INL/CON-22-69200-Revision-0

A laser ultrasonics-based approach for rapid screening of high entropy alloys

September 2022

Changing the World's Energy Future

Amey Rajendra Khanolkar, Subhashish Meher, David H Hurley

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Idaho Falls, Idaho 83415

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September 14th, 2022

Amey Khanolkar
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Energy & Environment Science & Technology Directorate

A laser ultrasonics-based approach for rapid screening of high entropy alloys

Amey Khanolkar[1], Subhashish Meher[2] & David Hurley[1]

[1]Condensed Matter & Materials Physics Group
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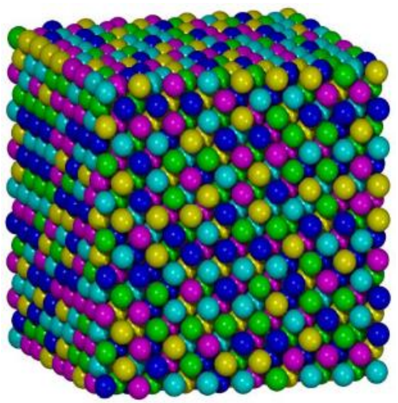
Sixth International Workshop on Structural Materials for Innovative Nuclear Systems (SMINS-6)
September 12-15, 2022, Idaho Falls, ID

High Entropy Alloys: Structural Materials for Extreme Environments

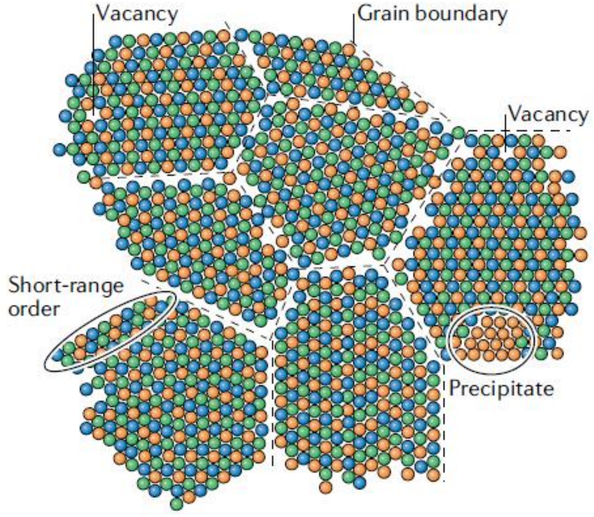
- Contain five or more elements in near-equiatomic proportions
- Presence of 4-5 principal elements can increase the configurational entropy of mixing sufficiently to overcome enthalpies of compound formation
- Possible to stabilize solid solutions at the expense of intermetallics that can be brittle and hinder mechanical performance
- Unconventional compositions and chemical structures → unprecedented combinations of mechanical properties

Atomic structure model of fcc CoCrFeMnNi

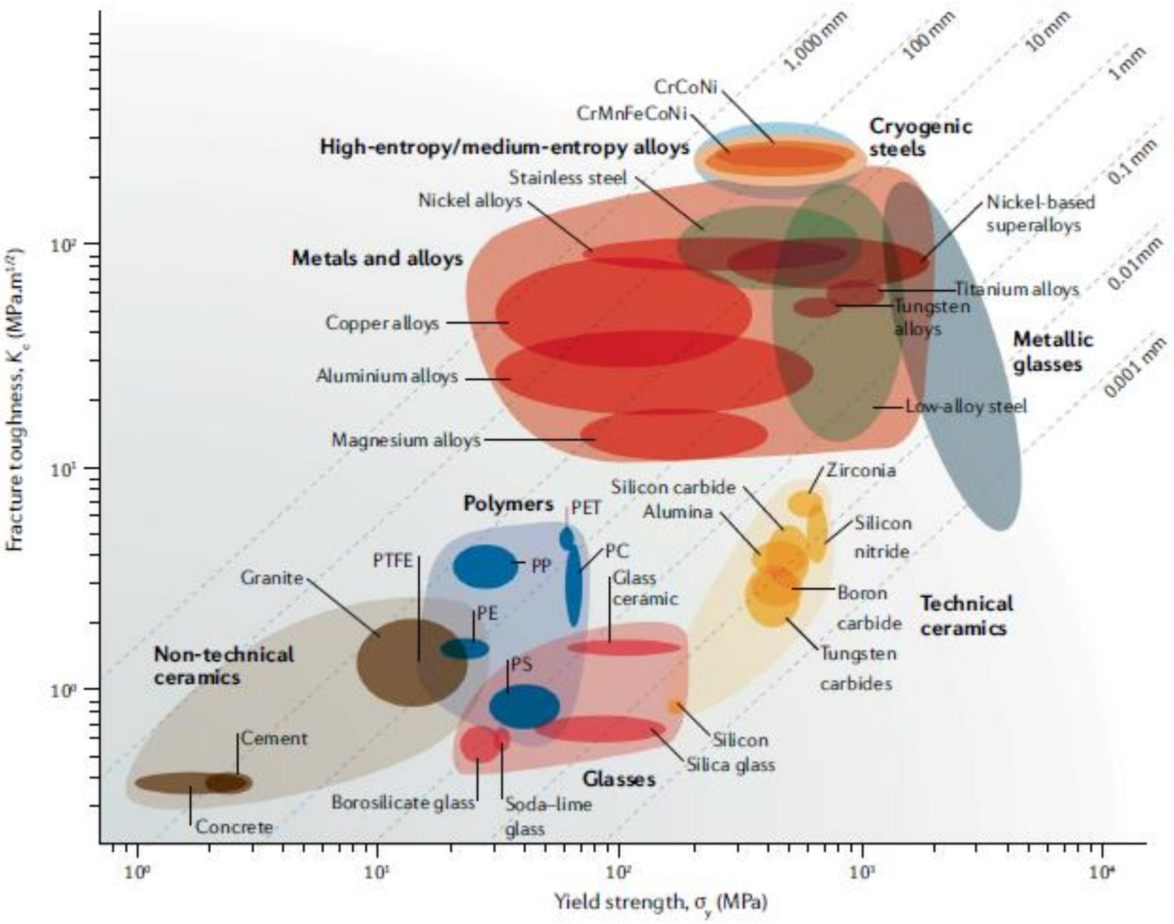


S. Wang, Entropy, 15(12), 5536-5548 (2013).

Tailored microstructure in HEAs



E.P. George, D. Raabe, & R.O. Ritchie, Nature Reviews Materials, 4(8), 515-534 (2019).



E.P. George, D. Raabe, & R.O. Ritchie, Nature Reviews Materials, 4(8), 515-534 (2019).

Research & Development Needs for High Entropy Alloys

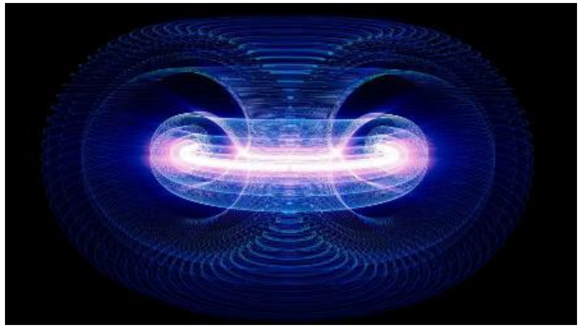
Basic Science

- Bridge the gap between the relatively well understood dilute solid solutions and the poorly understood concentrated solid solutions.
- Solid solution hardening, short-range atomic order and phase stability, dislocation mobility.
- **Microstructure-property relationships** – oxidation behavior, irradiation & corrosion resistance.

Technological Applications

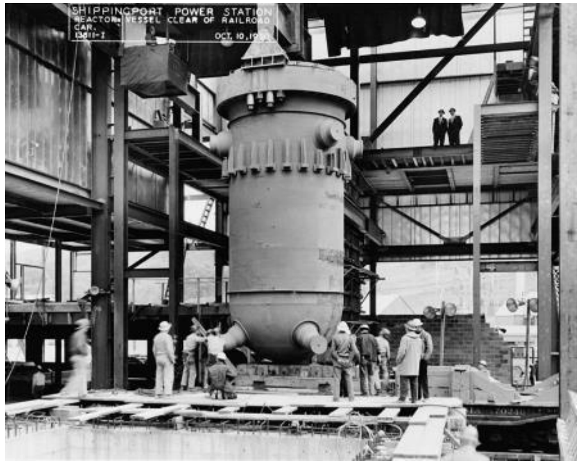
- High temperature structural materials:
  - Transportation industry & land-based gas turbine power generation
  - Energy generation systems (nuclear, fossil, concentrated solar power plants)
- Irradiation resistant materials:
  - Components in advanced fission & fusion reactors
- Corrosion resistant materials
- Functional materials
  - Catalytic materials for hydrogen generation
  - Thermoelectric materials
  - Cryogenic steels

Fusion reactor structural materials



American Nuclear Society

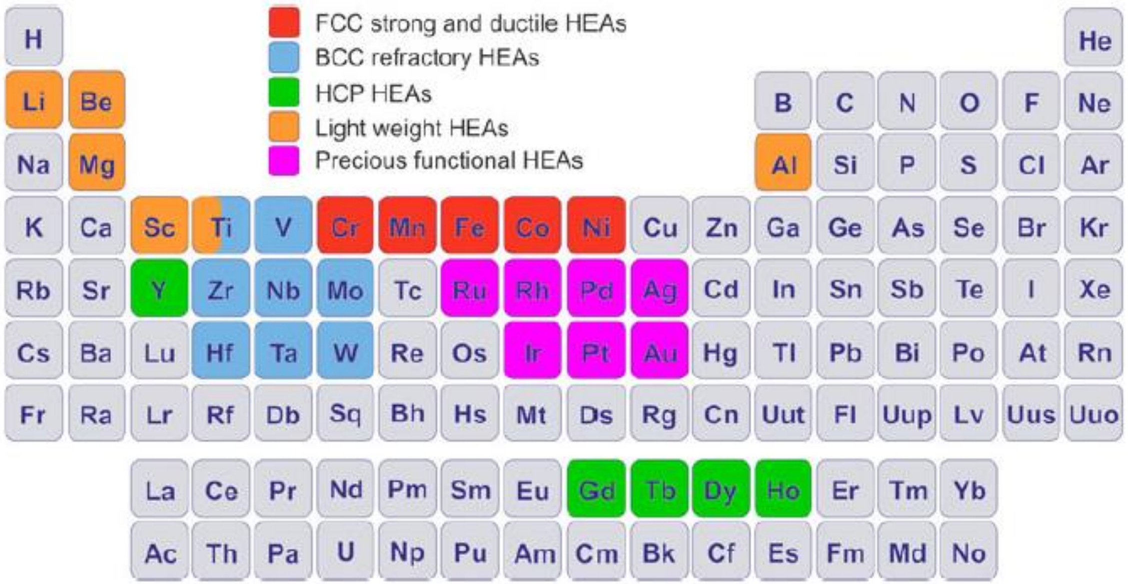
Fission reactor structural materials



U.S. Department of Energy, Naval Reactors Program

The Challenge with High Entropy Alloy – A Vast Compositional Space

Design Space with Principal Element Bases



Z. Li, A. Ludwig, A. Savan, H. Springer, & D. Raabe, Journal of Materials Research, 33(19), 3156-3169 (2018).

- Miracle (2019) estimated ~592 billion new HEA bases with 3-6 principal elements
- Prohibitively expensive & impractical to study using conventional methods

D. Miracle, Nat. Comm., 10: 1805 (2019)

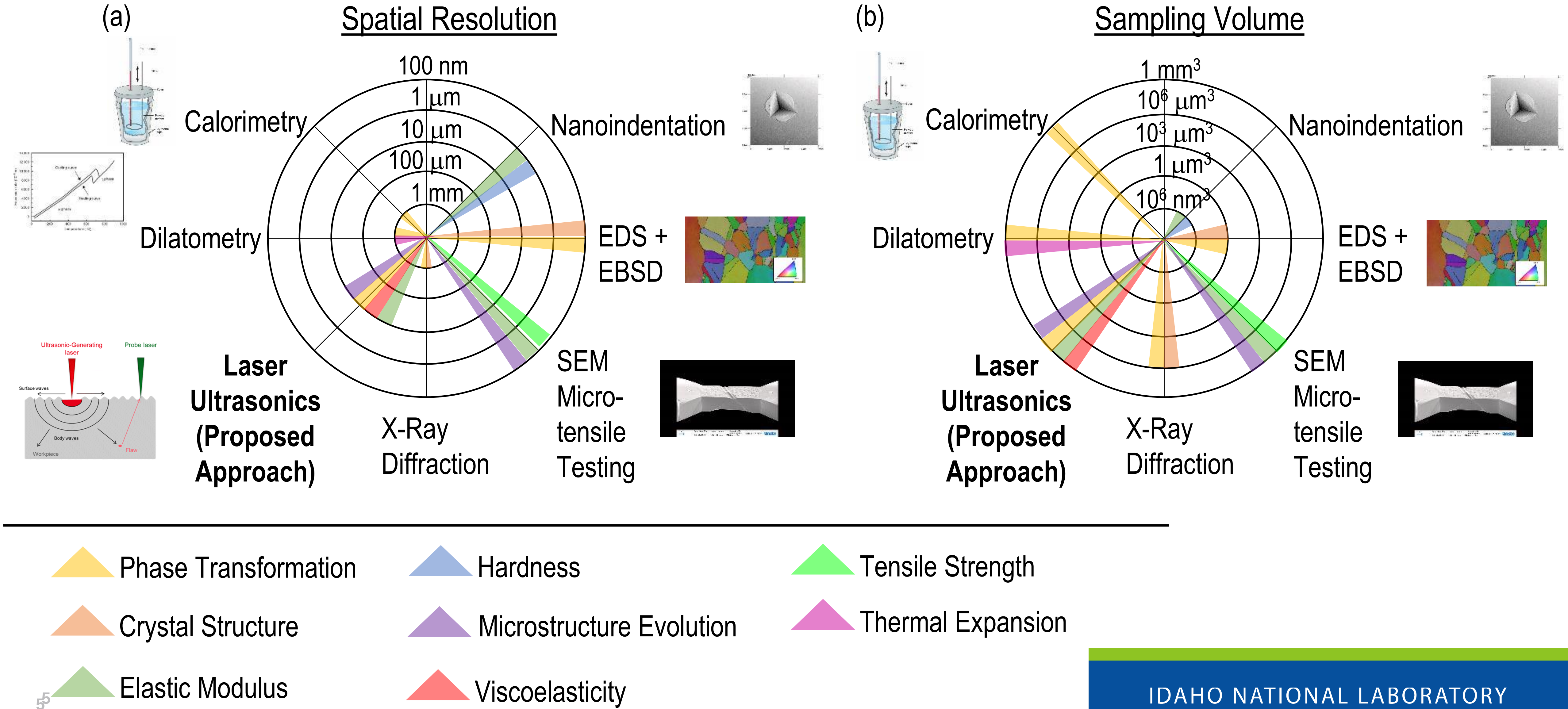
The needle in the haystack problem!

