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Laser Engineered Net Shaping (LENS) of High Entropy Alloys

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U.S. DEPARTMENT OF ENERGY NNSA National Nuclear Security Administration

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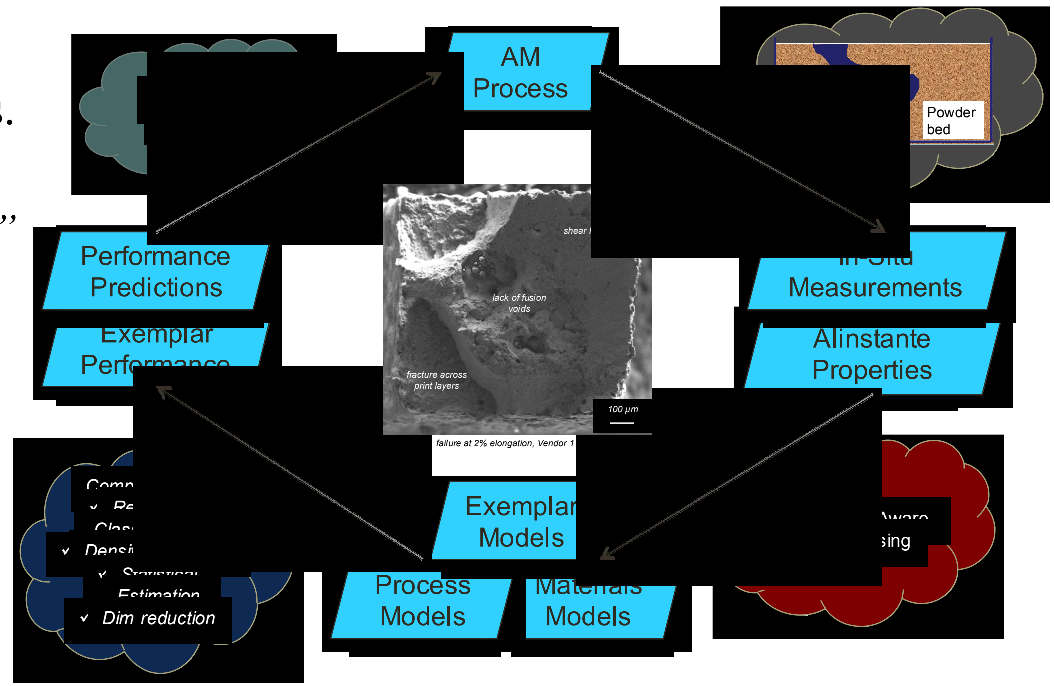
Sandia AM Program

Goal: 15+ year vision to combine promise of **metal additive manufacturing (AM)** with **deep materials & process understanding** to revolutionize design, manufacturing, & qualification paradigms.

- ➢ Materials, designs, and ultimately components are "*Born Qualified/Certified*"



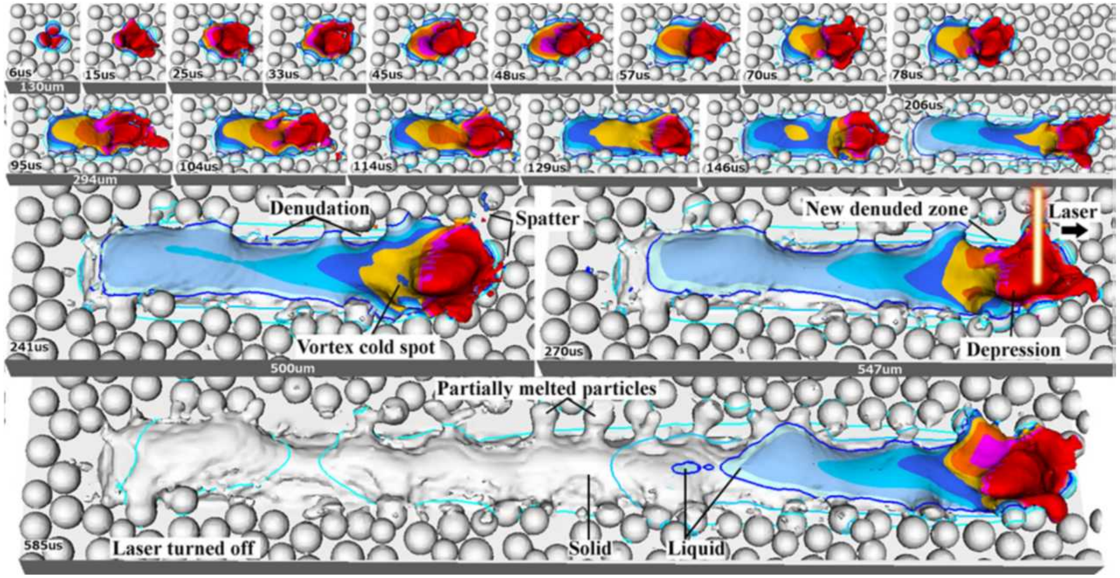
Promise of metals AM:

- Disruptive technology that allows simultaneous creation of optimized part geometries and materials-by-design
- Ideal for low volume, high value, high consequence, complex parts
- Inherently flexible and agile
- Ability to create near-net shape parts

Rather than focusing on process-modification to print materials, apply a materials-centric approach to enable desired outcome with available tools.

