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In-situ Ion Beam Induced Luminescence/Imaging of Scintillating Materials

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Presented at In-Situ RAD TEM Workshop

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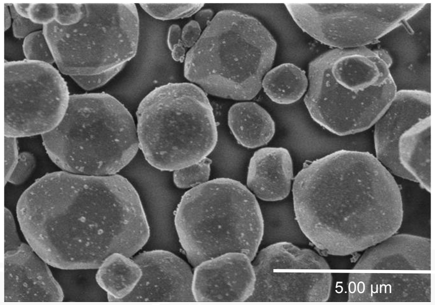
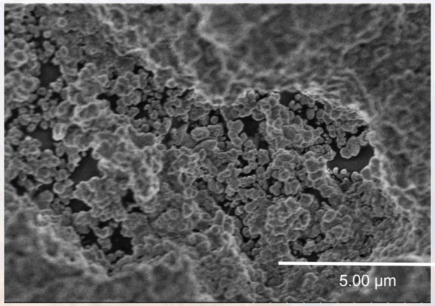


SEM: Commercial $CaWO_4$ – large grains



SEM: Synthesized $CaWO_4$ – nanograins

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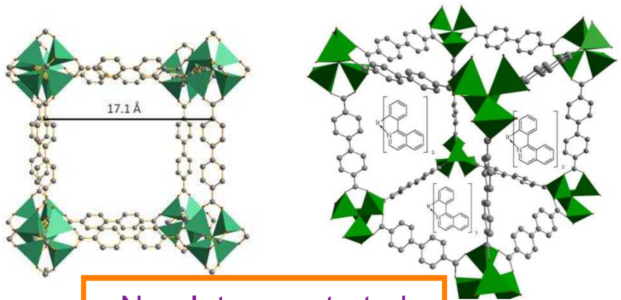
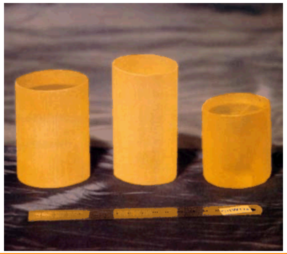
Introduction – Interest in Scintillators

Crucial to understand radiation-solid interactions on multiple length scales



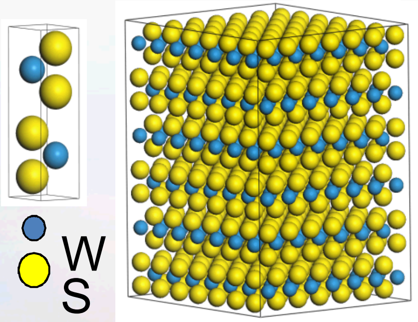
Non-Interpenetrated IRMOF-10



Single Crystal $CdWO_4$

Plastic Scintillators

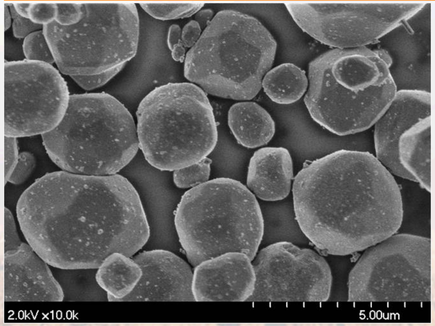
Nano **Micro** **Meso**



Tungsten (IV) Sulfide WS_2

Structure of High-Z ME_x Nanoscintillator

Commercial $CaWO_4$ Scintillating Powder

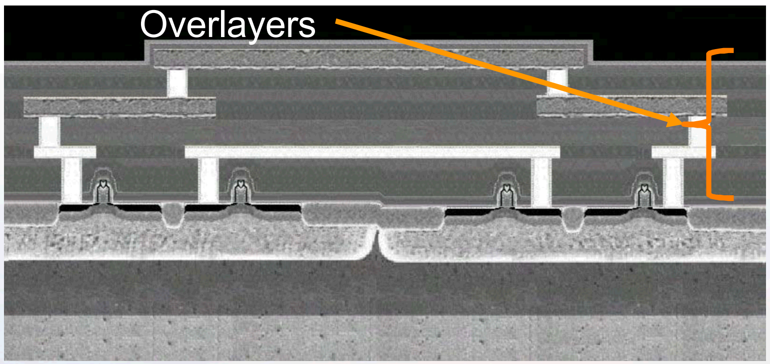


High-Z Nanocomposite

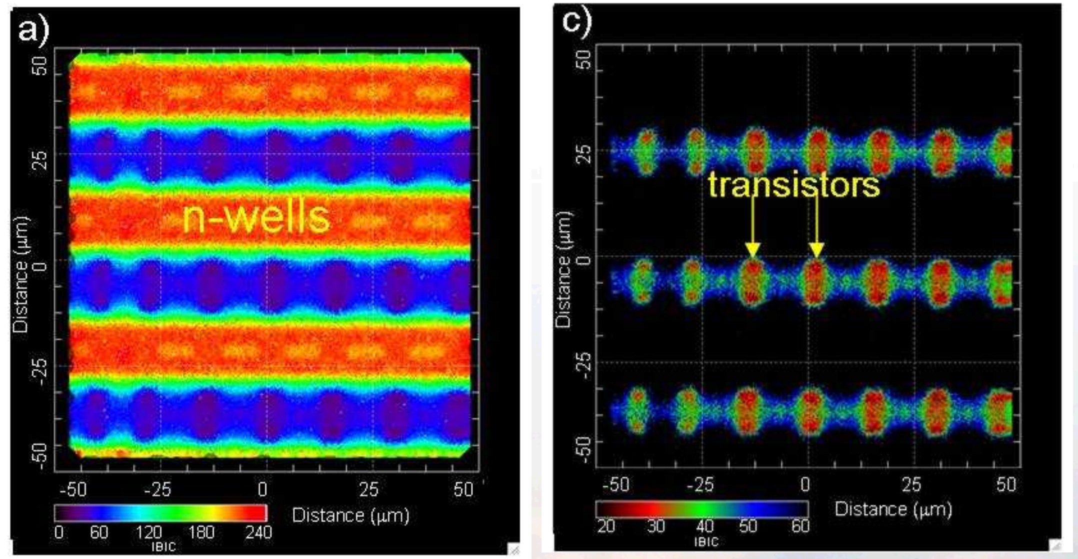
Advanced Applications of Scintillators

Novel radiation effects testing method

- Film of scintillating material required to create ion beam-induced luminescence (IBIL)
  1) IBIL creates signal corresponding to location of ion strike
  2) Radiation tolerance of material critical to experiment
  3) Must maintain optical properties as exposed to GeV-energy heavy ion beams
  4) In-situ ion irradiation TEM will allow for microstructual studies of radiation tolerance for these materials



SEM image of CMOS transistor from Analytical Solutions, Inc.



Ion beam induced charge (IBIC) imaging of Sandia SRAM device

Advanced Applications of Scintillators

Existing single crystal γ-ray scintillators suffer from performance and reliability issues

- Current Issues
  1) Long decay times
  2) Crystal anisotropy
  3) Low energy resolution (low luminosity & poor linearity)
  4) Complicated synthesis (single crystal growth)
  5) Chemical instability (hygroscopic)
- Scintillators with low energy resolution & detection efficiency cannot distinguish radiation type of quantify radiation
- Discovery of new scintillating materials needed

