Stony Brook University

# A Multimodal Approach:
## How to Apply Synchrotron X-ray Characterization and Beyond to Tackle YOUR Research Challenges?


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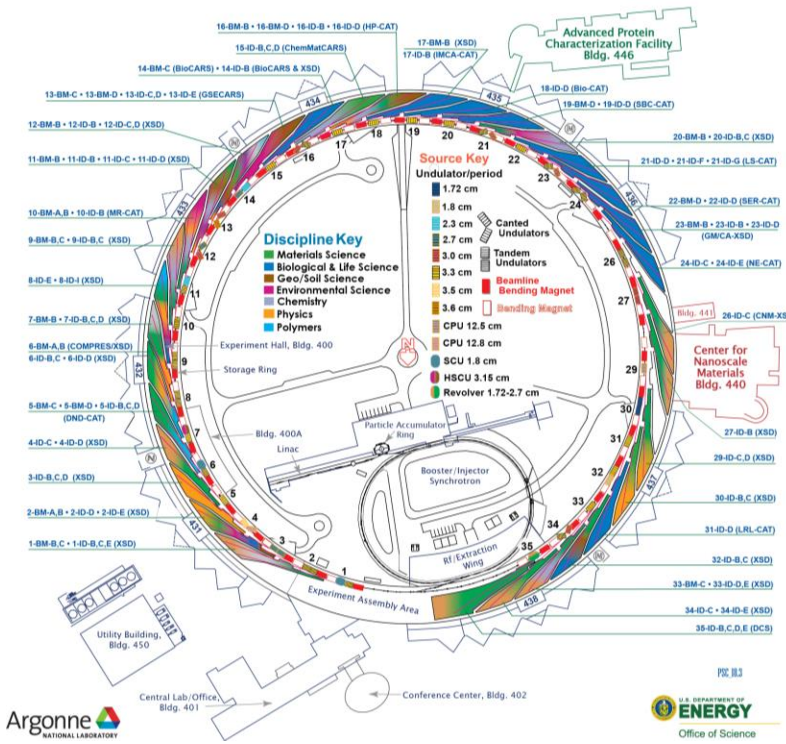
Brookhaven National Laboratory


25 th National School on Neutron and X-ray Scattering, August 6 – August 18, 2023
Thursday August 17 th, Multi-Modal Experiments

FAR BEYOND

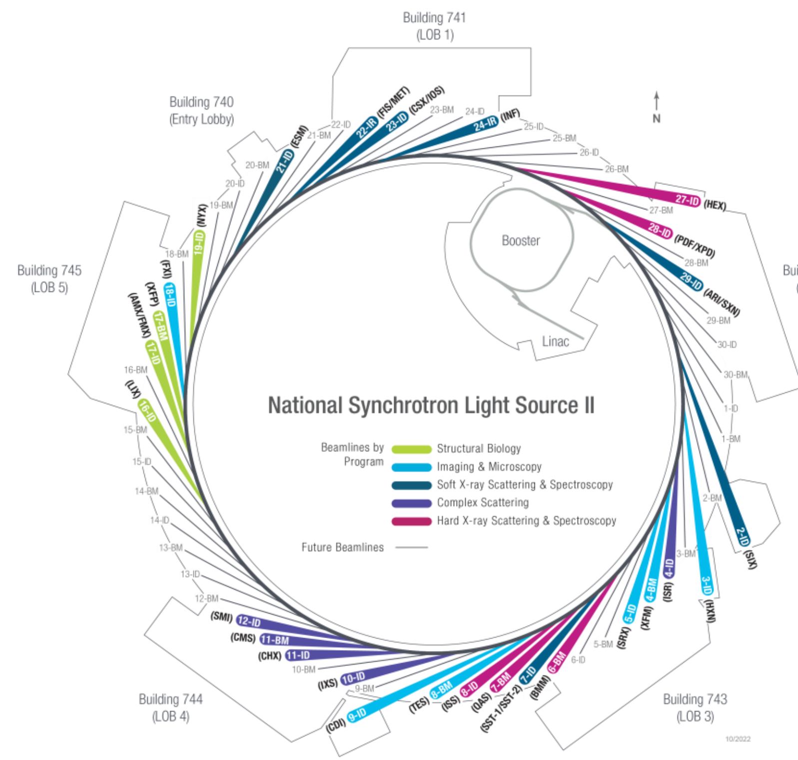
# A suite of cutting-edge characterization tools!

**Advanced Photon Source (APS)**




https://www.aps.anl.gov/Beamlines/Beamlines-Map

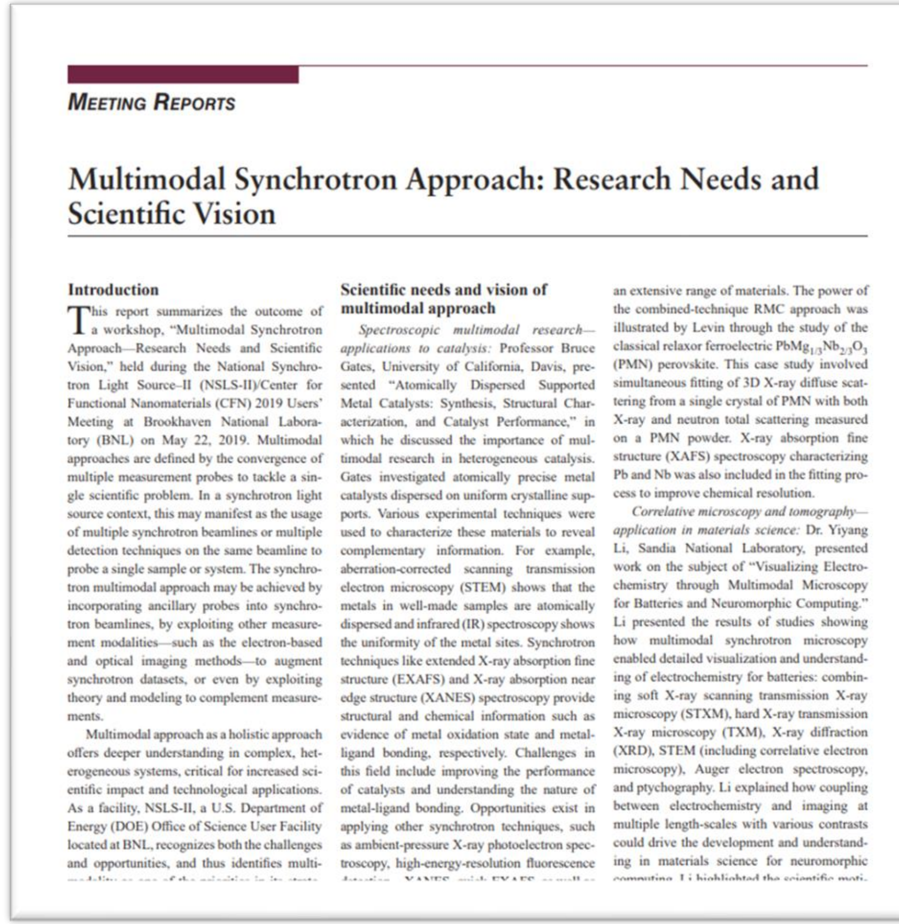
**National Synchrotron Light Source II (NSLS-II)**




https://www.bnl.gov/nsls2/about-nsls-ii.php

# A good reference: 2020 Workshop Report

- **"Multimodal Synchrotron Approach: Research Needs and Scientific Vision"** Yu-Chen Karen Chen-Wiegart, Iradwikanari Waluyo, Andrew Kiss, Stuart Campbell, Lin Yang, Eric Dooryhee, Jason R. Trelewicz, Yiyang Li, Bruce Gates, Mark Rivers, Kevin G. Yager *Synchrotron Radiation News (2020) DOI: 10.1080/08940886.2020.1701380*



MEETING REPORTS

## Multimodal Synchrotron Approach: Research Needs and Scientific Vision

### Introduction

This report summarizes the outcome of a workshop, "Multimodal Synchrotron Approach—Research Needs and Scientific Vision," held during the National Synchrotron Light Source–II (NSLS-II)/Center for Functional Nanomaterials (CFN) 2019 Users' Meeting at Brookhaven National Laboratory (BNL) on May 22, 2019. Multimodal approaches are defined by the convergence of multiple measurement probes to tackle a single scientific problem. In a synchrotron light source context, this may manifest as the usage of multiple synchrotron beamlines or multiple detection techniques on the same beamline to probe a single sample or system. The synchrotron multimodal approach may be achieved by incorporating ancillary probes into synchrotron beamlines, by exploiting other measurement modalities—such as the electron-based and optical imaging methods—to augment synchrotron datasets, or even by exploiting theory and modeling to complement measurements.

Multimodal approach as a holistic approach offers deeper understanding in complex, heterogeneous systems, critical for increased scientific impact and technological applications. As a facility, NSLS-II, a U.S. Department of Energy (DOE) Office of Science User Facility located at BNL, recognizes both the challenges and opportunities, and thus identifies multi-

### Scientific needs and vision of multimodal approach

*Spectroscopic multimodal research—applications to catalysis:* Professor Bruce Gates, University of California, Davis, presented "Atomically Dispersed Supported Metal Catalysts: Synthesis, Structural Characterization, and Catalyst Performance," in which he discussed the importance of multimodal research in heterogeneous catalysis. Gates investigated atomically precise metal catalysts dispersed on uniform crystalline supports. Various experimental techniques were used to characterize these materials to reveal complementary information. For example, aberration-corrected scanning transmission electron microscopy (STEM) shows that the metals in well-made samples are atomically dispersed and infrared (IR) spectroscopy shows the uniformity of the metal sites. Synchrotron techniques like extended X-ray absorption fine structure (EXAFS) and X-ray absorption near edge structure (XANES) spectroscopy provide structural and chemical information such as evidence of metal oxidation state and metal-ligand bonding, respectively. Challenges in this field include improving the performance of catalysts and understanding the nature of metal-ligand bonding. Opportunities exist in applying other synchrotron techniques, such as ambient-pressure X-ray photoelectron spectroscopy, high-energy-resolution fluorescence

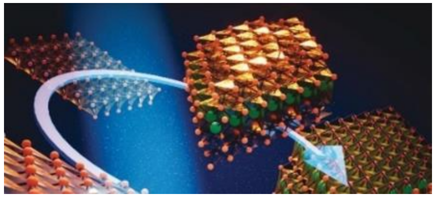
an extensive range of materials. The power of the combined-technique RMC approach was illustrated by Levin through the study of the classical relaxor ferroelectric $PbMg_{1/3}Nb_{2/3}O_3$ (PMN) perovskite. This case study involved simultaneous fitting of 3D X-ray diffuse scattering from a single crystal of PMN with both X-ray and neutron total scattering measured on a PMN powder. X-ray absorption fine structure (XAFS) spectroscopy characterizing Pb and Nb was also included in the fitting process to improve chemical resolution.

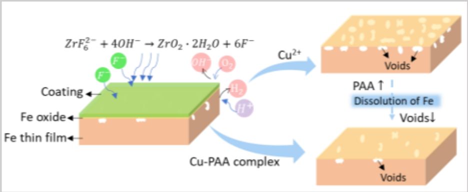
*Correlative microscopy and tomography—application in materials science:* Dr. Yiyang Li, Sandia National Laboratory, presented work on the subject of "Visualizing Electrochemistry through Multimodal Microscopy for Batteries and Neuromorphic Computing." Li presented the results of studies showing how multimodal synchrotron microscopy enabled detailed visualization and understanding of electrochemistry for batteries: combining soft X-ray scanning transmission X-ray microscopy (STXM), hard X-ray transmission X-ray microscopy (TXM), X-ray diffraction (XRD), STEM (including correlative electron microscopy), Auger electron spectroscopy, and ptychography. Li explained how coupling between electrochemistry and imaging at multiple length-scales with various contrasts could drive the development and understanding in materials science for neuromorphic

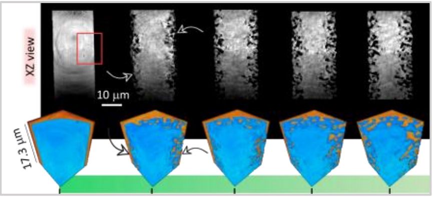
# Our Research Program on Functional Materials with Synchrotron X-ray Analysis

**1) Materials by Design:**
Nano-Architectured Porous Metals and Metallic Composites

**2) Sustainable Energy:**
Multimodal Synchrotron Studies of Energy Storage






**4) Sustainable Manufacturing:**
Environmentally friendly functional coating

**3) Sustainable Energy:**
Interfacial Processes of Materials in Molten Salts and Extreme Environments

# WIIFM?
# What's in it for me?

- What is a multimodal approach?
- Why we care about it?

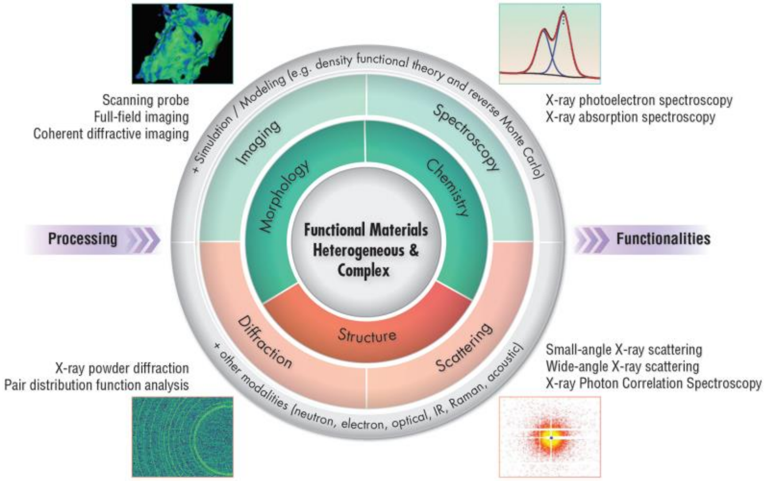
\- *Research example: Conversion coating*

- Ways to frame multimodal analysis.

\- *Research example: Battery*

- Beyond synchrotron
  - Other experimental modalities
  - Experiment – simulation feedback loop
  - Data science opportunities

\- *Research example: Nanoporous metals*



# 2-Min You Talk!

## *Talk to your neighbor(s):*

1) **What is your research topic?** (An "elevator pitch")
2) What are the **main techniques (2-5 of them)** you use to characterize them? (Name at least one X-ray or neutron technique, if possible!)

