# PNNL capabilities for reducing the impact of emissions from Mo-99 production

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2018 Mo-99 Topical Meeting – Knoxville, TN

# Outline

- PNNL's policy for engaging on medical isotope production
- PNNL Background
  - Our Identity
  - PNNL's involvement in medical isotope production (MIP) community
  - Adsorbent materials research
- Emissions reduction – what can be done?
- Current progress
  - Work performed under current project for NNSA

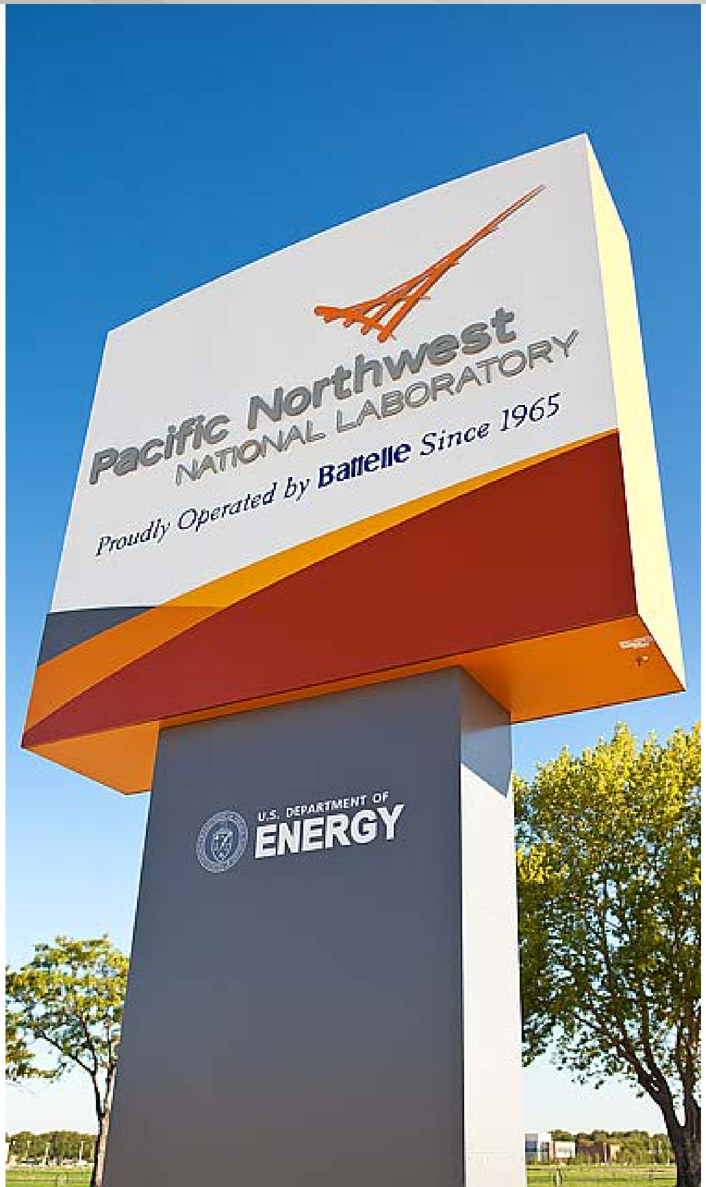
# PNNL Policy for Engaging on Medical Isotope Production

- PNNL has been involved with the mitigation of the effects of medical isotope production on the non-proliferation environment
  - In particular, emissions from fission-based Mo-99 production are known to be the dominant source of airborne radioactive isotope background
  - Emissions of xenon isotopes are particularly problematic for the detection of nuclear explosions

# PNNL Policy for Engaging on Medical Isotope Production

- To avoid conflicts of missions, we will continue to work with isotope producers with the following caveats. PNNL will:
  - Not permit the use of its facilities for the commercial production of Mo-99
  - Engage with domestic and international producers of medical isotopes to reduce the impact of production emissions on nuclear explosion monitoring systems through the use of:
    - Emissions control systems
    - Stack monitoring
    - Other activities that may reduce emissions
  - Be open and transparent and provide all of our R&D on an equal basis to any producers interested in those results
    - Proprietary issues, will of course, be honored

# Our Identity




## 3-PRONGED

### PNNL

A research laboratory that addresses many of America's most pressing challenges in energy, the environment, and national security through advances in basic and applied science.

### U.S. Department of Energy

PNNL is a U.S. Department of Energy government research laboratory managed by the Office of Science.

### Battelle

We have been operated by Battelle Memorial Institute since our inception in 1965. Battelle is a private nonprofit applied science and technology development company headquartered in Columbus, Ohio.