Challenges of Metals AM

- Conventional alloy AM parts have (mostly) highly variable and sub-optimal mechanical properties, prohibiting their widespread use.
- Current processing-based qualification approaches focus on optimizing process parameters to improve performance and reliability of conventional alloys (e.g., steels)



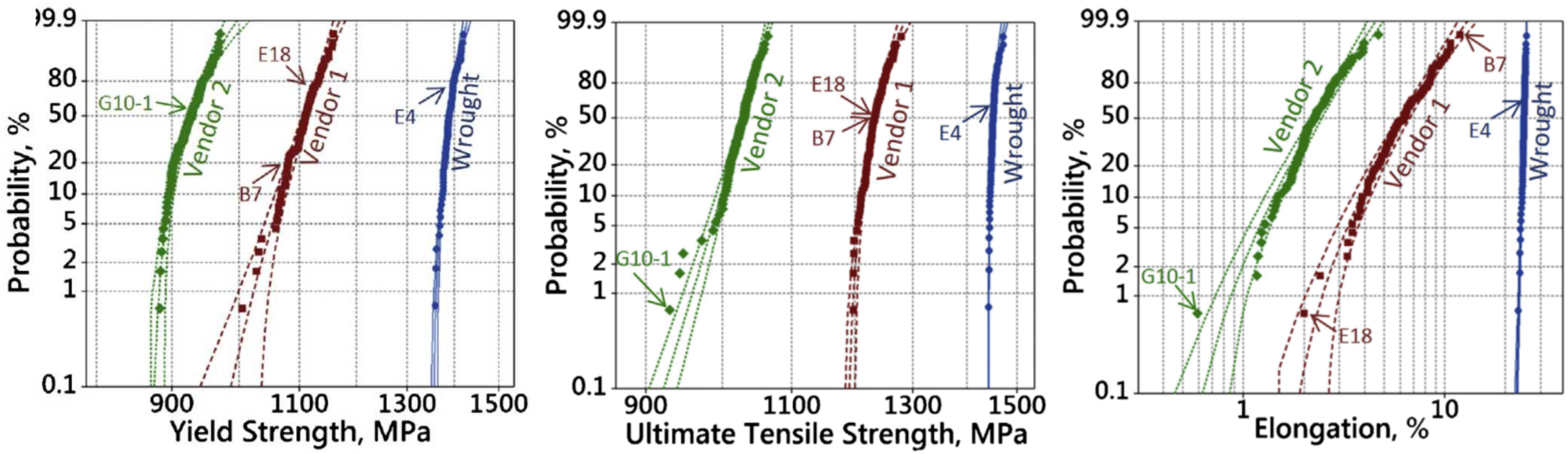
S. A. Khairallah, et al., Acta Mater., 2016

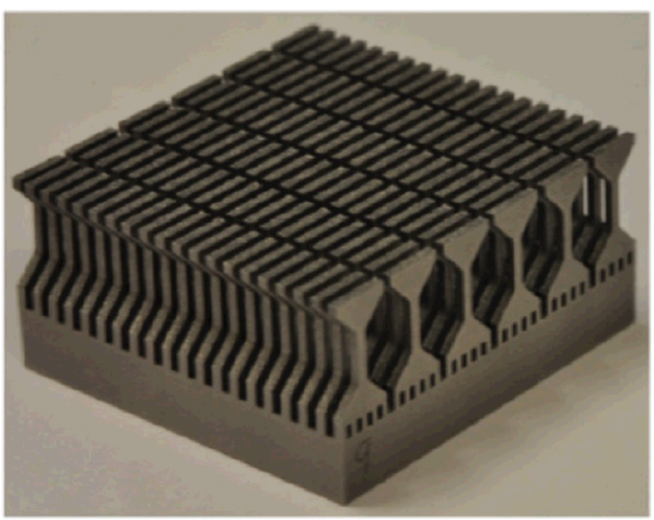
This approach has only had limited success…. Why?

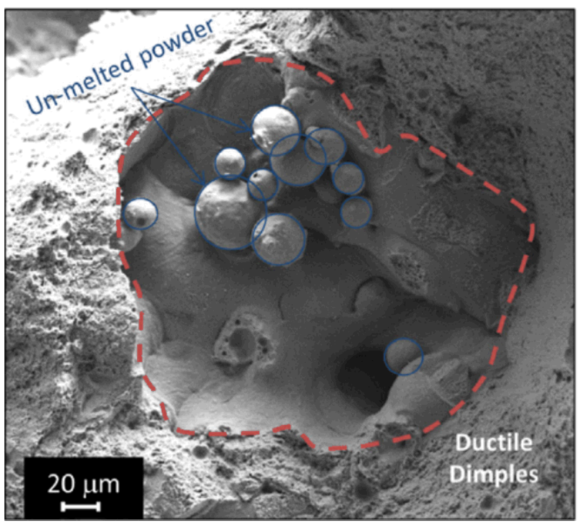
Conventional alloys degrade from melt/re-solidification processes and were never intended to be structural components in cast forms!

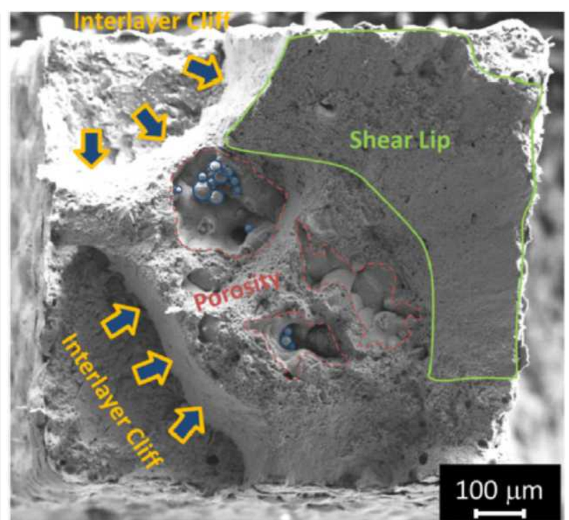
Challenges of Metals AM (17-4PH steel)