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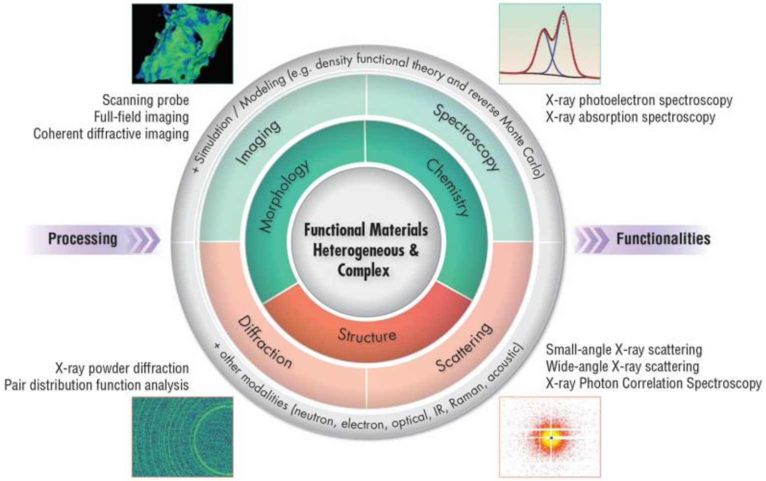
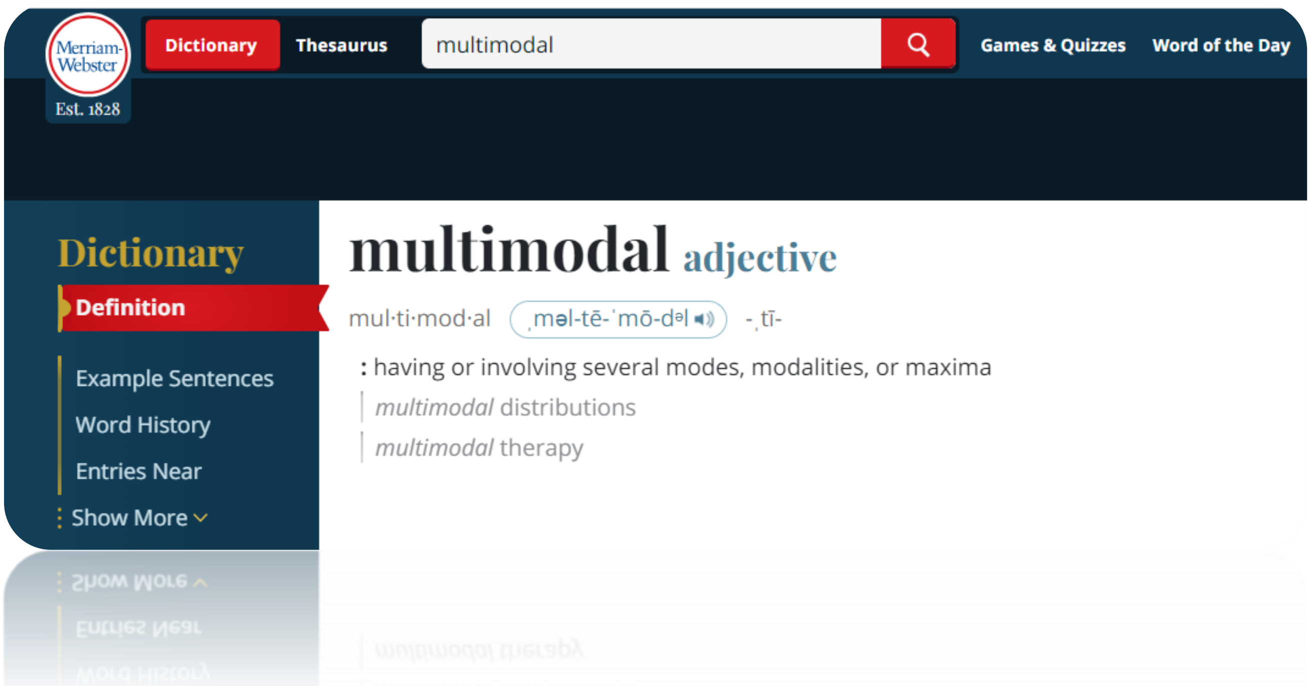


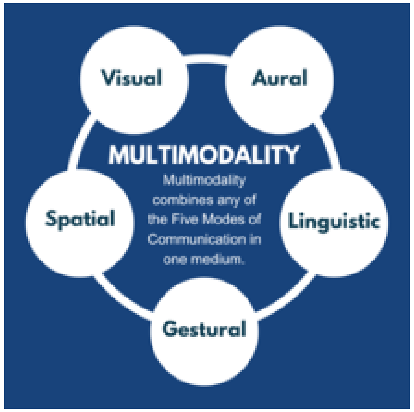
# What is multimodal?

- Dictionary definition:
  - https://www.merriam-webster.com/dictionary/multimodal

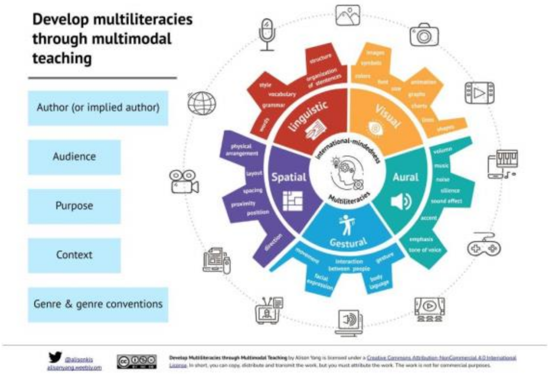


# Multimodality everywhere!

## Multimodal Pedagogy/Teaching

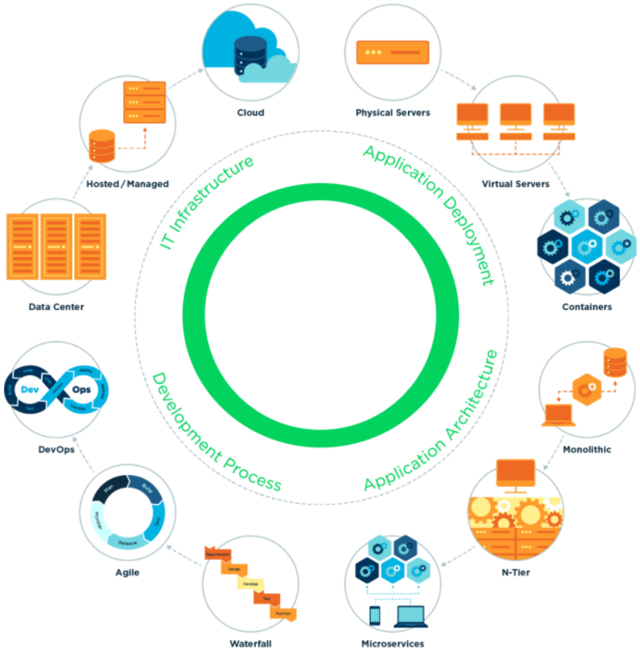



https://en.wikipedia.org/wiki/Multimodal_pedagogy




https://alisonyang.com/multimodal-teaching/

## Multimodal IT




https://www.suse.com/c/the-rise-of-multimodal-it-and-what-it-means-to-you/

## Multimodal Customer Experience

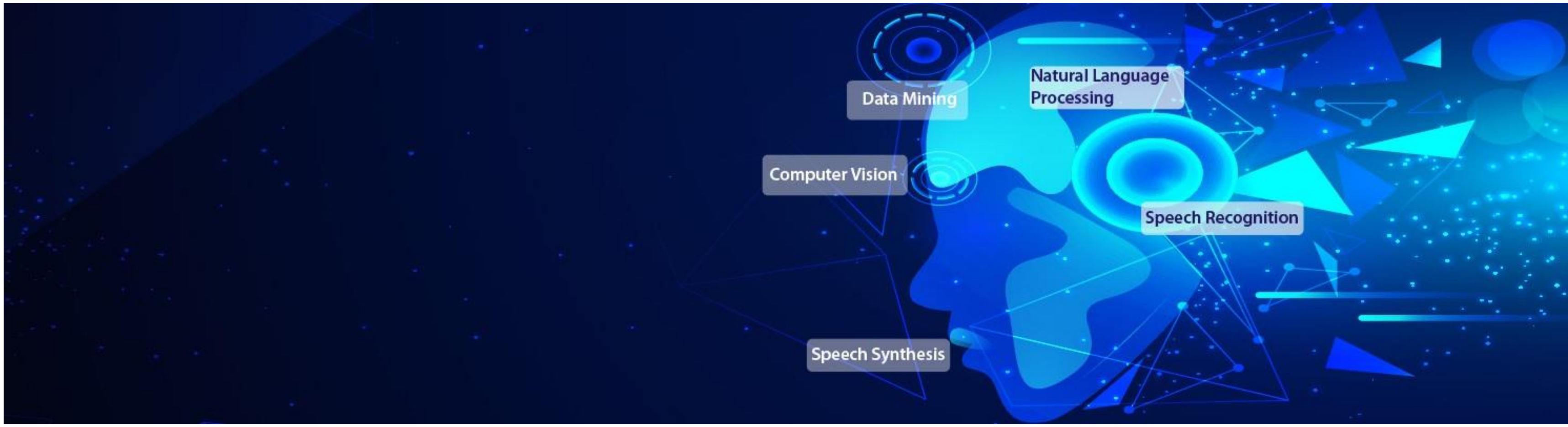

https://www.uniphore.com/blog/what-s-a-multimodal-customer-experience/

## Multimodal Transport

*Multimodal transport, also known as combined transport, is a transport system that involves the movement of goods using multiple modes of transport such as trucks, rail, air and ships.*

https://www.morethanshipping.com/what-is-multimodal-transport/

# Multimodal Artificial Intelligence!




https://www.aimesoft.com/multimodalai.html

Multimodal AI is a new AI paradigm, in which various data types (image, text, speech, numerical data) are combined with multiple intelligence processing algorithms to achieve higher performances. Multimodal AI often outperforms single modal AI in many real-world problems.

## Ecosystem!!

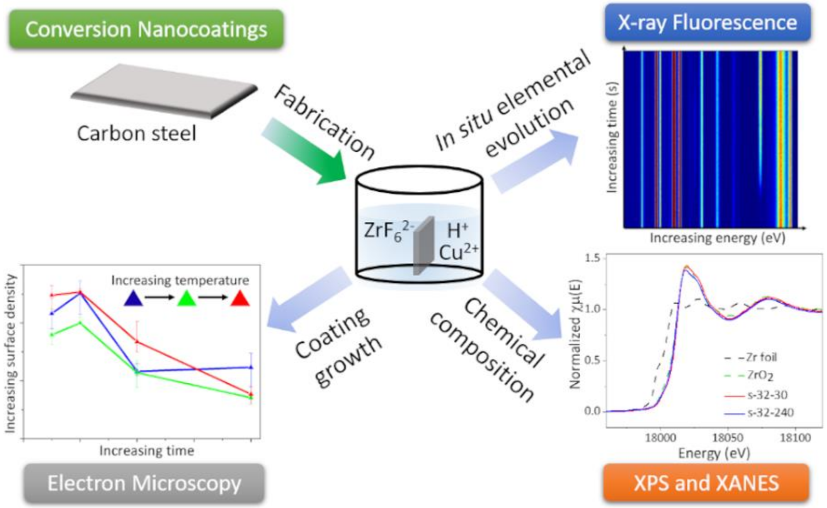
*What is the ecosystem of synchrotron (and neutron) characterization?*

# Multimodality – In the context of scientific research

- *"Multimodal approaches are defined by the convergence of multiple measurement probes to tackle a single scientific problem."*

*Karen Chen-Wiegart et al., Synchrotron Radiation News (2020)*

We have already been applying multimodal characterization from the beginning!




*Xiaoyang Liu, ACS Applied Nanomaterials, 2019*

# WIIFM?
# What's in it for me?

- What is a multimodal approach?
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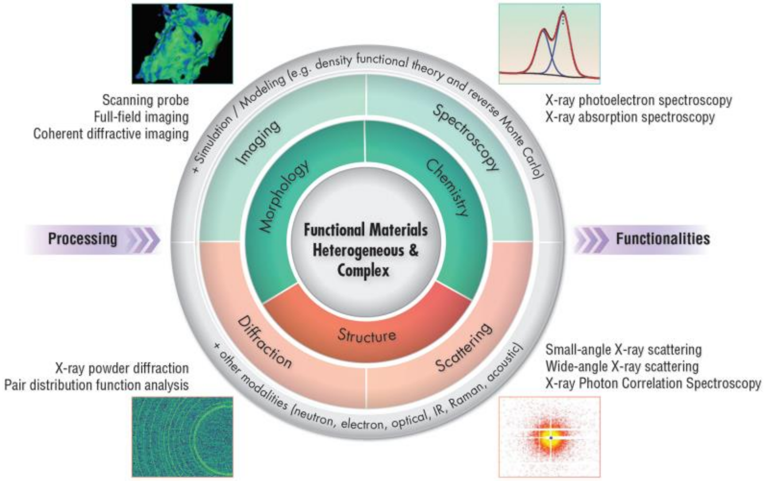
*- Research example: Conversion coating*

- **Ways to frame multimodal analysis.**

***- Research example: Battery***




- Beyond synchrotron
  - Other experimental modalities
  - Experiment – simulation feedback loop
  - Data science opportunities

*- Research example: Nanoporous metals*

# Why taking a multimodal approach?

Synchrotron offers a suite of the amazing characterization tools!!

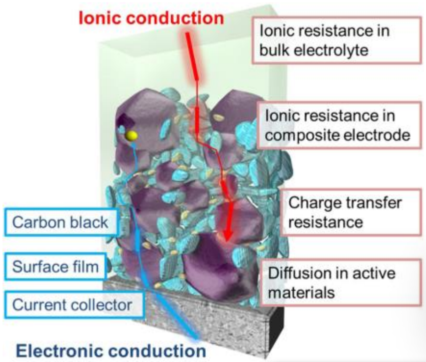
Awesome playground!!
Lots of opportunities!!
Unprecedent information!!

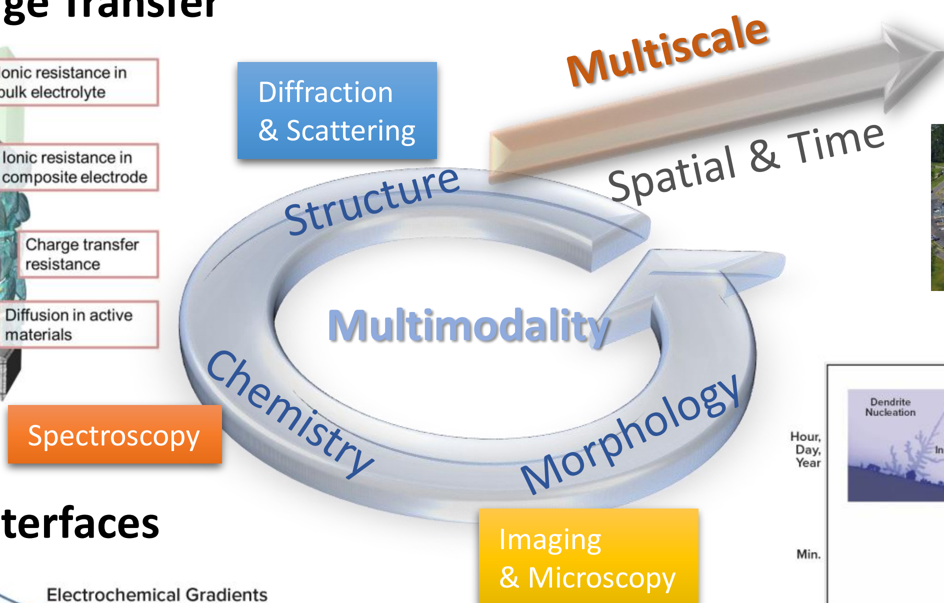
Easily get lost
– why am I doing what I am doing?

- Multimodal approach as a holistic approach offers deeper understanding in **complex, heterogeneous systems**, critical for increased scientific impact and technological applications.

