SAND2008-4954P

Office of Nonproliferation Research and Development

Mechanical Properties of $CeBr_3$ for Calibration of an Embedded-Ion Method Model

August 7, 2008

P. G. Allison
Mississippi State University

MECHANICAL ENGINEERING
BAGLEY
COLLEGE OF ENGINEERING
MISSISSIPPI STATE UNIVERSITY

Sandia National Laboratories

Advanced Inorganic Scintillators

Acknowledgements

This project is sponsored by DOE Office of Nonproliferation, NA22 Robert Mayo, Advanced Materials portfolio manager

Intern support thanks to Enabling & Predictive Simulation Research Institute (ESPRI), Jon Zimmerman

Sandia National Labs

F. P. Doty, PI
Xiaowang Zhou, modeling lead
Pin Yang, Thermo-mechanical properties
Paul Allison, MSU graduate student intern

Kansas State University

Prof. Doug McGregor
Mark Harrison, alloy crystal growth and characterization

Advanced Inorganic Scintillators

Project overview

- Determine cause of mechanical failure in LnX_3 scintillators;
- Develop practical strategies to scale detectors to large sizes

Goals:
Mitigate material limitations; Improve single crystal growth methods

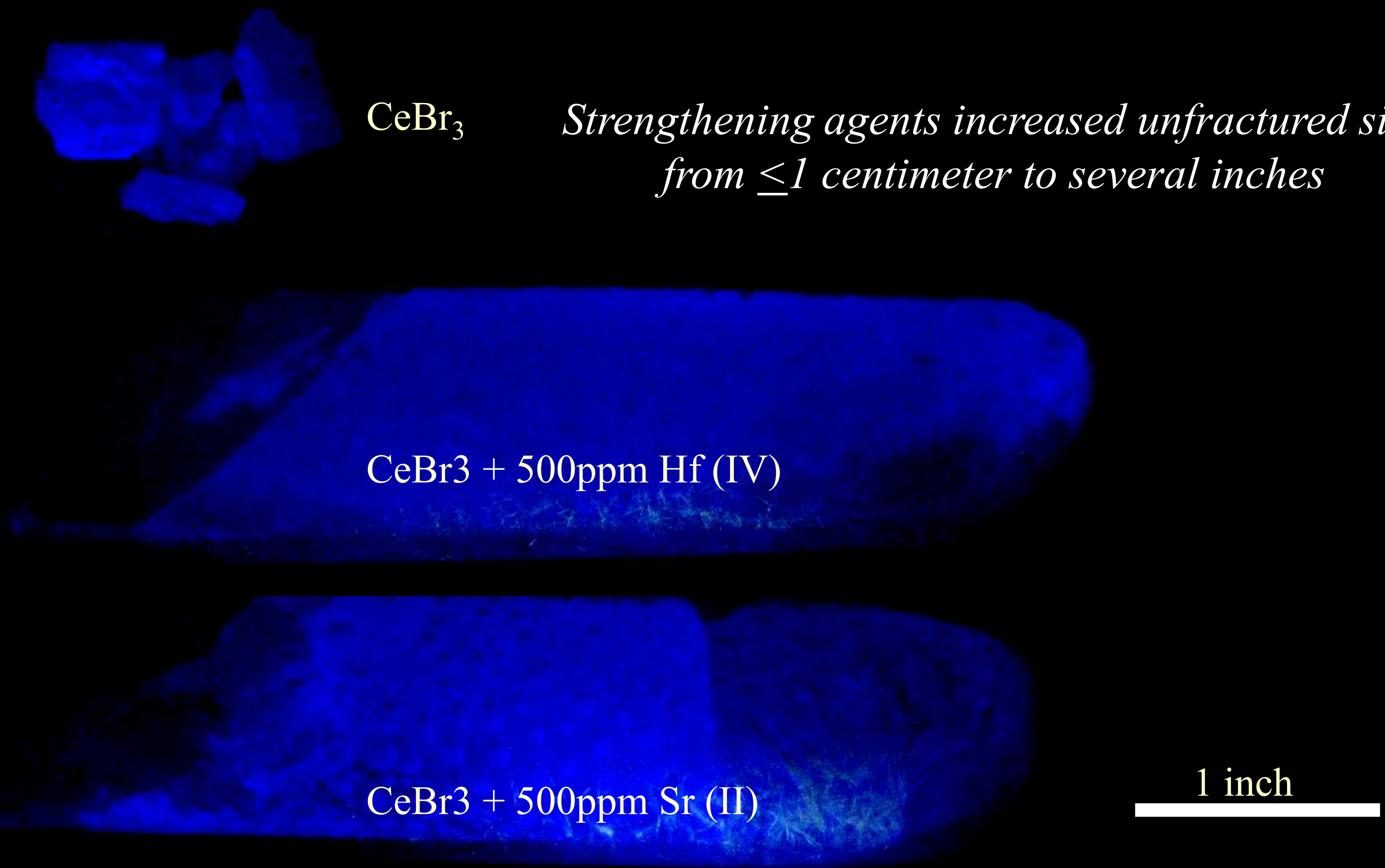
Technical approach:
1. *Measure thermal and mechanical properties*
2. *Model deformation and fracture mechanisms*
3. *Investigate alloy strengthening to improve fracture toughness*
4. *Develop seeded Bridgman growth to reduce thermal stresses*

