Nuclear Science and Security Consortium
Virtual Scholar Showcase 2020

Metal-Organic Frameworks as a Platform for Probing Transplutonium Electronic Structure and Signatures

June 3rd, 2020

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The George Washington University

NSSC
NUCLEAR SCIENCE and SECURITY CONSORTIUM

June 2 - 3, 2020

Introduction

Dr. J. August Ridenour, PhD
Chemistry, completed this year

Advisor: Dr. Christopher L. Cahill, GWU
Lab Mentor: Dr. Robert Rundberg, LANL
Focus Area: Radiochemistry and Forensics
Crosscutting Area: Nuclear Security Policy

Brief Research Description: Fundamental exploration of nuclear relevant *f*-element materials; their structure and spectroscopic signatures.

Three highlights of graduate career:
1. Collaborative research projects facilitated and sponsored by the NSSC
2. Science and Nuclear Materials – Teaching Assistantship through NSSC
3. People, connections, and experiences

Participated in projects at LANL and collaborative efforts between GWU, UNLV, and PNNL

GW-UNLV-PNNL Collaboration

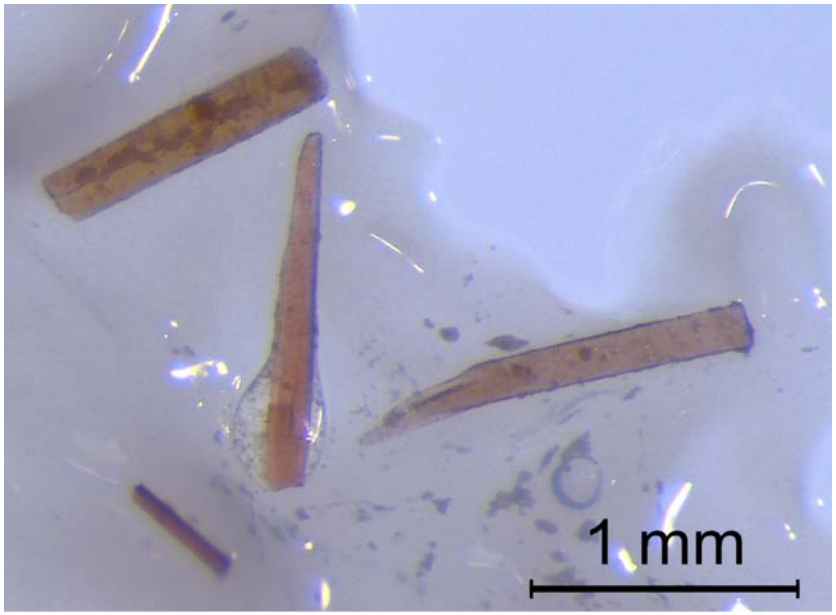
Transplutonium MOF

Deutsche Ausgabe: DOI: 10.1002/ange.201909988
Internationale Ausgabe: DOI: 10.1002/anie.201909988

An Americium-Containing Metal–Organic Framework: A Platform for Studying Transplutonium Elements

*J. August Ridenour, Robert G. Surbella III, Artem V. Gelis, Daniel Koury, Frederic Poineau, Kenneth R. Czerwinski, and Christopher L. Cahill**

Ridenour *et al.* Angew.Chem.Int. Ed. 2019, 58,16508 –16511.