# Research challenges complex, heterogeneous systems

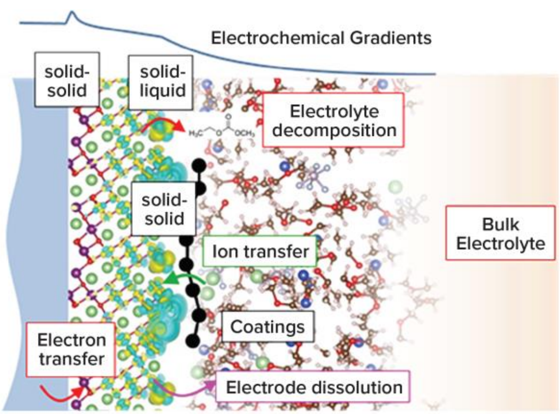
## Mass and Charge Transfer

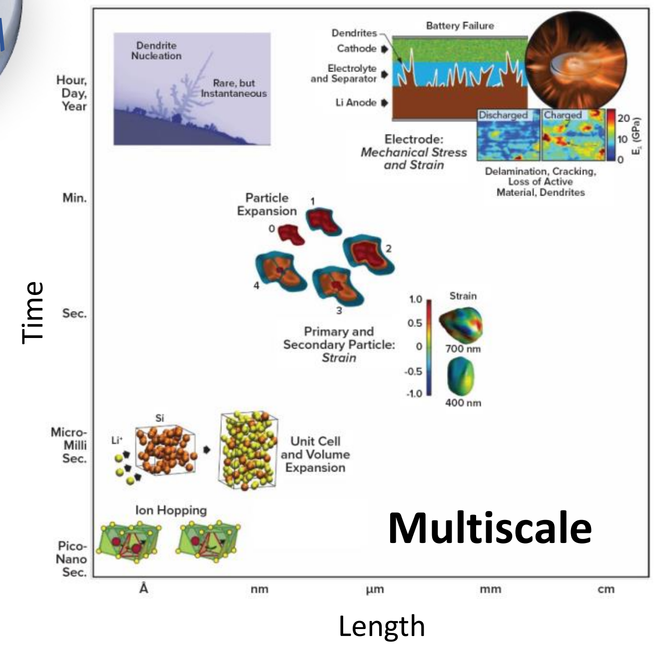



**Multimodal Synchrotron X-ray Characterization**

## Bulk and Interfaces



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1) What is your research topic? (An "elevator pitch")
2) What are the main techniques (2-5 of them) you use to characterize them? (Name at least one X-ray or neutron technique, if possible!)
3) **Why are you using them?**

   **What information can you get out of each of the techniques?**

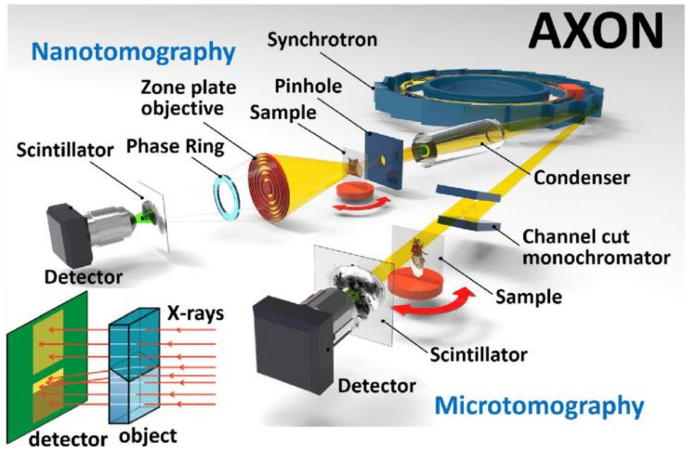
   **Are they complementary to each other?**

# Multimodal Synchrotron Approach

- In a synchrotron light source context, this may manifest as the usage of

1) **Multiple synchrotron beamlines** or
2) **Multiple detection techniques on the same beamline**

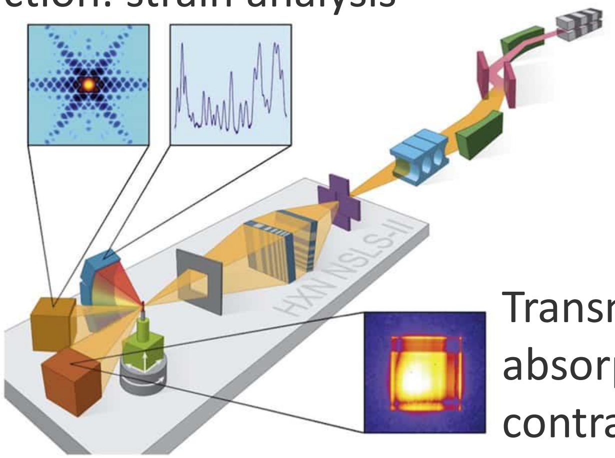
to probe a single sample or system.




Hwu, Y et al., BMC Biol 15, 122 (2017). https://doi.org/10.1186/s12915-017-0461-8

Fluorescence: elemental mapping

Diffraction: strain analysis




Transmission: absorption/phase-contrast imaging

Hanfei Yan et al., 2018 Nano Futures 2 011001 DOI 10.1088/2399-1984/aab25d

# Why using different beamlines?

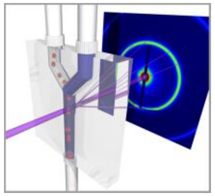
## Suite of beamlines with complementary techniques - enabling time-resolved, *operando*, multi-modal and multi-dimensional studies

**Scattering** | **Diffraction** | **Spectroscopy** | **Imaging**

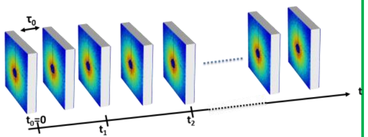
Structure

Chemistry

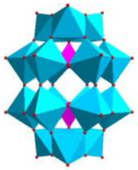
Morphology



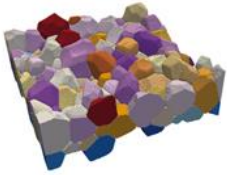
CMS



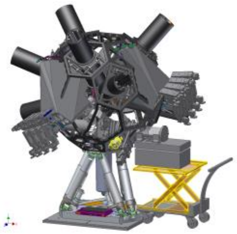
CHX

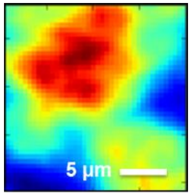


XPD

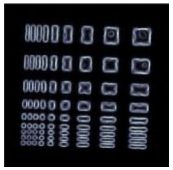


ISS



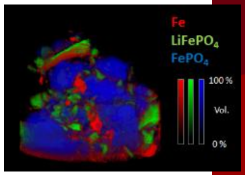



SRX



HXN

FXI




(XPD): X-ray Powder Diffraction

(ISS): Inner Shell Spectroscopy

(SRX): Sub-micron Resolution X-ray Spectroscopy

(CMS): Complex Materials Scattering

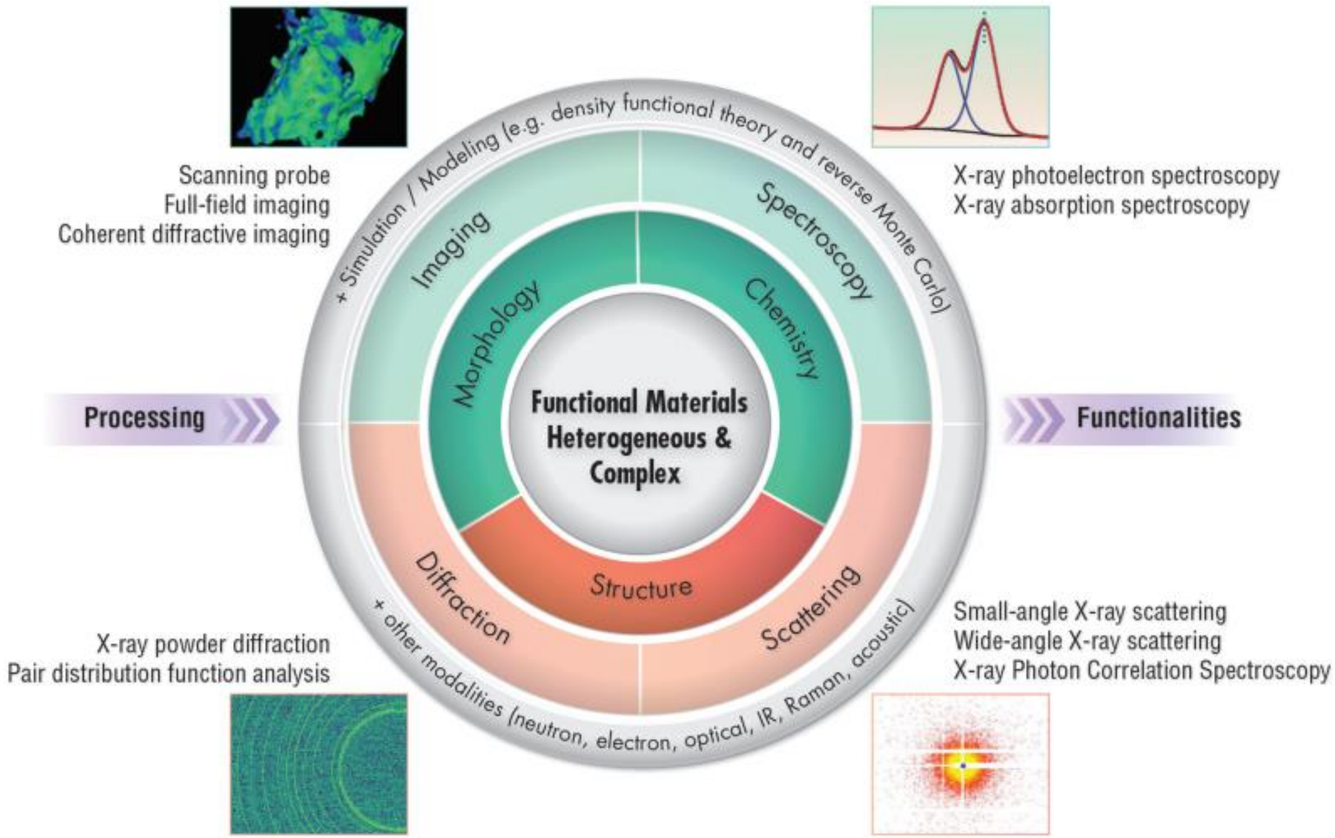
(HXN): Hard X-ray Nanoprobe

(CHX): Coherent Hard X-ray: XPCS

(FXI): Full-Field X-ray Imaging

**1. What is the processing – structure – property relationship? (How do we control the properties?)**

**2. How do the materials' morphology, chemistry and structure evolve as a function of time and processing/operating conditions?**

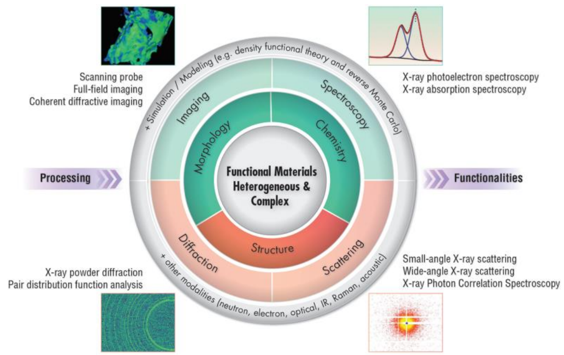



*Karen Chen-Wiegart et al., Synchrotron Radiation News (2020)*

# 2-Min You Talk!

*Talk to your neighbor(s):*

1) What is your res tch")
2) What are the ma you use to characterize them? (Name at echnique, if possible!)
3) Why are you usi
   What informatio the techniques?
   Are they comple

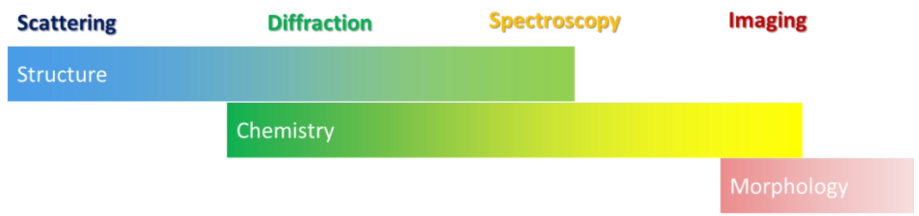



4) **Try to categorize them and see their connections:**

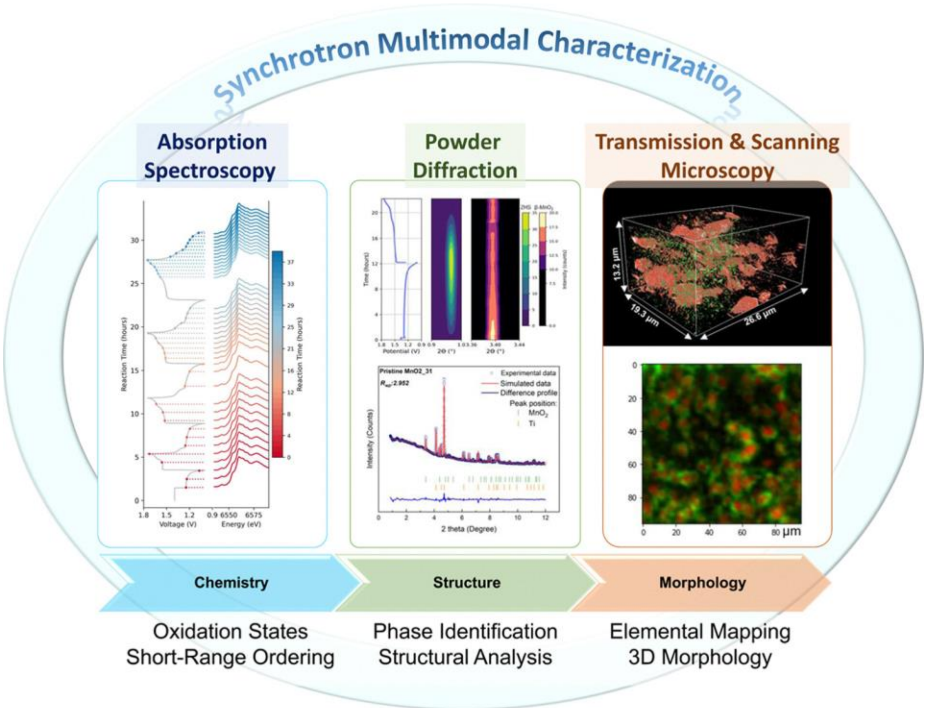
→ Building a mind-map/framework to think/plan your research

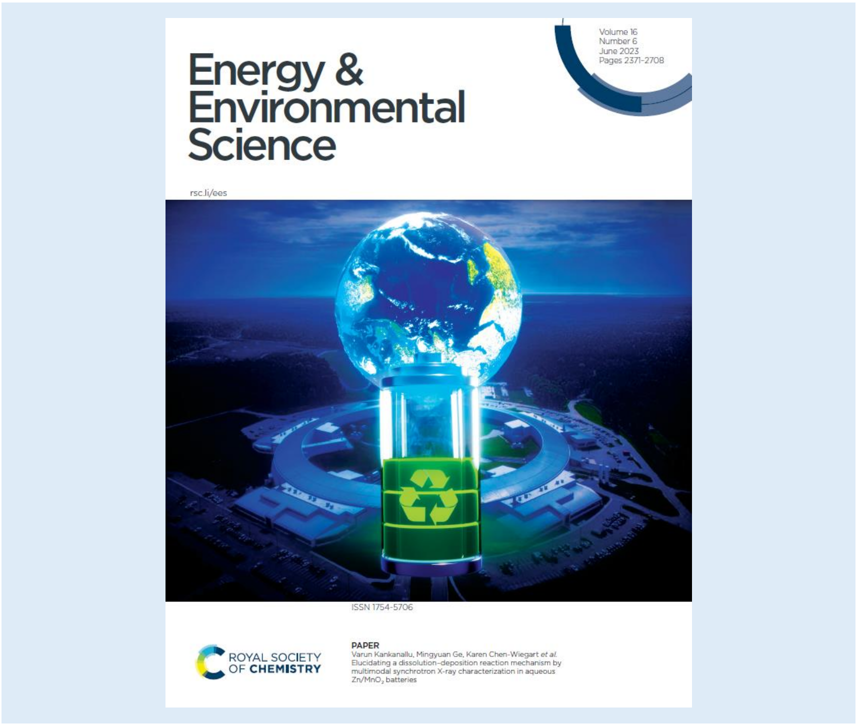
Avoid: I have a hammer, and thus everything looks like a nail!

Ask yourself: why am I using the technique, and what I am trying to get out of it?



# Towards better understanding of reaction mechanism by *operando* multi-modal X-ray synchrotron characterization

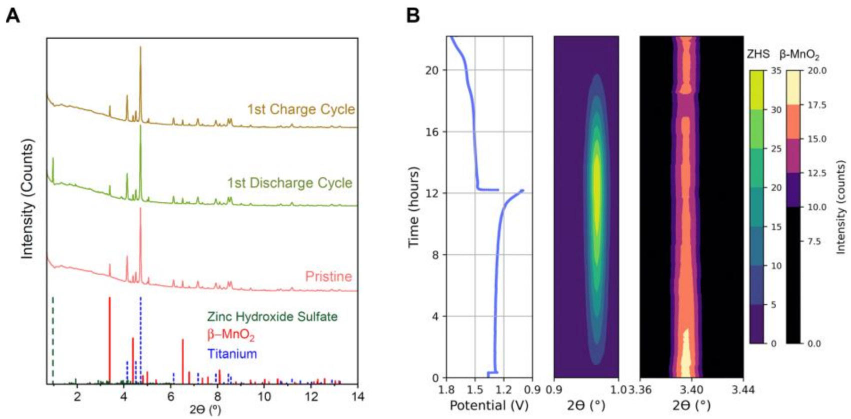



- *Varun R. Kankanallu, Xiaoyin Zheng, Denis Leschev, Nicole Zmich, Charles Clark, Cheng-Hung Lin, Hui Zhong, Sanjit Ghose, Andrew M. Kiss, Dmytro Nykypanchuk, Eli Stavitski, Esther S. Takeuchi, Amy C. Marschilok, Kenneth J. Takeuchi, Jianming Bai, Mingyuan Ge* and Yu-chen Karen Chen-Wiegart, Energy & Environmental Science (2023), DOI: 10.1039/D2EE03731A*

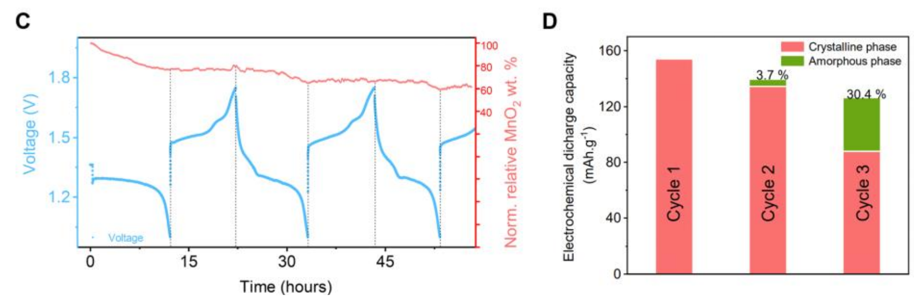
# *Operando* X-ray diffraction: Phase evolution

m2m#s
molecular to mesoscale




- Phase evolution of the β-$MnO_2$ electrode at the pristine, half-cycle and full-cycle states.
- The galvanostatic discharge–charge profile for the first cycle and its corresponding waterfall plot indicate the formation and disappearance of the zinc hydroxy sulfate (ZHS) phase and gradual reduction in $MnO_2$ peak intensity.