US Ports: 2 Billion Metric Tons

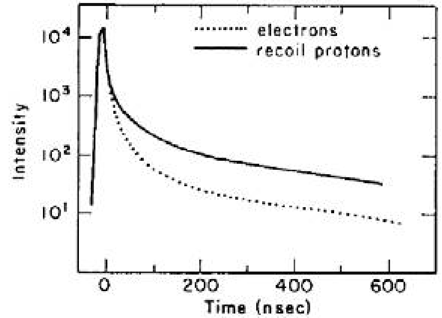
Introduction – Types of Scintillators

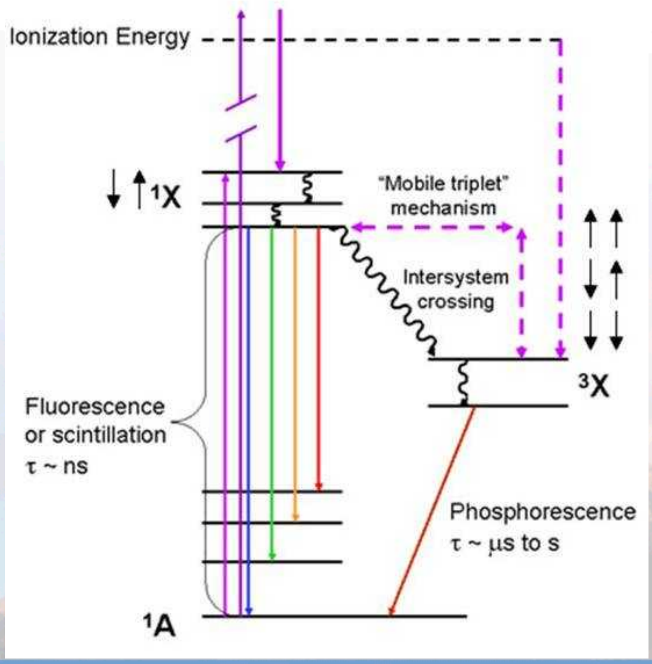
Want to achieve spectral discrimination with metal organic frameworks

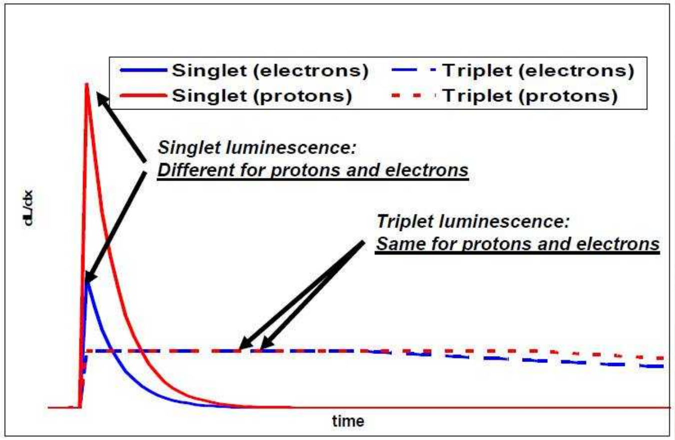
- Normal light emission is through singlet state, ~ns lifetime
- Triplets (*3/4 of the excitation)* normally a loss for scintillation
- Heavy metal fluor gives radiative path for triplets ~ μs time scale
- Triplet emission through SOC is:
  1. always longer wavelength than singlet emission
  2. always longer lifetime than singlet
  3. rate determined by exciton transport, density of states, and tau

Particle Discrimination possible if ONE pathway SATURATES

Pulse Shape Discrimination

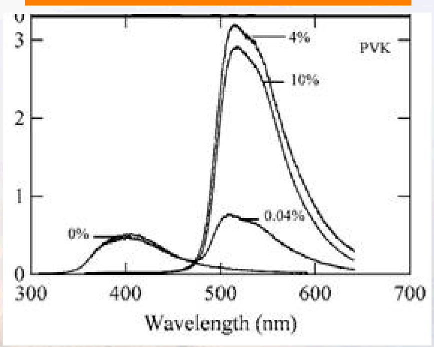






Singlet Emission dependent on dE/dx
Triplet Emission is not

Spectral Discrimination

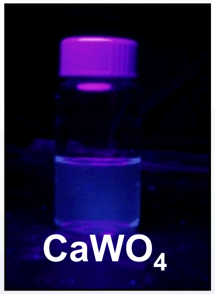


Introduction – Types of Scintillators

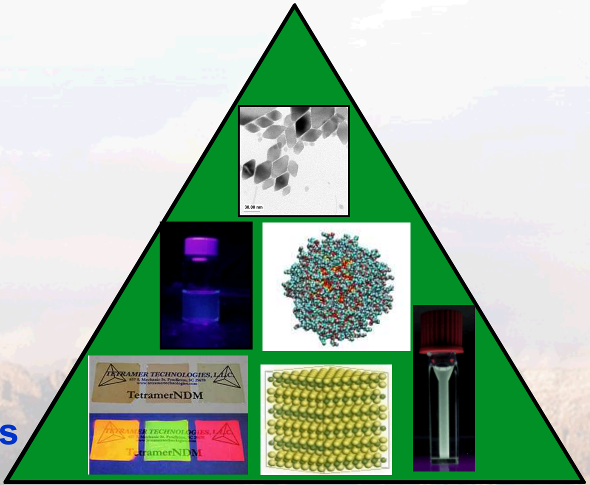
Want to explore potential of high-Z nanomaterials as scintillators

- Determine size effects (nano–meso) & activators of oxides (baseline)
- Develop novel size controlled scintillator materials based on unexplored high Z ME_x
- Investigate radiation Interaction (nano–meso)
- Improvements:
  - ↑*stopping power (higher density, less material)*
  - ↓*band gap (better luminosity emitted)*
  - ↓*non-radiative decay (noise reduction)*

Nano **Micro** **Meso**



Radiation Detectors Materials Characterization



Scintillating Materials Characterization

Radiation-Solid Interactions

Experimental – In-Situ Ion Beam Induced Luminescence

Ion Beam Induced Luminescence

- 3 MeV H^+ beam used as excitation
- Scintillation light collected as ion beam excites sample
- Light collected with OM-40 microscope or fiber optic mounted close to sample
- Avantes AvaSpec 2048 spectrometer

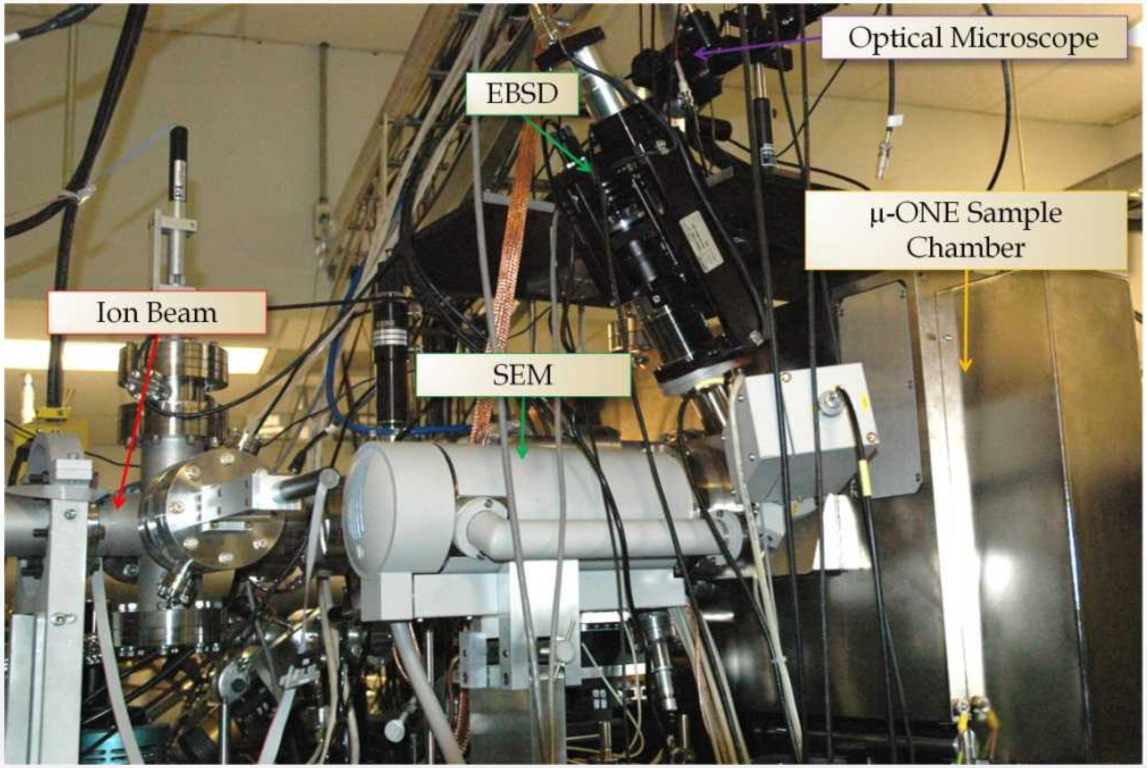
Decay Times

- 3 MeV H^+ beam
- Thin films of samples on PIN diodes
- Hamamatsu PMT run in photon-counting mode
- Light intensity measured as a function of time after ion strike