Crystalline material: ^{243}Am-containing GWMOF-6

Initial non-radioactive work completed at : GW

Synthesis and structural characterization undertaken at: UNLV

Spectroscopic analyses completed at: Pacific Northwest NATIONAL LABORATORY

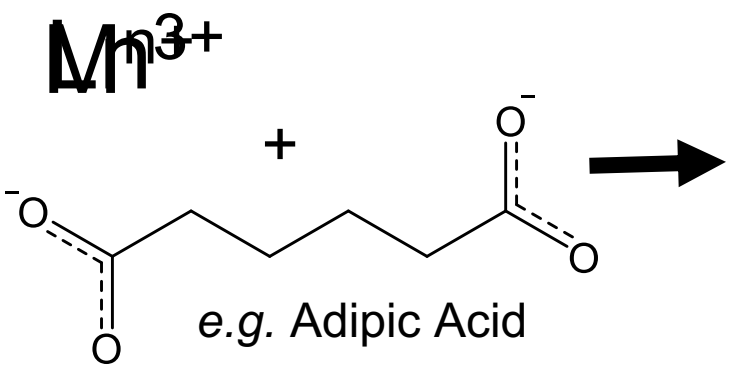
Relevance to the NNSA Mission

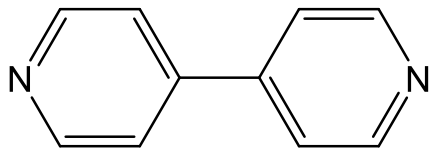
- Establishing knowledge and capabilities for synthesis and characterization of materials containing transplutonium elements as relevant to nuclear science and security

- Studying the spectroscopic signatures thereof to correlate luminescence profiles as a function of coordination environment and chemical make-up for spectroscopic identification

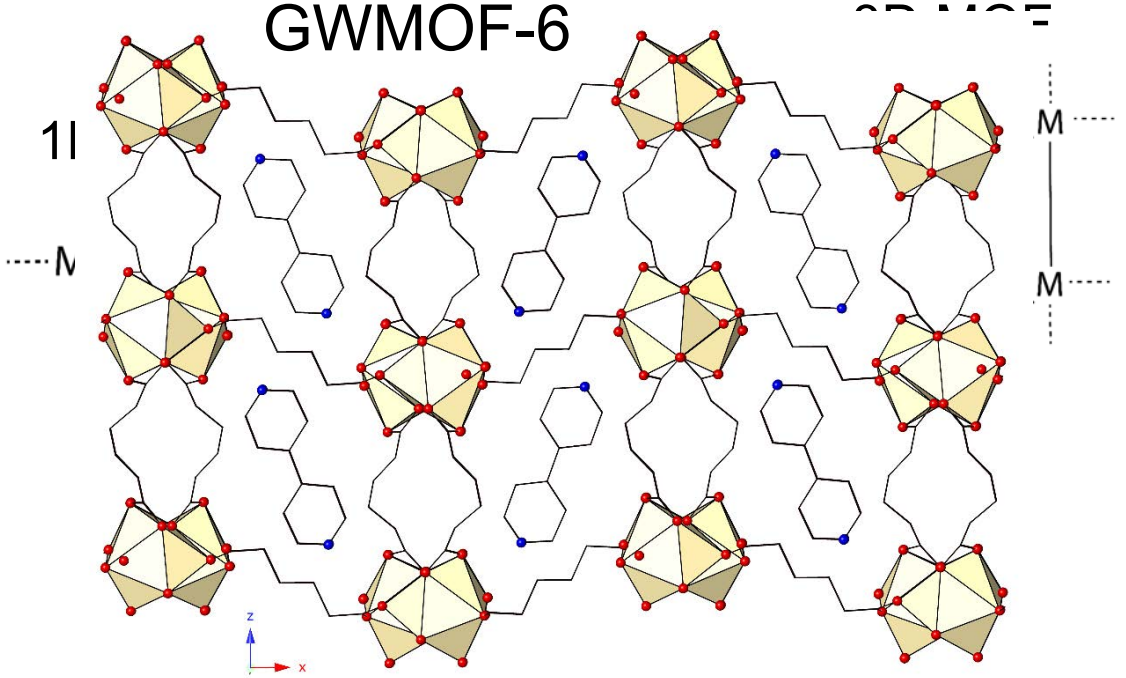
What are Metal-Organic Frameworks?

- Metal-Organic Frameworks (MOFs) are a class of compounds with a diverse array of properties
  - Applications in separations, gas sorption, nanotechnology, biomedicine, among many others
- MOFs provide a rigid, porous framework consisting of both metal center nodes and organic linkers