Along with high-fidelity models, there is a need for high-throughput, rapid characterization techniques for screening potential high entropy alloy compositions

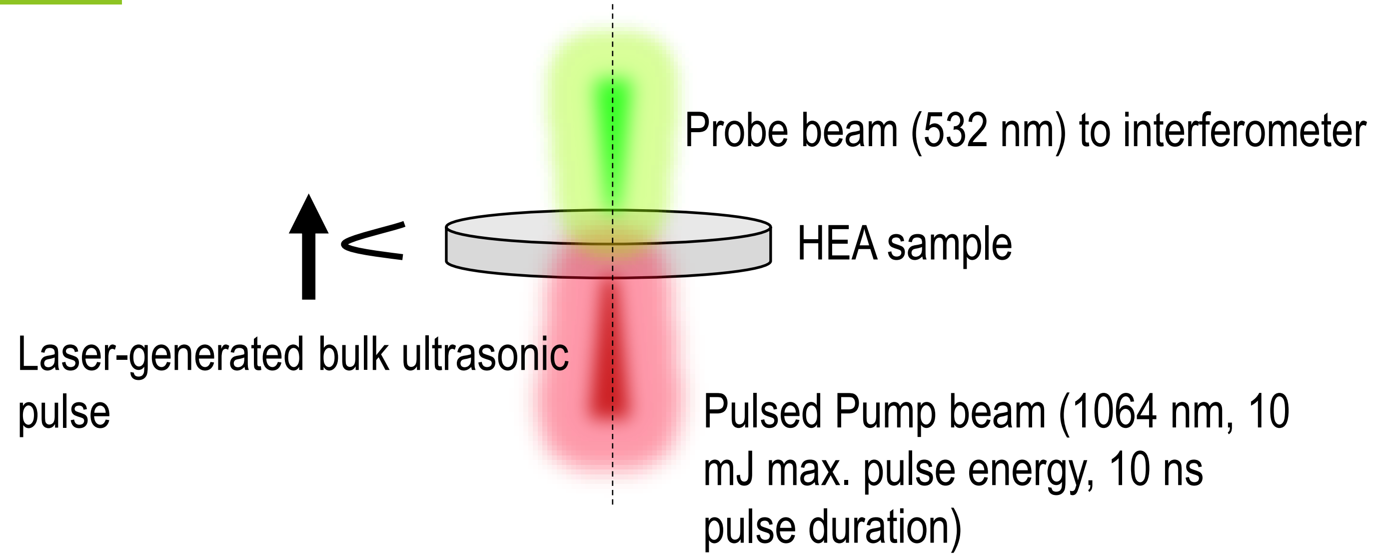
There are n = 75 stable elements that aren't toxic, radioactive or noble gases.
→ Over 219 million new CCA systems with $3 \leq r \leq 6$ principal elements.

By considering changes in composition, with every 10% change in concentration giving a new CCA base, this gives 592 billion possibilities for 3-6 principal elements.

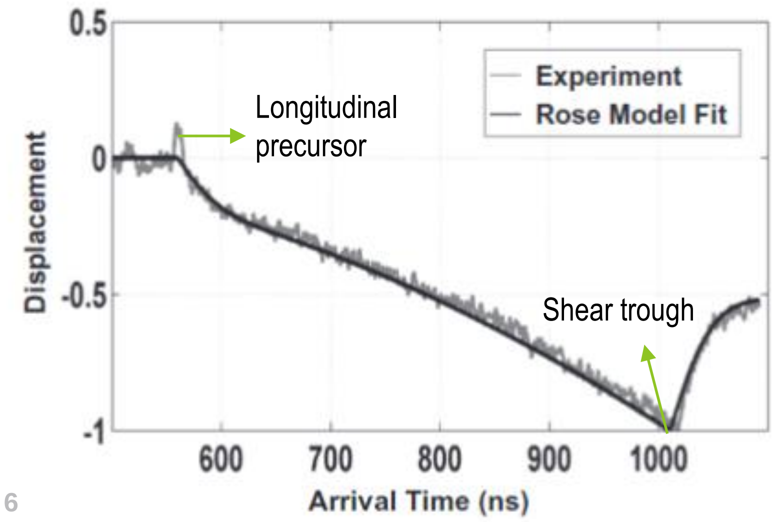
Candidate Techniques for Rapid Characterization

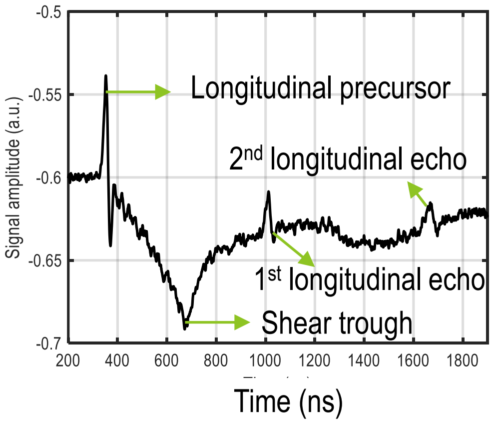


Laser Ultrasonics: A Non-Destructive Tool to Characterize Microstructure



Displacement waveform measured along the epicentral axis





S.J. Reese, Z.N. Utegulov, F. Farzbod, R.S. Schley, and D. H. Hurley. Ultrasonics 53, no. 3 (2013): 799-802.

- Ultrasonic waves are strongly affected by material microstructure, and therefore, serve as a facile means to probe elastic properties, phase content and their size distributions and volume fractions

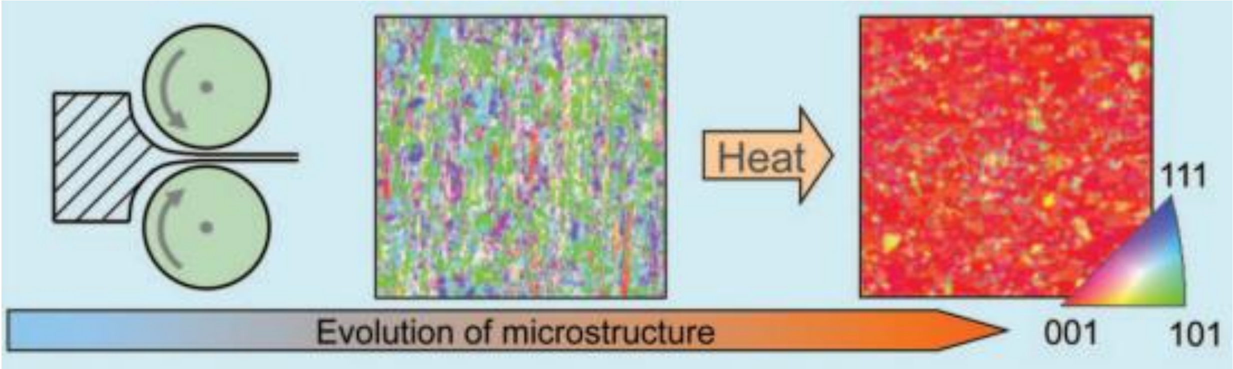
- Change in ultrasonic velocity w/ temperature → change in elastic moduli → change in phase content
- Ultrasonic scattering → change in elastic mismatch b/w domains
- Ultrasonic attenuation → viscoelasticity/ porosity, etc.

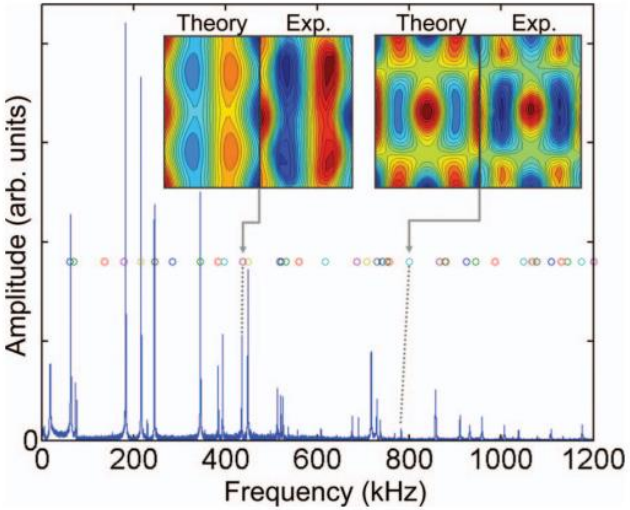
- Examples: $E(T) = \frac{\rho v_S^2 (3v_L^2 - 4v_S^2)}{v_L^2 - v_S^2}$ $\quad G(T) = \rho v_S^2$

  - Measurement of elastic stiffness tensor in anisotropic materials
  - Fatigue damage using laser-generated plate waves
  - *In situ* changes in grain microstructure in pure metals and alloys

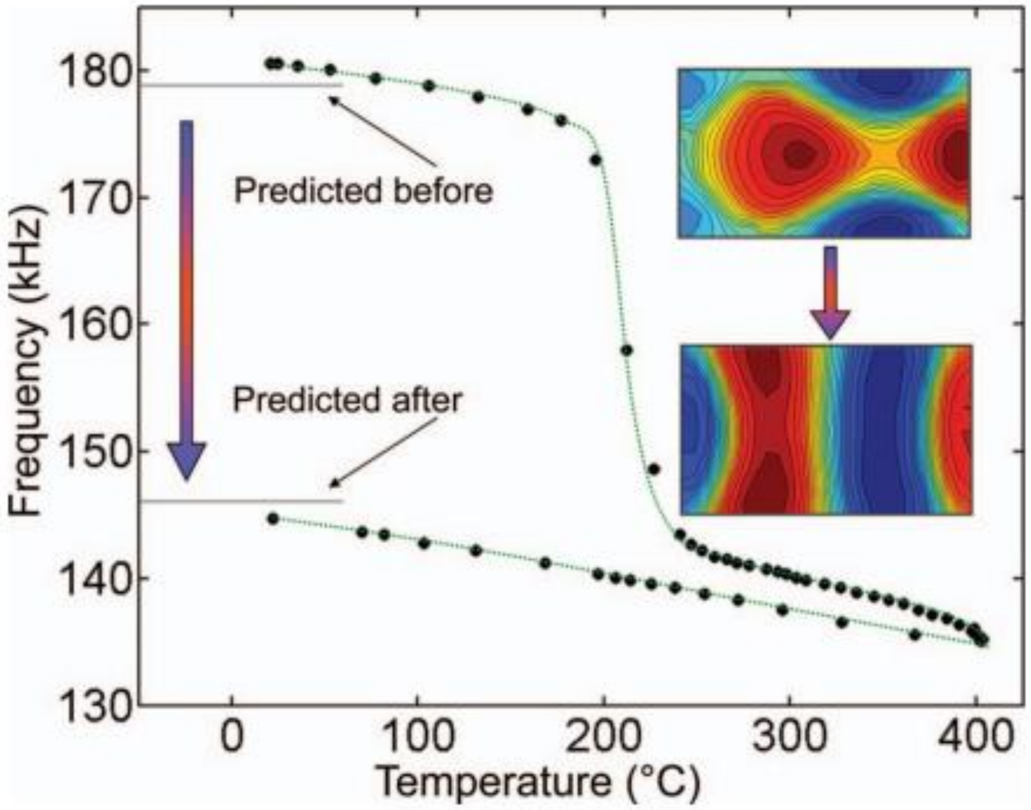
Laser Ultrasonics: A Non-Destructive Tool to Characterize Microstructure

- In situ laser-based resonant ultrasound measurements of microstructure mediated mechanical property evolution in Copper





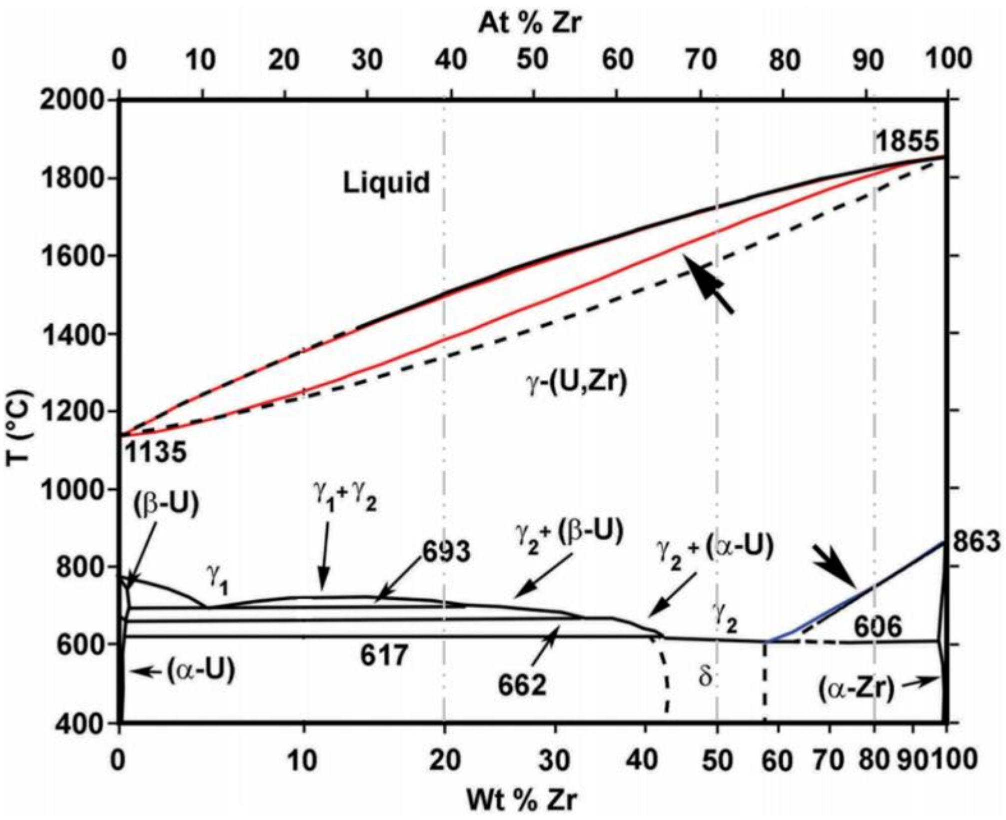
- Evolution of a resonant mode during annealing was examined.
- During recrystallization, the center frequency of this mode shifts by approximately 20% of the original value

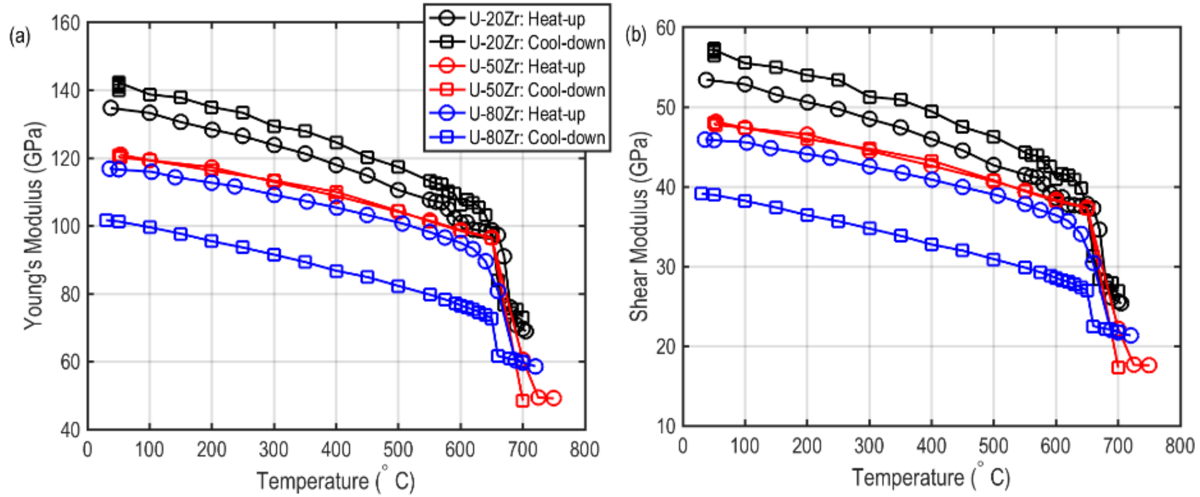


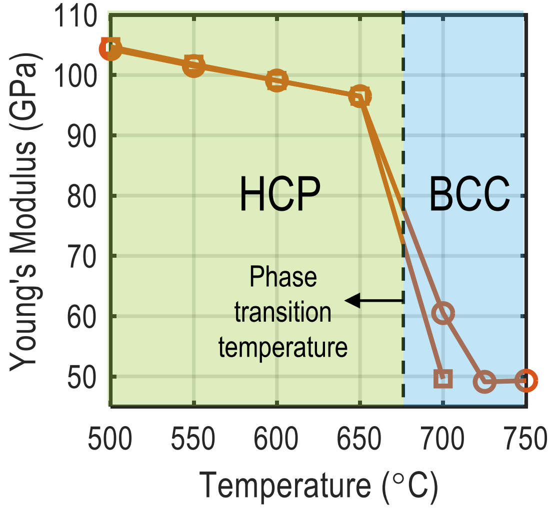
D.H. Hurley, S. J. Reese, S. K. Park, Z. Utegulov, J. R. Kennedy, and K. L. Telschow. Journal of Applied Physics 107, no. 6 (2010): 063510.