Radiation Hardness

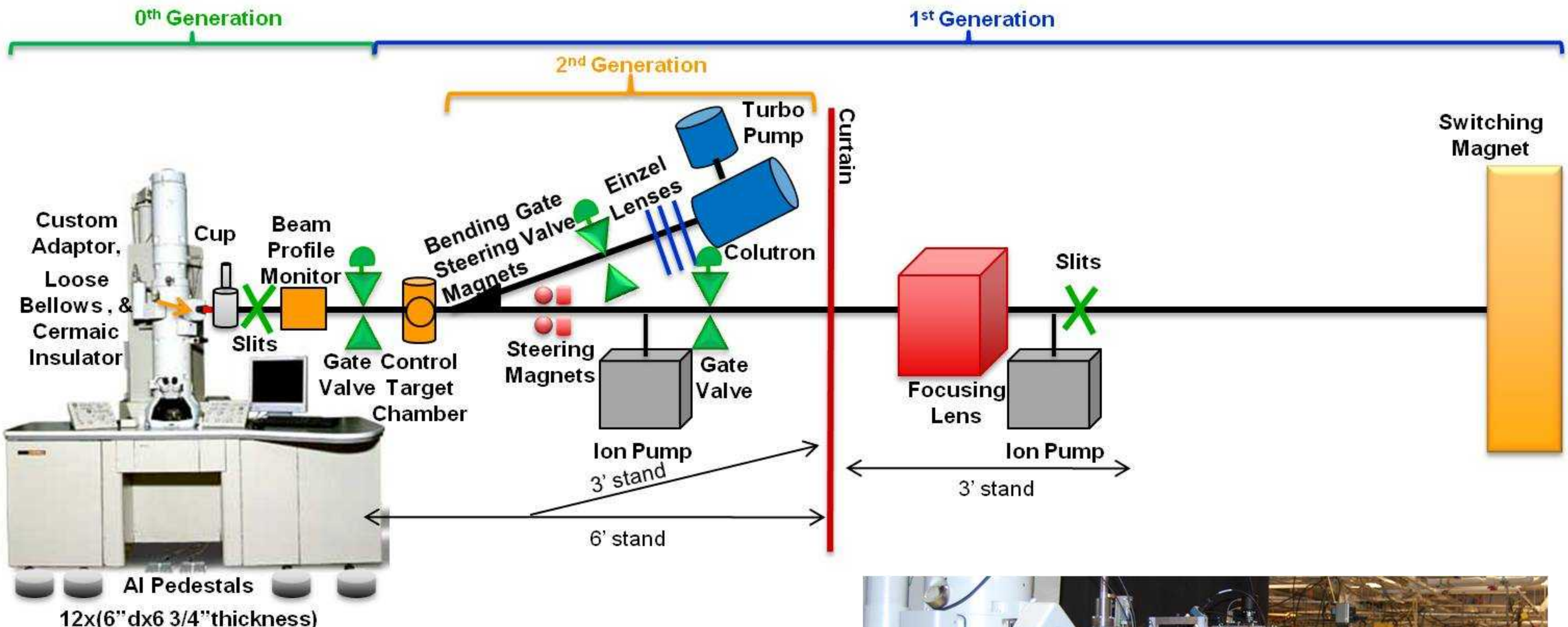
- Radiation hardness experiments performed with 3 MeV H^+ beam from tandem accelerator
- IBIL Spectra measured constantly as sample exposed to beam
- Overall decrease in emitted light observed as materials damages

Experimental – Nearly In-Situ SEM Ion Irradiation

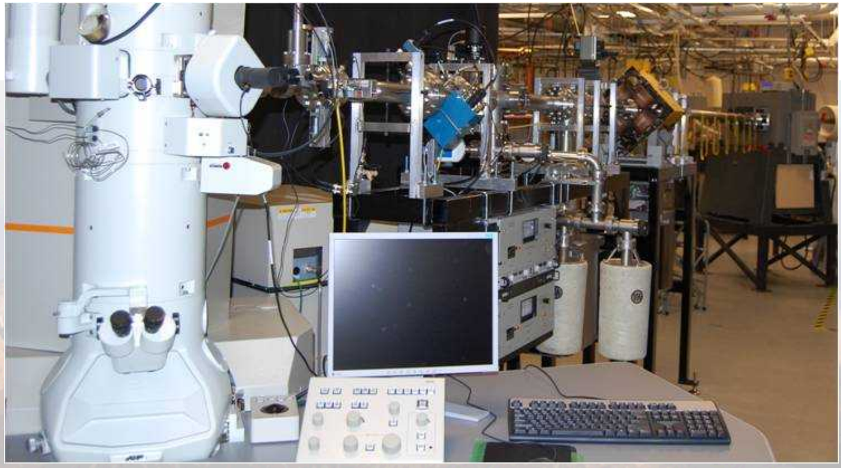


- Sample oriented and focused in front of ion beam
- Scintillation light collected as ion beam excites sample
- After irradiation, sample board translated in x-direction to orient in front of SEM
- Micrograph collected, then series repeated

Experimental Setup – In-Situ Experiments



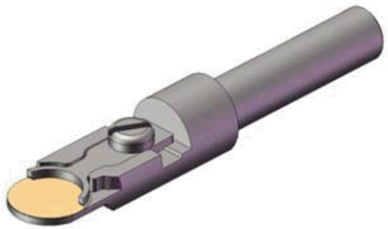
- 0th and 1st generation completed
  Ion beam effects have been observed
- 2nd generation initiated



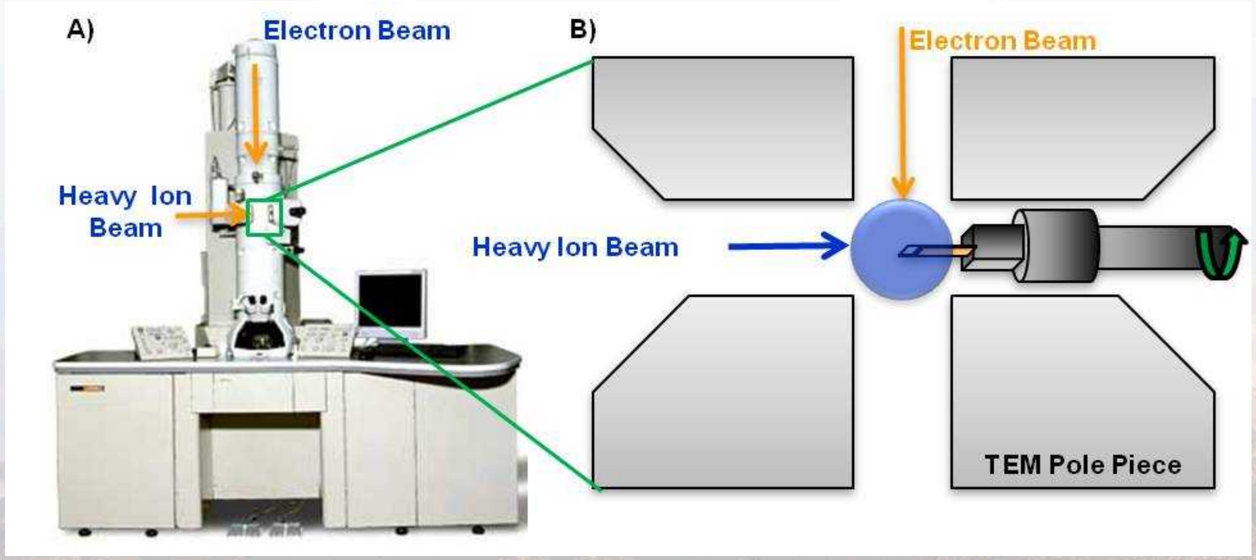
Experimental – TEM Tomography

Nanoscintillators observed with tomography technique

- Samples with interesting microstructures studied with tomography over range of tilt angles
- 3D reconstructions of particles obtained
- Sample irradiated inside in-situ TEM
- Tomography repeated to observe potential microstructural changes resulting from irradiation