# Our History

## 1940s

LAB BEGAN IN SUPPORT OF MANHATTAN PROJECT

## 1965

PACIFIC NORTHWEST LABORATORY IS ESTABLISHED

## 1980s

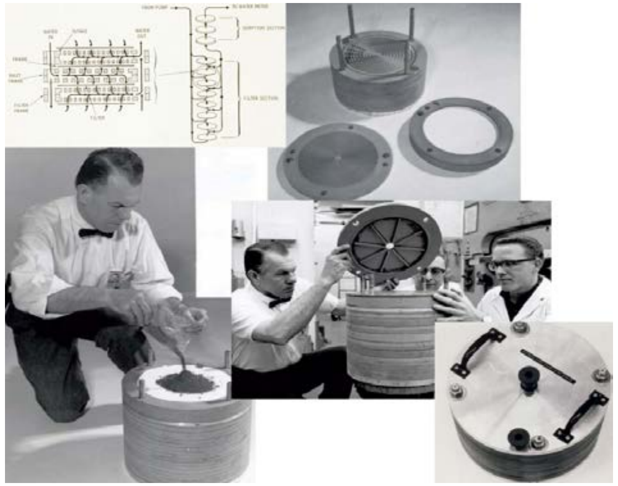
CLEANUP EFFORT BEGINS FOLLOWING THE SHUTDOWN OF THE LAST HANFORD REACTOR

## 1990s

TRACE DETECTION EXPERTISE IS LEVERAGED TO SUPPORT MODERN NUCLEAR EXPLOSION MONITORING

## Today

PNNL CONTINUES TO SUPPORT THE U.S. GOVERNMENT IN TRACE DETECTION OF RADIONUCLIDES IN THE ENVIRONMENT



# MIP COMMUNITY INVOLVMENT

## ABATEMENT RESEARCH

ABATEMENT, WHILE NOT ALWAYS FEASIBLE IN EVERY CASE, IS STILL THE MOST EFFECTIVE MEANS TO MITIGATE THE CHALLENGES OF RADIOXENON EMISSIONS

## WOSMIP

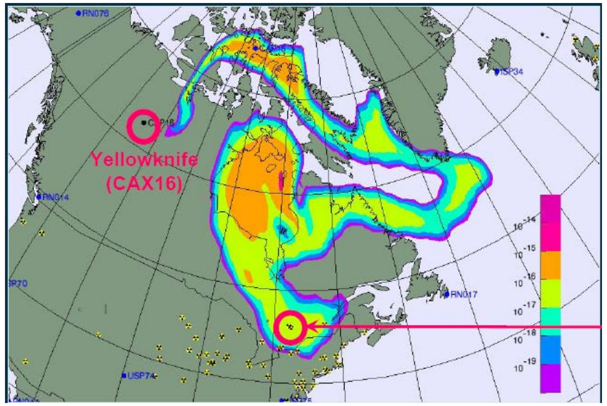
THE NEXT WOSMIP IS SCHEDULED FOR December 3-7, 2018

## UNDERSTANDING THE SCIENCE

WHETHER THOUGH EXAMINING FACILITY RELEASE PATHWAYS OR ATMOSPHERIC TRANSPORT, WORK REMAINS ON UNDERSTANDING THE SCIENCE

## STAX PROJECT

STAX AIMS TO PURCHASE AND INSTALL STACK MONITORING SYSTEMS AT COOPERATING FACILITIES TO AUGMENT THE CAPABILITY OF THE CTBT IDC TO DISTINGUISH EMISSIONS FROM ISOTOPE PRODUCTION



# Adsorbent Materials Research and Process Development

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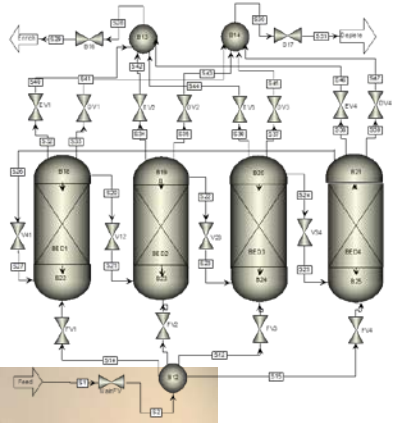
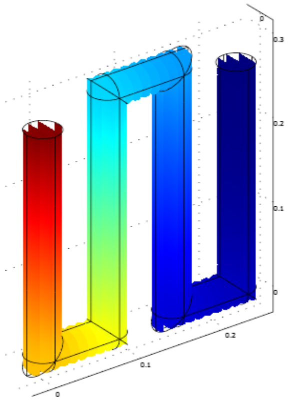
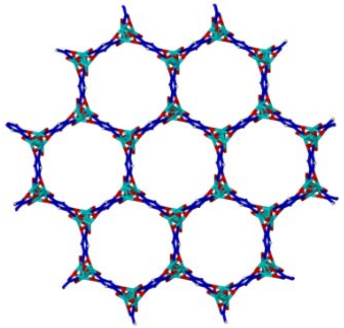
- **Adsorption Process Modeling**
  - Aspen Adsorption
  - COMSOL Multiphysics
- **Hiden Gravimetric Adsorption Analyzer**
  - Can output the rate of uptake, as well as the final amount adsorbed at a given pressure and temperature
- **Adsorption Breakthrough Instrument**
  - Can perform breakthrough experiments for gas mixtures (up to three gas species and a carrier).
- **Metal Organic Frameworks**
  - Constructed from precursor chemicals that in most cases are readily available
  - Promising xenon adsorption properties for gas separation (NiDOBDC)