B.C. Salzbrenner, et al., J. Mat. Proc. Tech., 2017



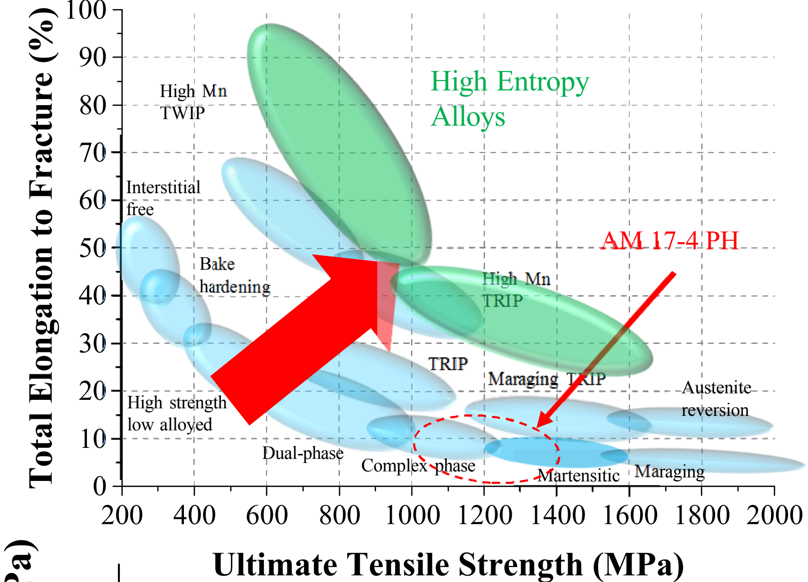


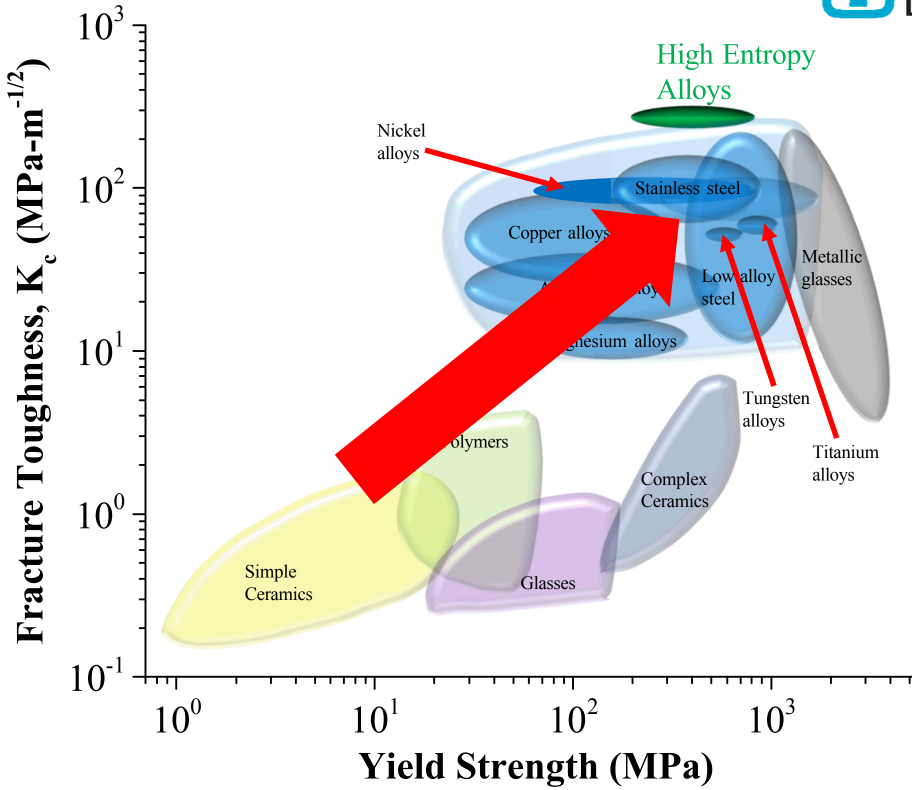




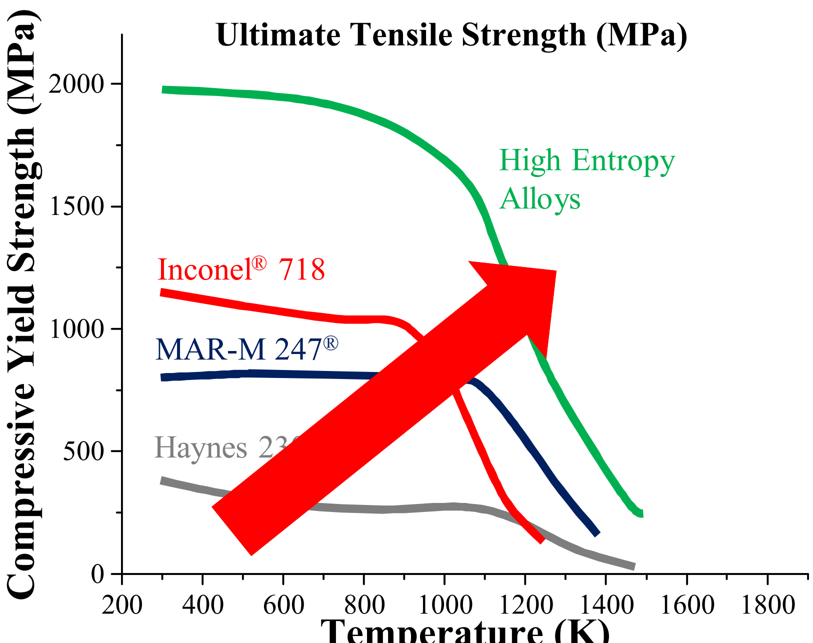
High Entropy Alloys: A Materials Approach to Metals AM?

D. Raabe, et al., Steel Res. Int., 2015





B. Gludovtaz, et al., Science., 2014



D. Miracle, et al., Acta Mater., 2017

HEAs have properties exceeding most conventional alloys, suggesting improved resistance to failures associated with defects in AM parts.