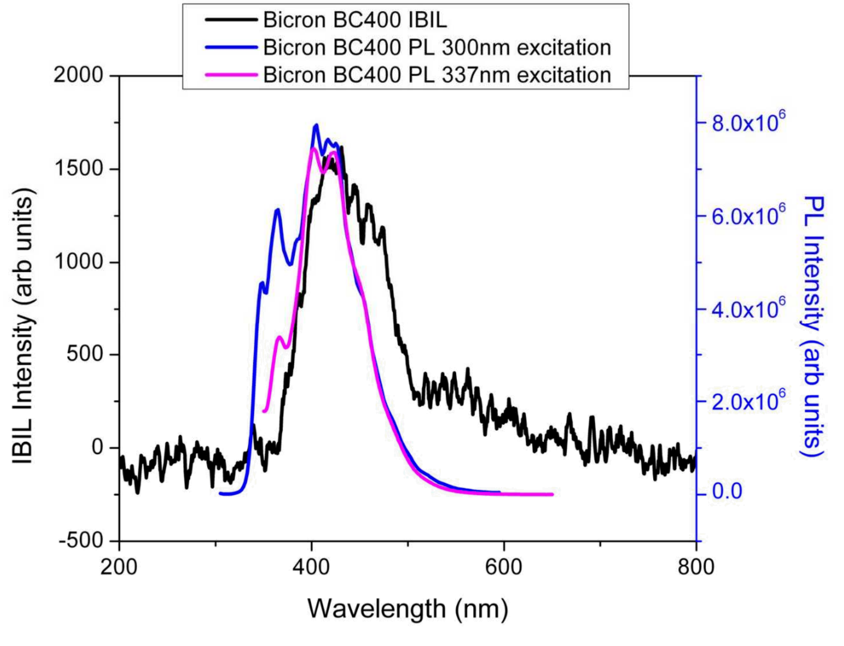
Hummingbird Tilt Stage
(-82° to +82°)



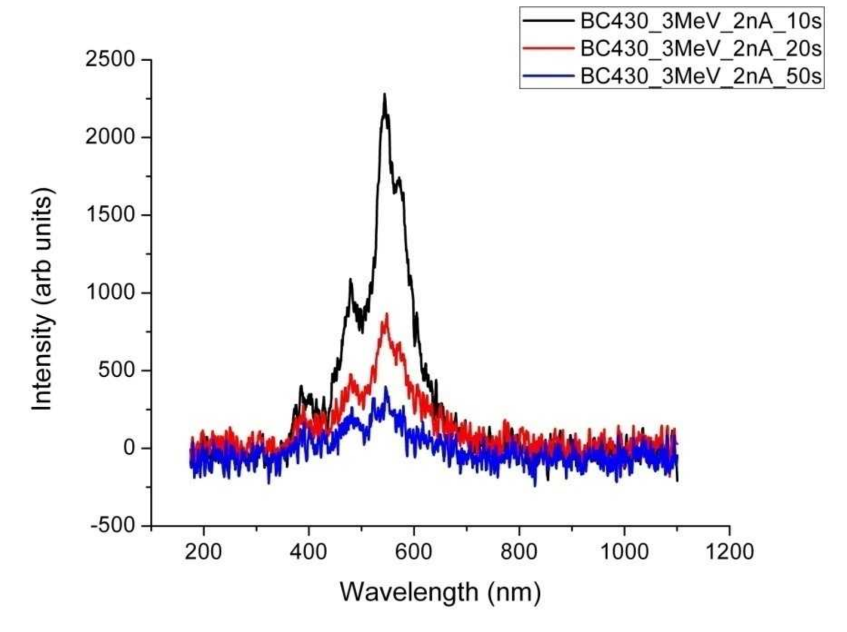
Results – IBIL of Organic Scintillators

Effects of excitation mechanism and radiation damage

- IBIL demonstrates different luminescence characteristics than PL
- Drastic degradation in optical properties as a result of irradiation observed in organic scintillators



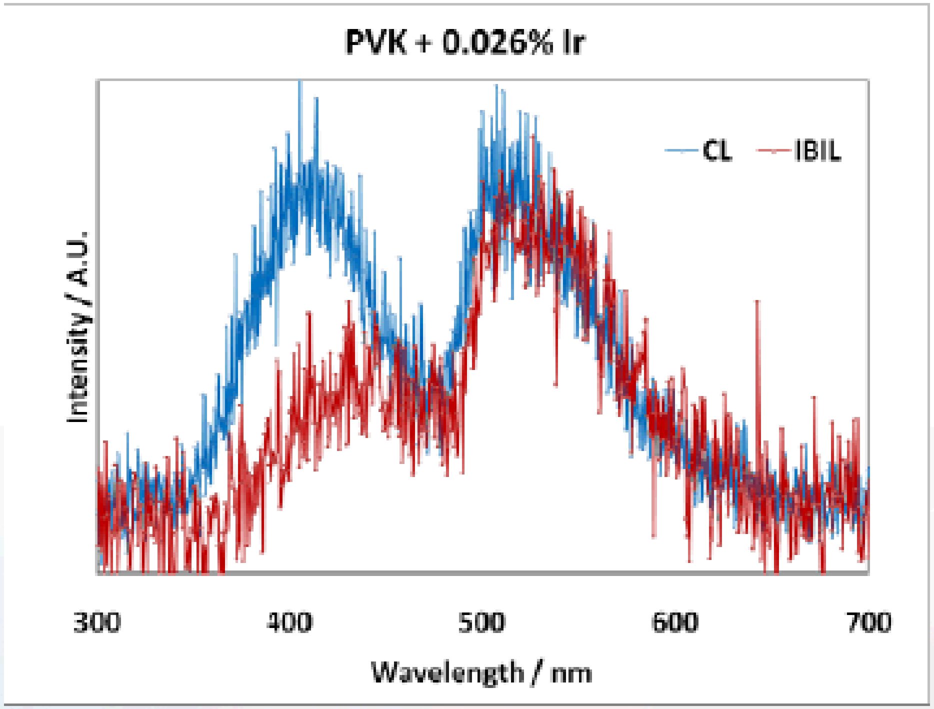
PL and IBIL of Commercial Organic Scintillator