Deliverables:
Improved alloys, growth methods, and predictive model for elasticity and fracture of lanthanide halide scintillator materials

Capability improvement addressed:
Large, low cost gamma spectrometers for detection and monitoring of SNM

Alloys enable successful HGF crystals

UV luminescence images



Strengthening agents increased unfractured size from ≤1 centimeter to several inches

MECHANICAL

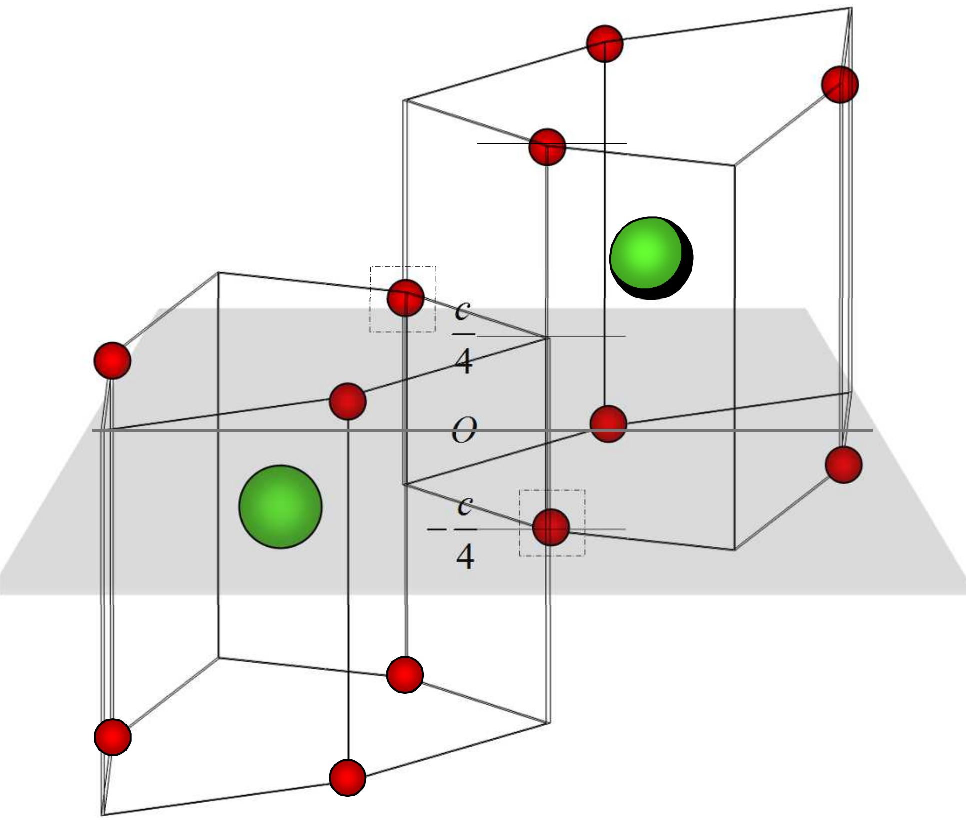
BAGLEY COLLEGE OF ENGINEERING MISSISSIPPI STATE UNIVERSITY

Sandia National Laboratories

Advanced Inorganic Scintillators

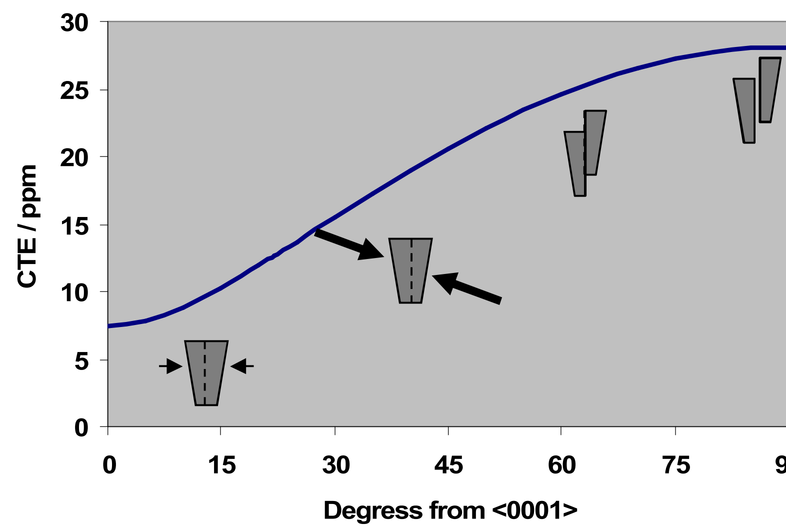
Model Development: Motivation

Limited and anisotropic slip result in formation of <u>critical flaws</u> on cleavage planes



Large anisotropy of thermal expansion results in <u>shear forces</u> on cleavage planes if gradient is not aligned with c-axis