Goal: demonstrate these alloys as a materials-based approach to ***achieve the promise of metals AM, i.e. insertion into structural applications***.

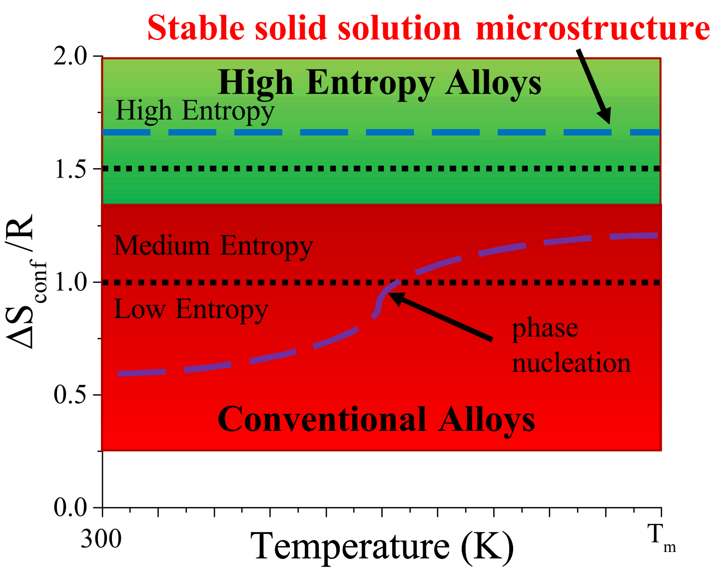
Why might HEAs be a materials-based approach?

High Entropy Alloys: primarily solid solutions containing 5+ alloying constituents, where the solutions have high configurational entropy ($\Delta S_{conf} > 1.4R$, approx. 12 J/mol-K) .

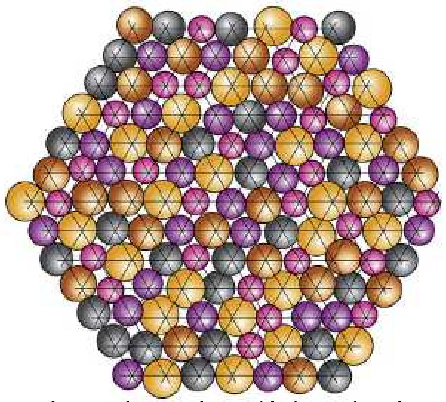
High configurational entropy is believed to thermodynamically suppresses phase separation, a primary route for degradation of mechanical properties.

Competition between Gibbs energy for solid solution and intermetallic formation

$$\Delta G^{SS} < \Delta G^{IM} \quad \rightarrow \quad \Delta S^{SS} > \frac{\Delta H^{IM} - \Delta H^{SS}}{T}$$



D. Miracle et al., Entropy, 2014



Disordered solid solution

D. Miracle et al., Acta Mat., 2017

Thermodynamically stable and predictable solid solution microstructure, independent of processing!

Ideal for layer-by-layer melting/re-melting of AM…

This hypothesis remains controversial and highly-debated, and why the proposed work has high scientific impact potential.

Reports suggest HEAs are better as AM materials!

- Due to their stable structures and remarkable properties ***in as-cast form***, HEAs are uniquely suited for AM processing..
- Recent literature has shown proof-of-concept of AM consolidation of HEAs but detailed investigations into the fundamental process-structure-property relationships are lacking.

Table I – Strength and ductility of conventional and AM processed stainless steel (red) and a comparable HEA (blue). Unlike the steel, the HEA shows insensitivity to processing method, with superior mechanical properties and lower variability.

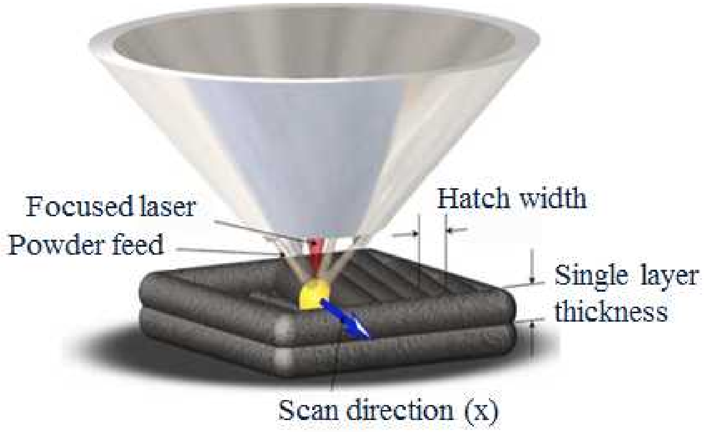
Condition	Ultimate Strength (MPa)	Strain-to-Failure (%)
Conventional 17-4 PH	1450 +/- 1%	22 +/- 5%
Laser AM 17-4 PH	1125 +/- 16%	5.5 +/- 82%
Conventional AlCoCrFeNi HE alloy	1426 +/- 9%	5.6 +/- 34%
E-beam AM AlCoCrFeNi HE alloy	1670 +/- 4%	26.5 +/- 25%

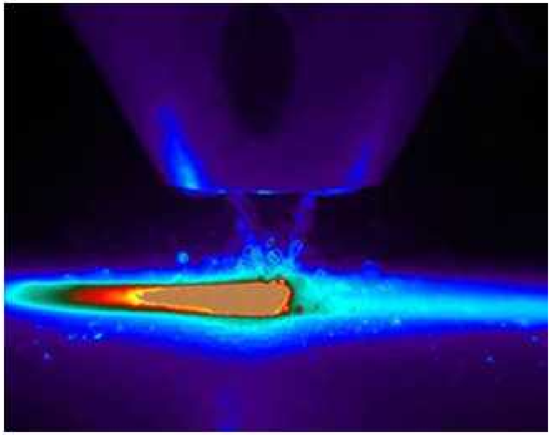
B.C. Salzbrenner, et al., J. Mat. Proc. Tech., 2017