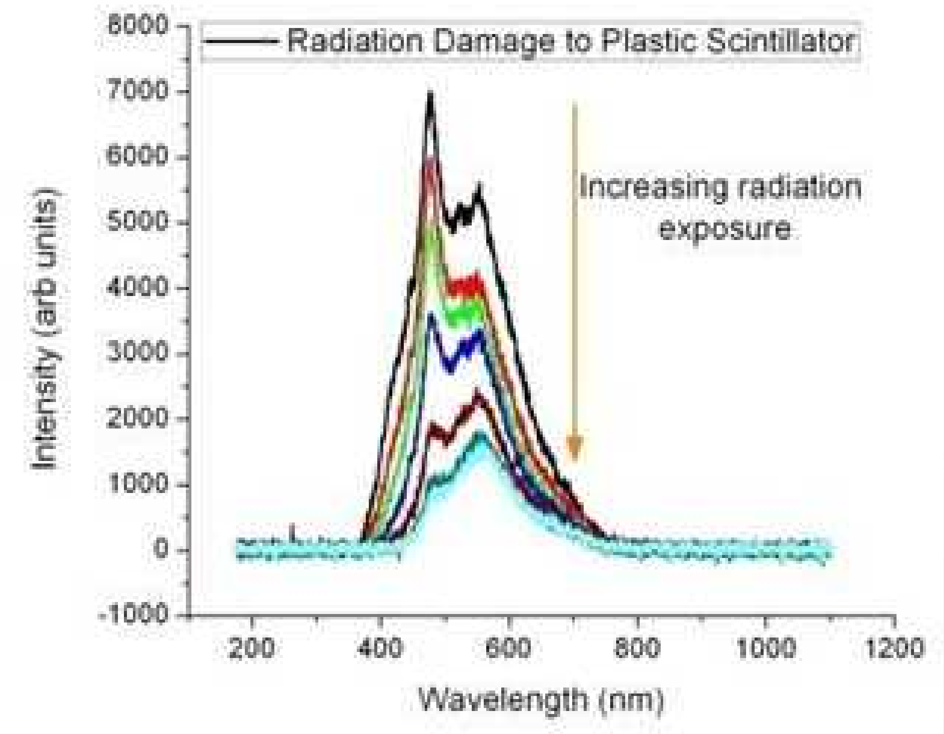


IBIL Decay of Organic Scintillator with Irradiation

Results – IBIL of MOFs

Metal-organic frameworks demonstrate spectral discrimination with IBIL/CL



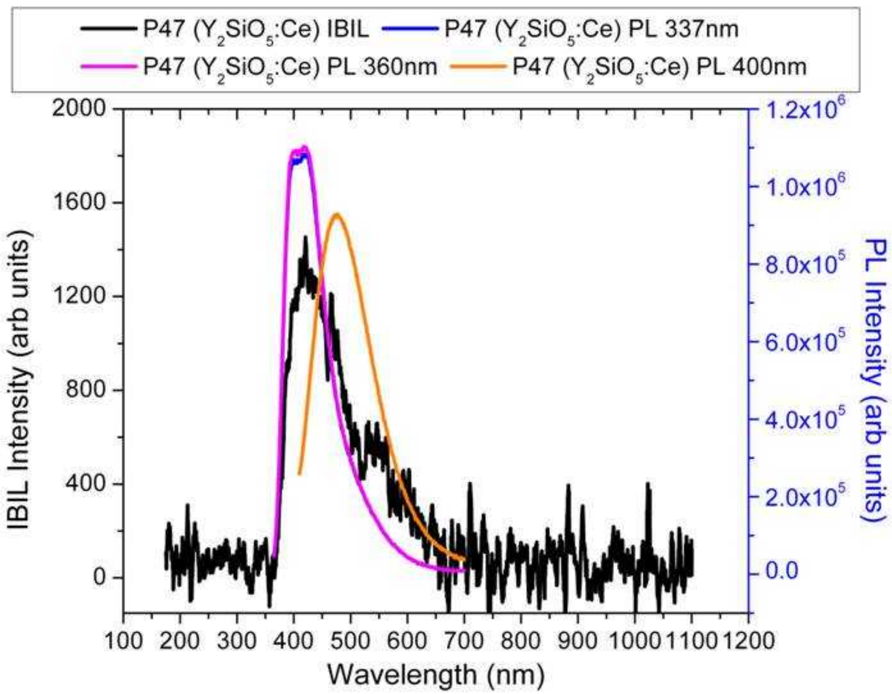


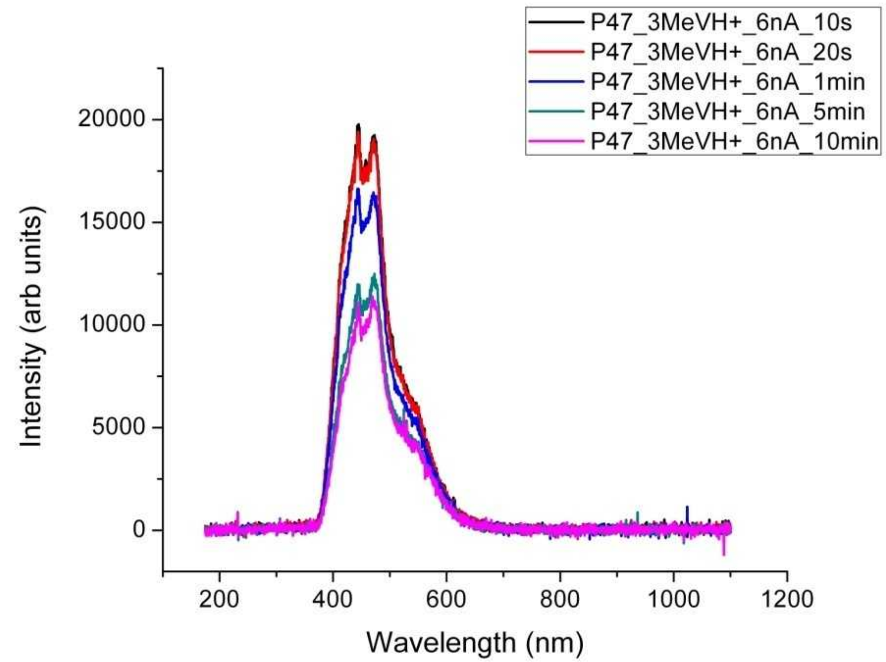
- Spectral discrimination
- CL simulates response to gamma rays
- IBIL simulates response to neutrons

- PL and IBIL of MOF demonstrating spectral discrimination
- IBIL decay of MOFs with irradiation – changes observed in relative peak height

Results – IBIL of Oxides

P47 phosphor studied for potential in radiation effects microscopy

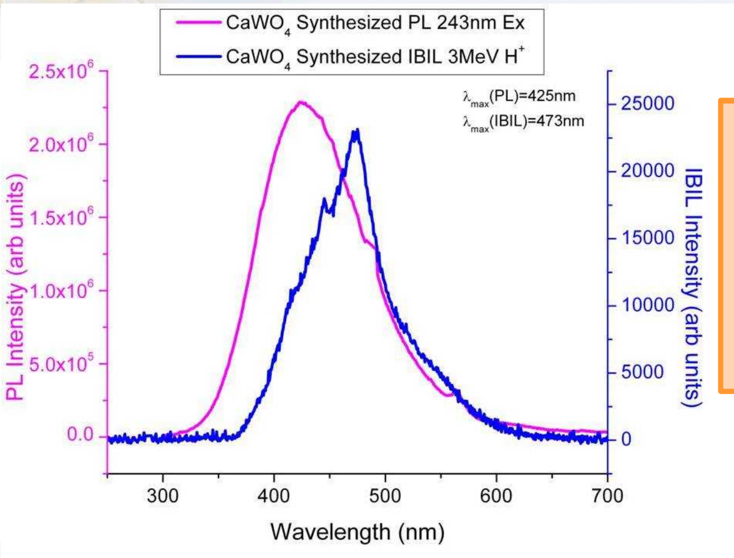




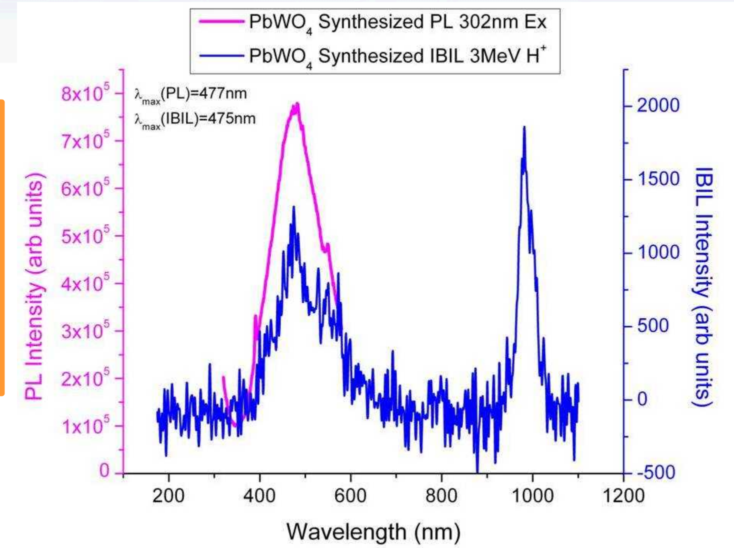
- P47 is effective phosphor – PL and IBIL similar
- Peak emission dependent on excitation wavelength

- Degradation in optical properties also observed in P47
- Oxides demonstrate improved radiation tolerance compared to organic scintillators