- Pure compression for c-axis T gradient
- Both magnitude and orientation of shears increase rapidly for off-axis



Advanced Inorganic Scintillators

National Nuclear Security Administration

Model Development: Limitations of current approaches

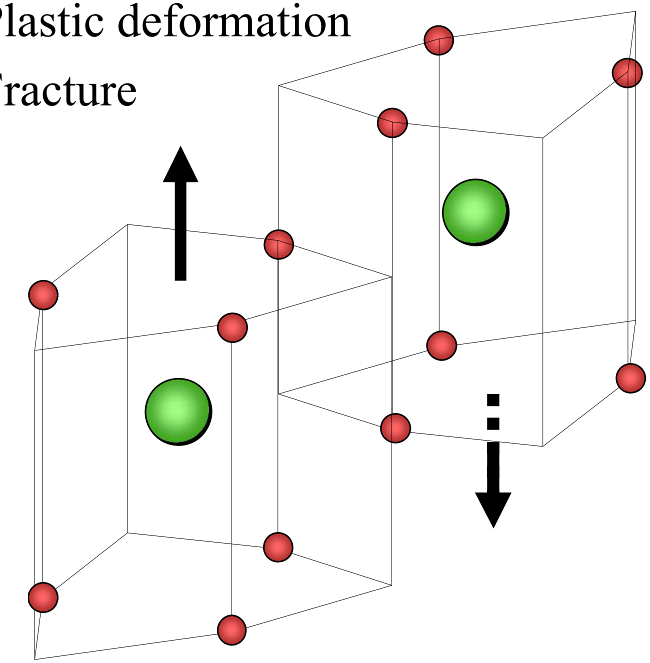
Problem: Develop modeling approach to simulate mechanics of LnX_3 crystals

•Thermal stress during growth

•Slip systems and dislocation reactions

•Plastic deformation

•Fracture



Slip and Fracture in LnX_3

1st generation ionic potentials: Fixed Charge
- Overestimates cohesive energy by factor of four!
- Stoichiometric compositions ONLY
- Can not treat defects

2nd generation: Variable Charge
- Charges solved by minimizing potential for each δt
- *Computationally VERY expensive*.
- Energy is not rigorously conserved!