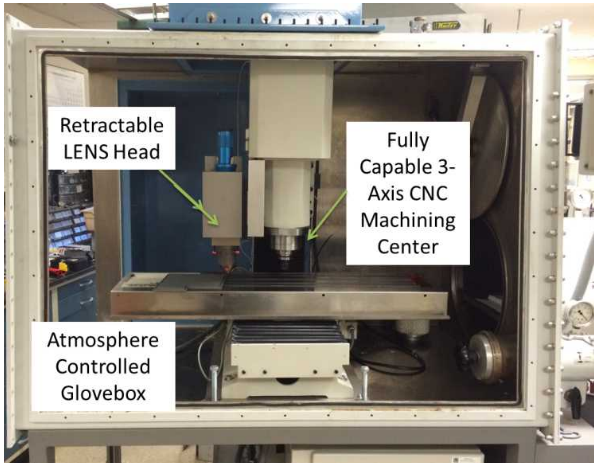
H. Shiratori, et al, Mater. Sci. Eng. A, 2016

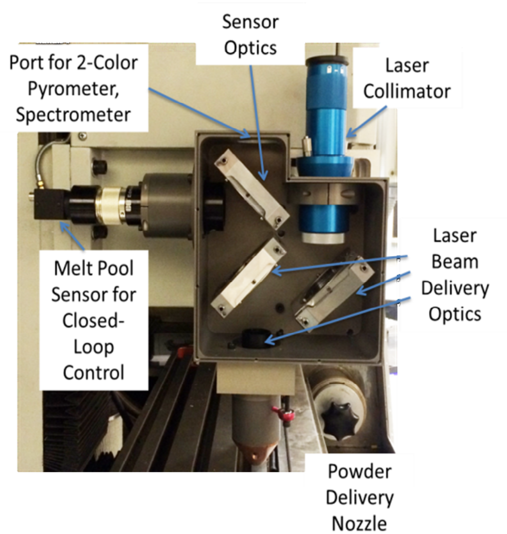
In-house processing capabilities:

Laser Engineered Net Shaping (LENS)