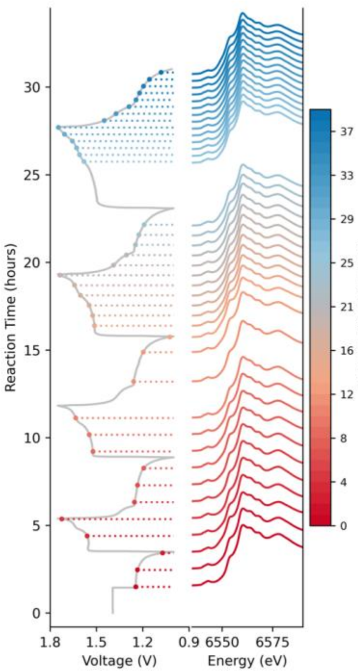
- Normalized relative $MnO_2$ weight percentage vs. the electrochemical potential for the first ~ 3 cycles.
- Relative capacity contribution by the amorphous phase in the 2nd and 3rd cycles.



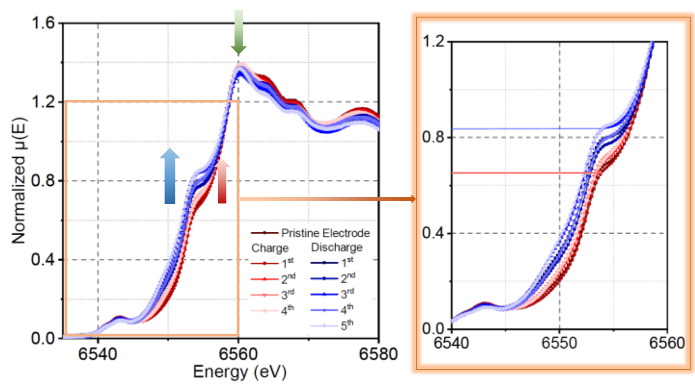


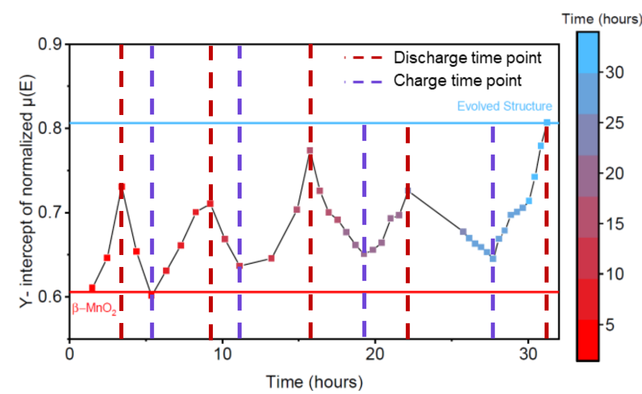
- *Varun R. Kankanallu, Xiaoyin Zheng, Denis Leschev, Nicole Zmich, Charles Clark, Cheng-Hung Lin, Hui Zhong, Sanjit Ghose, Andrew M. Kiss, Dmytro Nykypanchuk, Eli Stavitski, Esther S. Takeuchi, Amy C. Marschilok, Kenneth J. Takeuchi, Jianming Bai, Mingyuan Ge* and Yu-chen Karen Chen-Wiegart, Energy & Environmental Science (2023), DOI: 10.1039/D2EE03731A*

# *Operando* X-ray Absorption Spectroscopy (XAS): Gradual conversion of β-$MnO_2$ structure




- *Operando* X-ray absorption near edge structure (XANES) vs. the electrochemical potential and reaction time.




- Selected spectra points taken at the end of discharge and charge profiles: the variation in the pre-edge feature
- The Y-intercept of normalized XAS spectra near the pre-edge feature indicating the evolution of structure

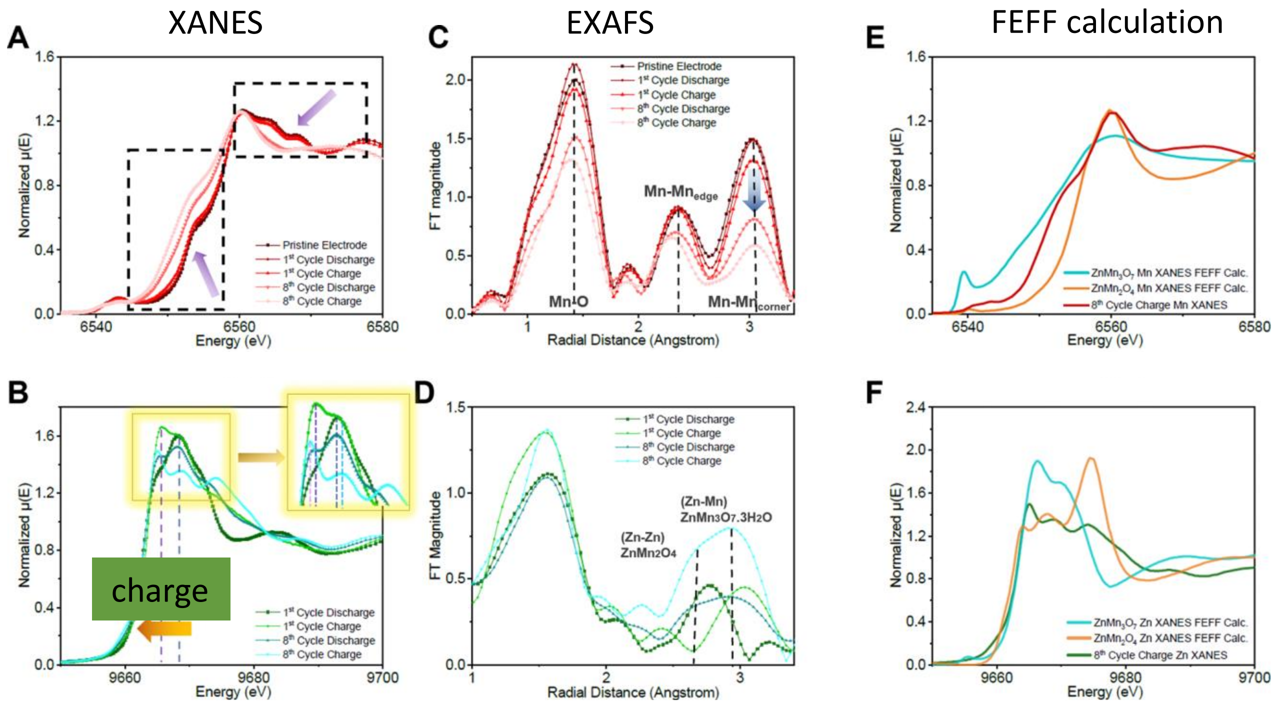

- *Varun R. Kankanallu, Xiaoyin Zheng, Denis Leschev, Nicole Zmich, Charles Clark, Cheng-Hung Lin, Hui Zhong, Sanjit Ghose, Andrew M. Kiss, Dmytro Nykypanchuk, Eli Stavitski, Esther S. Takeuchi, Amy C. Marschilok, Kenneth J. Takeuchi, Jianming Bai, Mingyuan Ge* and Yu-chen Karen Chen-Wiegart, Energy & Environmental Science (2023), DOI: 10.1039/D2EE03731A*

# *Ex situ* XAS of first and eight cycle: discharge and charge

## XANES & Extended X-ray Absorption Fine Structure (EXAFS)

Mn K-edge

Zn K-edge

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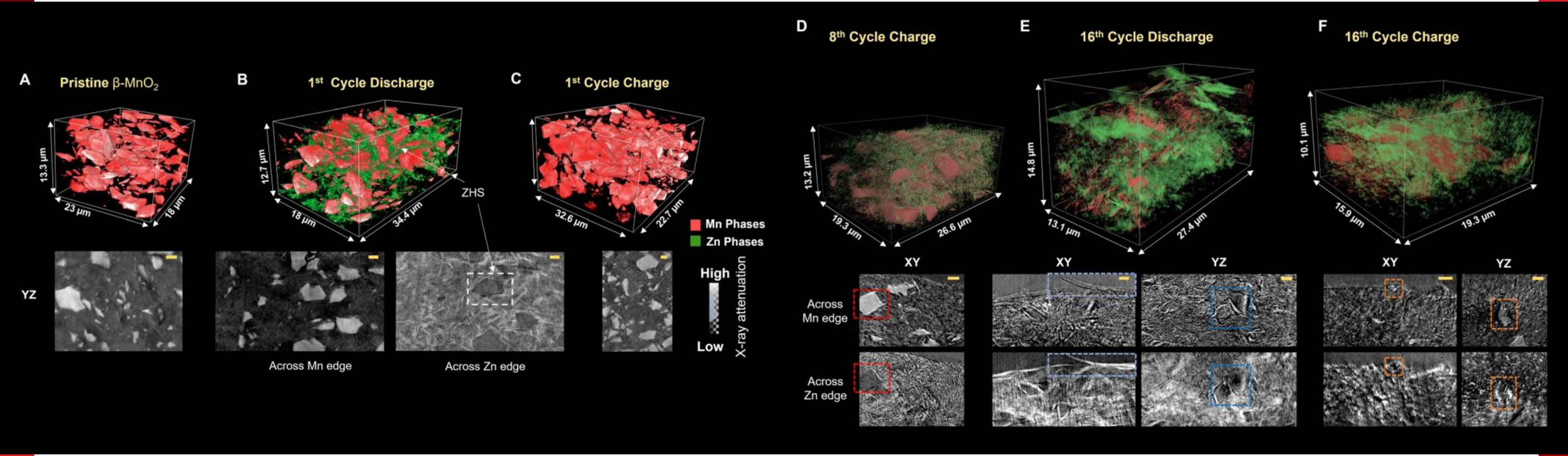
# Key morphological features of β-MnO2 electrodes

- **1st cycle discharge:** A dense growth of ZHS precipitate → **reversible upon charge**
- **8th cycle charge:** partial dissolution of β-$MnO_2$ particles and the Zn–Mn amorphous complex phase
- **16th cycle:** discharge and charge: dissolution of β -$MnO_2$ and dense growth of Zn phases throughout



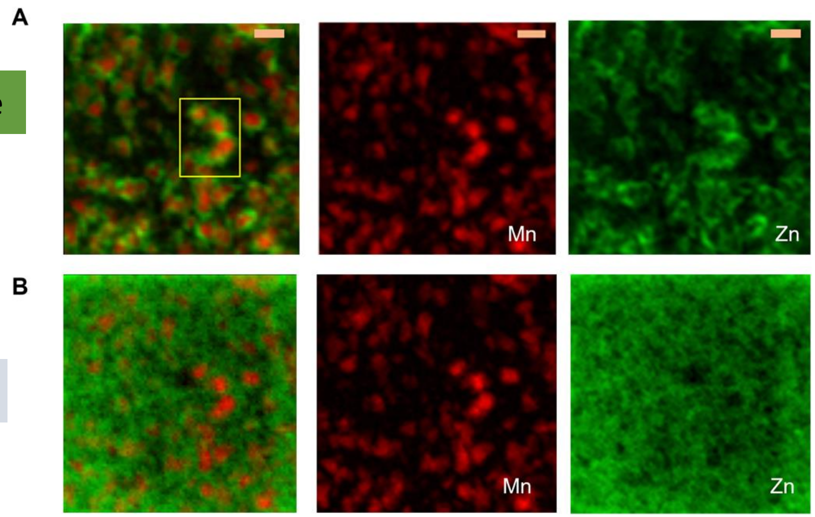


- *Varun R. Kankanallu, Xiaoyin Zheng, Denis Leschev, Nicole Zmich, Charles Clark, Cheng-Hung Lin, Hui Zhong, Sanjit Ghose, Andrew M. Kiss, Dmytro Nykypanchuk, Eli Stavitski, Esther S. Takeuchi, Amy C. Marschilok, Kenneth J. Takeuchi, Jianming Bai, Mingyuan Ge* and Yu-chen Karen Chen-Wiegart, Energy & Environmental Science (2023), DOI: 10.1039/D2EE03731A*

# Colocalization of the Zn and Mn phase around the electrode

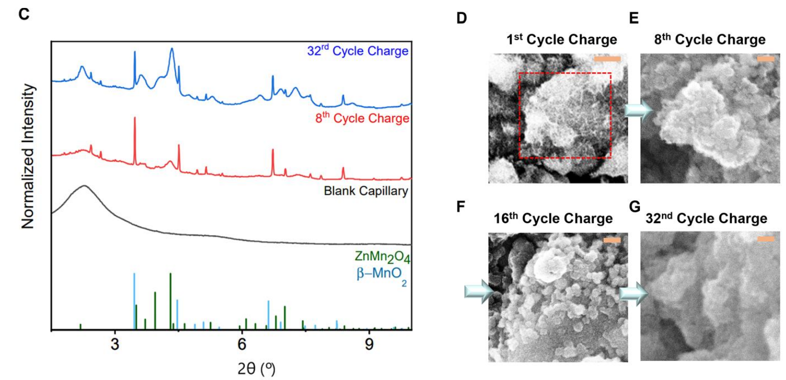
discharge

charge




- Colocalization of the Zn phase over the $MnO_2$ particles (scale bar = 5 micron): ZHS phase formation and reversibility

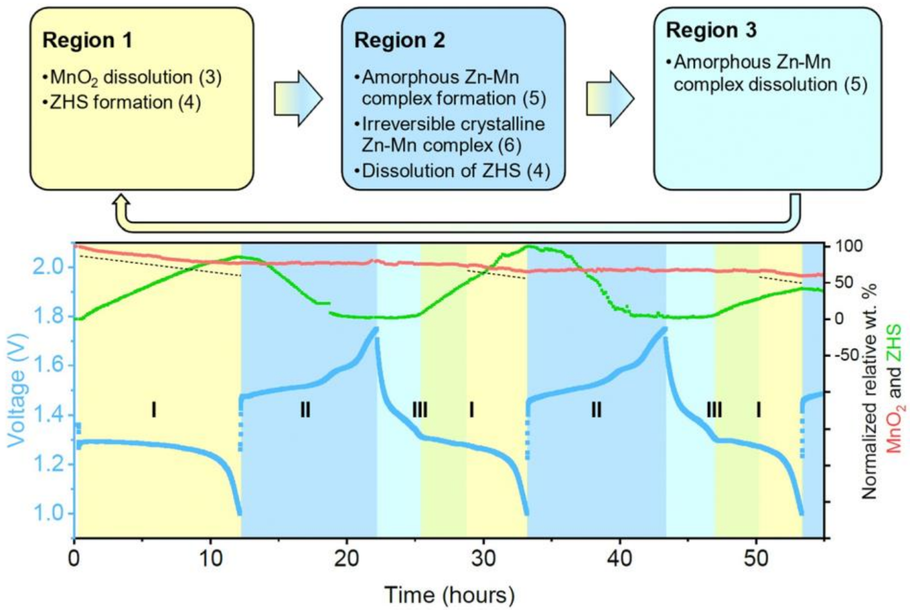
- Growth of the $ZnMn_2O_4$ phase obtained at the end of 8th and 32nd cycle.
- SEM of 1st cycle at the charged state having a flower like deposition over the $MnO_2$ particle.
- Growth of spherical round feature, (scale bar = 500 nm, for D = 100 nm)