3rd generation: This Work
The embedded ion method that we are developing solves these problems

•Equilibrium charges are analytically integrated:

•No energy minimization is required

•Energy is rigorously conserved

100 ns time scale, 1,000,000 atoms

.

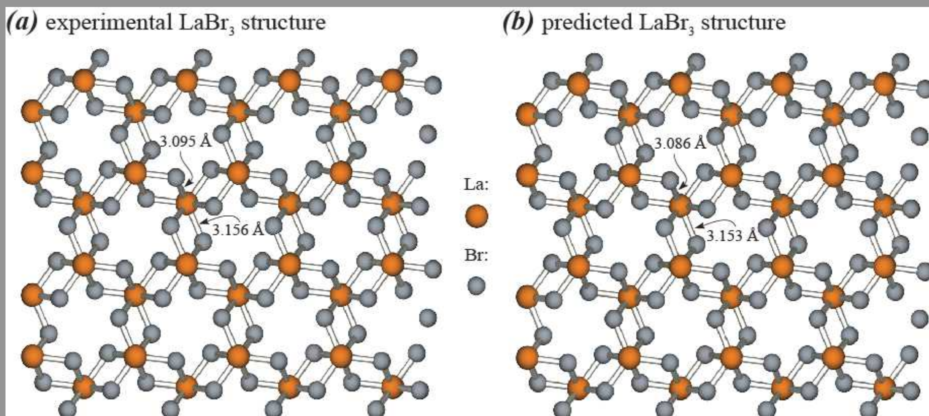
MECHANICAL ENGINEERING
BAGLEY
COLLEGE OF ENGINEERING
MISSISSIPPI STATE UNIVERSITY

Sandia National Laboratories

Advanced Inorganic Scintillators

Model Development: Molecular Dynamics Simulations

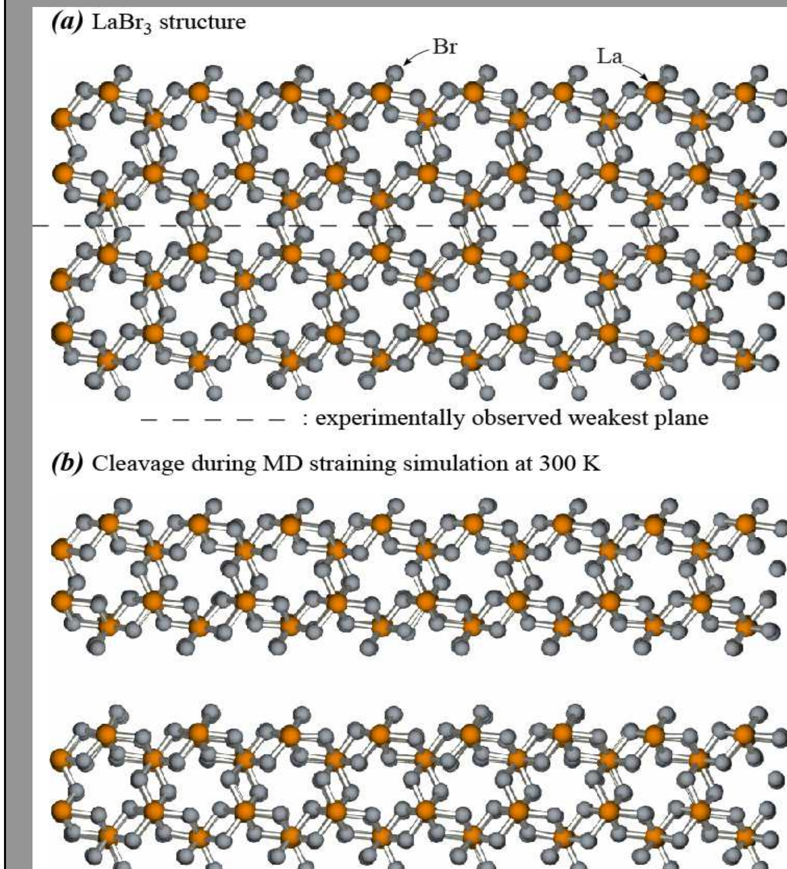
Bond Length Prediction



$LaBr_3$ Property Prediction

lattice constants (Å)				cohesive energy (eV/atom)		thermal expansion coefficient ($\times 10^{-8}$)			
a		c				a-axis		c-axis	
cal.	exp.	cal.	exp.	cal.	exp.	cal.	exp.	cal.	exp.
8.0	8.0	4.5	4.5	-4.3	-4.3	750	2800	860	800