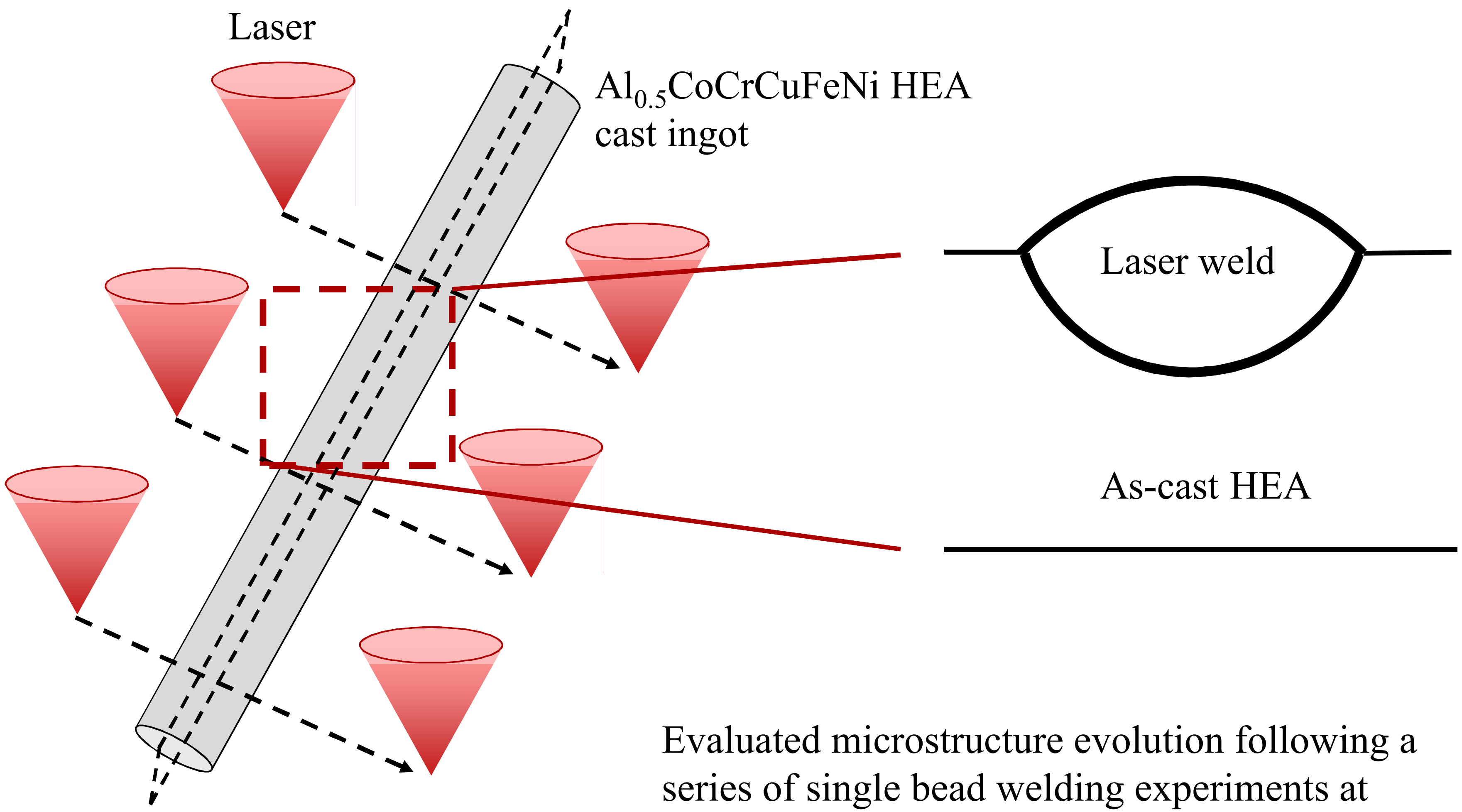






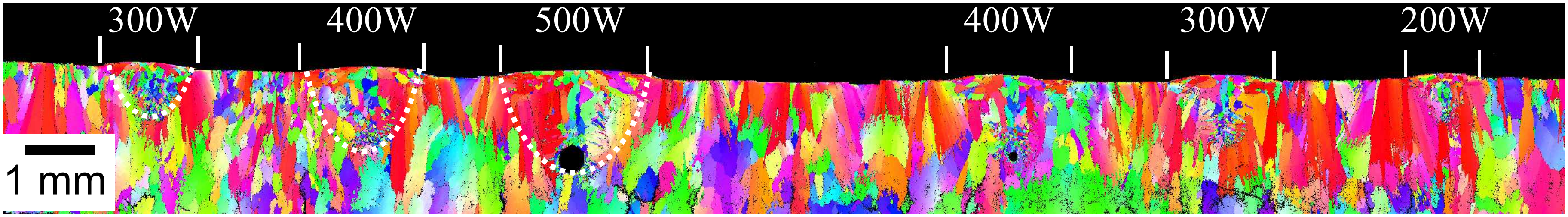
- Open architecture Laser Engineered Net Shaping (LENS) apparatus for multi-material and custom alloy printing.
- 2-color pyrometer and FLIR cameras for in situ melt pool geometry and temperature measurements.
- High temperature induction coil for in situ annealing.
- Hybrid AM and subtractive processing.
- Controlled powder feed rate with up to 5 independent powder chemistries – enable in situ alloy design studies.

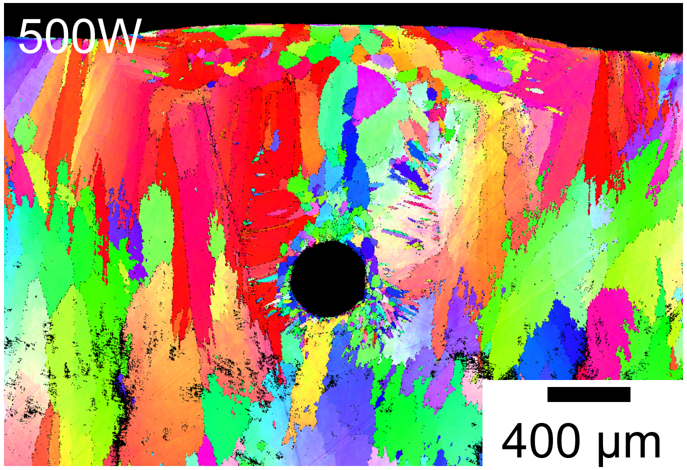
Microstructural stability analysis of HEAs during rapid solidification of AM

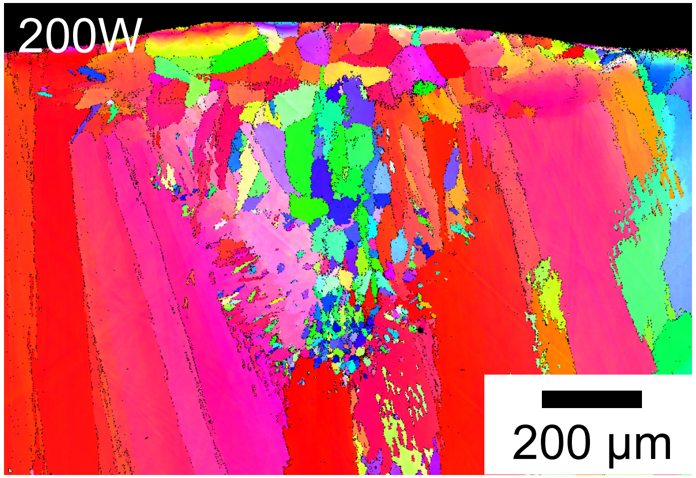


Evaluated microstructure evolution following a series of single bead welding experiments at varying laser power

Stable single-phase microstructures retained following laser melting experiments





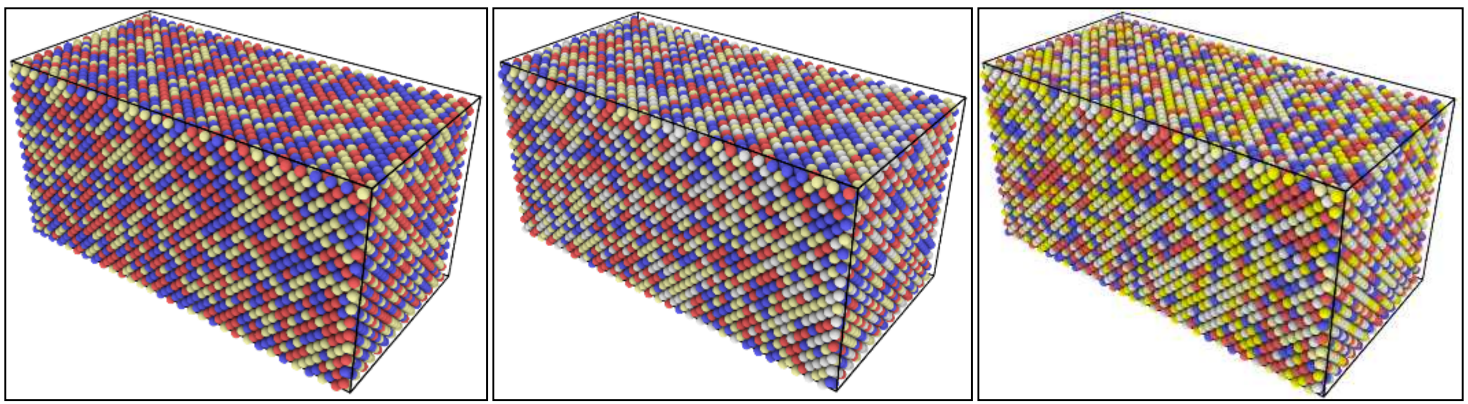


Composition unaffected, and single phase microstructure maintained: validation of hypothesis!