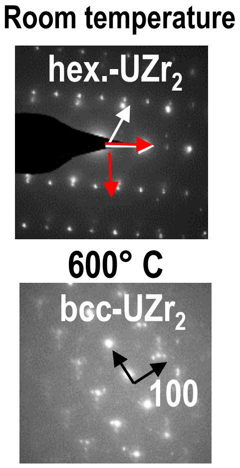
Laser Ultrasonics: A Non-Destructive Tool to Characterize Microstructure

- *In situ* monitoring of microstructure evolution during thermal processing of **uranium-zirconium alloys** using laser-generated ultrasound









A. Khanolkar, A., T. Yao, Z. Hua, C.A. Dennett, … D.H. Hurley, Journal of Nuclear Materials 553 (2021): 153005.

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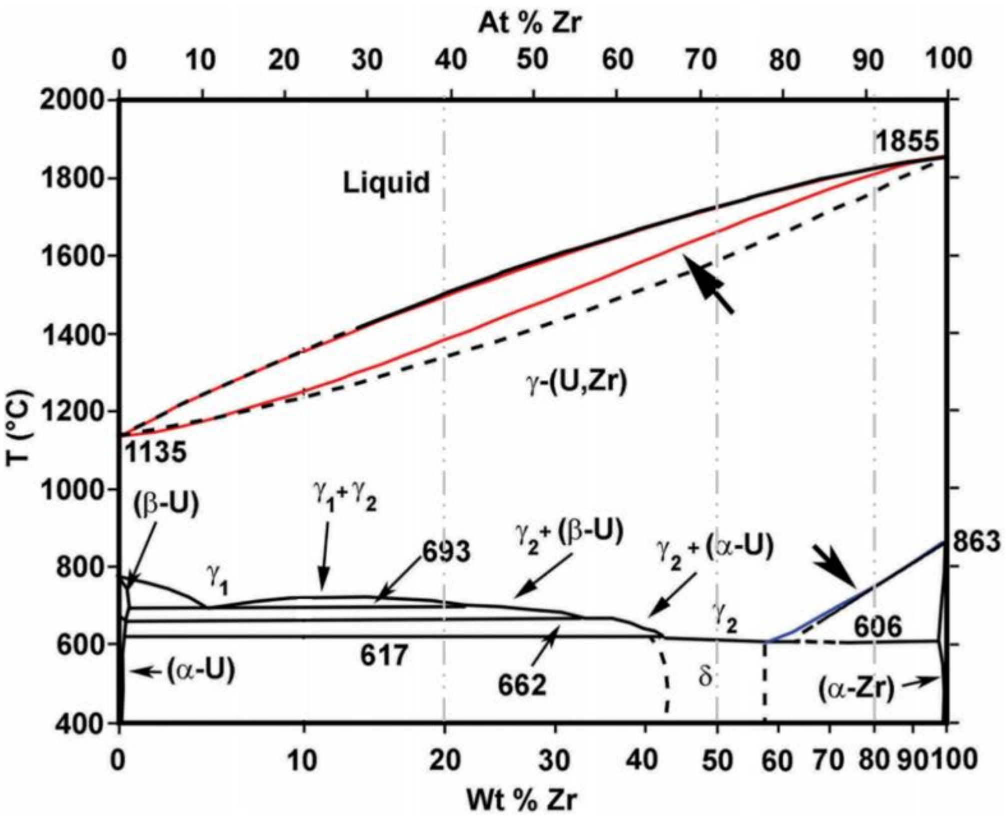

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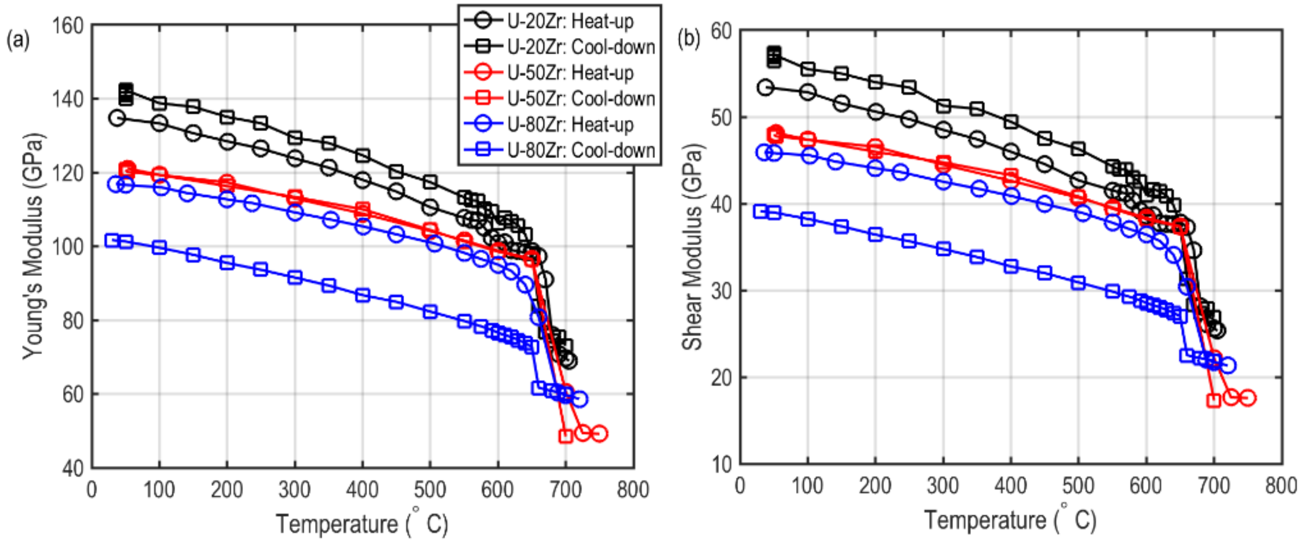
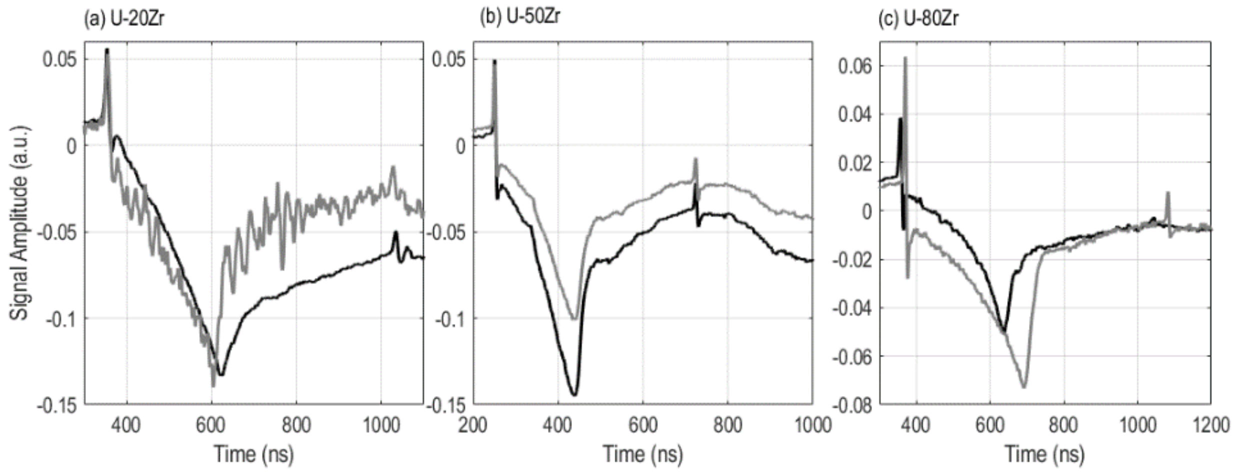




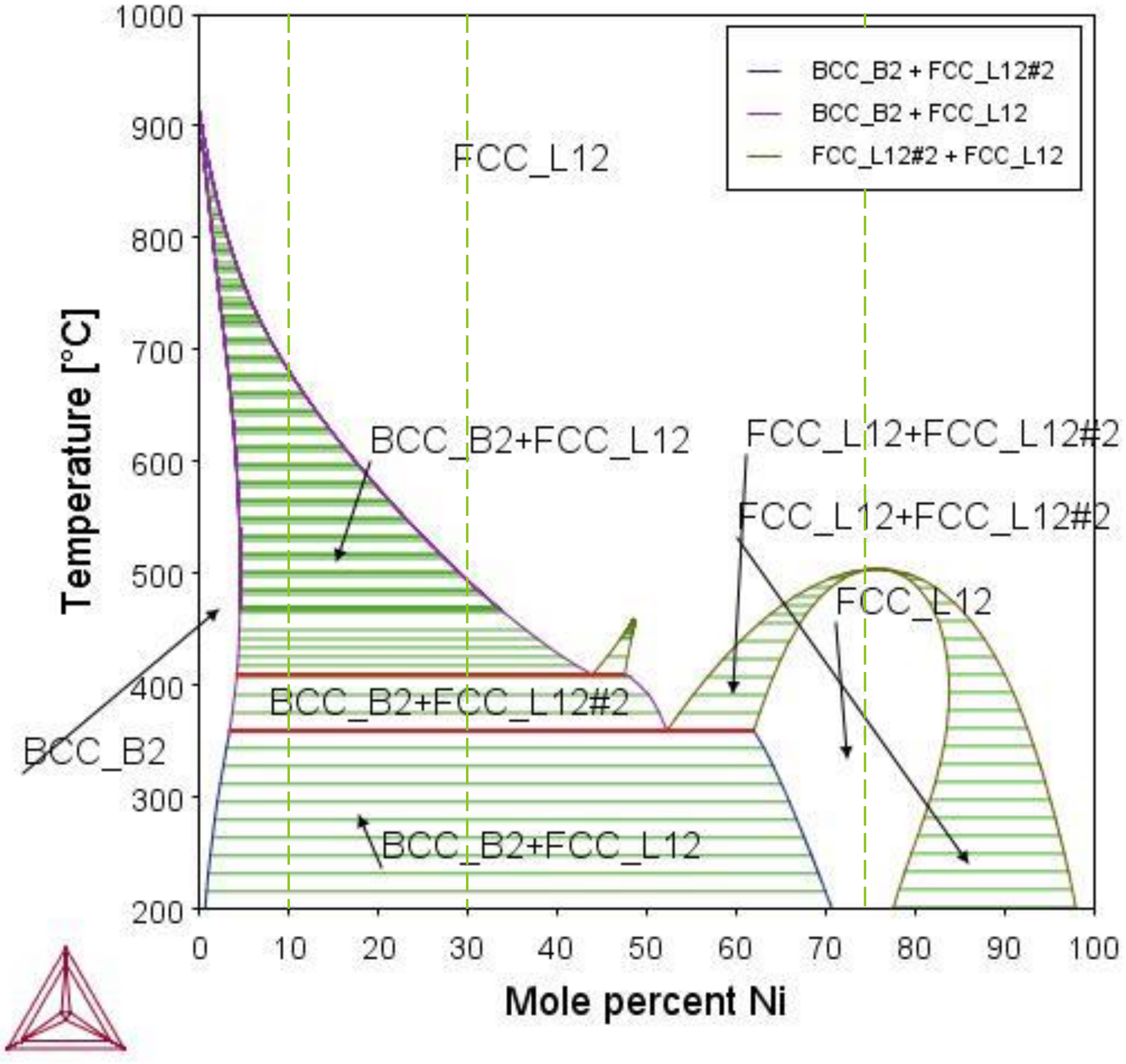
Outline

- **Motivation**
  - Need for High-throughput, rapid characterization tools to screen high entropy alloy compositions
  - Develop microstructure → property relationships

- **Objective**
  - Investigate the applicability of the laser ultrasonics technique as a high-throughput tool for screening HEA compositions

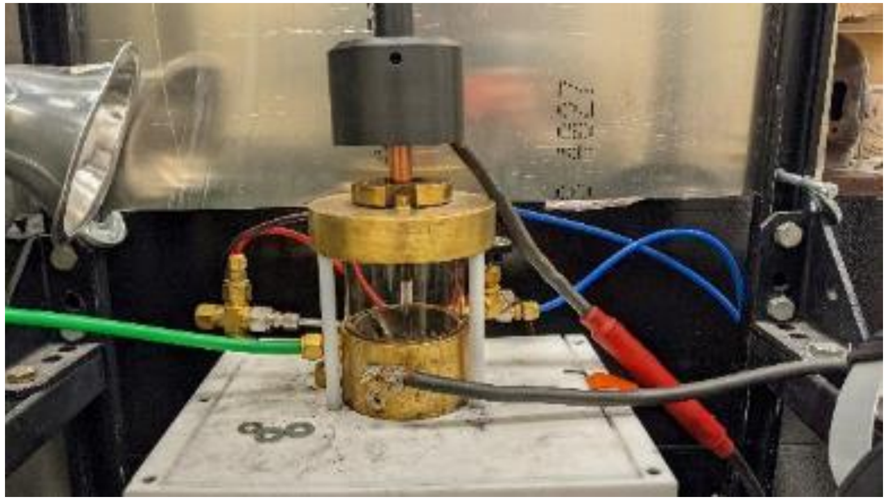
- **Approach**
  - Start with Binary Fe-Ni alloy system
  - Demonstrate on an HEA composition