Cleavage Prediction



Advanced Inorganic Scintillators

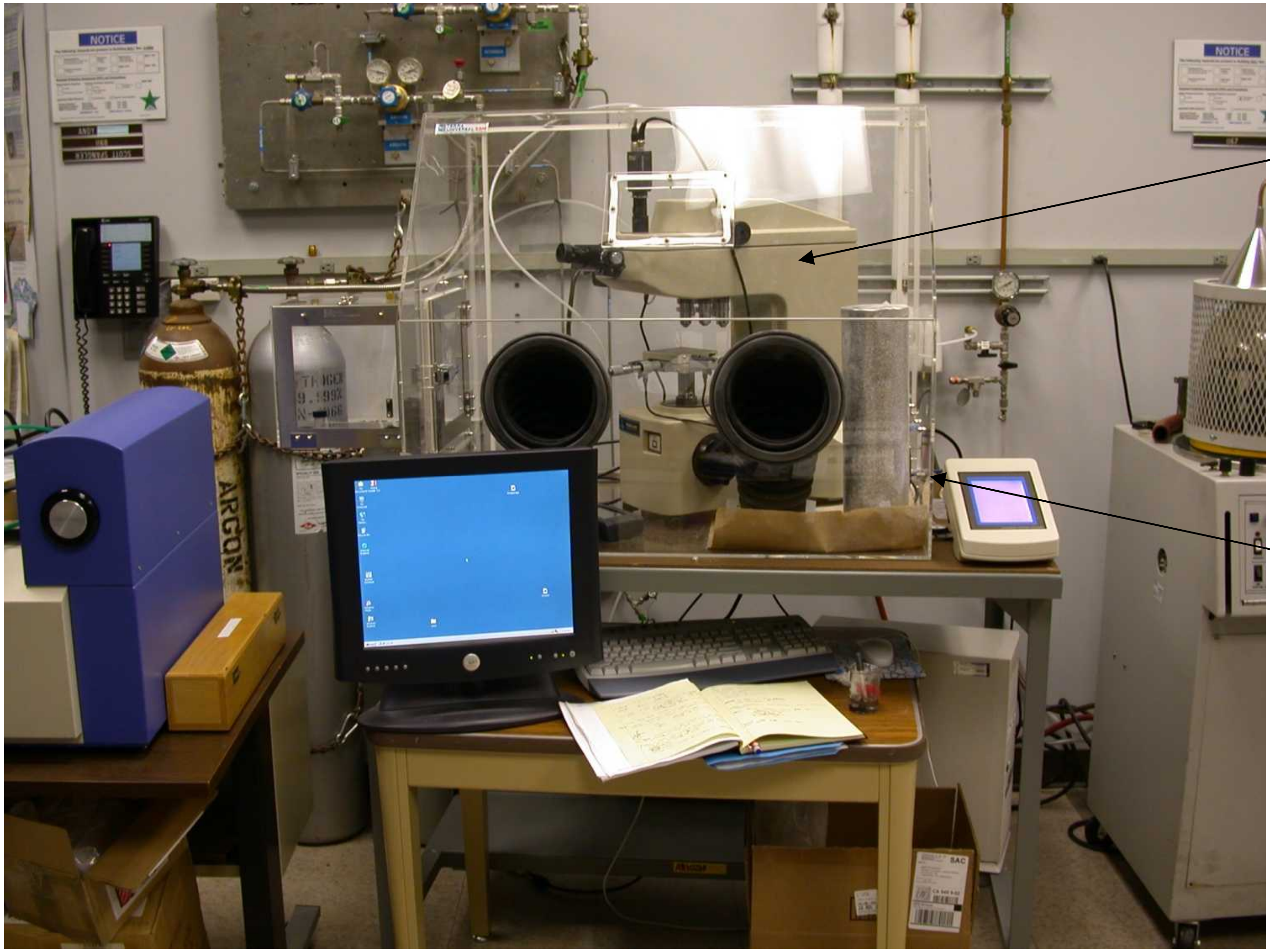
Summer overview

- **Calibration of embedded-ion method (EIM) model with mechanical property data**