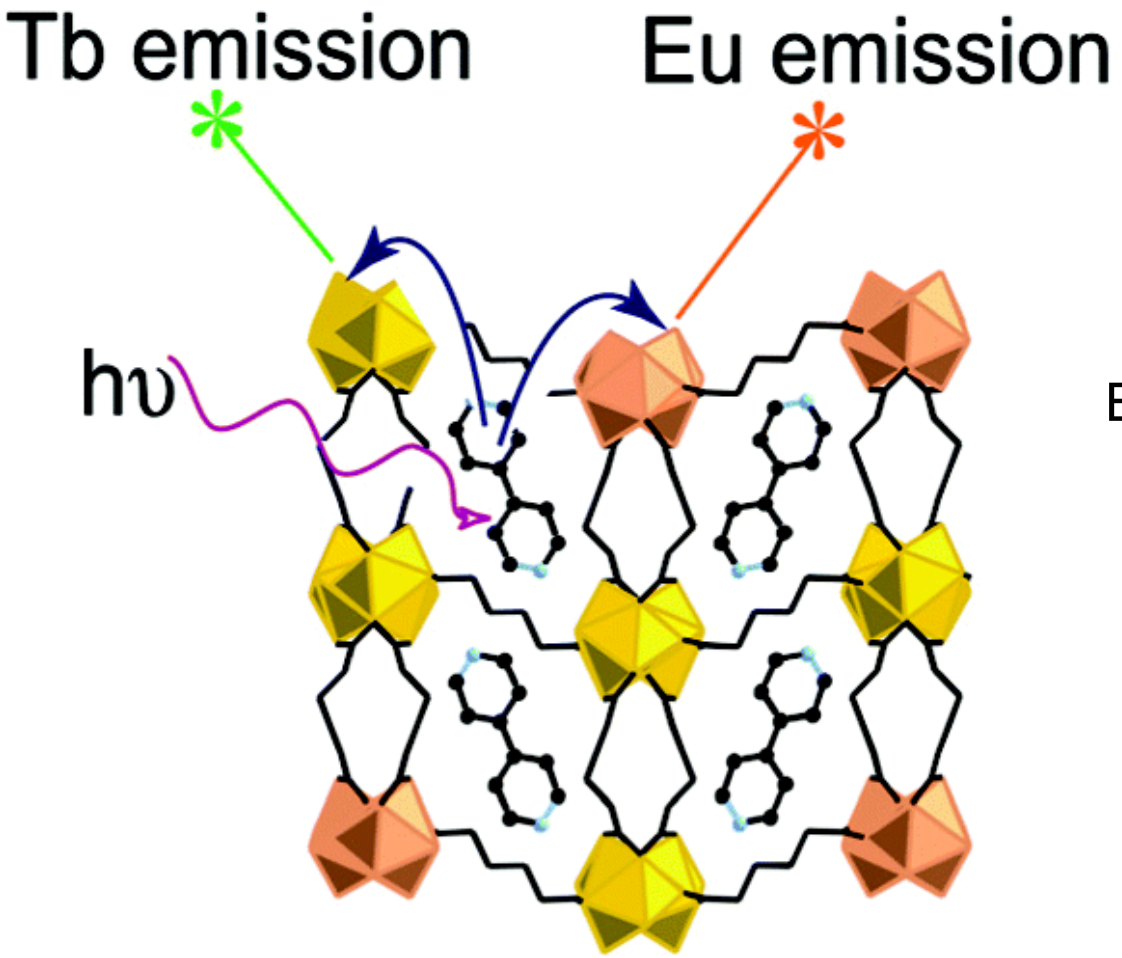


4,4'-bipyridine



De Lill, Cahill, Inorg. Chem., 2005, 44, 2, 258-266.

Lanthanide GWMOF-6



Guest-enhanced luminescence *via* the antenna effect

Broadly: A platform for studying Transplutonium elements

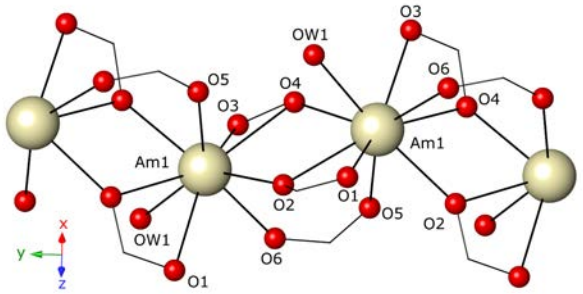
1. Provides a rigid framework for the incorporation of actinides to study their properties

2. Utilizes the properties of the MOF to *enhance* the properties of the actinide for more efficient study

De Lill, de Bettencourt-Dias, Cahill, Inorg. Chem., 2007, 46, 10, 3960-3965.