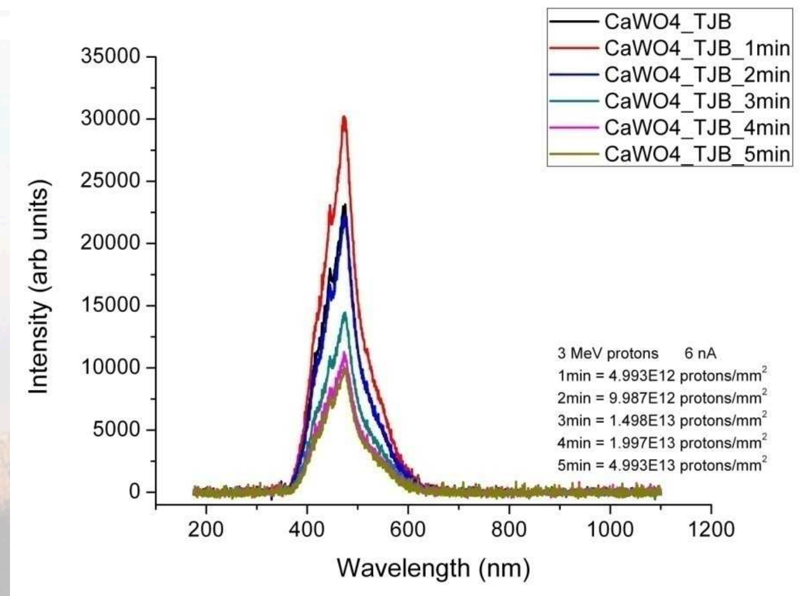
Results – IBIL of Nanoscintillators



Luminescence with proton excitation demonstrates different properties than with UV excitation



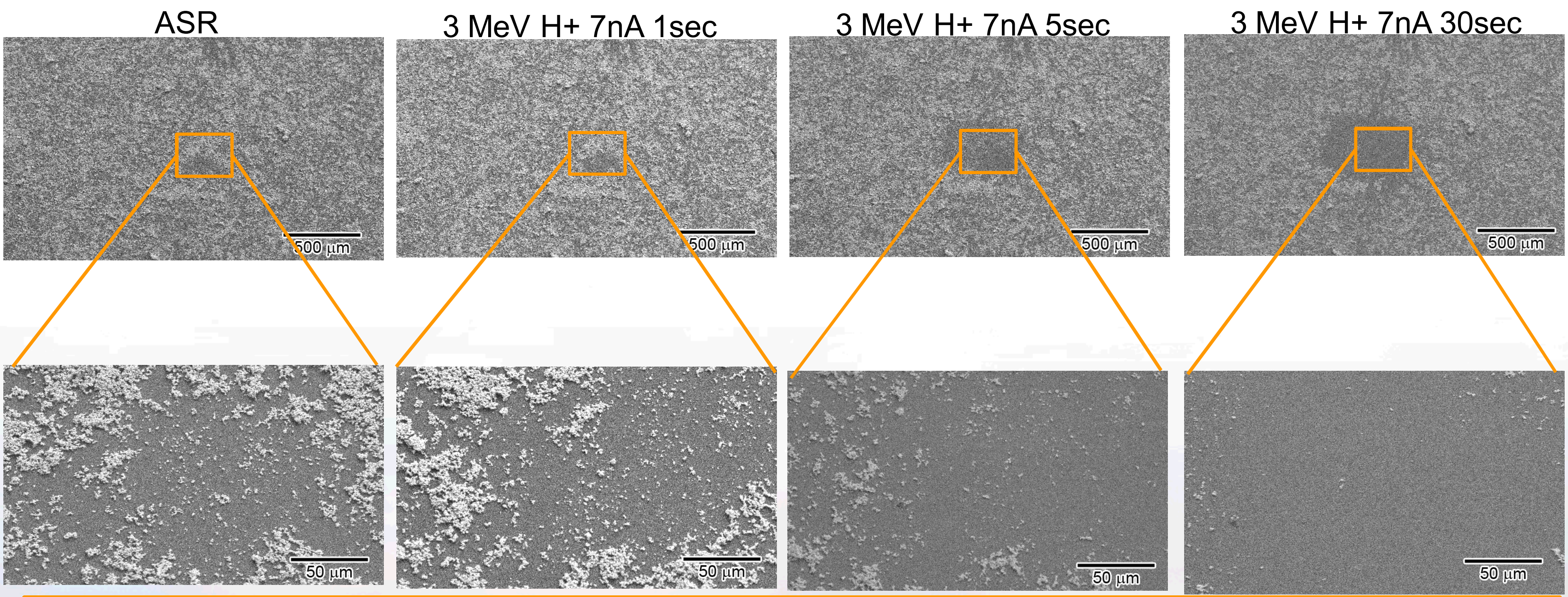
Crucial to study materials with various excitation mechanisms to fully understand luminescent properties



Most materials demonstrate a degradation in optical properties with irradiation – want to understand fundamental mechanism

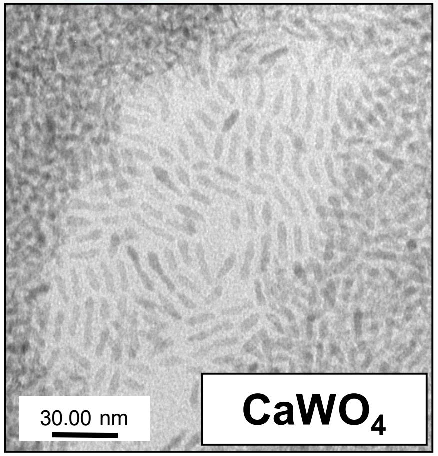
Results - Nanoscintillators

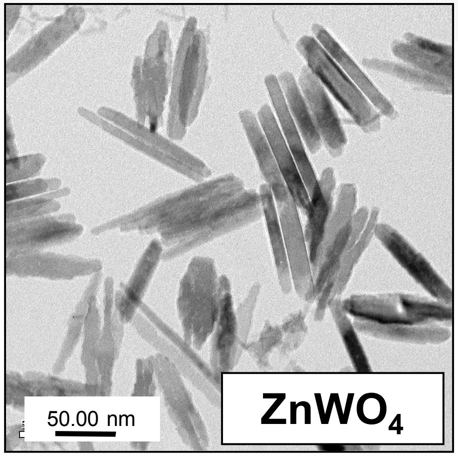
Nearly In-Situ SEM Ion Irradiation of Nanoscintillators

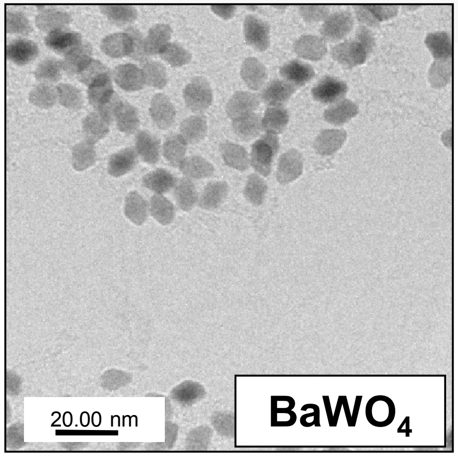


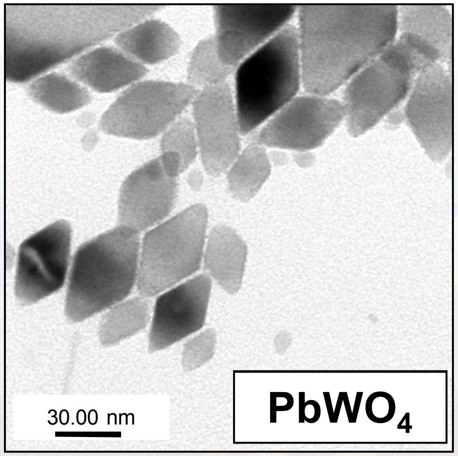
- Want to understand if microstructure is affected by irradiation and how that influences optical properties
- Drop cast films of $PbWO_4$ nanoscintillators irradiated with 3 MeV proton beam, then imaged with SEM
- Material being ablated off of the surface – need better technique to study microstructural changes