Novel Simulations Tools for Alloy Design and Optimization

- Molecular Dynamics (MD) effort to develop “**big data**” tool to enable parametric **alloy optimization**
- These tools can also enable new alloy development, and insights about the stability of hyper-dimensional alloys with high configurational entropy

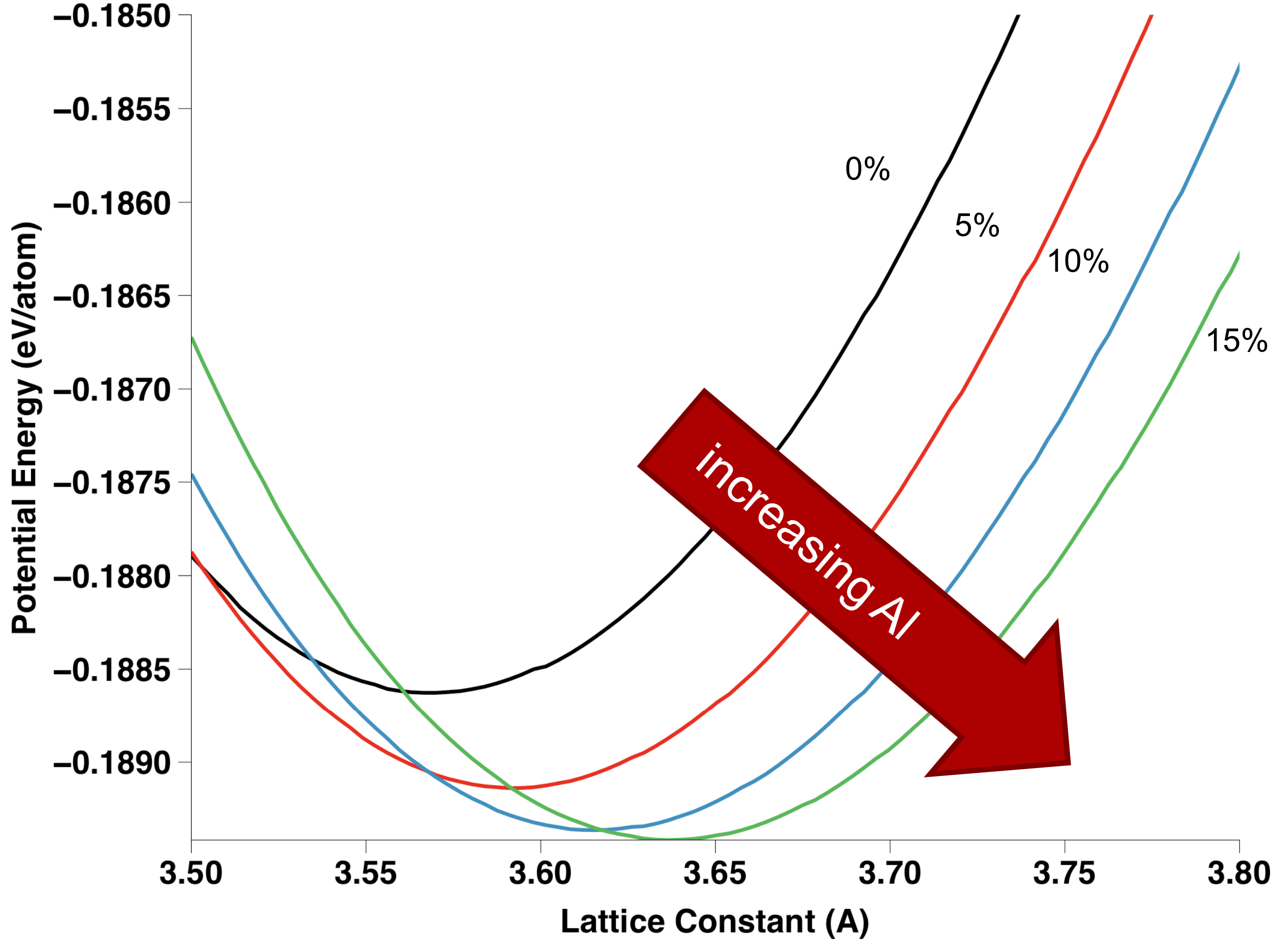
increasing entropy →



FeNiCo | FeNiCoMn | FeNiCoCrMn

Atomistic (MD) simulations snapshots from investigations of HE alloy stability

Novel Tools for Alloy Design and Optimization

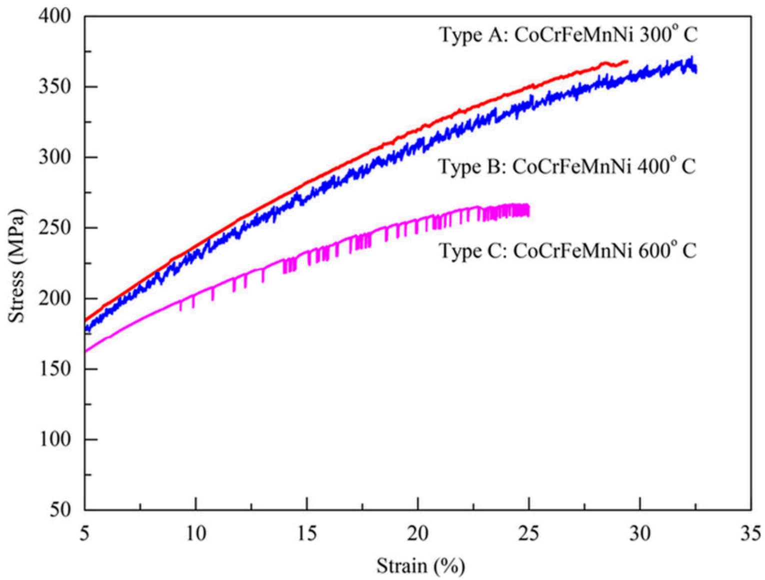


Atomistic (MD) simulations will enable new thermodynamic predictions of stable HEAs