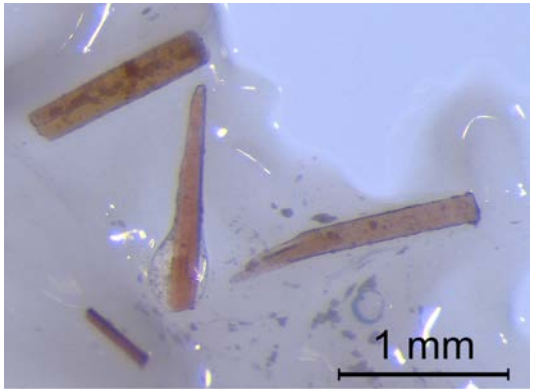
Why GWMOF-6 for Am^{3+}?

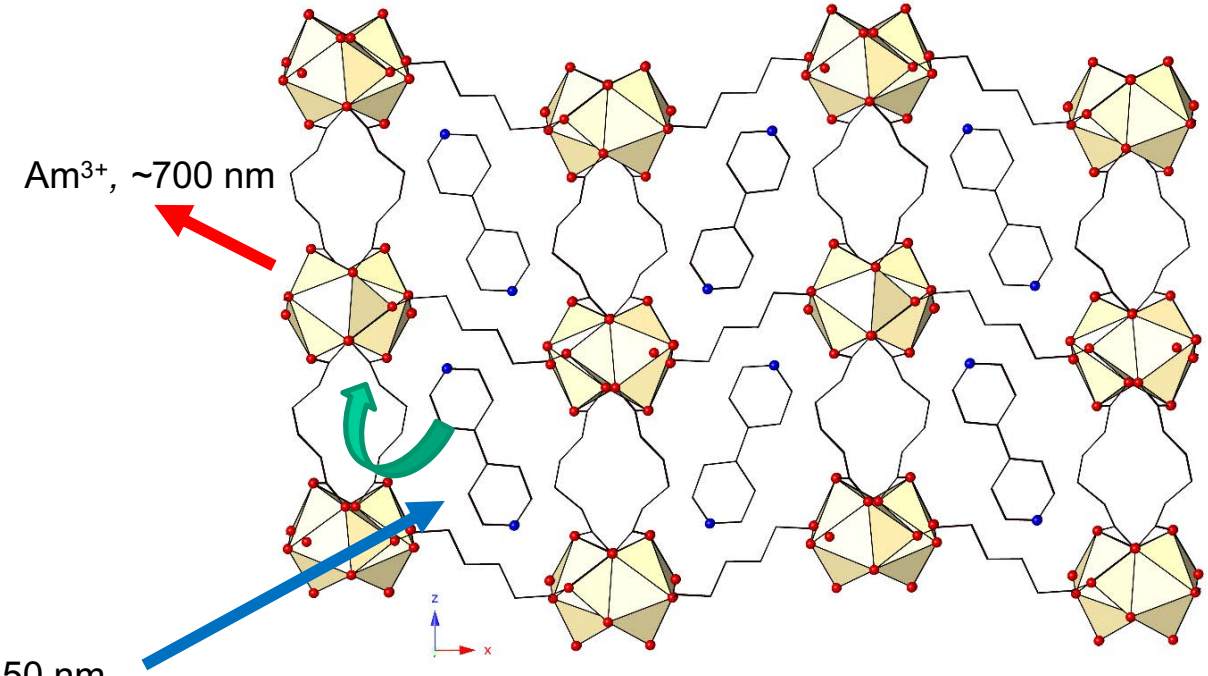
- Specifically: An^{3+} into the GWMOF-6 architecture, a trivalent *lanthanide*-containing MOF,
  - Chemical similarities to trivalent actinides
  - Scalable synthesis, with large single crystalline material
  - **Aromatic guest molecules for ligand-to-metal energy transfer**