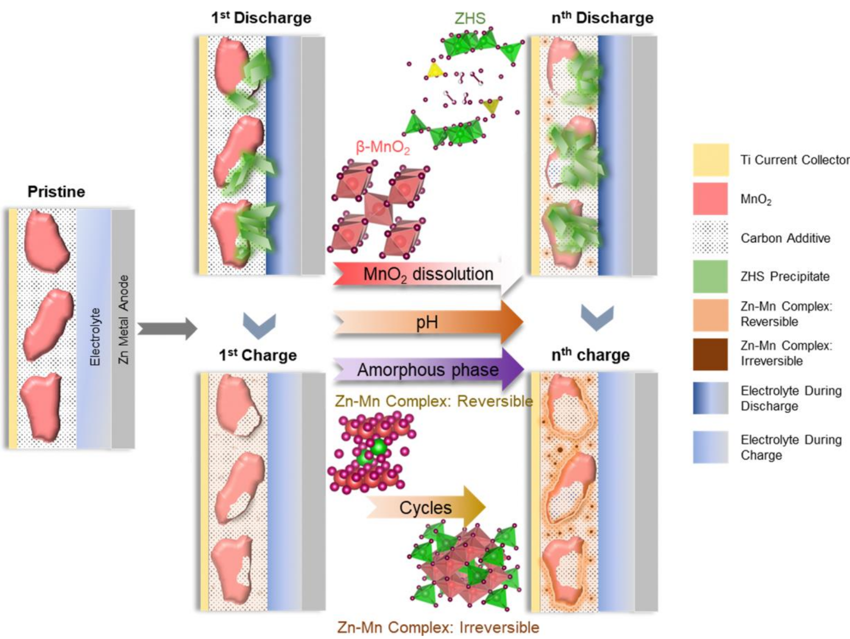




- *Varun R. Kankanallu, Xiaoyin Zheng, Denis Leschev, Nicole Zmich, Charles Clark, Cheng-Hung Lin, Hui Zhong, Sanjit Ghose, Andrew M. Kiss, Dmytro Nykypanchuk, Eli Stavitski, Esther S. Takeuchi, Amy C. Marschilok, Kenneth J. Takeuchi, Jianming Bai, Mingyuan Ge* and Yu-chen Karen Chen-Wiegart, Energy & Environmental Science (2023), DOI: 10.1039/D2EE03731A*

# Proposed reaction mechanism

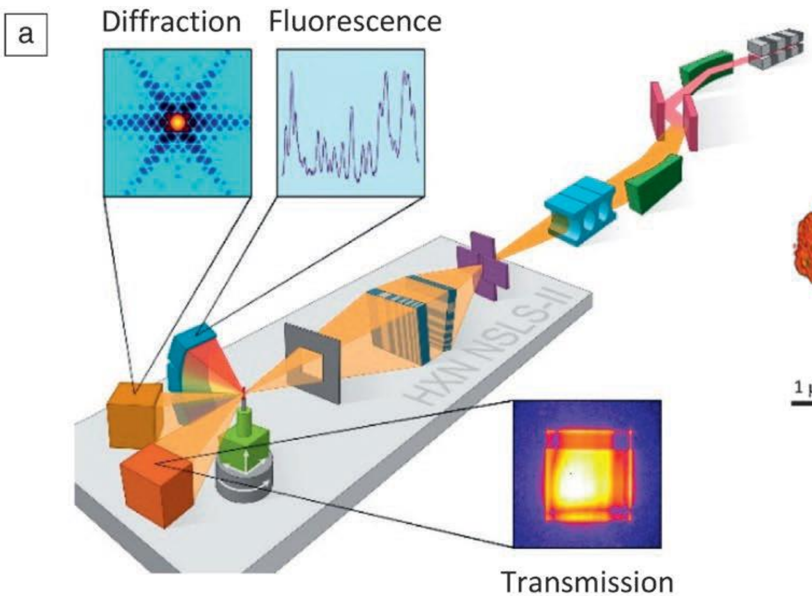


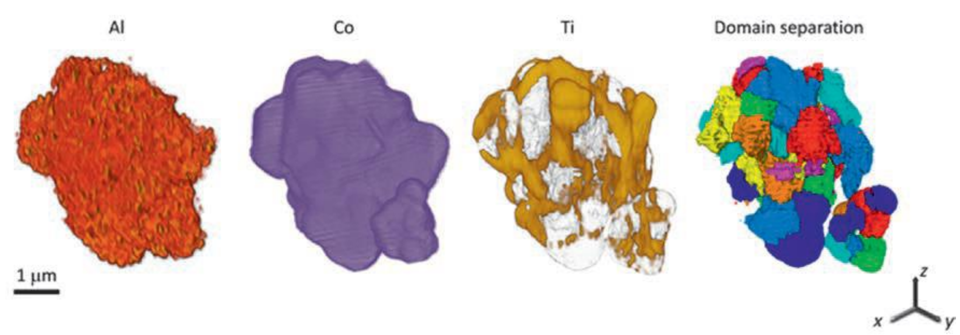

- *Varun R. Kankanallu, Xiaoyin Zheng, Denis Leschev, Nicole Zmich, Charles Clark, Cheng-Hung Lin, Hui Zhong, Sanjit Ghose, Andrew M. Kiss, Dmytro Nykypanchuk, Eli Stavitski, Esther S. Takeuchi, Amy C. Marschilok, Kenneth J. Takeuchi, Jianming Bai, Mingyuan Ge* and Yu-chen Karen Chen-Wiegart, Energy & Environmental Science (2023), DOI: 10.1039/D2EE03731A*

# How about for one type of technique?

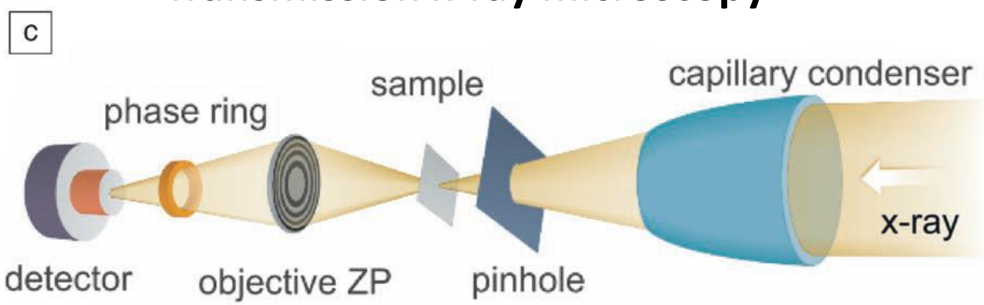
## Scanning x-ray microscopy



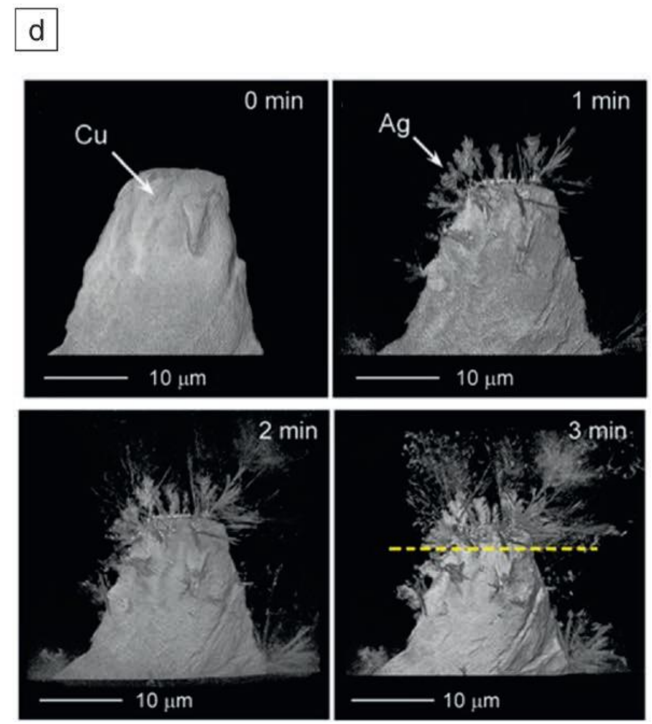

From HXN, 3-ID, NSLS-II




## Transmission x-ray microscopy



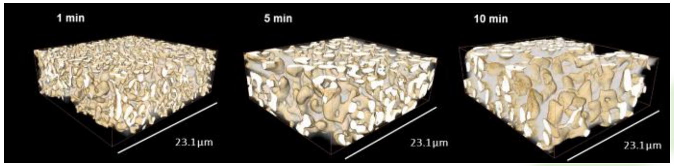

*2020 MRS Bulletin, Nanoscale x-ray and electron tomography, Hanfei Yan , Peter W. Voorhees , and Huolin L. Xin , Guest Editors*



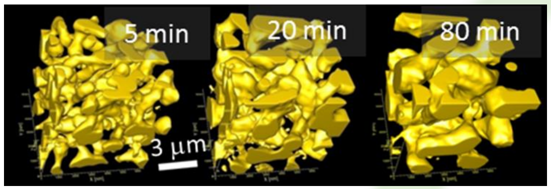

From FXI, 18-ID, NSLS-II

# Modern X-ray Imaging: Multi-dimensional

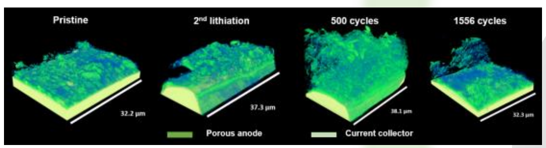
*X-ray Imaging & Microscopy*




Zhao et al., 2017, ACS App. Mat. & Int.




Chen Wiegart et al., 2012 Acta Mat.




Zhao et al., 2018, Nano Energy

**Need 1**

**3 Dimensional (Spatial) Characterization**

Tomography

*In situ operando*

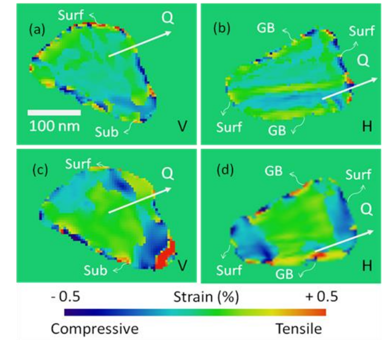
Spectroscopic Imaging

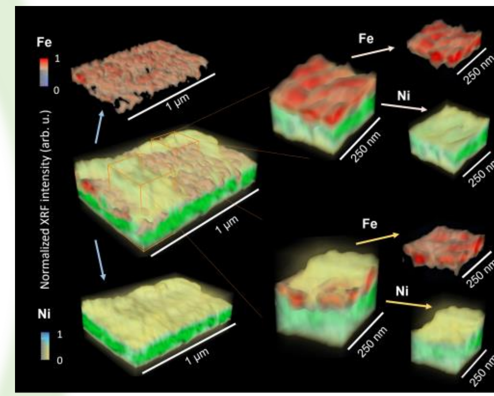
Diffraction Imaging

**Need 2**

**Time-Resolved with Real Environment**

Chen Wiegart et al., 2018 Nanoscale




Zhao et al., Materials Horizons, 2019

**Need 3**

**Correlation between Morphology and Other Characteristics: Elemental Distribution, Chemical States, Strain Distribution**

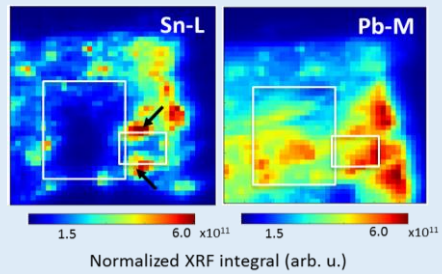
# X-ray Microscopy at NSLS-II: A Suite of Tools for Scientific Discovery

Hard

Scanning probe

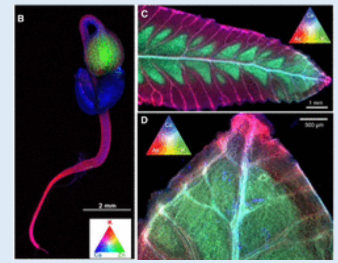
HXN

SRX




*Chen-Wiegart, et al., Scientific Reports (2017)*

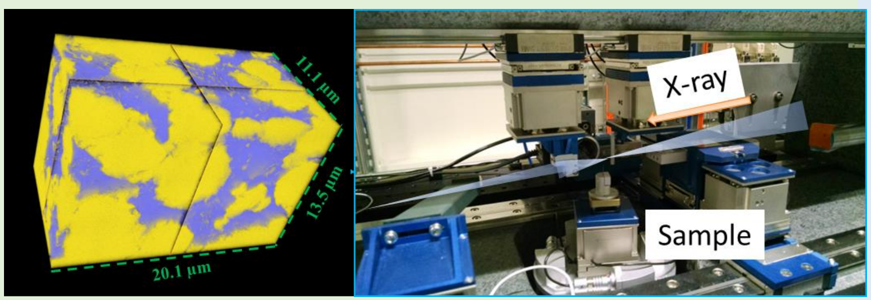
XFM



*P Kopittke, et al, Plant Physiol., (2018)*

*nanometers* **resolution** *microns*

Full Field Imaging




FXI

TES

Tender

- Complementary in resolution, field of view, energy range
- Combination w/ spectroscopy and diffraction analysis

# 2-Min You Talk!

## *Talk to your neighbor(s):*

1) What is your research topic? (An "elevator pitch")
2) What are the main techniques (2-5 of them) you use to characterize them? (Name at least one X-ray or neutron technique, if possible!)
3) Why are you using them?

   What information can you get out of each of the techniques?

   Are they complementary to each other?
4) Try to categorize them and see their connections:

→ Building a mind-map/framework to think/plan your research

Avoid: I have a hammer, and thus everything looks like a nail!

Ask yourself: why am I using the technique, and what I am trying to get out of it?

5) **Think of other techniques that you may be using in the future that you learned during the X-ray and neutron summer school?**

# WIIFM?
# What's in it for me?

- What is a multimodal approach?
- Why we care about it?

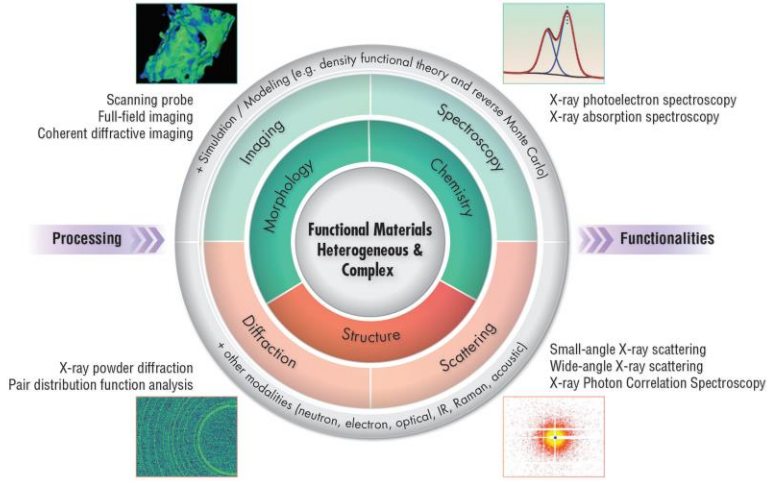
- *Research example: Conversion coating*

- Ways to frame multimodal analysis.