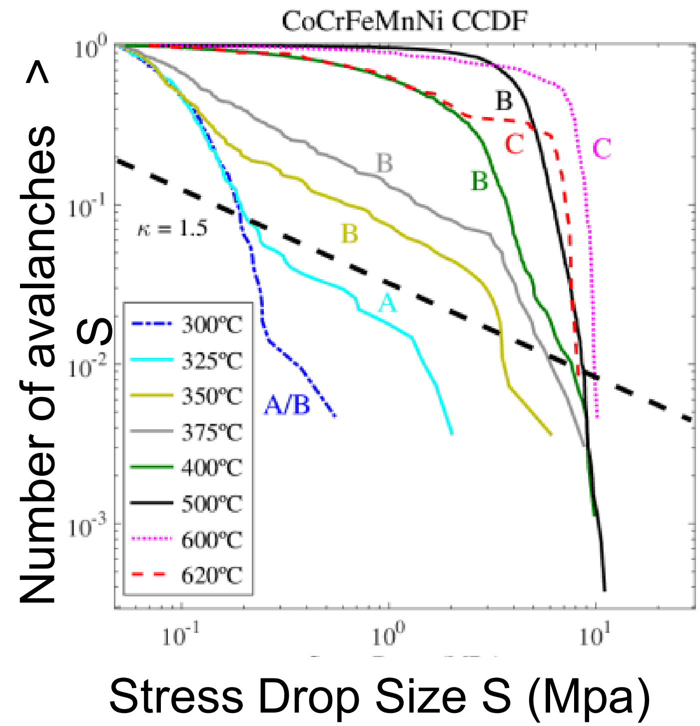
Mechanical Properties Characterization

Sandia National Laboratories

- Collaboration with Prof. Karin Dahmen at UIUC.
- Support model development for deformation behavior of HE alloys.



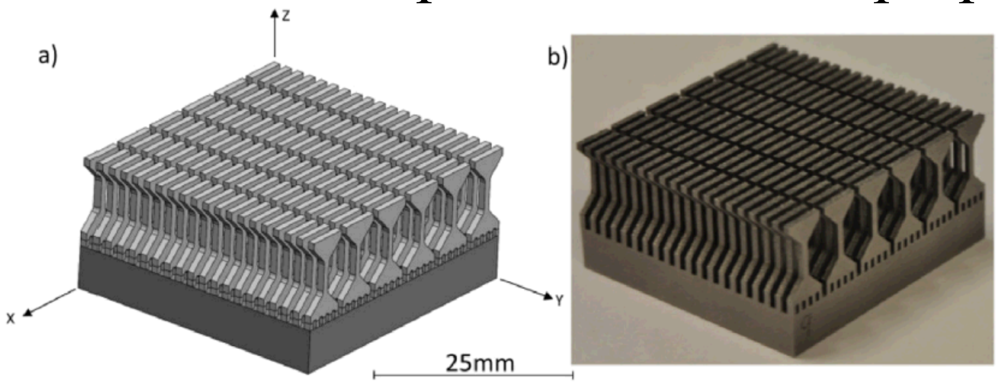
Previous work has shown that serrations in stress-strain curves correlate predictively with temperature-dependent deformation modes in HE alloys.



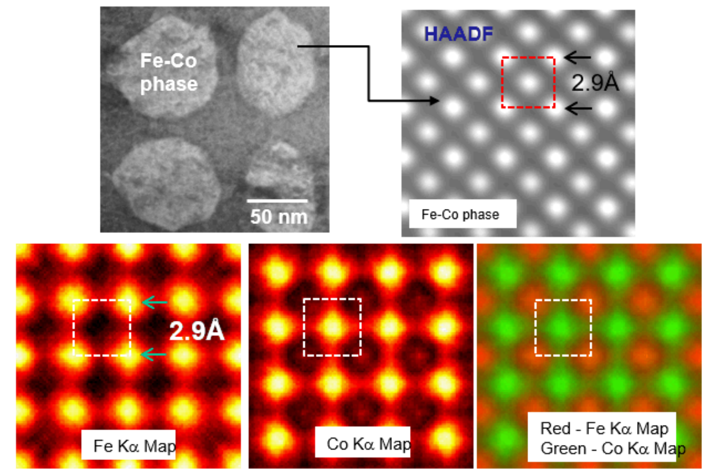
Non-destructive method of predicting strength in HEAs

Future Work for TMS 2019!

Consolidate pre-alloyed CoCrFeMnNi HEA powders using LENS and determine process-structure-properties relationships



Fe-Co intermetallic phase



AC-STEM with atomic-scale EDS (Lu *et al.*, *Sci. Rep.* 2014)

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Pre-alloyed CoCrFeMnNi powder

Co	21.40%
Cr	18.38%
Fe	19.87%
Mn	19.23%
Ni	21.09%

Summary

Materials-centric approach to additive manufacturing through High Entropy Alloys – method for studying HEA solidification behavior.

Laser melting experiments on $Al_{0.5}CoCrCuFeNi$ HEA cast ingot showed a retention of the single phase structure, suggesting no significant change in composition.

Molecular Dynamics simulations are enabling novel routes for high entropy alloy design and optimization – rapid exploration of composition space.

Future goals: Process and characterize samples of consolidated CoCrFeMnNi pre-alloyed powders