Ionic Radii

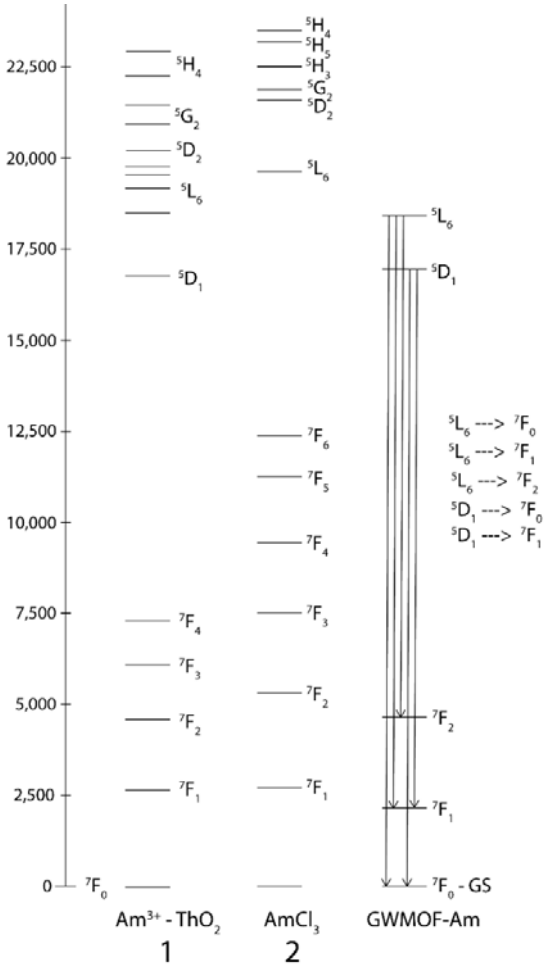
Lanthanide	Americium
Pr^{3+}: 1.126 Å	Am^{3+}: 1.09 Å
Nd^{3+}: 1.109 Å	

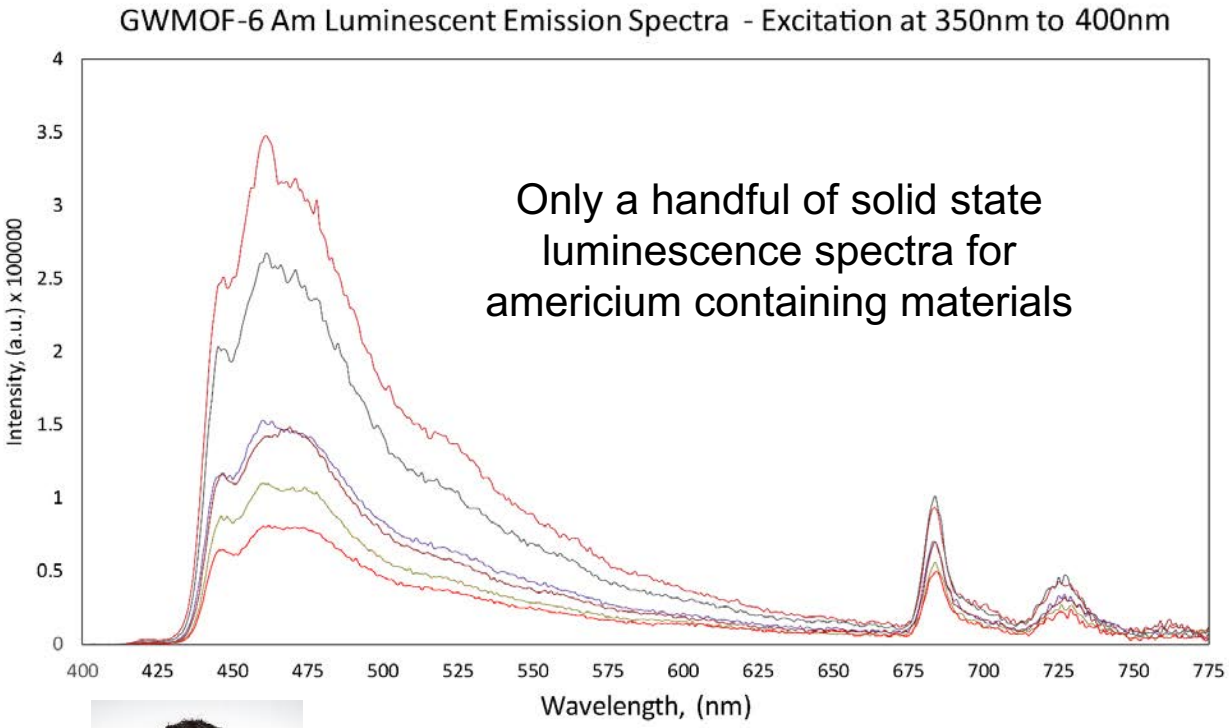




Spectroscopic Analysis

- Americium(III) luminescence signature
  - The 4,4'bipyridine provides an energy pathway for guest-enhance luminescent emission