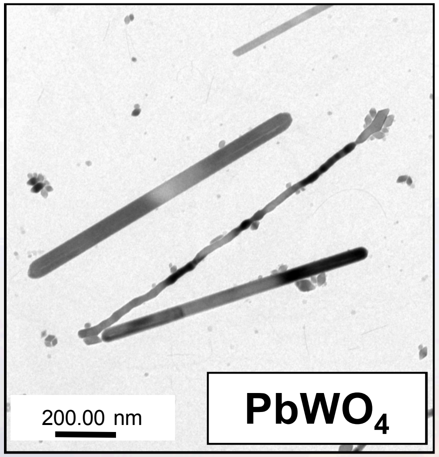
Results – Microstructure







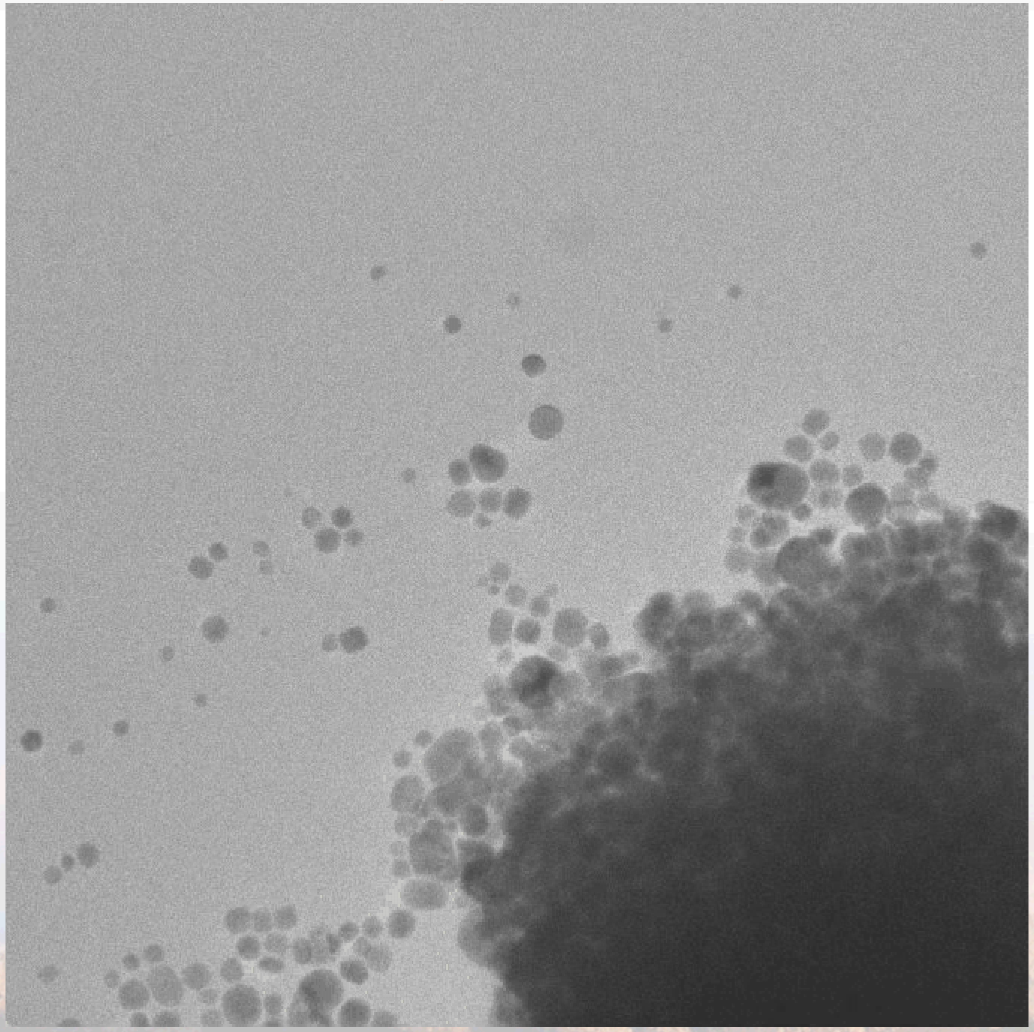




- Entire family of MWO_4 materials synthesized:
  M = Ca, Cd, Pb, Sr, Ba, Zn, Na
- Varying composition and synthesis method results in a range of interesting morphologies

Results - Tomography

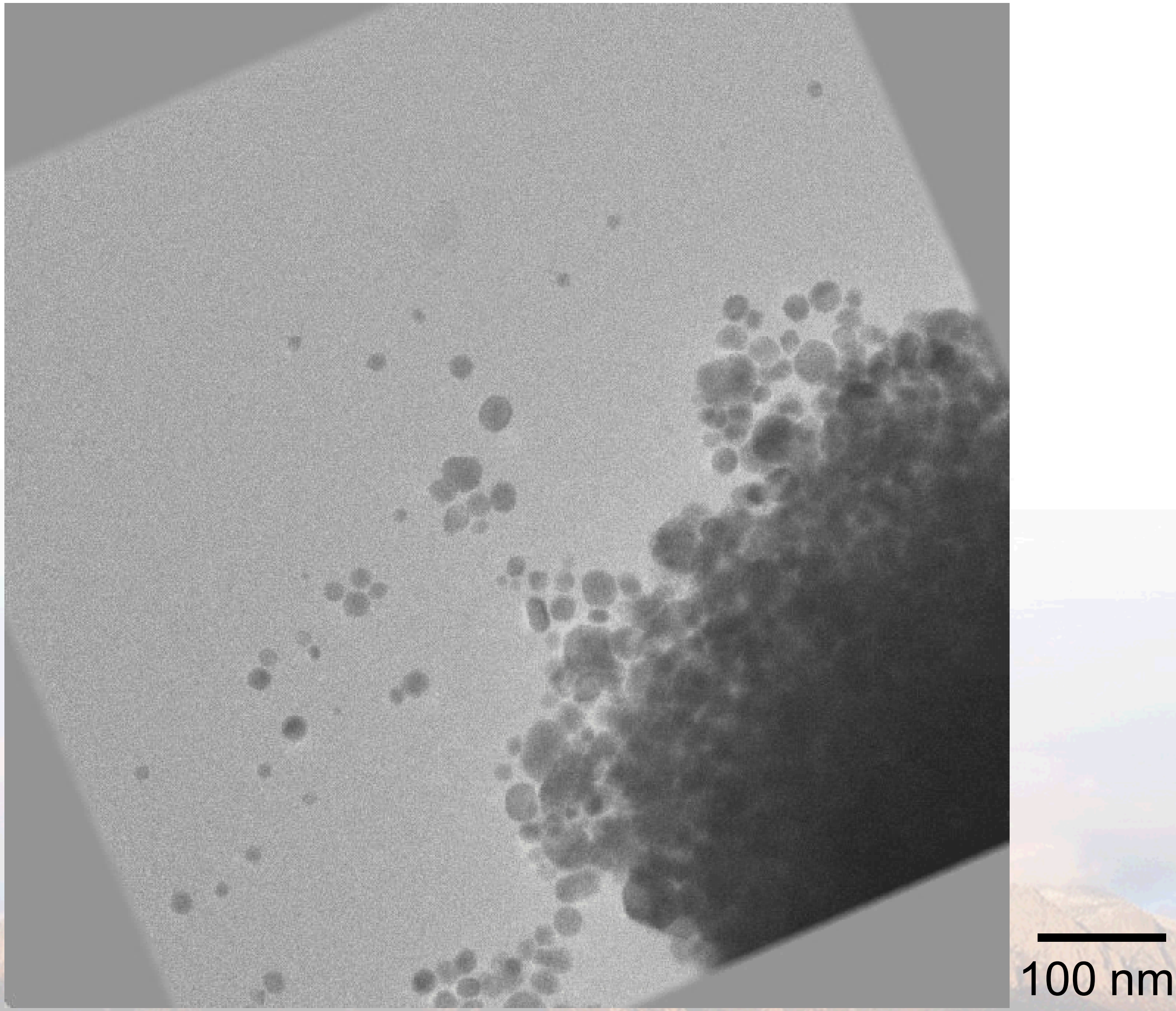
Raw stack of $CdWO_4$ nanoparticles at 60,000X