Fe-Ni Binary Alloy System: Calculated Phase Diagram



CALPHAD calculations using Thermo-Calc software

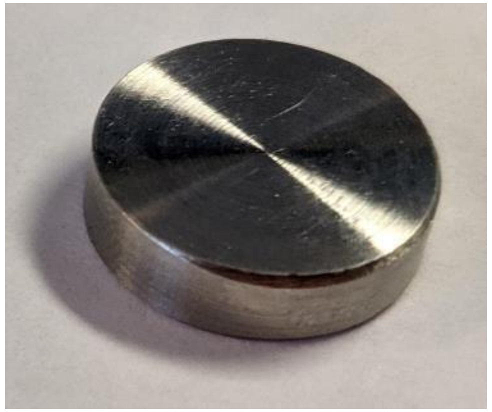
Fe-Ni Alloy Fabrication using Arc-Melting



As-cast Fe-Ni alloy immediately after arc melting

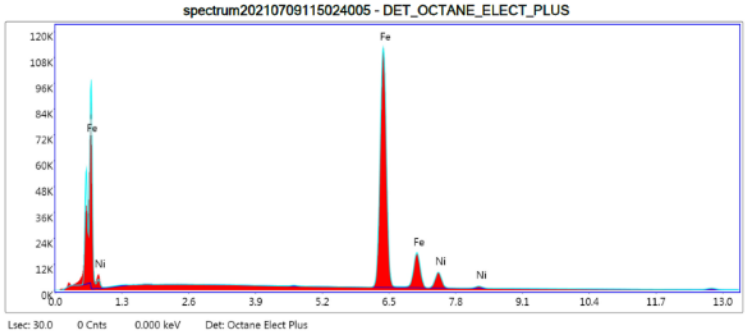
Fe-Ni alloy after annealing at 1100°C for 2 hours, and then naturally cooling

Samples machined to cylindrical geometry with flat & parallel faces

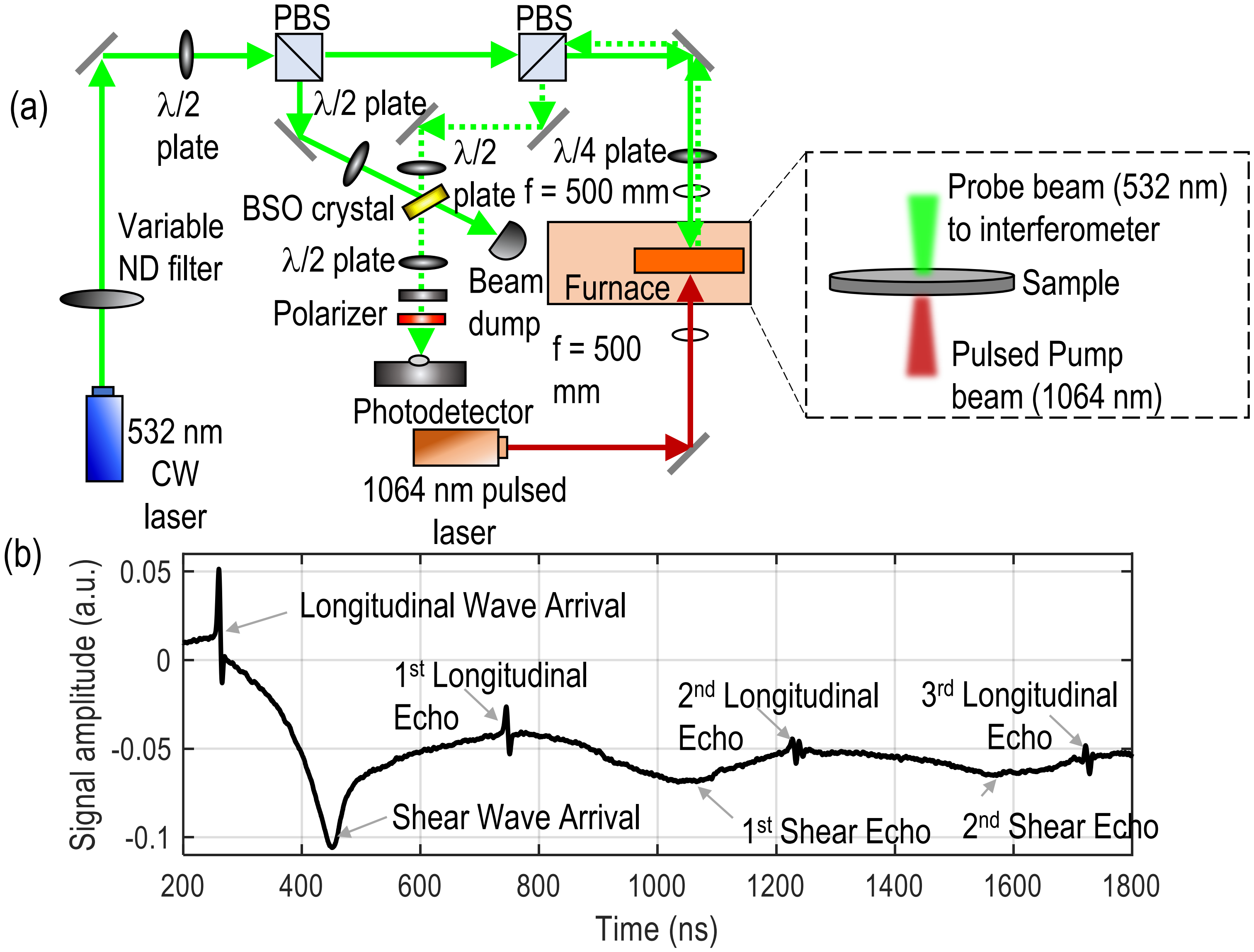


Sample	Element	Weight %	Atomic %	Net Int.	Error %	K ratio	Z	A	F
Fe-10Ni	Fe	91.33	91.72	54502.91	1.54	0.8430	0.9107	1.0043	1.0092
	Ni	8.67	8.28	3819.89	2.86	0.0762	0.9304	0.9399	1.0044
Fe-30Ni	Fe	72.66	73.64	41522.71	1.64	0.6811	0.9076	1.0034	1.0293
	Ni	27.34	26.36	11462.54	2.26	0.2424	0.9273	0.9527	1.0034
Fe-75Ni	Fe	27.40	28.40	16601.33	2.02	0.2698	0.8998	1.0014	1.0928
	Ni	72.60	71.60	31416.26	1.78	0.6581	0.9198	0.9843	1.0011

EDS spectrum

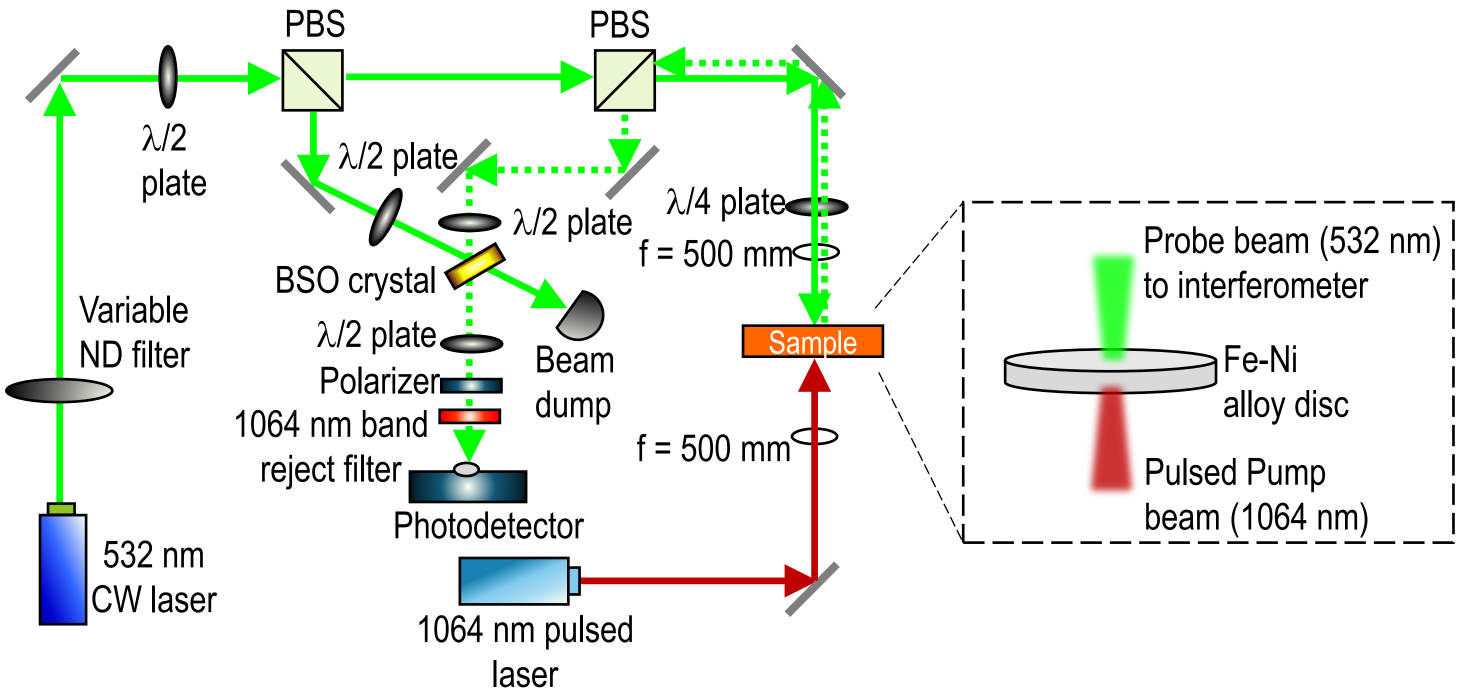


Laser Ultrasonics Setup at INL

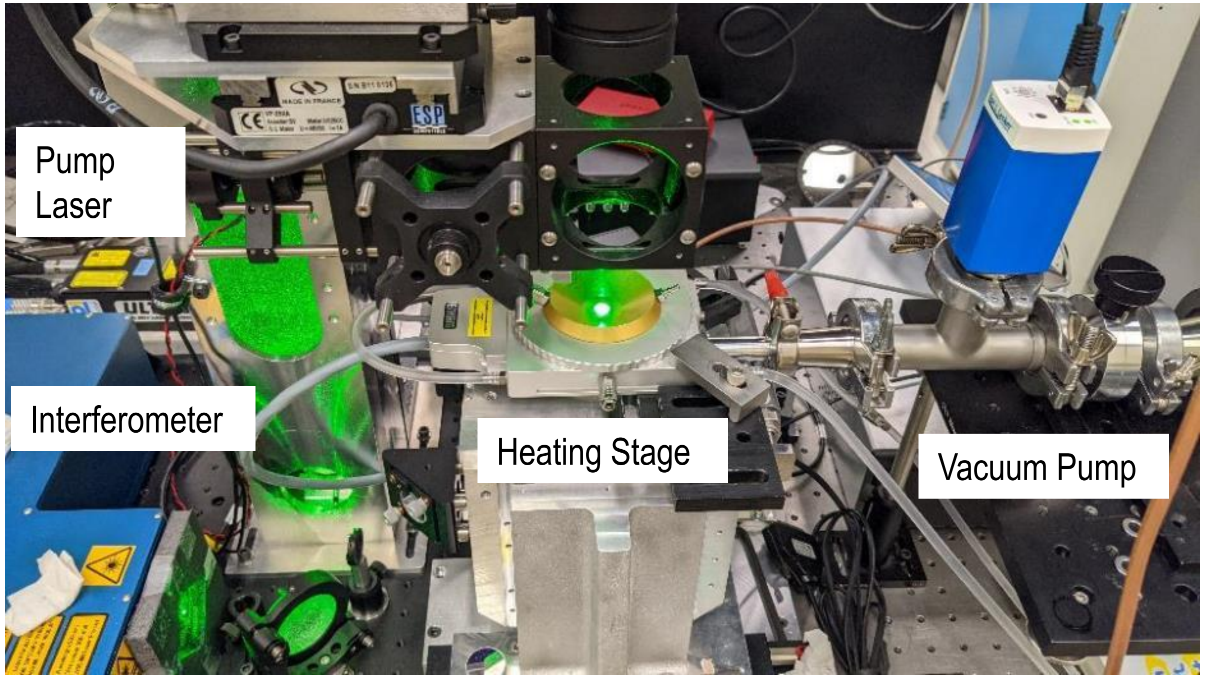


Laser Ultrasonics Setup at INL

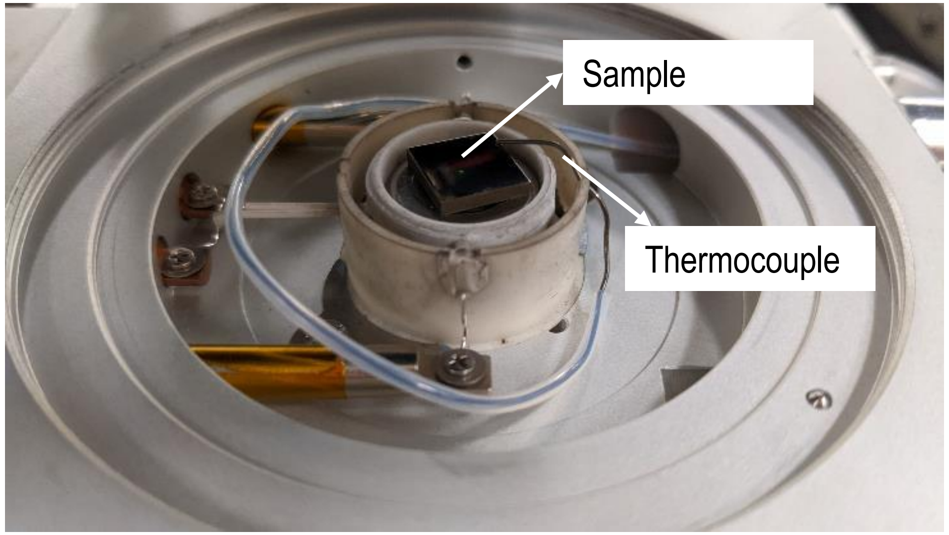
(a)