- *Research example: Battery*

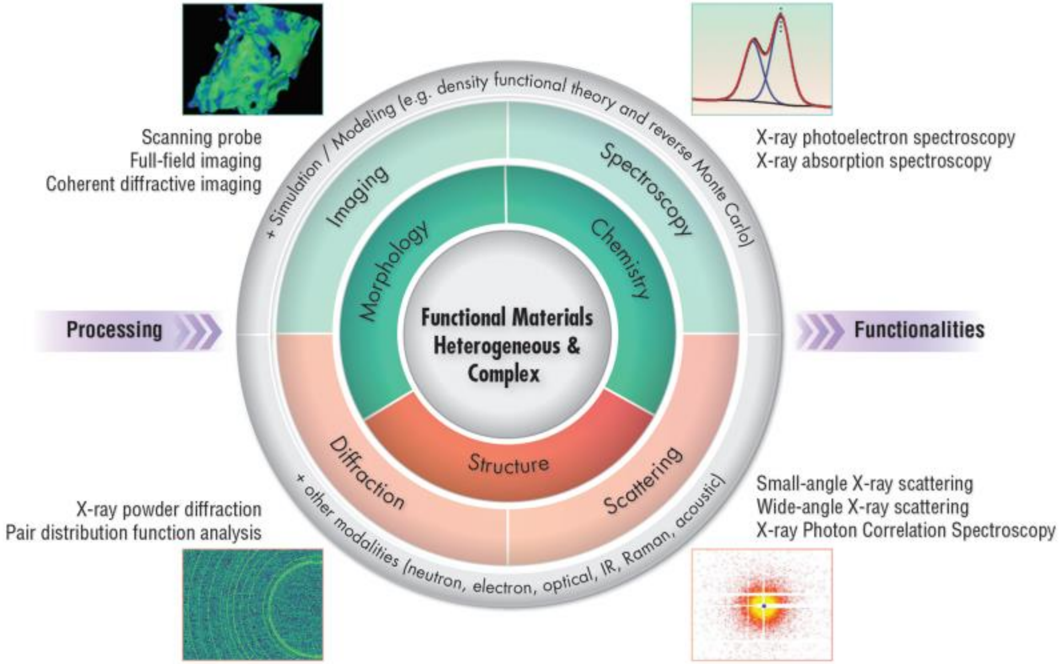
- **Beyond synchrotron**
  - **Other experimental modalities**
  - **Experiment – simulation feedback loop**
  - **Data science opportunities**

**- *Research example: Nanoporous metals***



# Beyond Synchrotron

- The synchrotron multimodal approach may be achieved by incorporating ancillary probes into synchrotron beamlines, by exploiting other measurement modalities—such as the electron-based and optical imaging methods—to augment synchrotron datasets, or even by exploiting theory and modeling to complement measurements



# Now broaden it a bit from synchrotron!

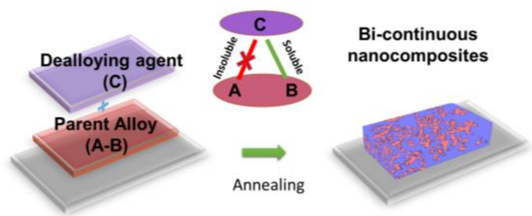
- How do I complement my synchrotron studies?

- Lab-based techniques?
  Pre-characterization?
  Ex-situ studies to complement the in-situ study?

- Other advanced characterizations?
  E.g. imaging: TEM, Atom-probe, etc.?

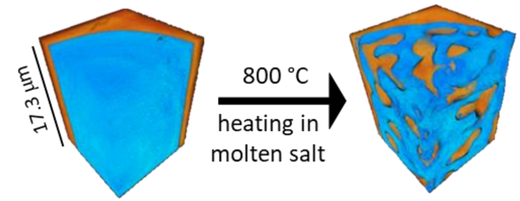
- Simulation/modeling/theory?

# Dealloying with different media




*Solid-state meta dealloying (SSMD)*

*Zhao et al., Nanoscale 2021*




*Molten salt dealloying (MSD)*

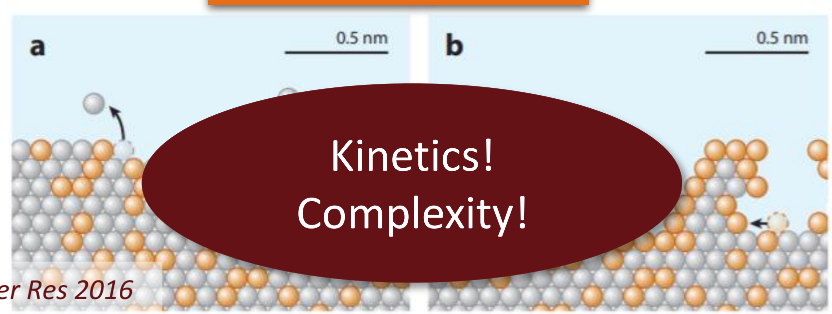
*Liu, et al. Nat Commun. 2021*

Dealloying

Processing

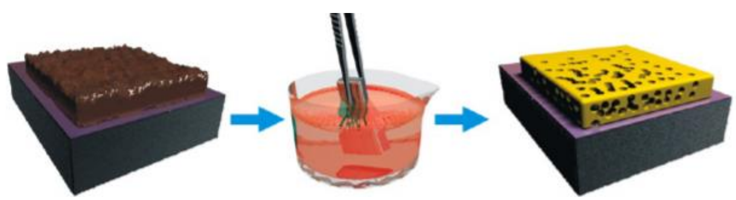
Property

Structure



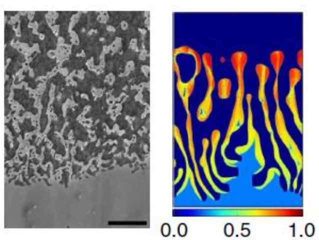

Kinetics!
Complexity!

*McCue et al. Annu Rev Mater Res 2016*



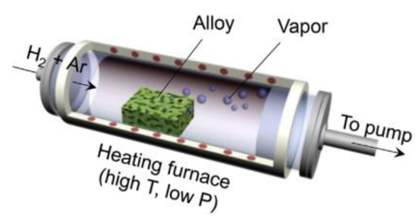
*Aqueous solution dealloying*

*Tessier et al., ACS AMI, 2015*




*Liquid metal dealloying*

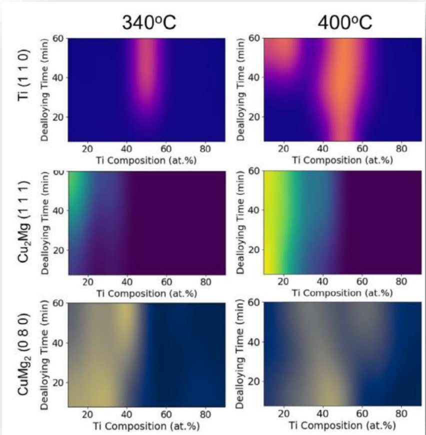
*Geslin, et al. Nat Commun. 2015*




*Vapor phase dealloying*

*Liu, et al. Nat Commun. 2018*

# Synchrotron X-ray Characterization




*Zhao et al., Acta Materialia, 2023*

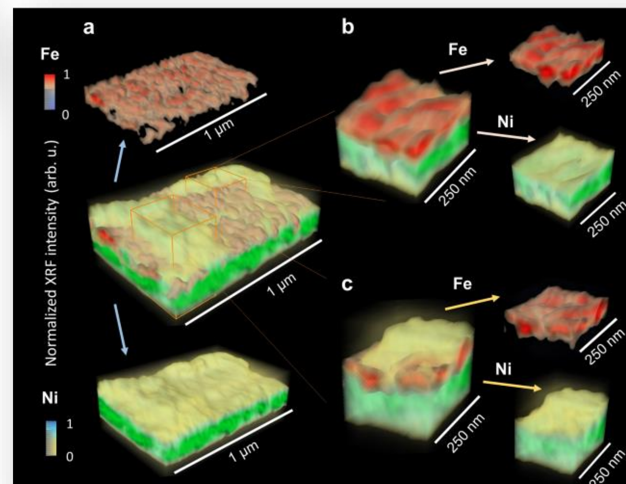
Crystalline structure
Phase identification

Diffraction
& Scattering

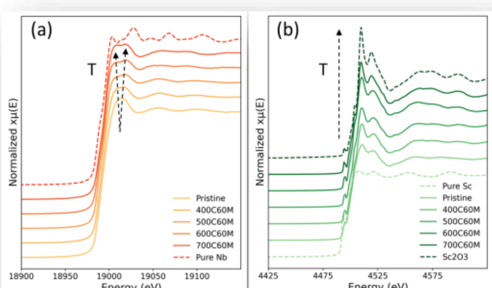

Synchrotron X-ray
Characterization

Chemical States
Local structures

Spectroscopy




*Zhao et al., Materials Horizons, 2019*



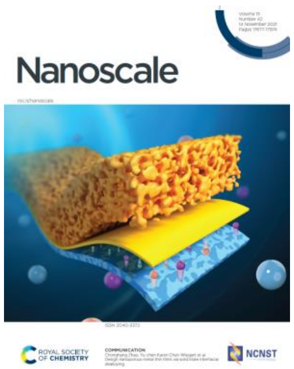

*Chung et al., Adv. Mat. Int. accepted*

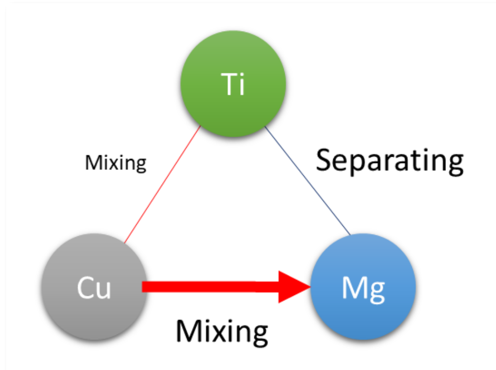
Imaging
& Microscopy

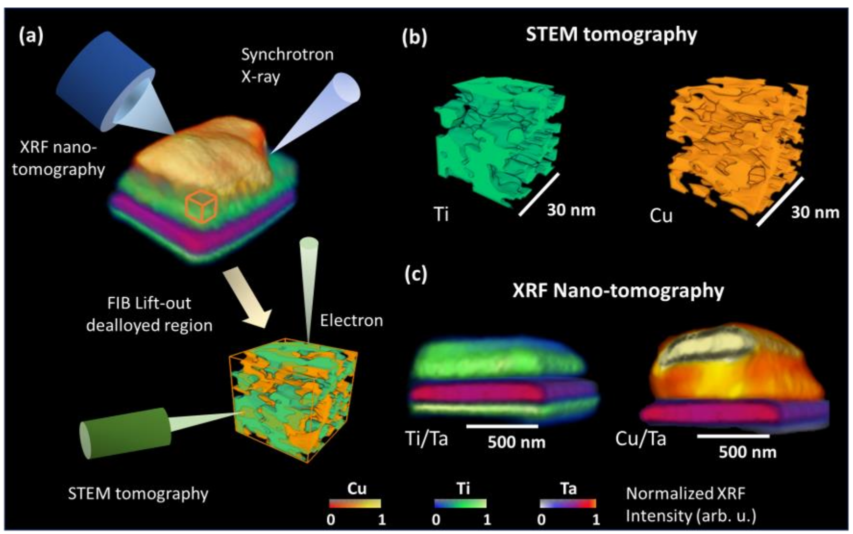
Morphology
Elemental, chemical,
and strain mapping