- Images acquired with SerialEM
- 120° of total tilt
- Each frame collected at 1.5° of tilt

Results - Tomography

Aligned and rotated stack of $CdWO_4$ nanoparticles at 60,000X

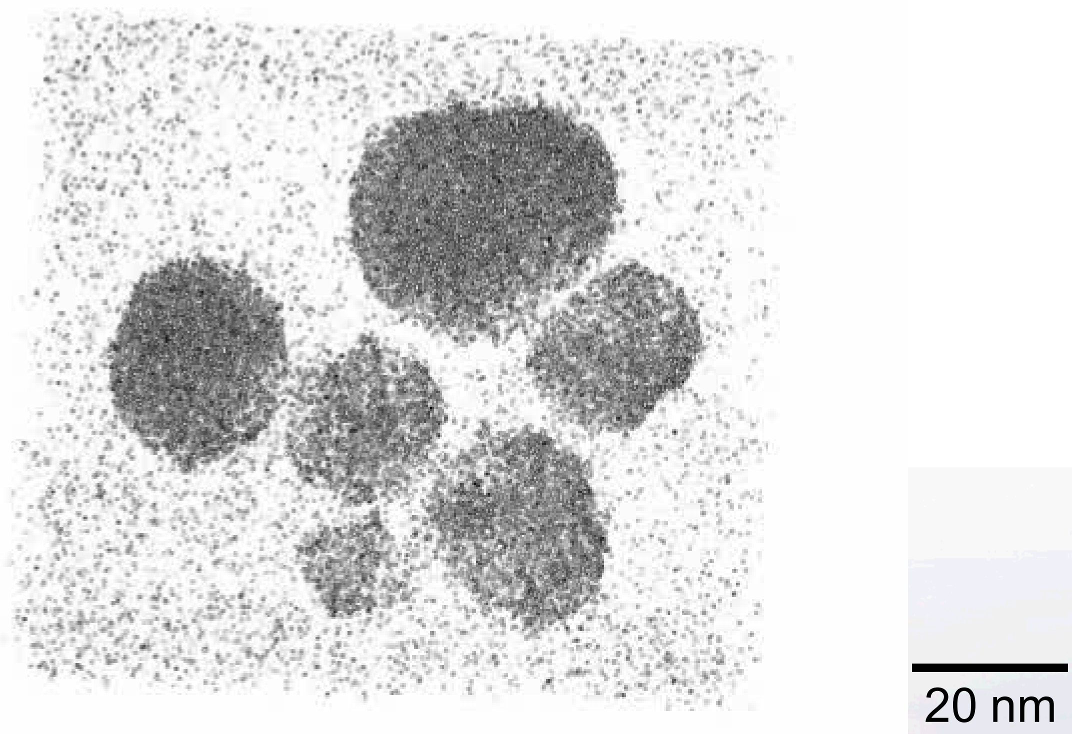


- Aligned to remove jitter
- Rotated so tilt axis is vertical

Results - Tomography

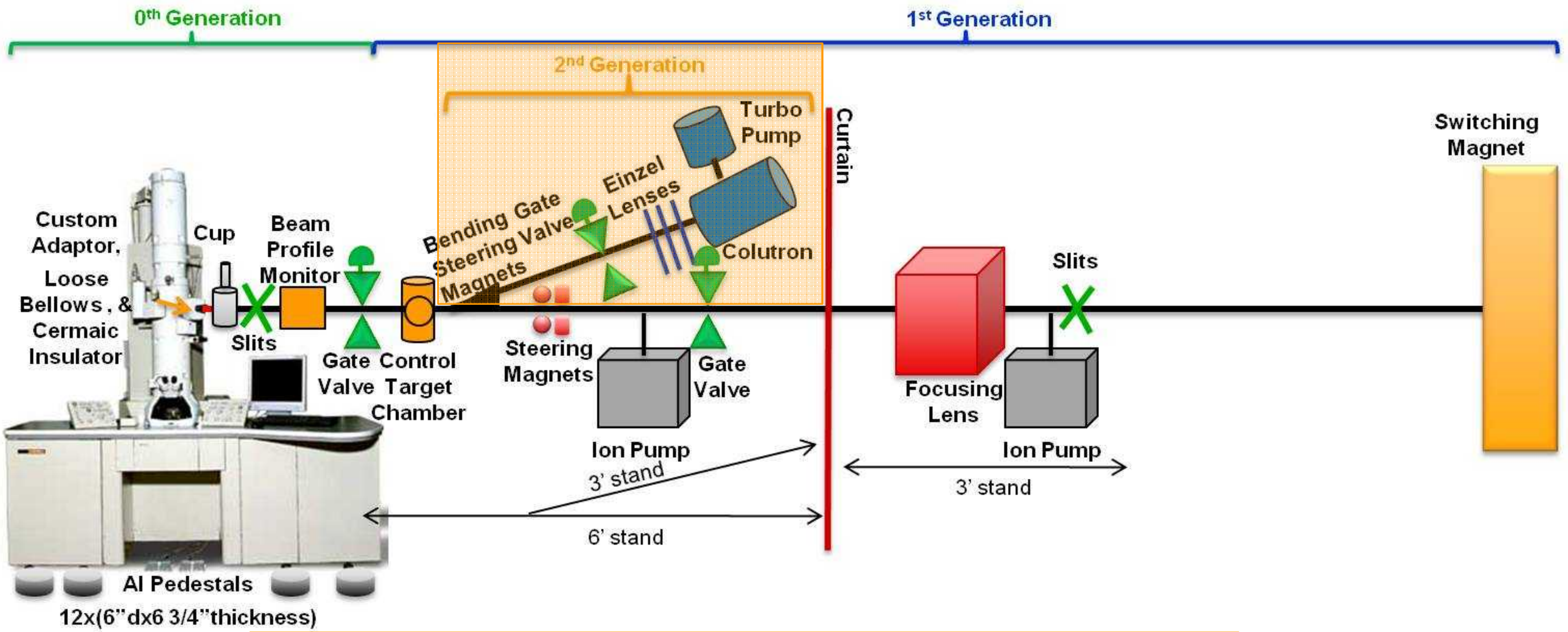
Processed tomography data of $CdWO_4$ nanoparticles at 60,000X

- Processing done in eTomo as part of the IMOD imaging processing package
- Particle size: ~20-50 nm

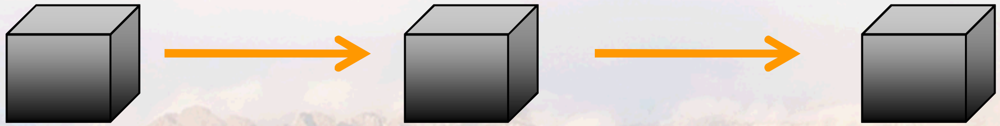


Kremer J.R., D.N. Mastronarde and J.R. McIntosh (1996) Computer visualization of three-dimensional image data using IMOD. J. Struct. Biol. 116:71-76.
Mastronarde, D. N. (1997) Dual-axis tomography: an approach with alignment methods that preserve resolution. J. Struct. Biol. 120:343-352.

Future Work



3D tomo → Irradiation → 3D tomo →Irradiation → 3D tomo…



- Develop 2nd generation of TEM ion beam line at Sandia
- 4D (in-situ TEM tomography)

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Conclusions & Acknowledgements

- In-situ TEM Ion Irradiation is proving to be a useful technique to study potential novel scintillating materials
- Radiation-solid interactions of nanomaterials is not well-understood
- Observing scintillating materials during irradiation is crucial to further developing them for application
- Further studies will give insight into if/how microstructural changes under irradiation affect optical properties

- All staff and technologists of Ion Beam Lab at SNL, especially Dan Buller
- Mark Kinnan & Steve Thoma for PL measurements; PinYang for CL measurements
- Tim Boyle, Sarah Hoppe, Thu Doan, Derek Wichhart, Alia Saad for nanoparticle synthesis
- Patrick Feng for MOF synthesis and characterization
- Science of Extreme Environments Research Institute (SEERI) for support and funding.
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