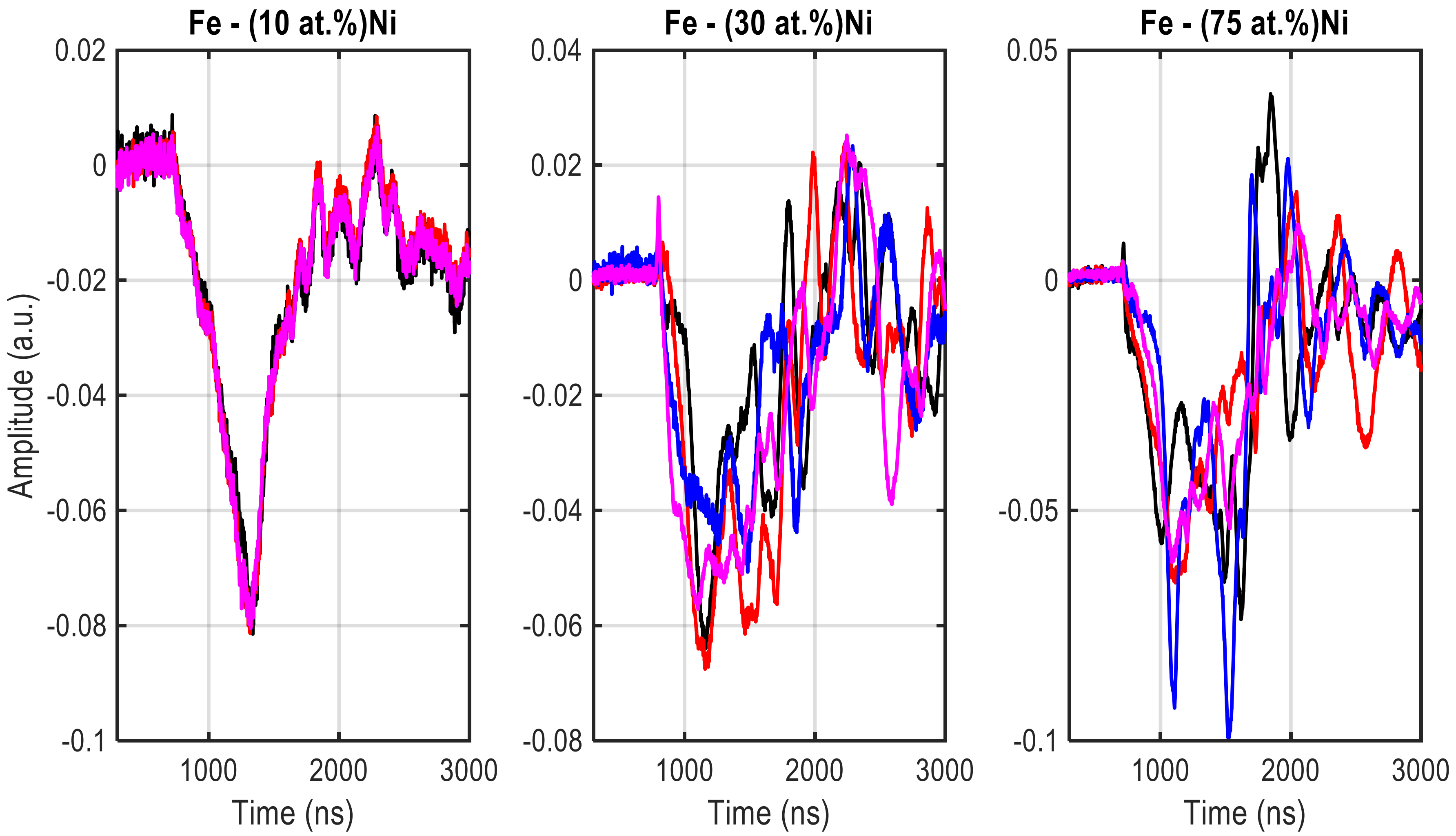
(b)



(c)



Epicentral Ultrasonic Waveforms at Room Temperature



Strong ultrasonic scattering from microstructural heterogeneities in Fe-30Ni and Fe-75Ni

Metallographic Examination of Grain Size Distribution

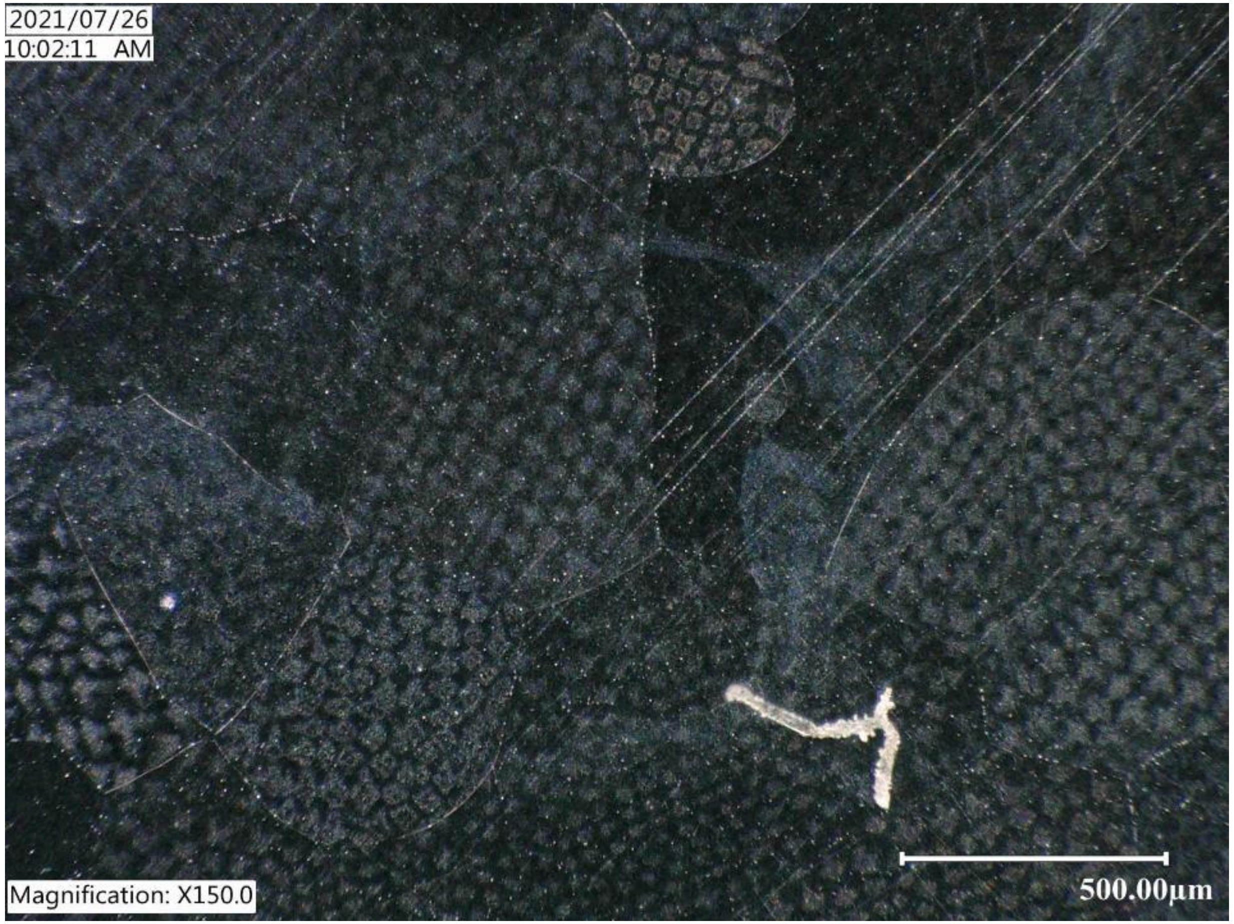
Fe-10Ni

Fe-30Ni

Strong ultrasonic scattering due to elastic anisotropy/ mismatch b/w grains

Metallographic Examination of Grain Size Distribution

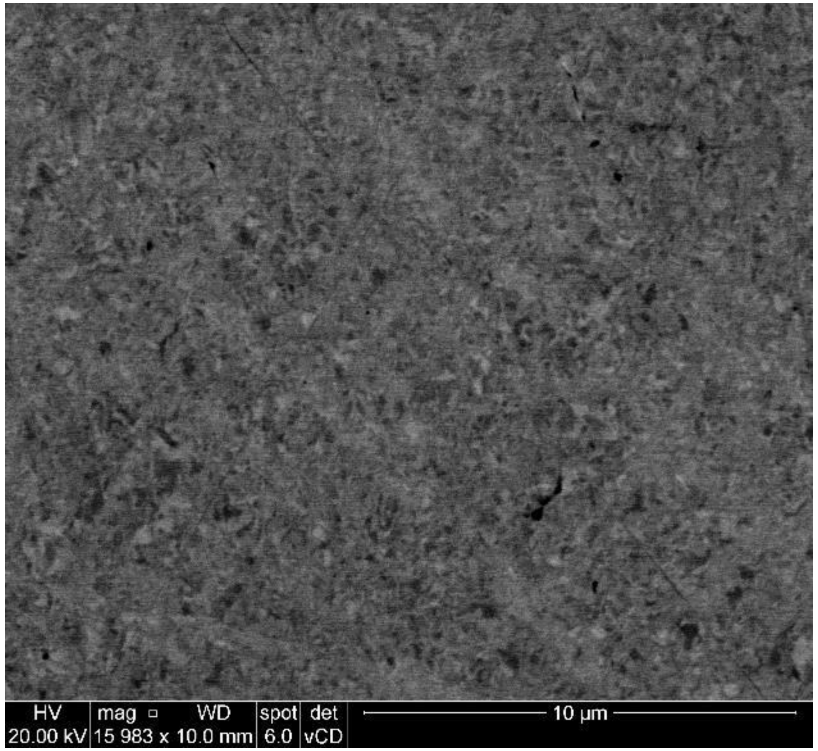
Fe-30Ni (zoomed-in)