# Thin Film Solid-State Metal Dealloying (SSMD):

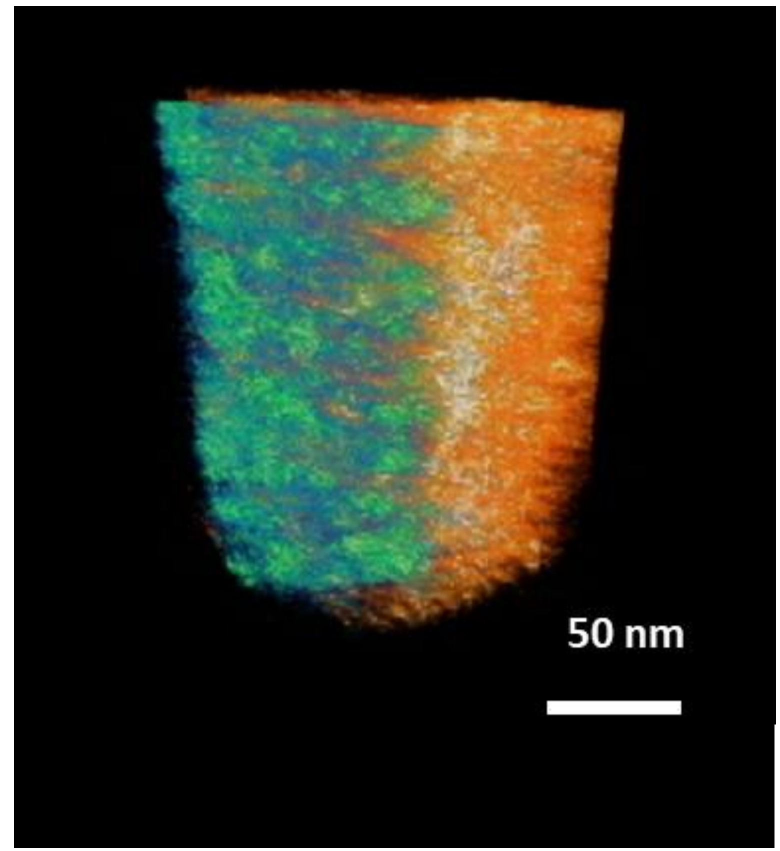
# Ti-Cu/Mg System: 3D Morphology



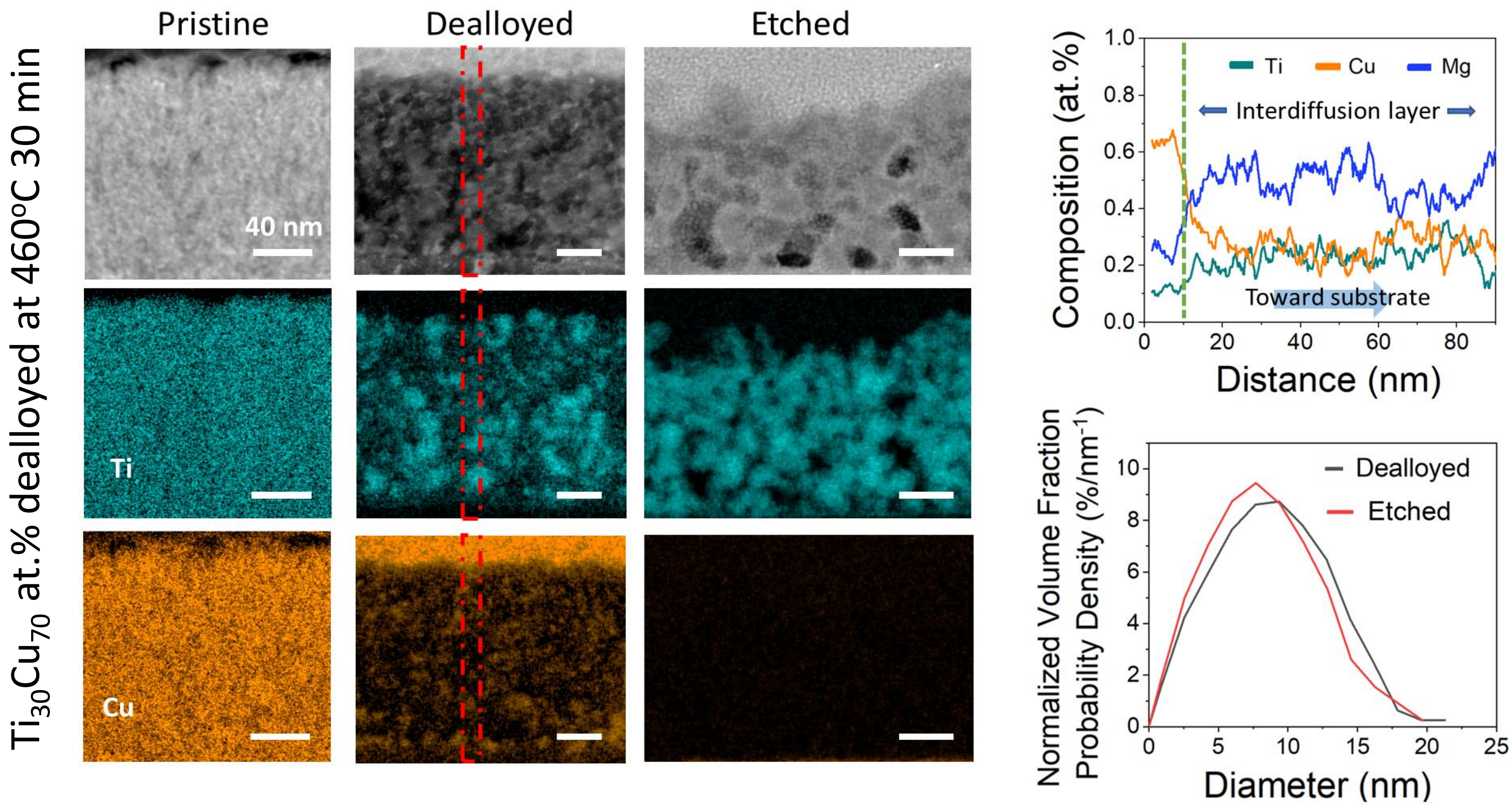




*Hard X-ray Nanoprobe (HXN) 3-ID, NSLS-II*




*STEM (Talos), CFN*

# Thin film SSMD Ti-Cu/Mg System: **Nanoporous Structure**



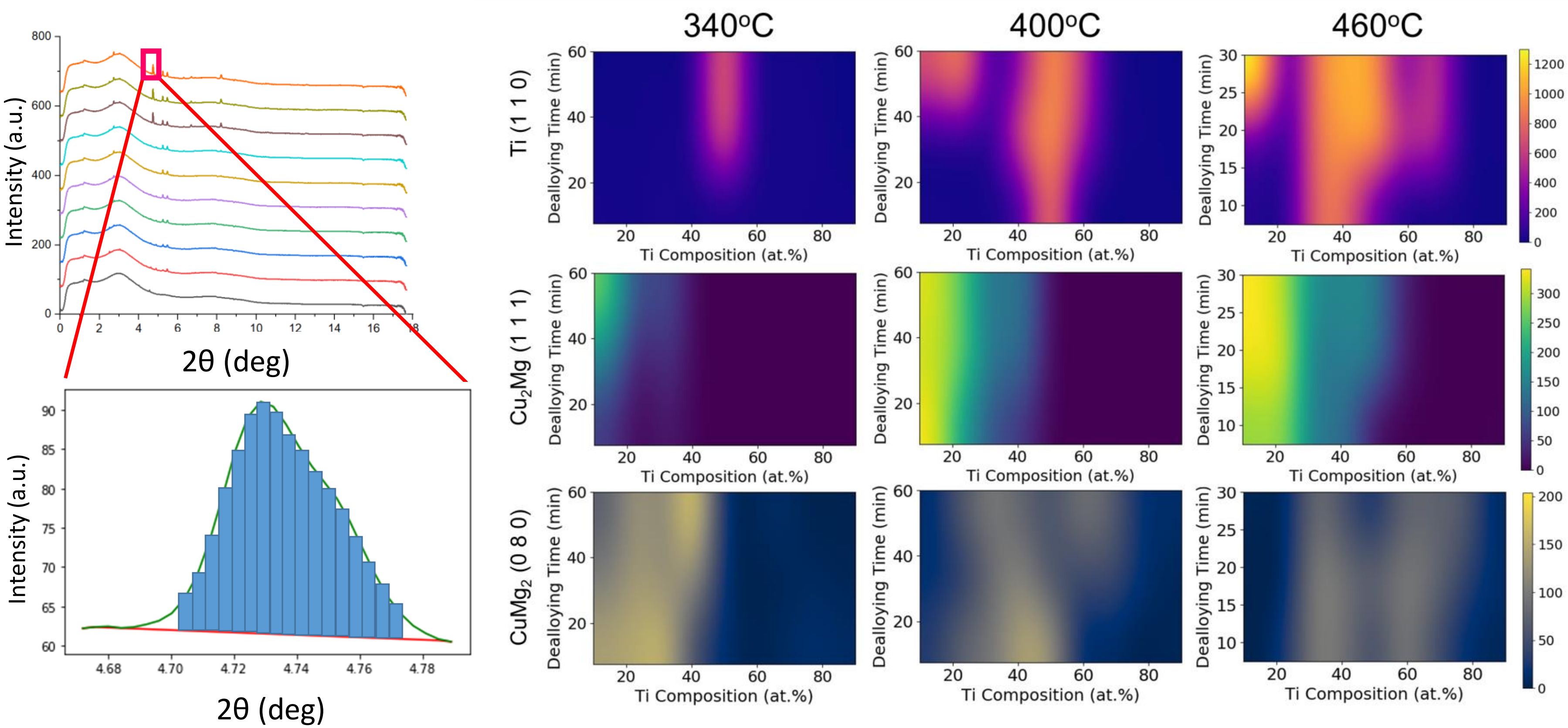

- Creating a nanoporous structure by the thin-film SSID.
- The Ti ligament size is ~5-15 nm, quite fine for structures created by metal dealloying.
- Consistent morphology observed in the etching process

Stony Brook University

*C. Zhao, K. Chen-Wiegart et al. (2021), Nanoscale*

Brookhaven National Laboratory

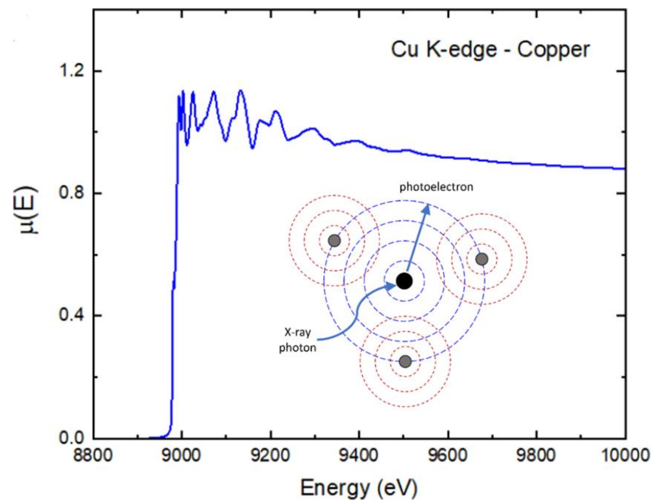
# Thin film SSMD Ti-Cu/Mg:
# **Structural Evolution with Time, Temperature and Composition**




- The dealloying rate varies with the parent alloy composition.
- The $CuMg_2$ phase → $Cu_2Mg$ at 400ºC
- Crystallization sequence: $CuMg_2$,$Cu_2Mg$ first, followed by Ti

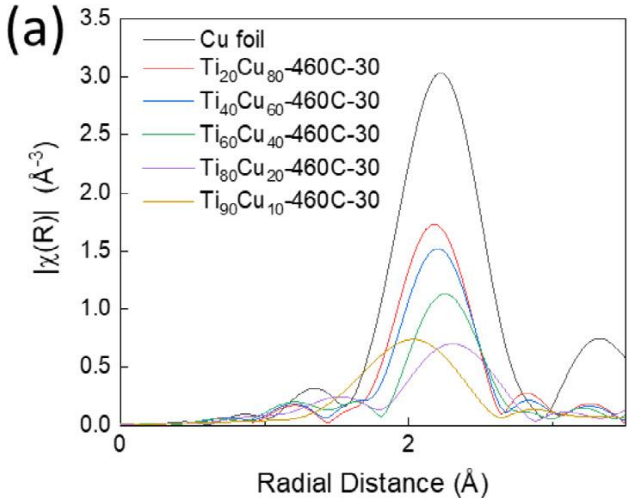
# Thin film SSMD Ti-Cu/Mg: **Local Bonding Formation**

- **Bonding:** Cu-Cu, Cu-Ti, Cu-Mg; Ti-Ti, Ti-Cu, Ti-O
  - Bond length and coordination numbers
- XRD: The order of crystalline phase formation: $CuMg_2$ , $Cu_2Mg$, and Ti
- EXAFS: The **Ti phase first shows self-reorganization during dealloying**, earlier than the crystallization process.

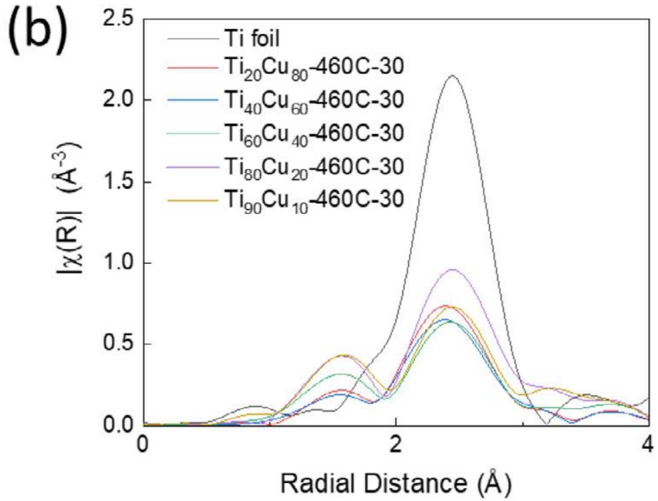



*Sanson, Microstructures 2021*

**Cu EXAFS**



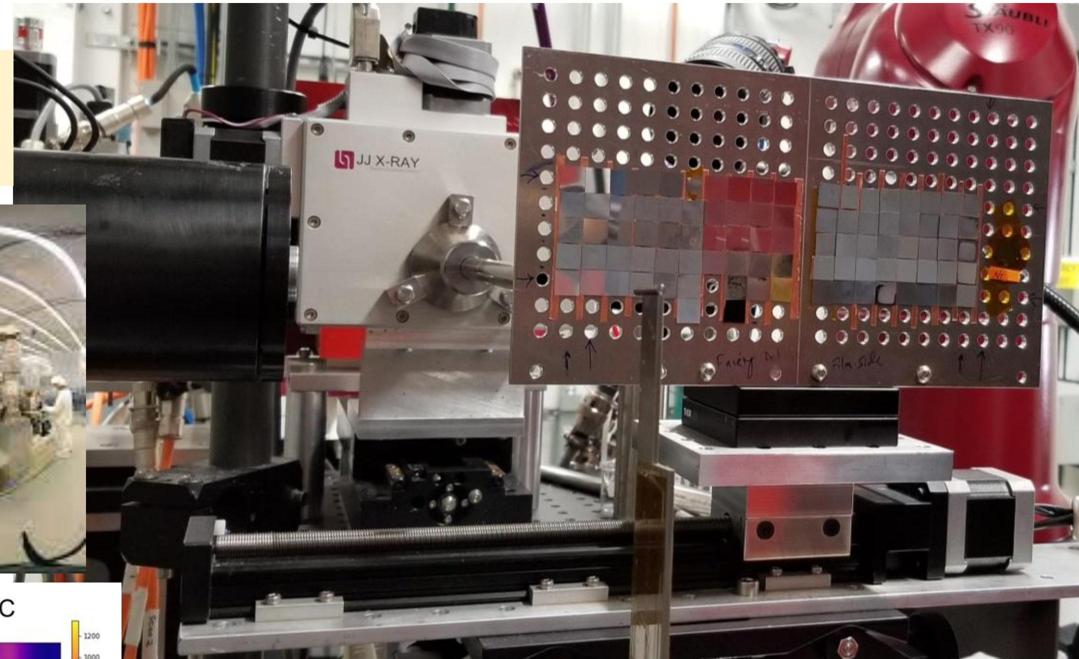

**Ti EXAFS**

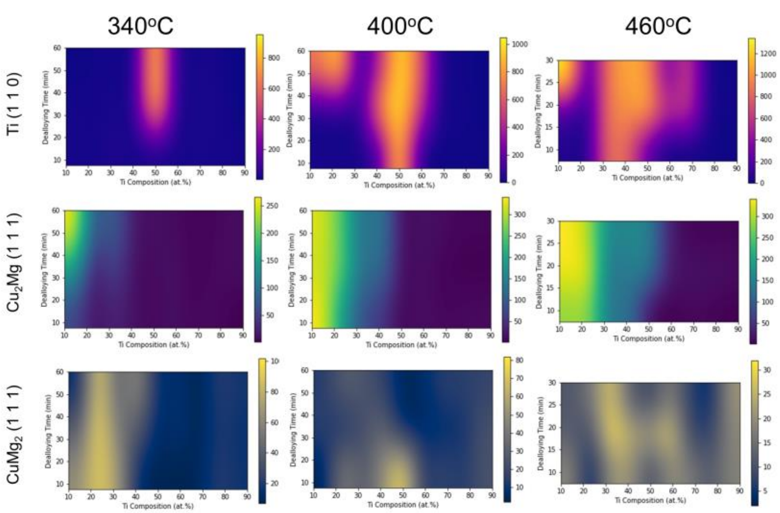


# .....A heroic but unsustainable act!

CFN clean room

Nov, 2019
XPD, NSLS-II




90 individual thin film samples prepared from different alloy compositions, dealloying time and temperature!

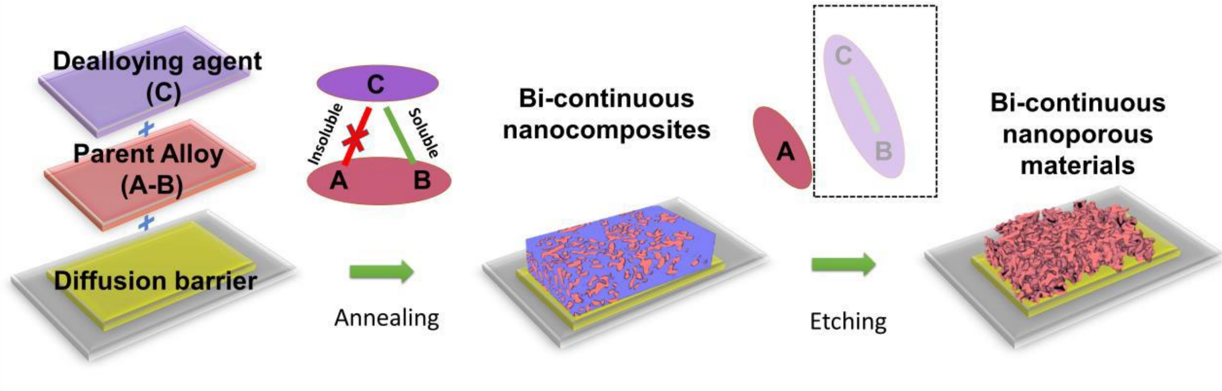
For just one (Ti-Cu) system!!

# The challenge that we're all facing as experimentalists…

# **Large parameter space!**

I want to understand how to control / design the materials!

**Solid State Metal Dealloying**




**Engineering** Processing parameters

- Materials composition
- Materials thickness
- Processing time
- Processing temperature
- ……

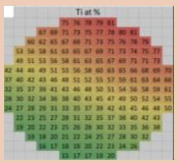
**Science** Physical / chemical parameters

- Elemental properties, valence shell attributes, ionicity attributes
- Alloy mixing enthalpy & entropy
- Atomic configuration
- Kinetics pathways / mechanisms
- …….