I. Set-up a Vickers hardness tester with a moisture free environment
II. Validate indenter set-up and specimen preparation procedure
III. Analyze alloy strengthening with hardness and fracture toughness data
IV. Determine if non-destructive testing method is applicable for lanthanide halide alloys

Advanced Inorganic Scintillators

I. Vickers micro-indentation test set-up in controlled atmosphere

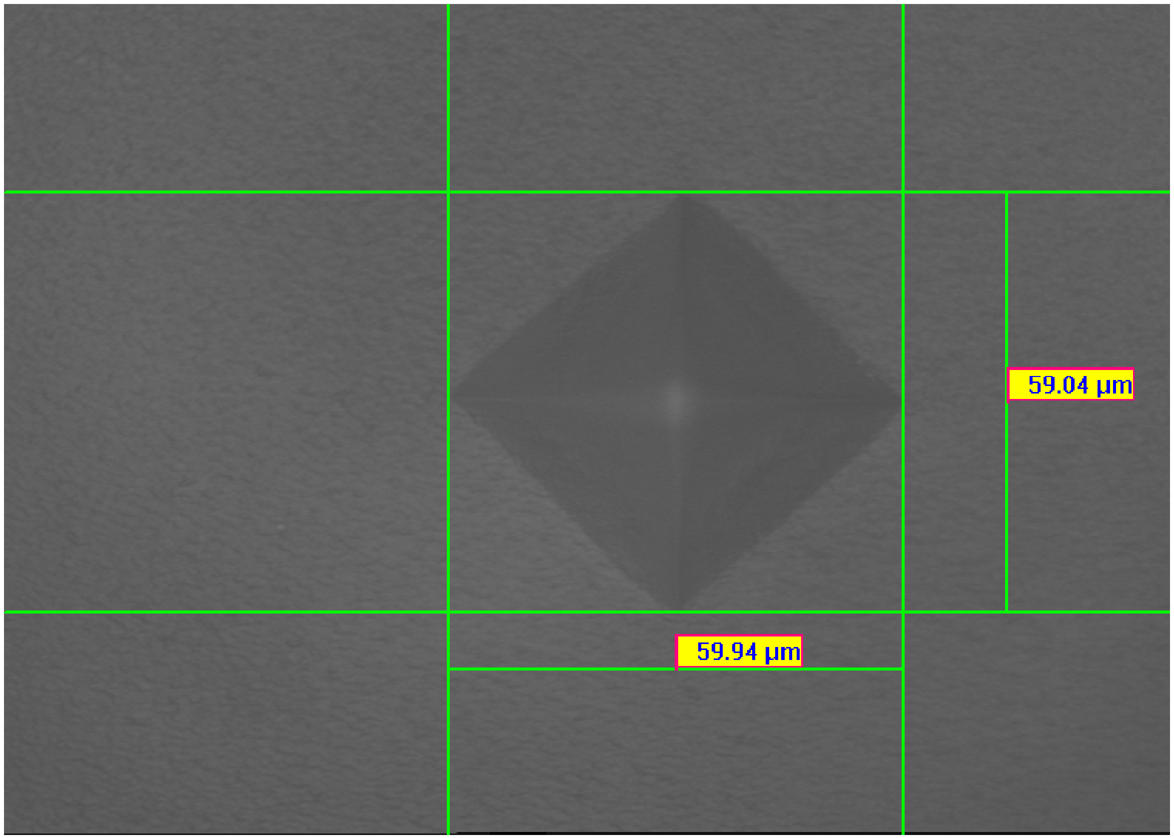


Vickers hardness tester

Dry nitrogen atmosphere filled glove box

Advanced Inorganic Scintillators

II. Validation of Vickers micro-indentation set-up and specimen preparation procedure



Lithium Fluoride hardness data cleaved {001} faces	Lithium Fluoride hardness data polished with 0.5 µm silica
HV0.2/20 (GPa) **Roberts and Page, 1986**	**HV0.2/20 (GPa)** **15 indents**
1.10 ± 0.04	**1.04 ± 0.01**