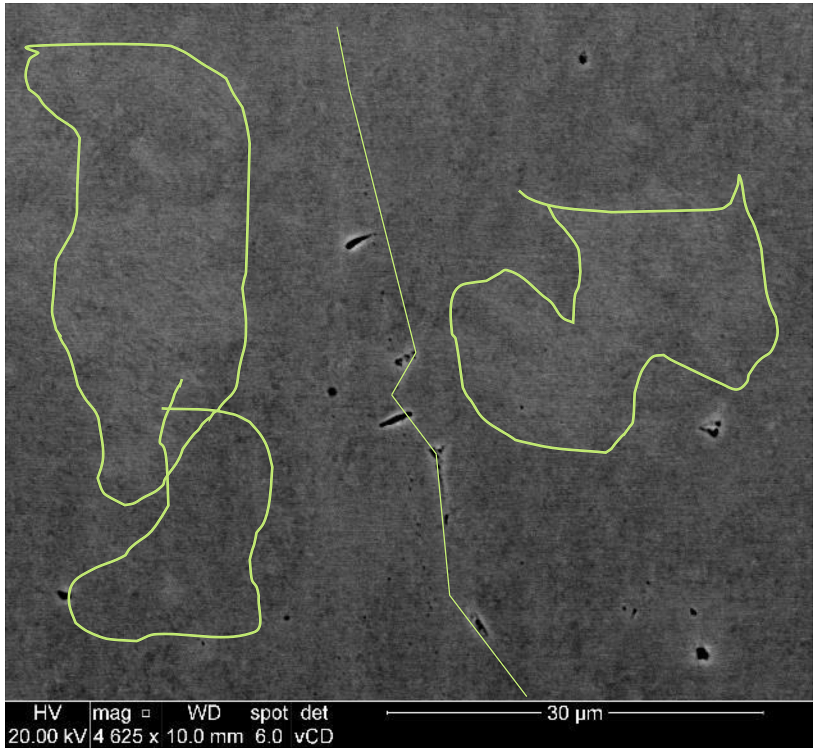
Fe-75Ni

Scanning Electron Microscopy Images of Fe-Ni Microstructure

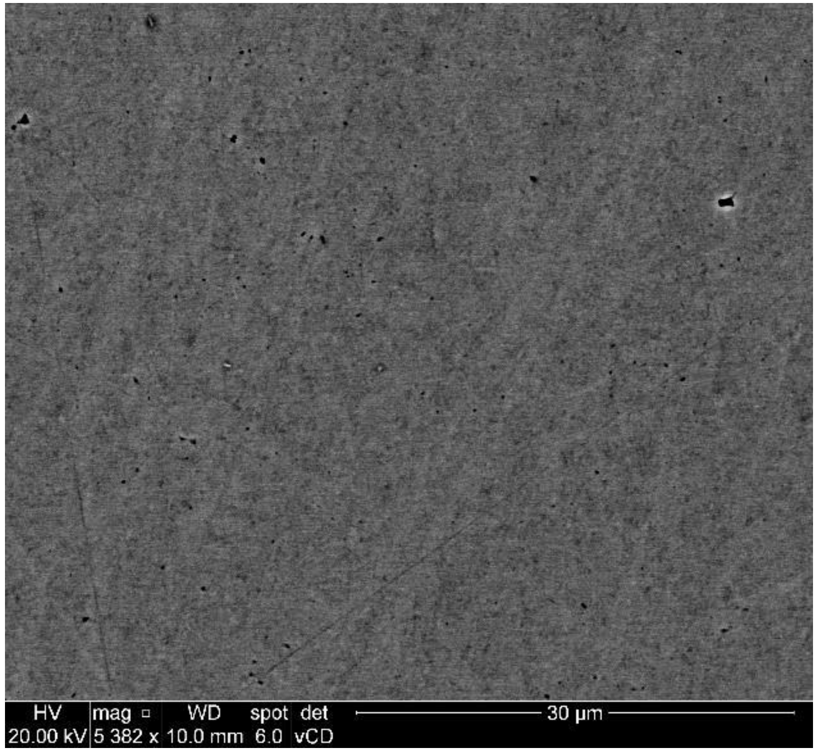
(a) Fe-10Ni



(b) Fe-30Ni

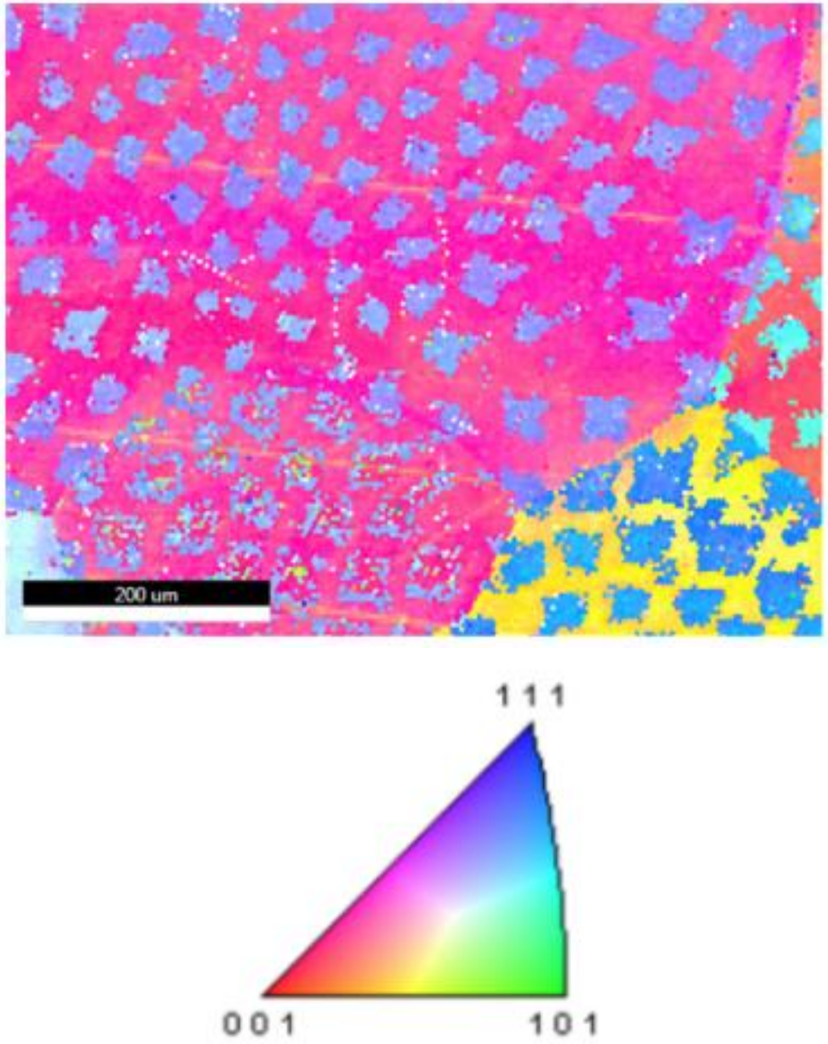


(c) Fe-75Ni

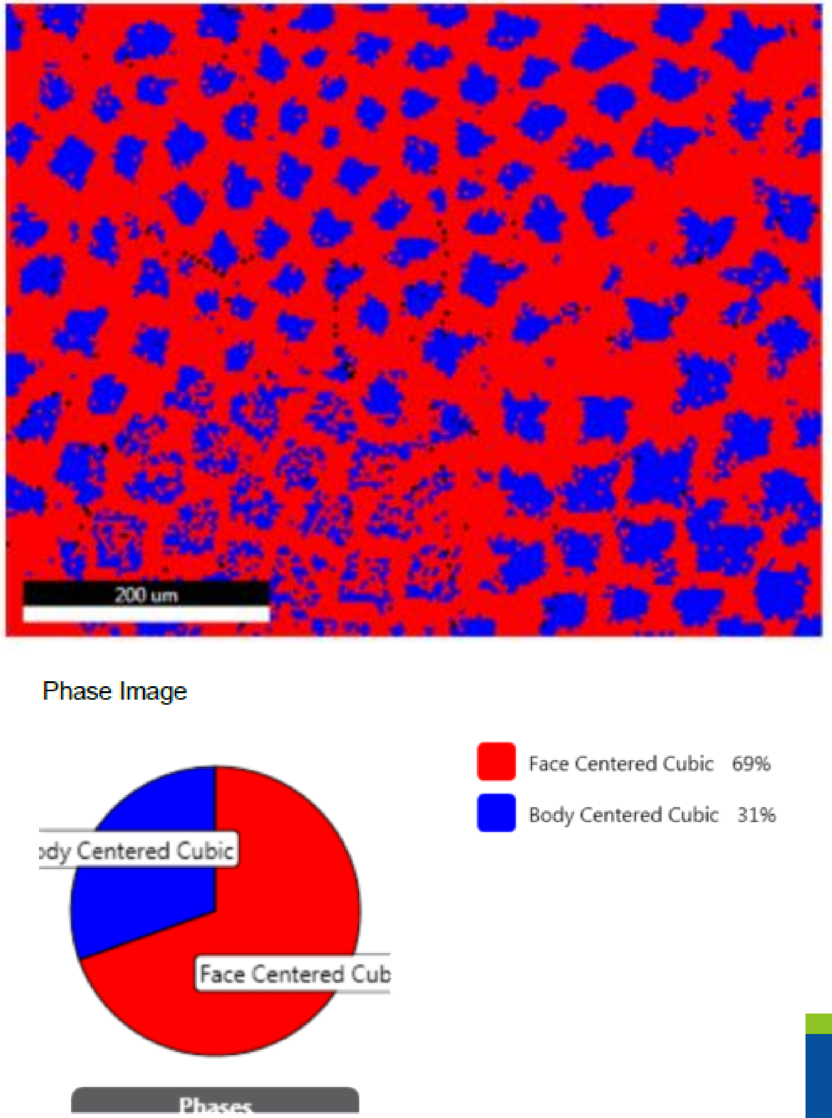


Electron Backscatter Diffraction Imaging of Cast Fe-30Ni

(a)

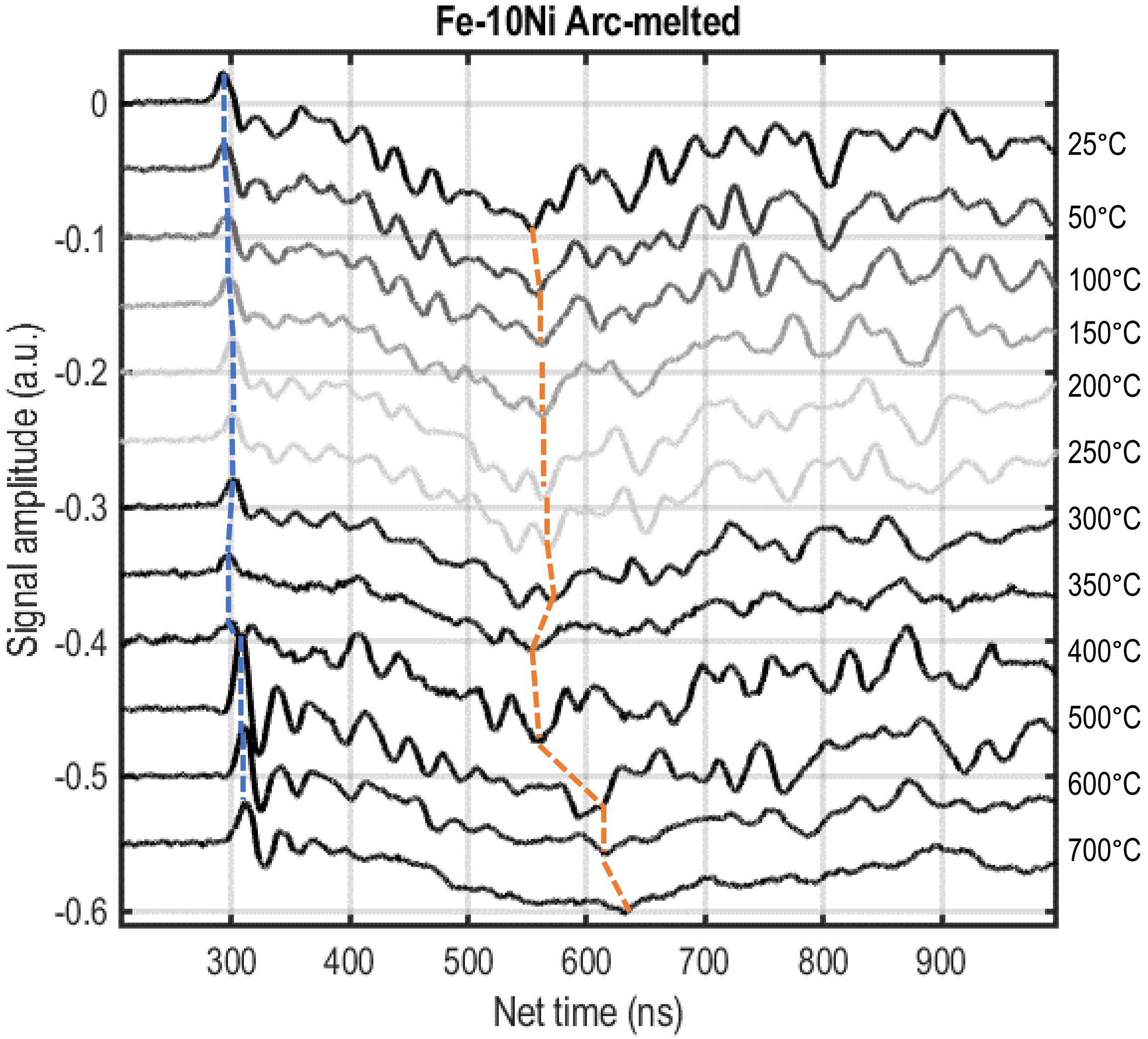


(b)

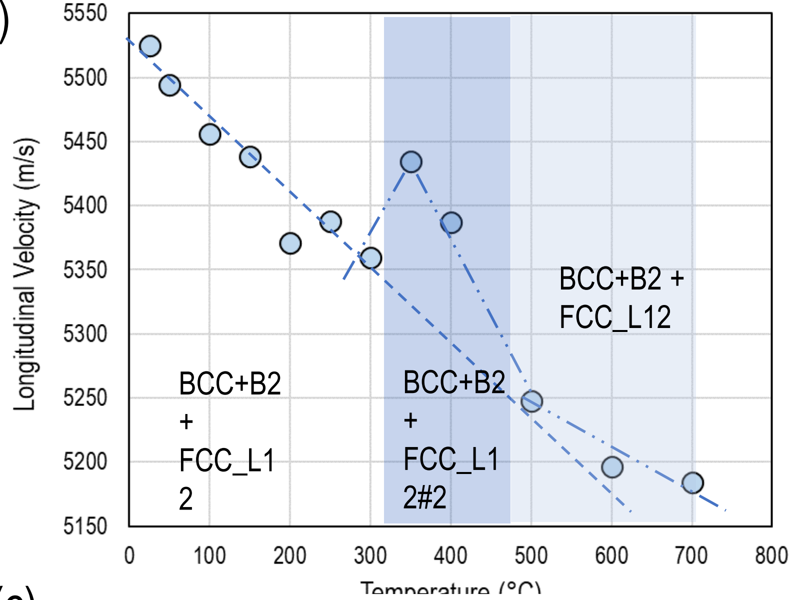


Mapping Phase Transition in Fe-10Ni using In situ Laser-generated Ultrasound

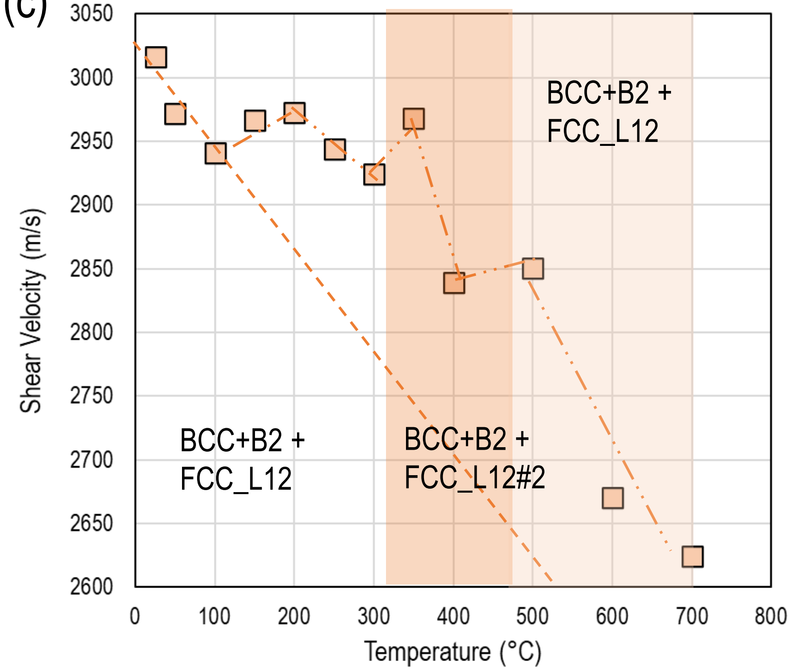
(a)



(b)



(c)



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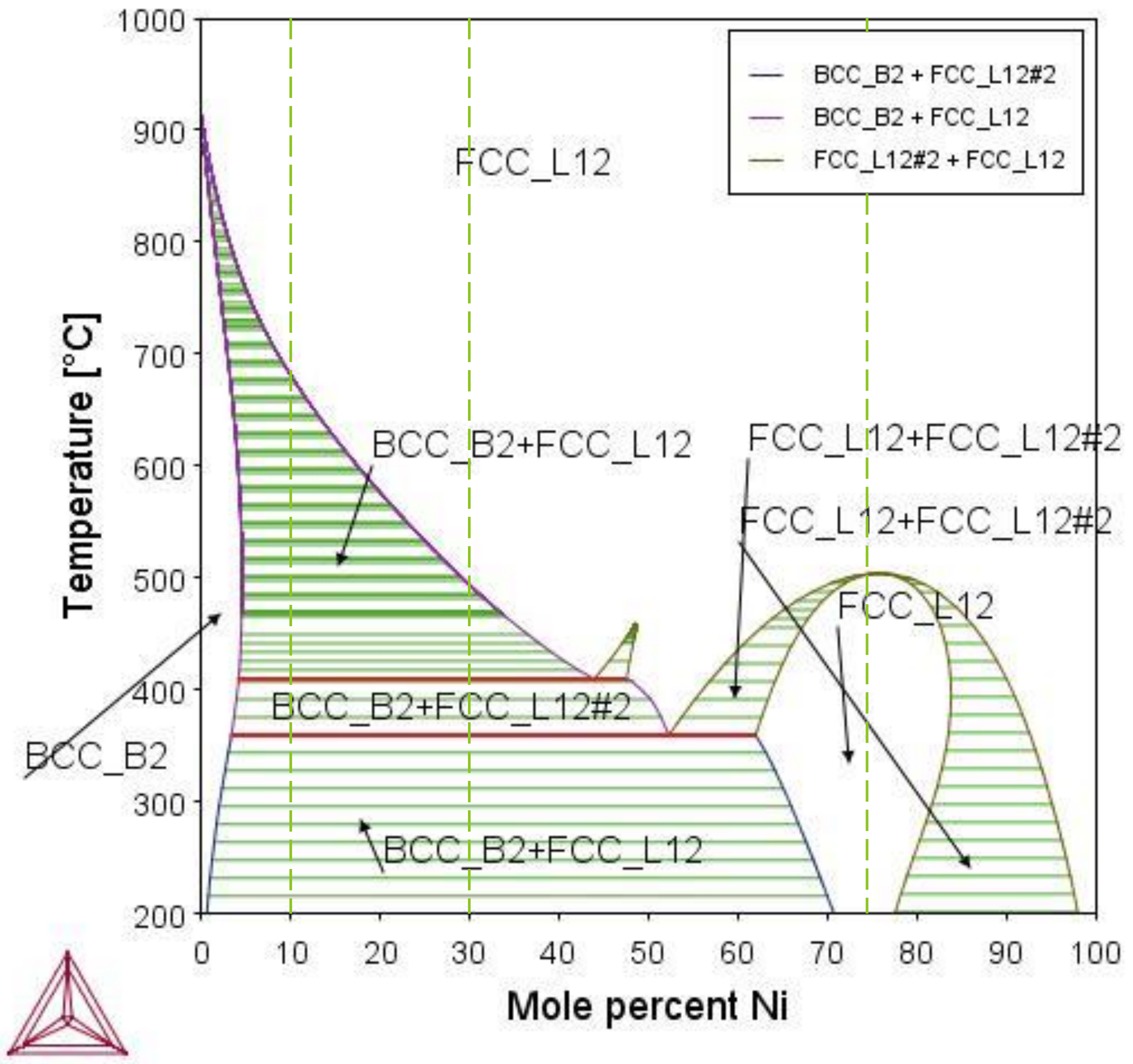