# Workflow of an ML-augmented framework to design SSID

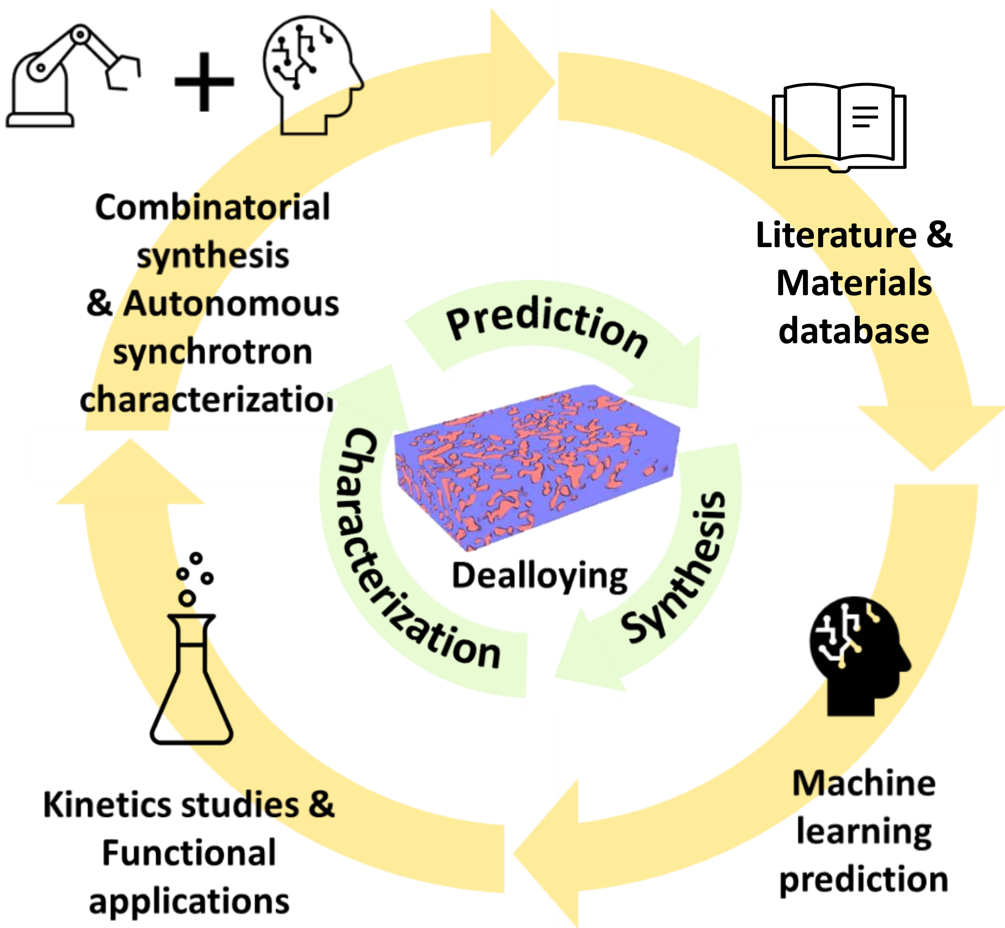
bluesky

gpCAM



- Combinatorial thin film
- Synchrotron X-ray Automatic pattern analysis (XCA)
- Autonomous decision-making (gpCAM)

- Crystal formation
- Processing parameters




matminer

pymatgen

XenonPy

- Reported dealloying systems

scikit learn

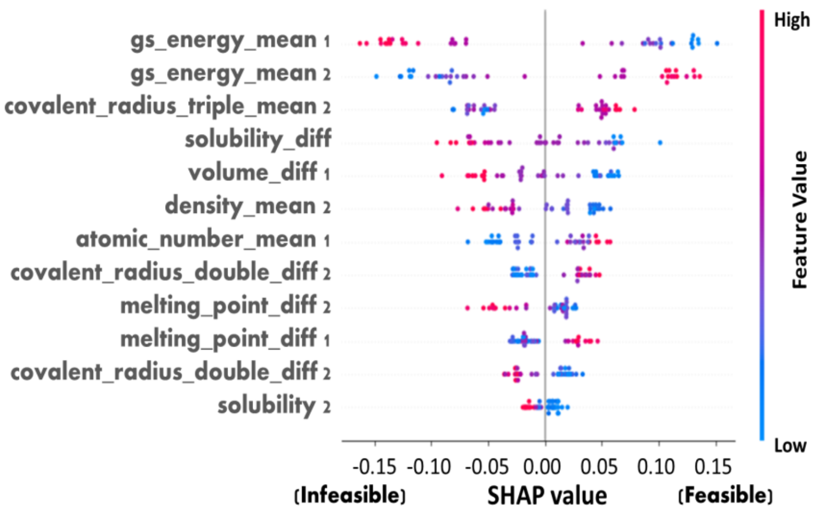
- Miscible & immiscible
- Dealloying systems



*Zhao, Chung, Chen-Wiegart et al., Communications Materials, 2022*

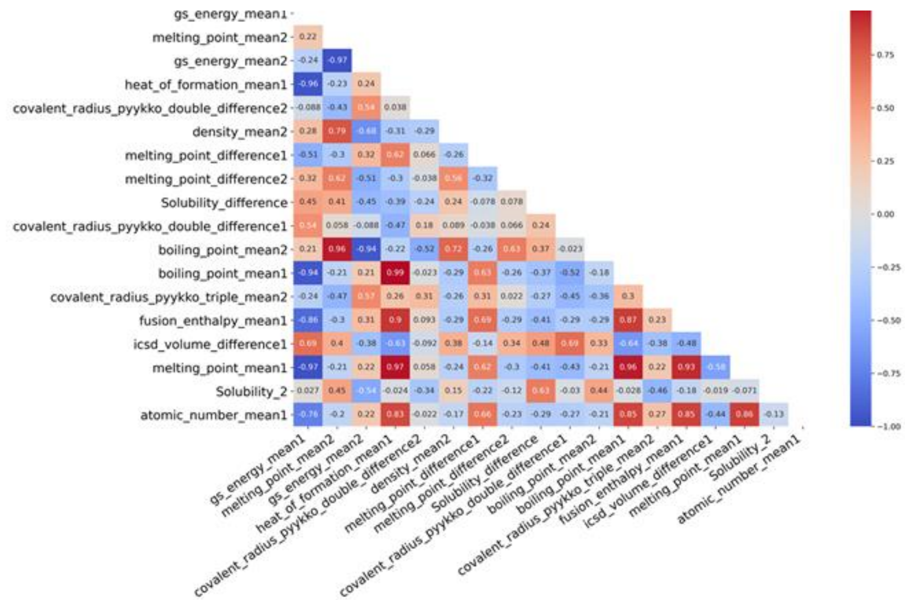
# Relevant parameters

Variables which are ranked by their **impacts on differentiating the dealloying agent** from the parent alloy




(sorted by the random forest method.)

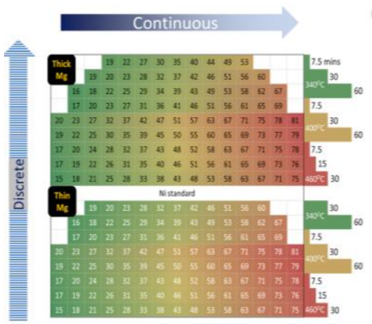
**The correlation matrix:** top key variables from each of the three ML methods, total of 18 variables.

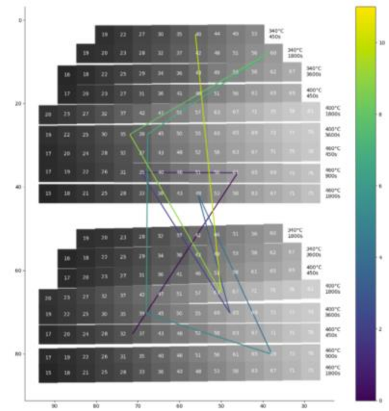


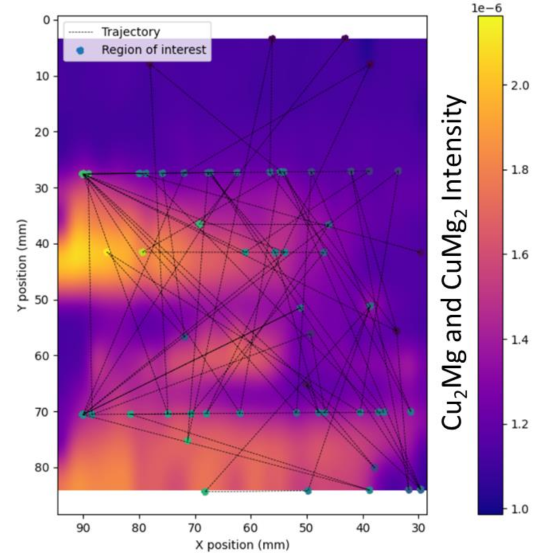

6 valuables with r > 0.8 were removed

**Variables ending with 1 represent the properties of the first two elements in a ternary system, and those ending with 2 represent the properties of the last two elements.*

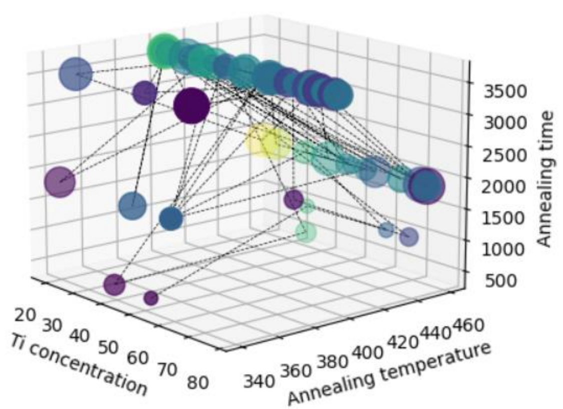
# Autonomous synchrotron analysis



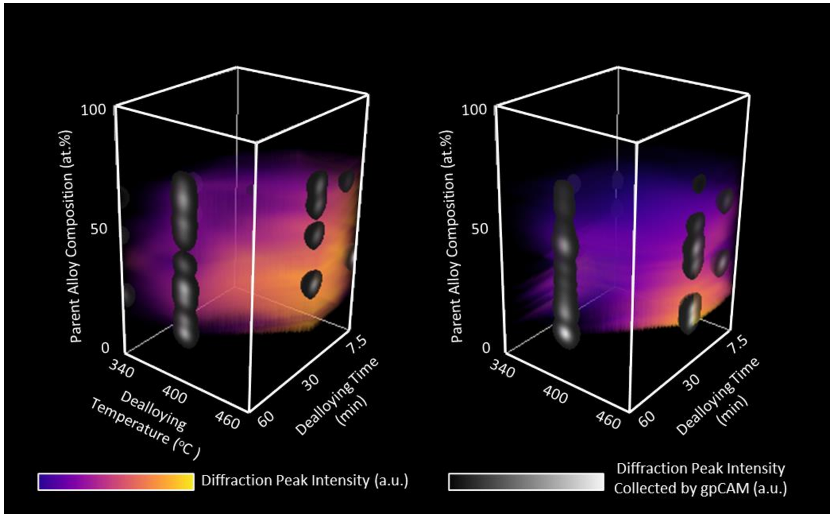






The 'beamline' coordinate (x, y)

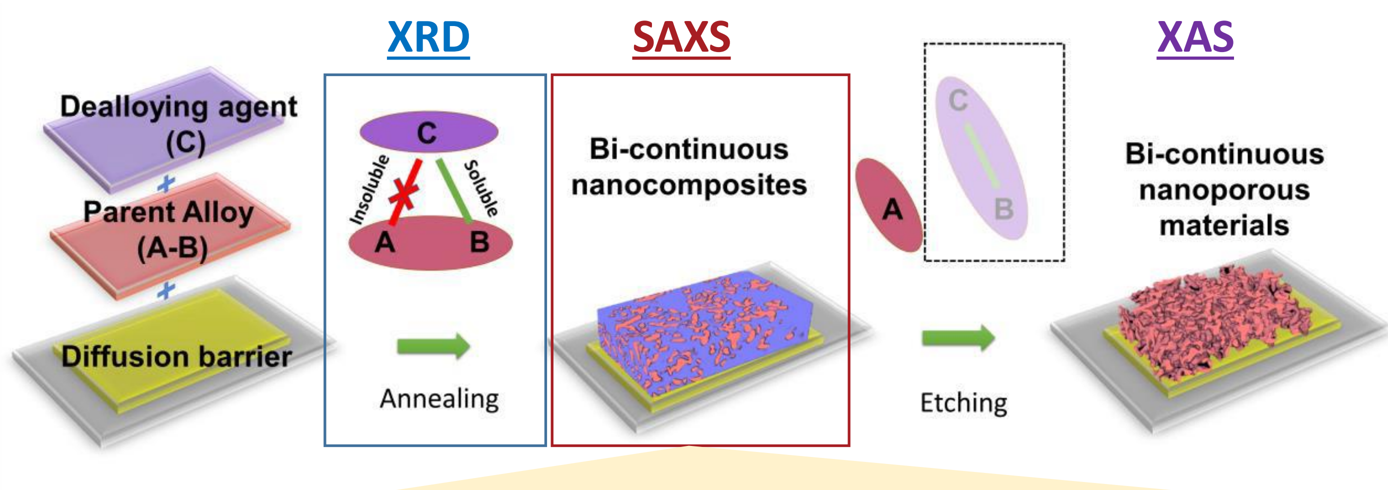



The physical parameter coordinate

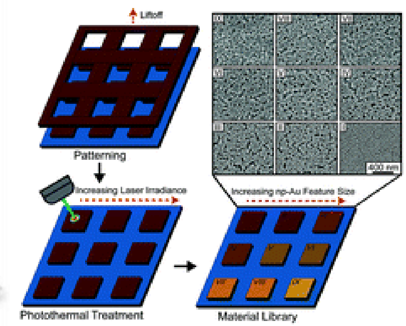


# Beyond phase determination…

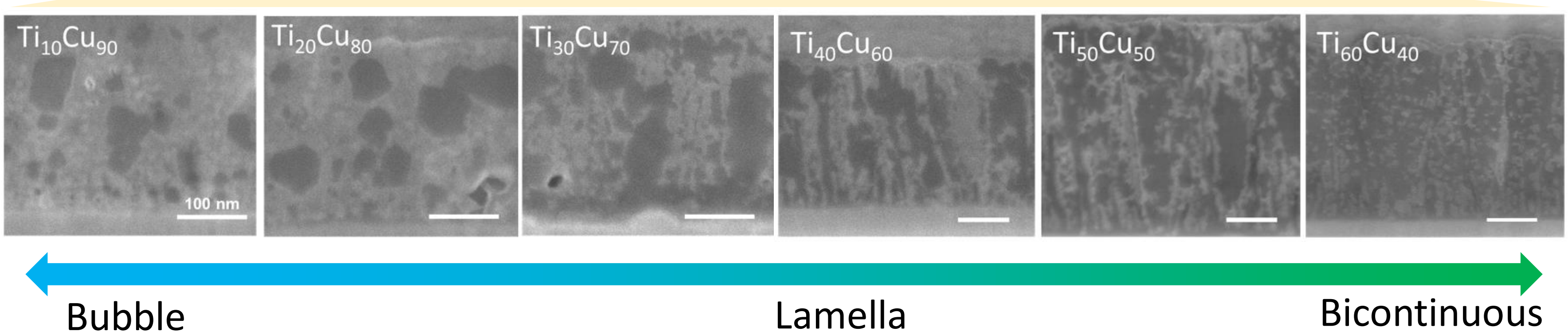
- How about the morphology & chemical speciation?



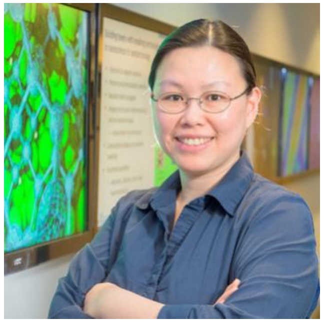

**Applications**




*Seker et al., Nanoscale*, 2016



# More references



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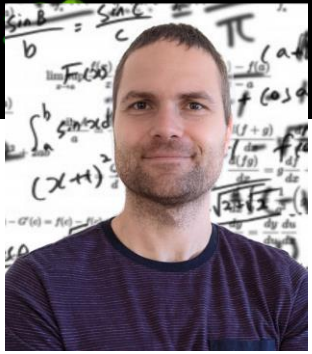

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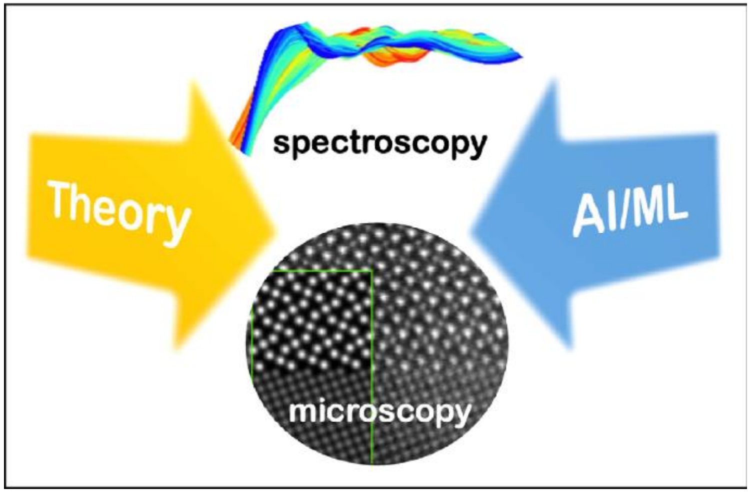


Theory+AI/ML for microscopy and spectroscopy: Challenges and opportunities Davis Unruh, Venkata Surya Chaitanya Kolluru, Arun Baskaran, Yiming Chen & Maria K. Y. Chan MRS Bulletin (2022) https://doi.org/10.1557/s43577-022-00446-8