Only a handful of solid state luminescence spectra for americium containing materials

Dr. Gian Surbella

Pacific Northwest
NATIONAL LABORATORY

An Americium Metal-Organic Framework (MOF)

Transplutonium MOF

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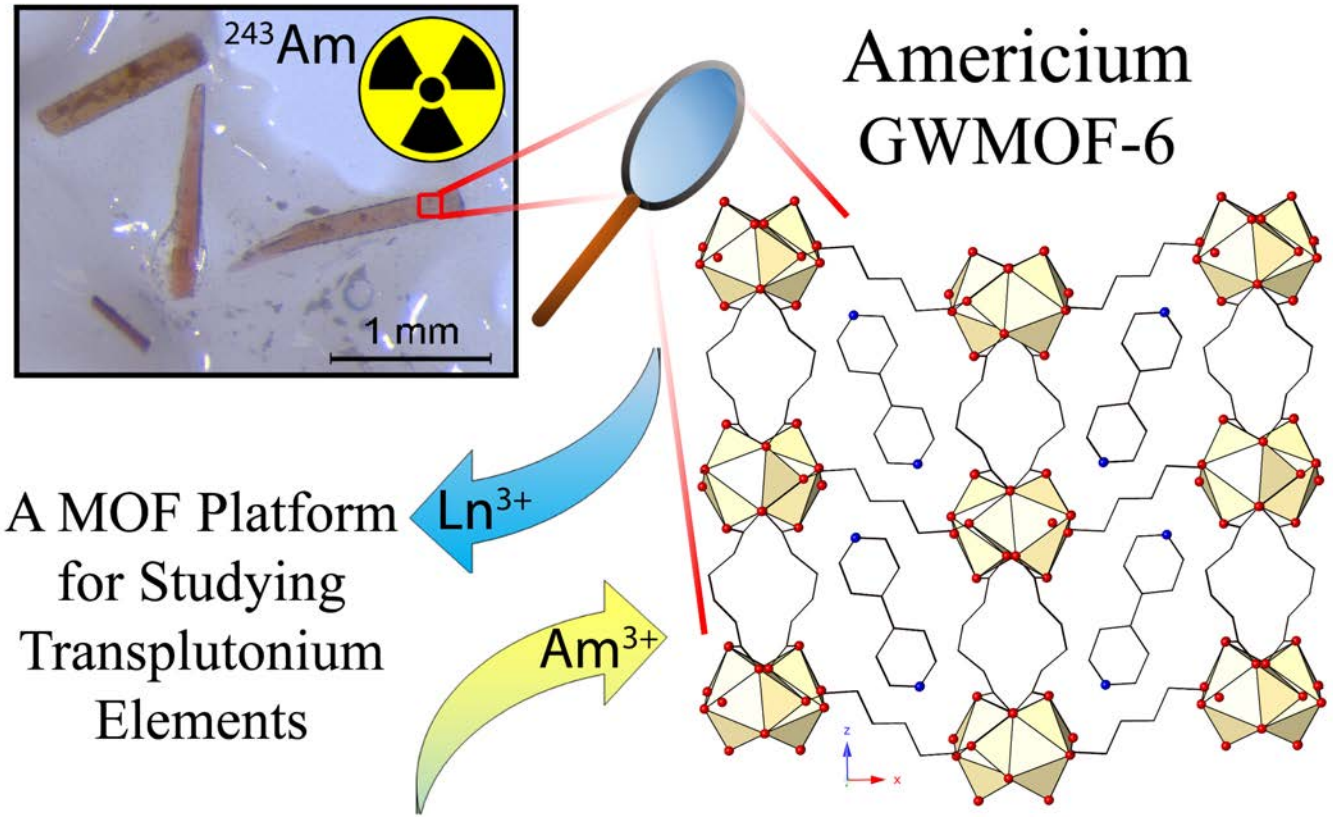
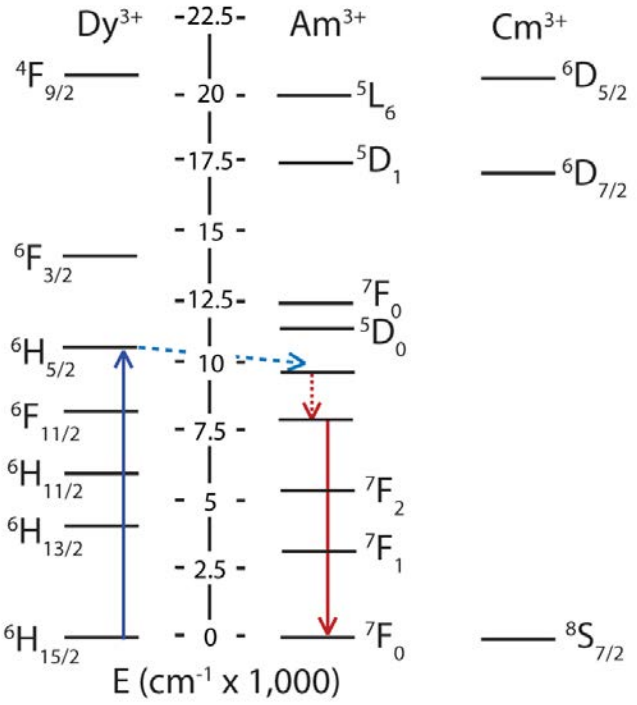