$HV = 0.0018544(P/d^2)$

P = load, N

d = average length of the two diagonals of the indentation, mm

Advanced Inorganic Scintillators

IV. Use of NDT methods

Use longitudinal (V_L) and shear (V_T) wave velocities for determination of:

a) Poisson's ratio
b) Elastic modulus
c) Shear modulus

$$Velocity = \frac{Thickness}{\frac{1}{2}(TOF)}$$

$$Poisson's\ Ratio,\ \mu = \frac{1-2\left(\frac{V_T}{V_L}\right)^2}{2-2\left(\frac{V_T}{V_L}\right)^2}$$

$$Modulus\ of\ Elasticity,\ E = \frac{V_L^2\rho(1-\mu)(1-2\mu)}{(1-\mu)}$$

$$Shear\ Modulus,\ G = V_T^2\rho$$

Advanced Inorganic Scintillators

IV. Use of NDT methods

Property values determined for $CeBr_3$ strengthened with Cd

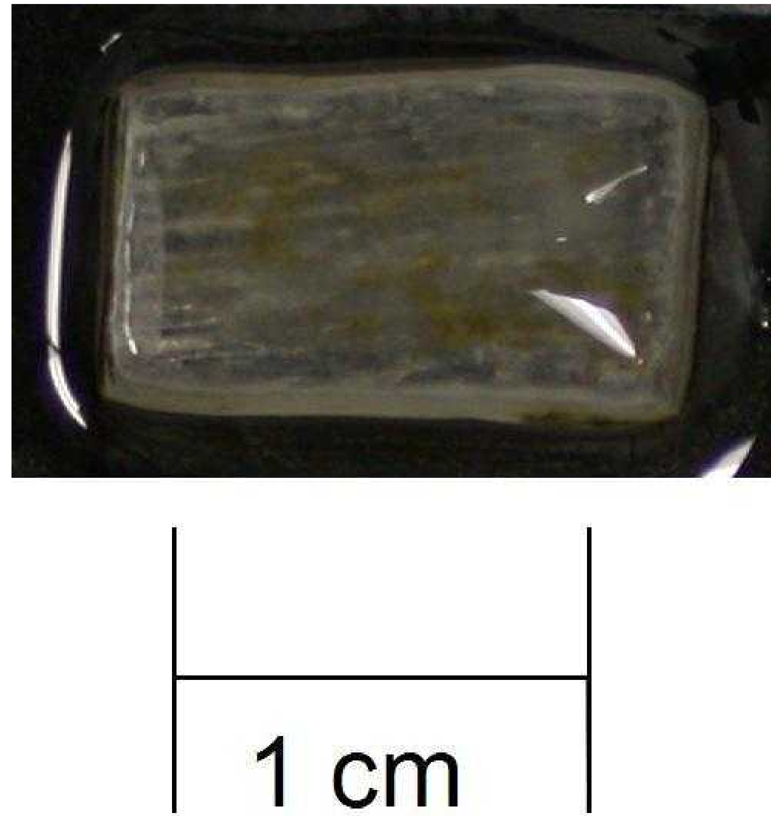
Longitudinal velocity, $V_L = 3.655 \times 10^5$ cm/s

Transverse velocity, $V_T = 2.851 \times 10^5$ cm/s

Poisson's ratio = -0.276

Elastic Modulus = 54 GPa

Bulk Modulus = 37 GPa



EIM Model Calibration

Micro-indentation

- Set-up Vickers micro-indenter with a controlled atmosphere
- Validated set-up and specimen prep procedure with LiF
- Expanded property database of lathanide halide alloys with additional hardness and fracture toughness data

Ultrasonic NDT

- Examined possibility of novel technique for lanthanide halide alloys
- Investigated appropriate coupling agents and transducers for determination of longitudinal and shear wave velocities.
- Begun NDT determination of mechanical properties (Poisson's ratio, elastic modulus, and bulk modulus) of strengthened lanthanide halide materials