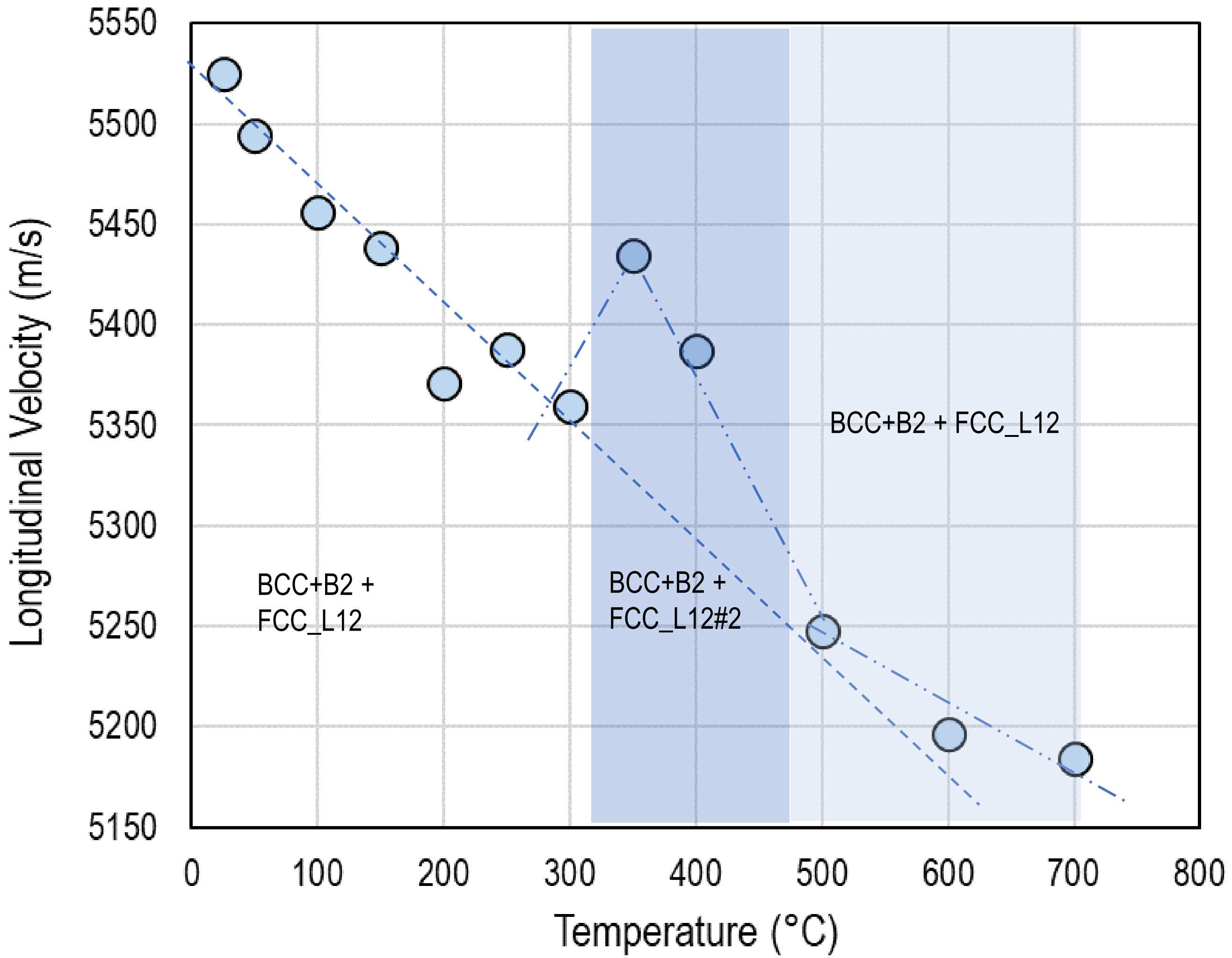

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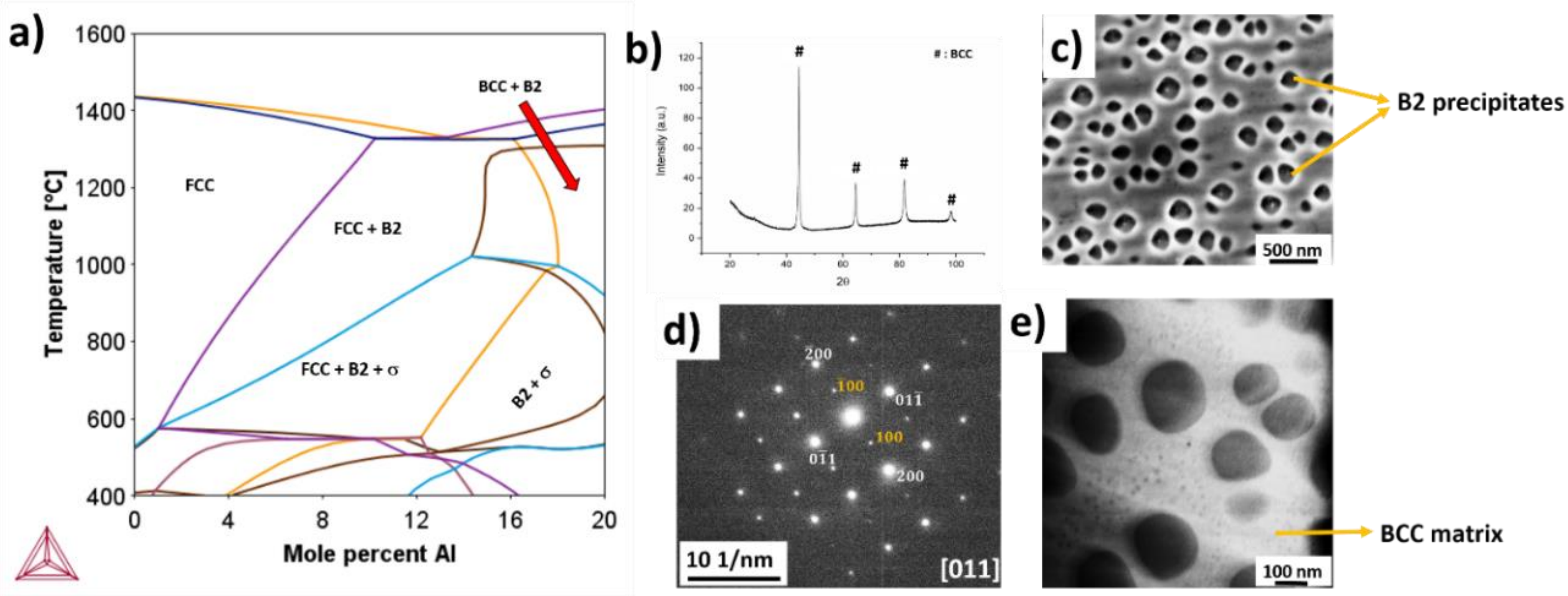
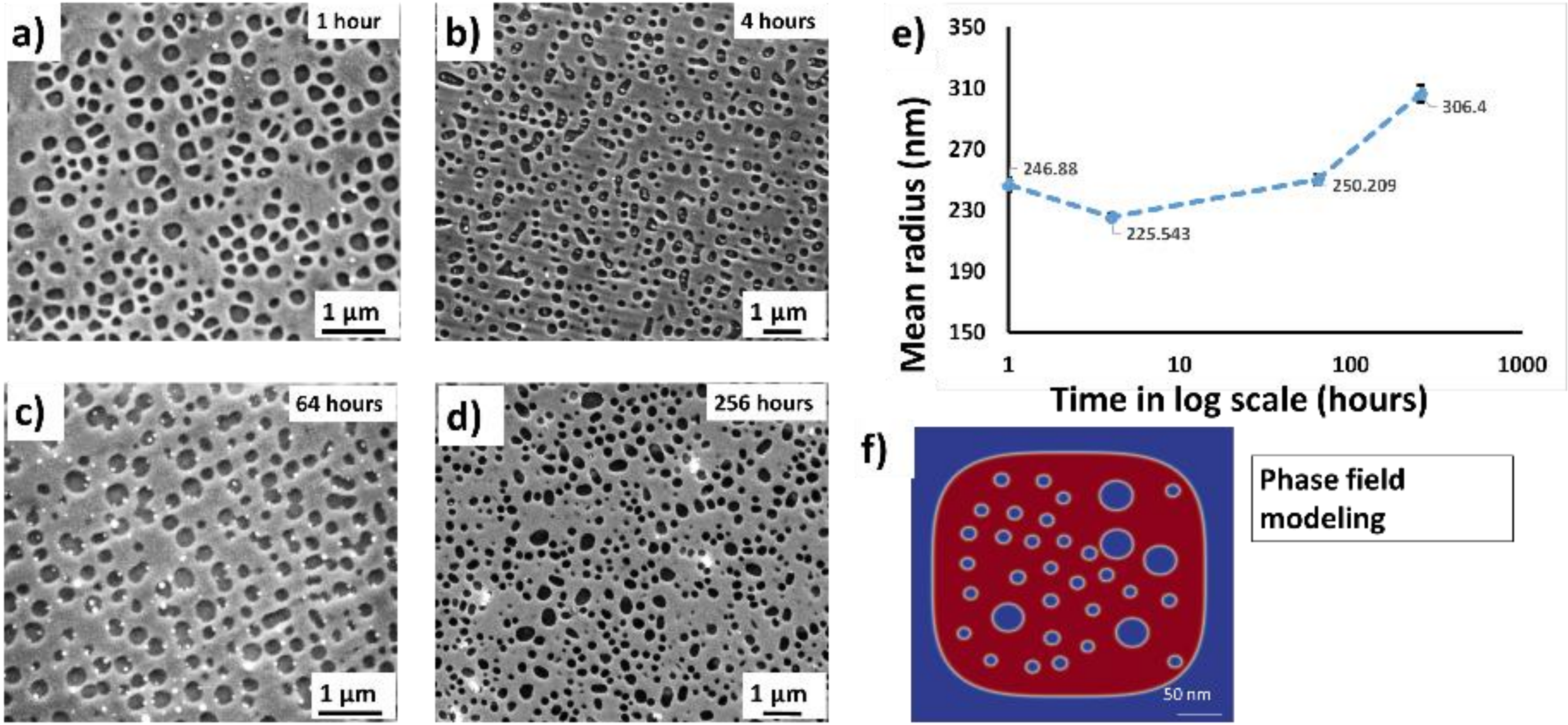
Demonstrate Approach on an HEA alloy (FeCrNiAlCo) sample

- A computationally designed HEA with bulk composition: Fe_{34}-Cr_{34}-Ni_{14}-Al_{14}-Co_{4} (in atomic percent) was fabricated using arc-melting process.



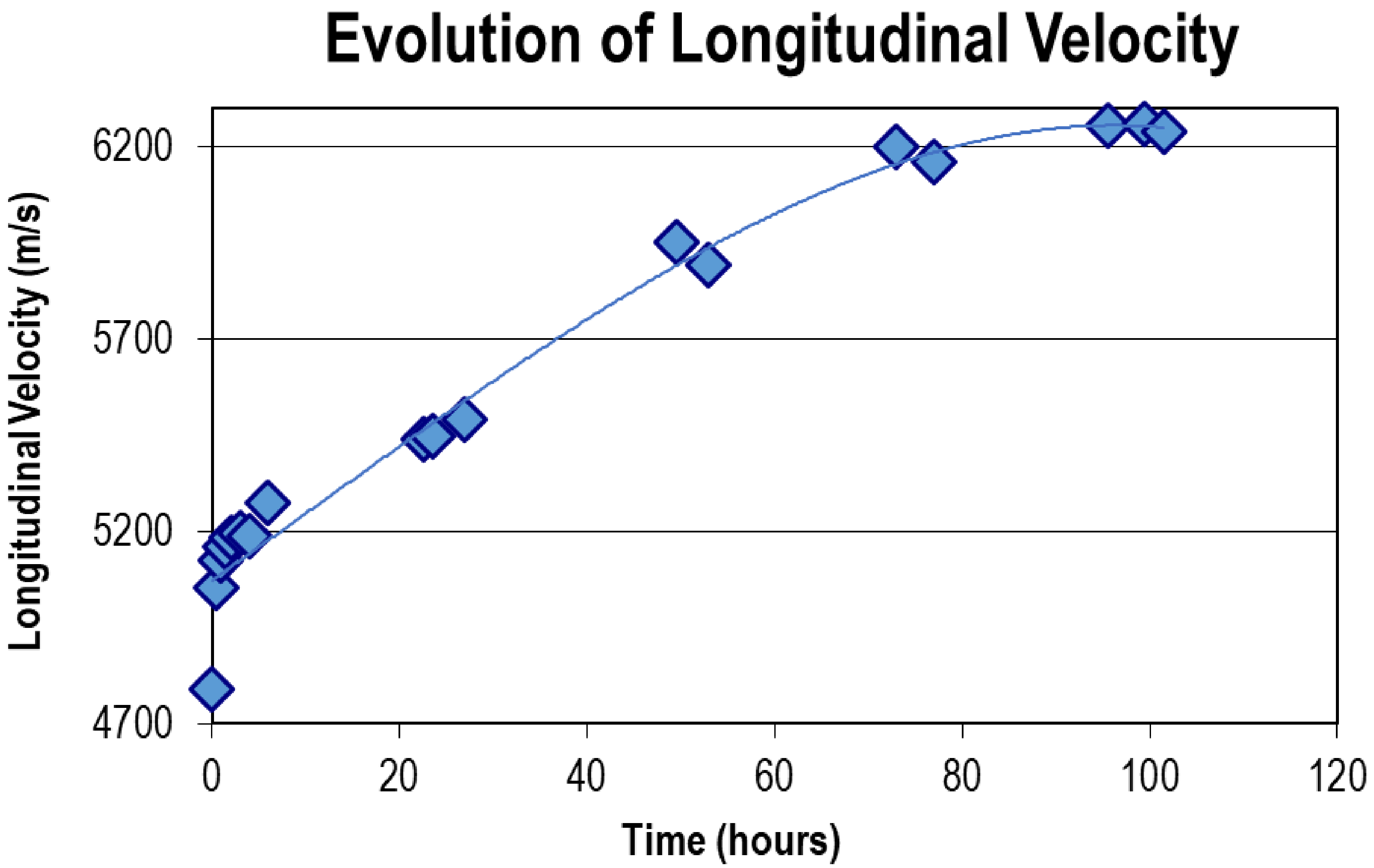
S. Meher, S. Kadambi, T.M. Lillo, in preparation (2022)

Coarsening Kinetics of Ordered B2 Precipitates



S. Meher, S. Kadambi, T.M. Lillo, in preparation (2022)

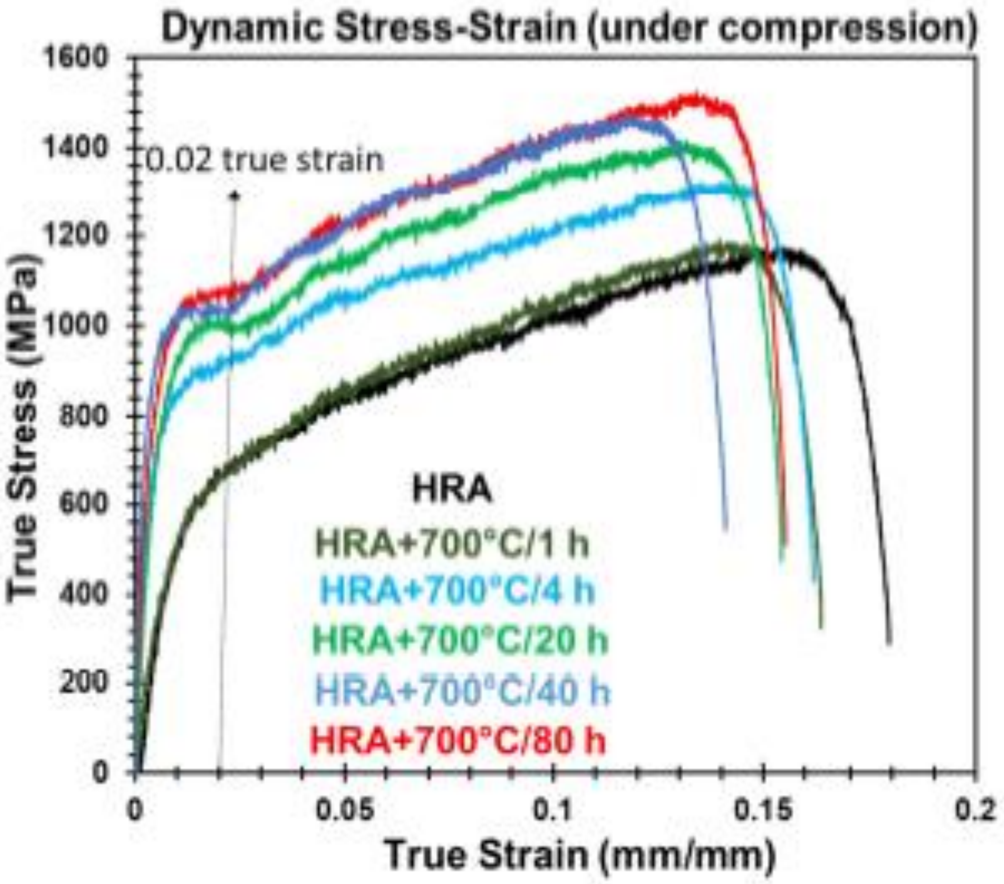
Using Laser Ultrasonics to Monitor Coarsening Kinetics