Future Directions

- Massive compendium of Ln^{3+}-based MOFs – can we find one that is more appropriate for harnessing and enhancing Americium spectroscopic properties?

- Explore the structural dependence (the coordination environment) of Am^{3+} luminescence

- Continue to explore spectroscopic signatures
  - Solid-state UV-Vis, luminescence lifetime, *etc*
  - Effect of water in the inner coordination sphere
  - Effect of An^{3+} concentration quenching
  - Metal center site symmetry
  - Energy transfer between 4*f* and 5*f* metal centers

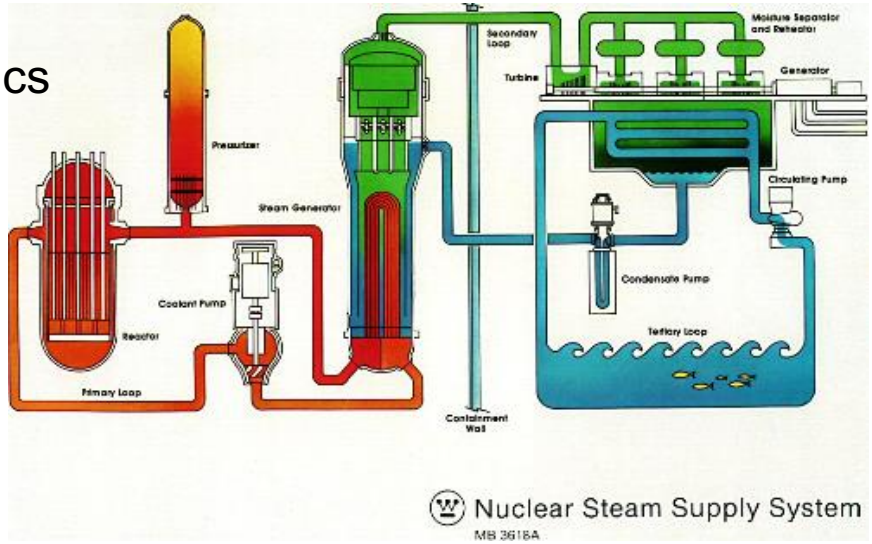


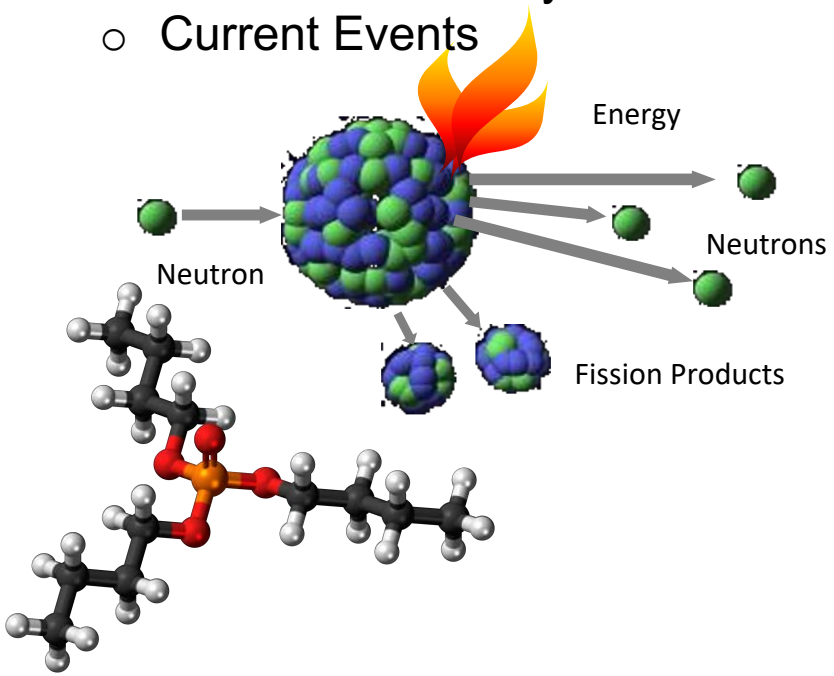
Theoretical 4*f*-5*f* energy transfer pathway

Science of Nuclear Materials (ScNM) Course
at the Elliot School of International Affairs
Co-sponsored by the NSSC

Teaching Assistant for the "Science of Nuclear Materials" at GW's Elliot School of International Affairs

- Basics of Nuclear Chemistry and Physics
- Fission Dynamics
- Nuclear Reactors
- History of Weapons
- Power and Legacy Nuclear Waste
- International Policy and Treaties
- Current Events



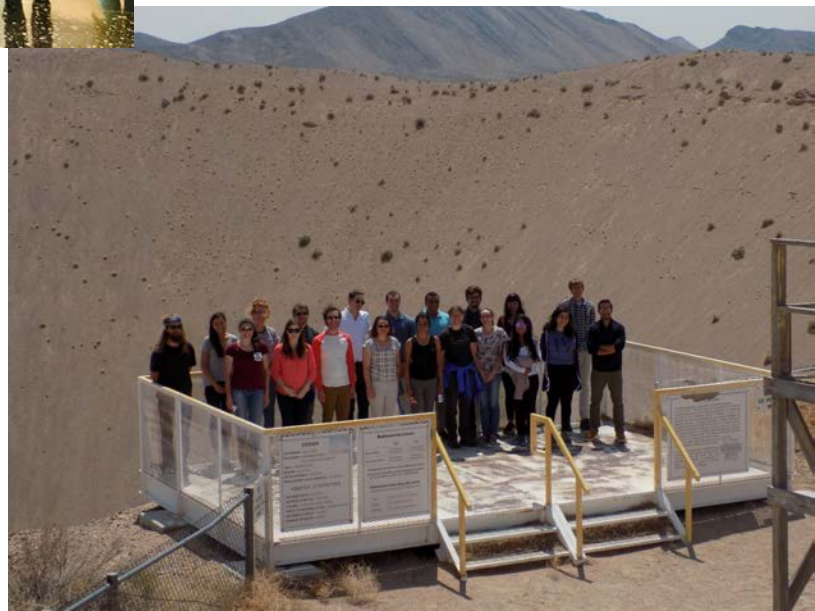


In my three years teaching…

- ~35 science or nuclear science policy graduate students
- Many were working in executive branch positions; e.g. State, NNSA, NRC
- I learned a different way to think and communicate within this nuclear security community

NSSC Experience

Some highlights: Trips to Los Alamos, Sandia, the State Department building in DC, and the Nevada National Security Site



NSSC Experience

2014, Dickinson College
BS in Chemistry and History

2015, Began at GWU
PhD, Chemistry

2017, Began Engagement with NSSC – Three Years of Teaching Assistantship

2018, Keepin Nonpro. Summer Program at LANL

2018, Participation in Schubert Review, UNLV

2019, GW Boot Camp on Nuclear Security Policy

2019, Transplutonium Research at UNLV and PNNL

2019, University Program Review – Raleigh

2019, NSSC Fall Workshop – Livermore

2020, March 3rd – Defended Dissertation

2020, May 17th – Graduated from GW

Acknowledgements

Dr. Christopher Cahill
Advisor, GWU

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PNNL

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UNLV

Dr. Artem Gelis
UNLV

Dr. Frederic Poineau
UNLV

Dr. Daniel Koury
UNLV

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