster heater idea for the control of liquid/solid interface shape. The interface is moved closer to the booster heater where it becomes convex, s predicted by the results of thermal simulations. Subsequent translation of the charge did not substantially move the location of interface relative to the booster position.

Control of interface shape: Convex interface by design

In-situ imaging enables the control of interface in 5-zone furnace.

During growth the temperature profile is optimized such that desired convex interface is maintained through the entire growth process, as observed through neutron imaging for certain materials. Recipes for new materials are being developed presently, including Tl_2LiYCl_6:Ce, Cs_2HfBr_6, $CsPbBr_3$ and others.

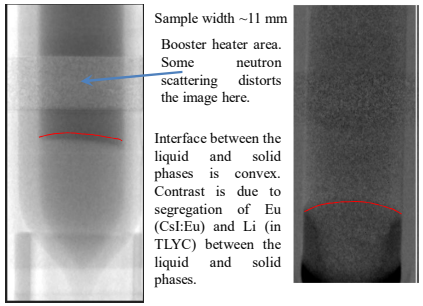

Fig. 6. Location and shape of liquid/solid interface accurately controlled in a 5-zone furnace for CsI:0.5%Eu (left) and Tl_2LiYCl_6:Ce (right) crystal growth.

Observation of crystal nucleation

The location of nucleation point is strongly affected by local heat extraction at the tip

The nucleation of crystal at the tip was observed in-situ, which was strongly affected by the temperature distribution within the tip. Careful design of temperature profile is required. Our imaging allows for a direct visualization of nucleation process on a macroscopic scale.

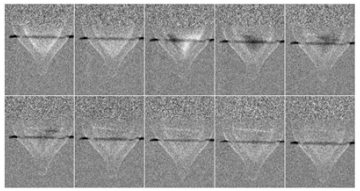

Fig. 7. Time sequence of images taken during the nucleation of CsI:0.5%Eu crystal at the tip of the ampule. The nucleation happens at the point of contact between the ampule and the holder, serving as a heat extractor in this case. The dark spot at the third image in the top row is where the crystal nucleation started.

Migration/diffusion of elements during crystal growth

The migration of certain elements both in the liquid and in the solid phases, as well as between them can be measured at high T

During growth of several materials the migration of Eu and Li within the solid phase was observed in-situ at high T for the first time.

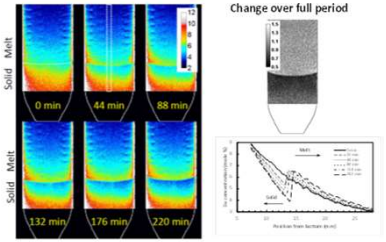

Fig. 8. Maps of Eu concentration measured during BaBrCl:5%Eu growth while the temperature was held constant. Migration of Eu within the solid phase and between the solid and liquid phases is observed quantitatively.

Conclusion

In-situ imaging can speed up introduction of new materials

- Accelerates introduction of new materials to industrial scale production.
- Interface shape can be optimized to remain convex during growth.
- Understanding of dynamics of solid-liquid interface can be used for the optimization of crystal growth in conjunction with simulations.
- Nucleation can be observed in-situ.
- Elemental mapping can be performed for elements, which have sufficiently large neutron attenuation cross sections at the resonance energies.

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