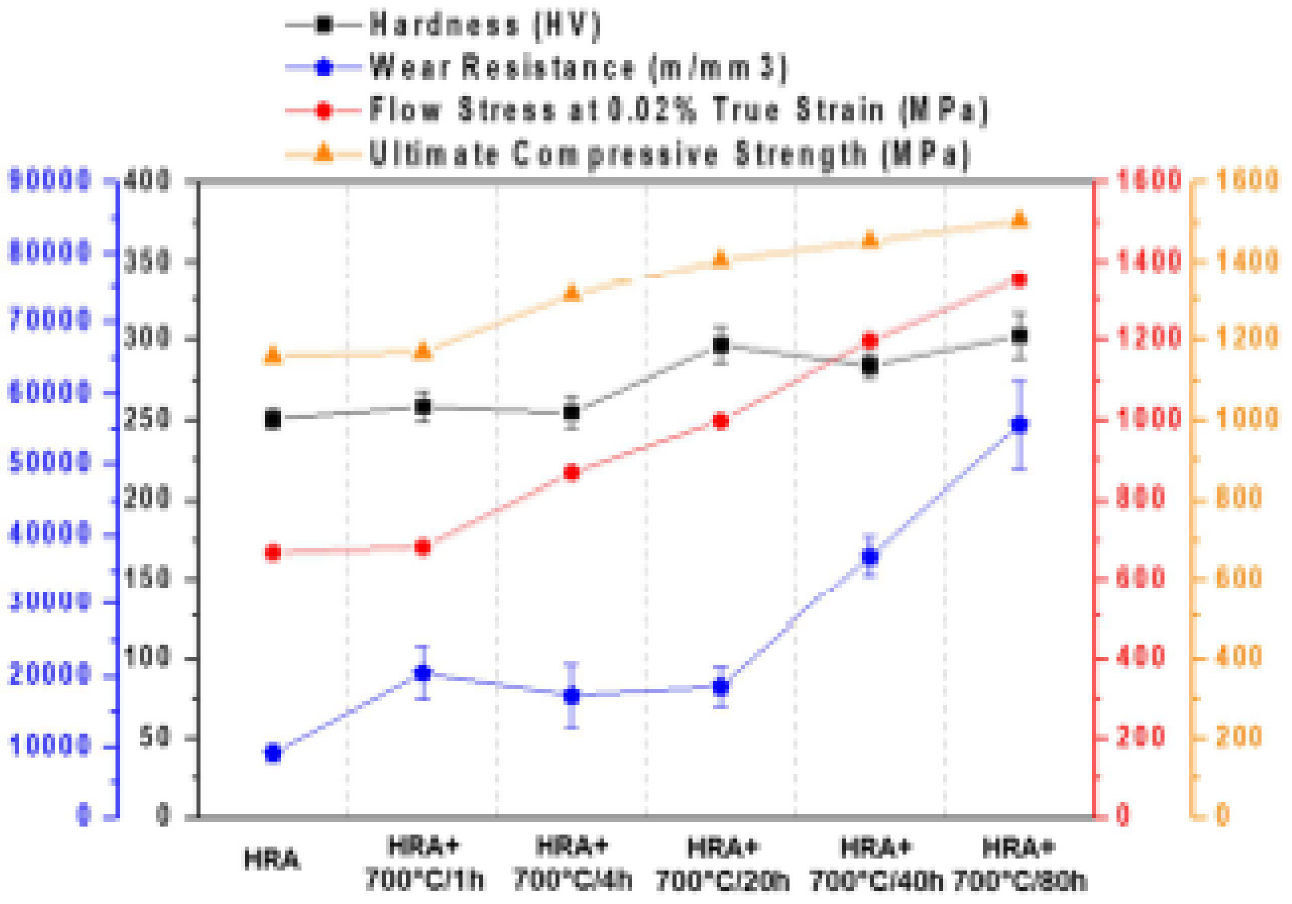


- Fe-Cr-Ni-Al-Co sample previously annealed at 750°C for 256 hours
- Same sample was held at 950°C in the optical heating stage
- Laser ultrasonic waveforms recorded periodically while holding sample at 950°C

Laser-generated ultrasound can monitor temporal evolution of precipitate-induced strengthening

Intragranular B2 precipitates positively influence tensile strength





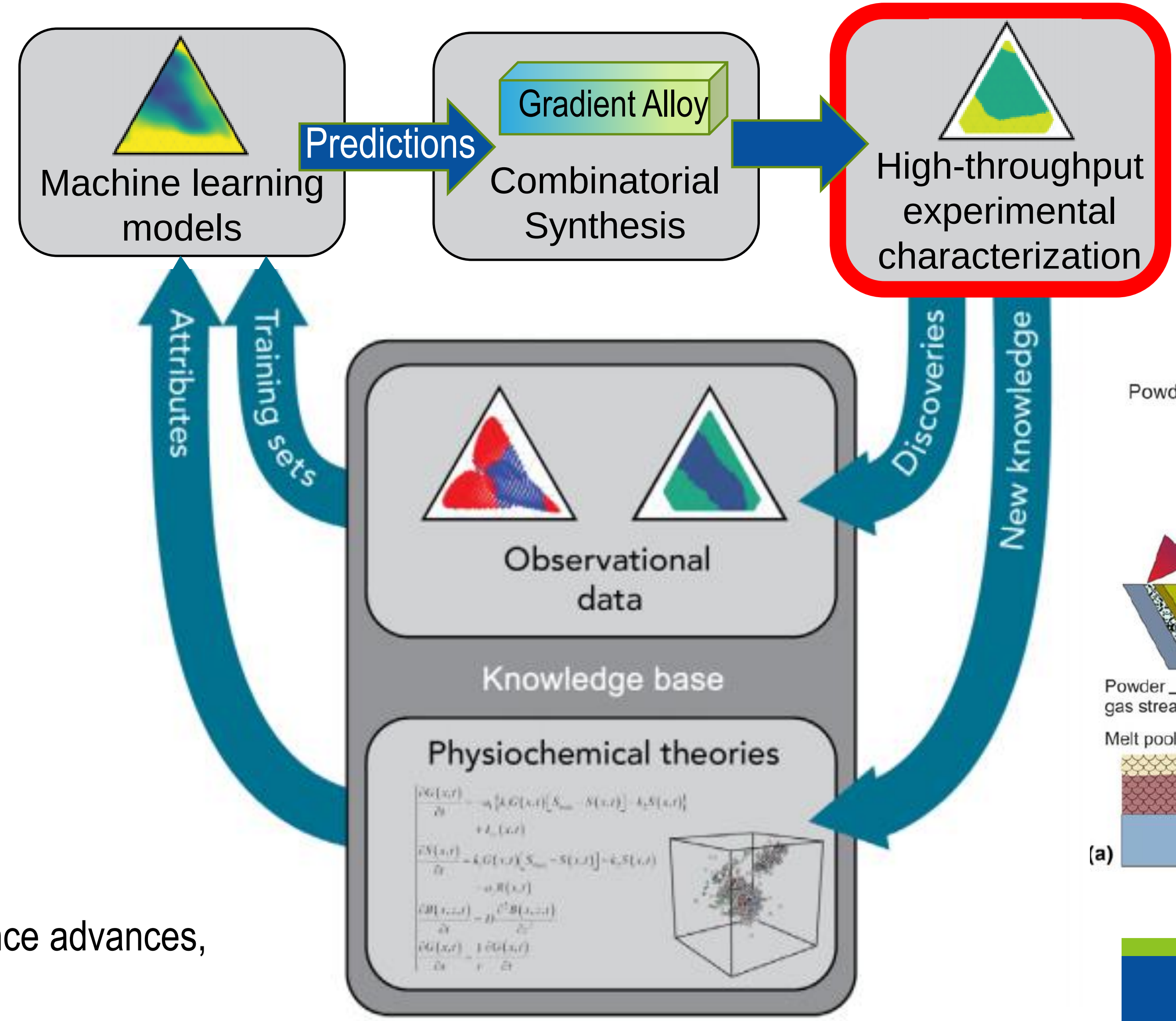
$Al_{0.5}CoCrFeNi$ hypoeutectic alloy consisting of an as-solidified FCC+B2 microstructure

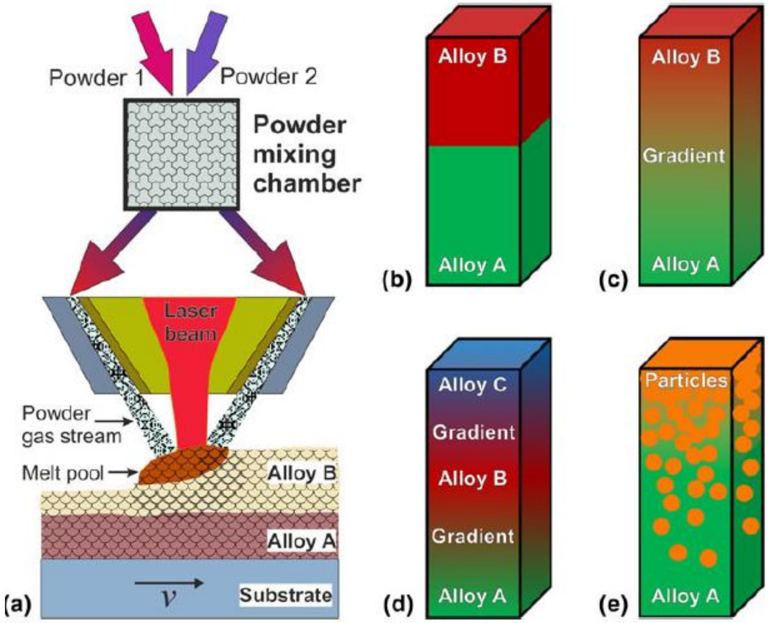
B. Gwalani, T. Torgerson, …, R. Banerjee. "Influence of fine-scale B2 precipitation on dynamic compression and wear properties in hypo-eutectic Al0. 5CoCrFeNi high-entropy alloy." Journal of Alloys and Compounds 853 (2021): 157126.

Summary

- Laser-generated ultrasonics shows potential in *in situ* (real-time) & non-destructively:
  - Monitoring grain growth
  - Phase transformation boundaries
  - Coarsening kinetics

A Holistic Approach Towards Material Discovery



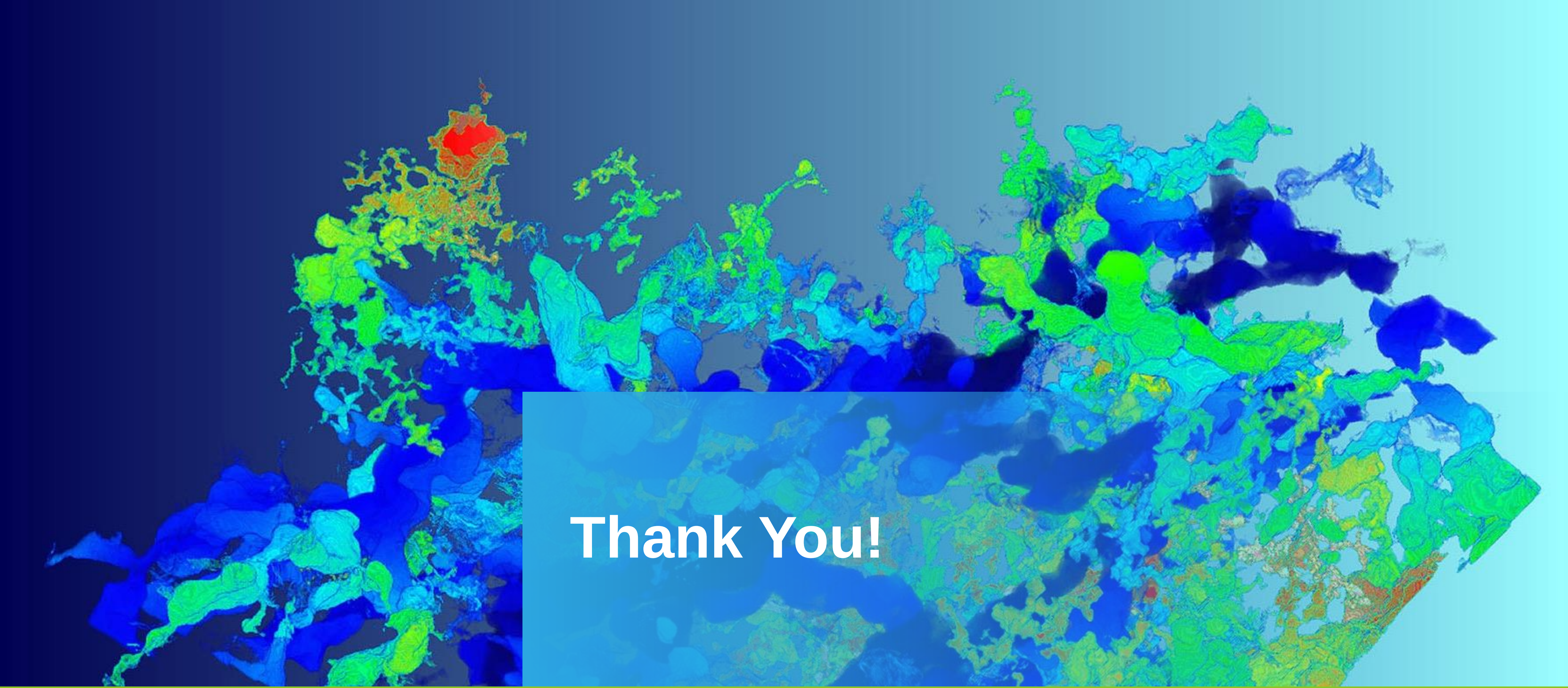


F. Ren, …, A. Mehta, Science advances, 4(4), 1566 (2018).

Acknowledgements

- Work supported through the ***INL Laboratory Directed Research and Development*** (LDRD) Program under DOE Idaho Operations Office Contract DE-AC07-05ID14517

- Assistance with sample fabrication and preparation:
  - Dennis Tucker
  - Austin Matthews
  - Wesley Jones



Thank You!

Battelle Energy Alliance manages INL for the
U.S. Department of Energy's Office of Nuclear Energy

INL Idaho National Laboratory

Idaho National Laboratory

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