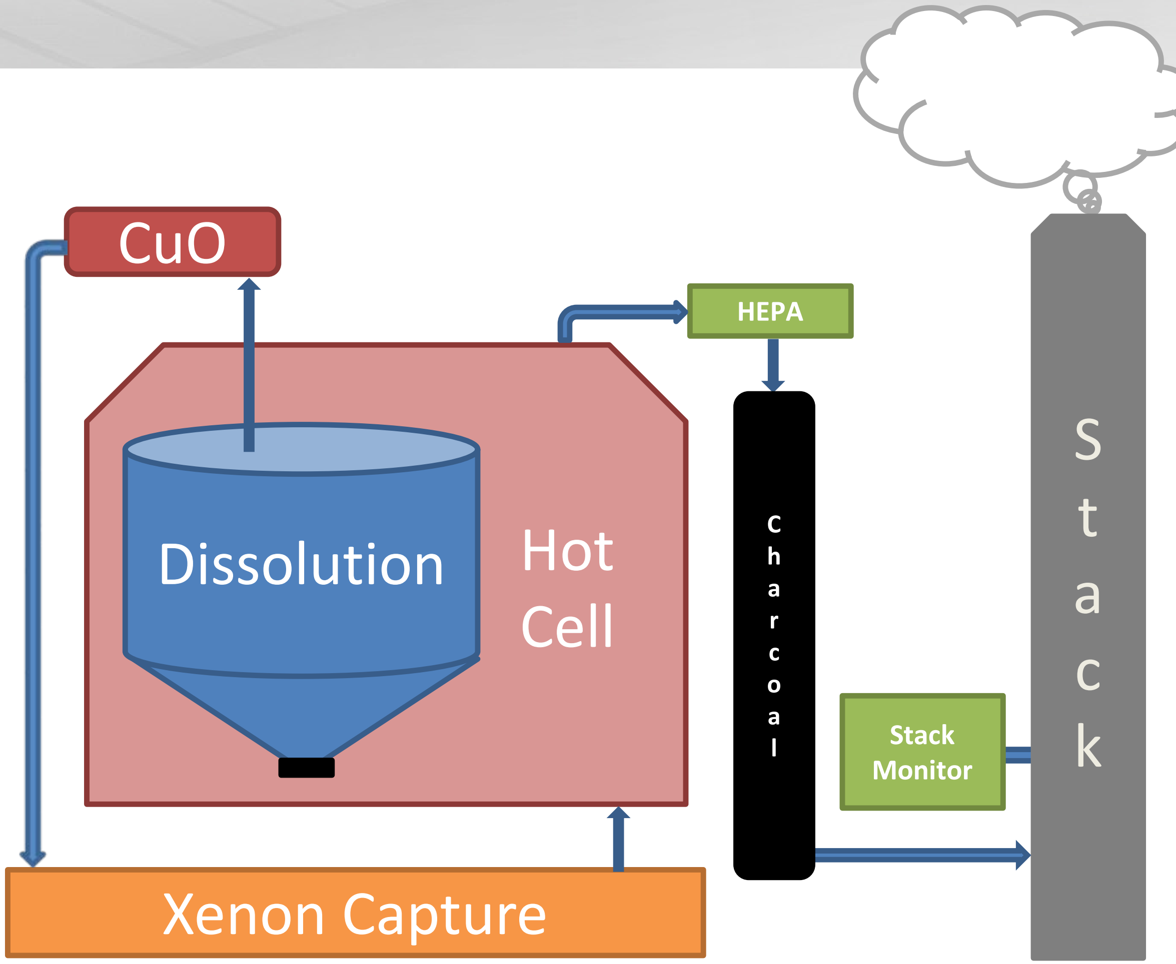


Cooper and Thallapally, Nature Materials 2014

# Emissions reduction – what can be done?

- Each facility is different
  - No single solution
- The level of emissions released from a facility is dependent on three principals.
  - Operational Sharpness
  - Chemistry
  - Abatement systems/engineered systems
- Stack Monitoring
  - Real time feedback




**Abatement system**. Diagram for description of facility abatement systems. This is a simplified diagram and actual systems would be engineered according to the chemistry and facility needs.

- Iodine
  - Charcoal Filters
  - Silver Zeolite Filters
  - Alkaline Scrubbing
  - Mercurex Process
  - Silver iodide formation
  - Containment/Delay Tanks
- Particulate
  - HEPA Filtration
- Xenon
  - Cooled charcoal beds
  - Containment/Delay Tanks
  - Silver Zeolite Filters

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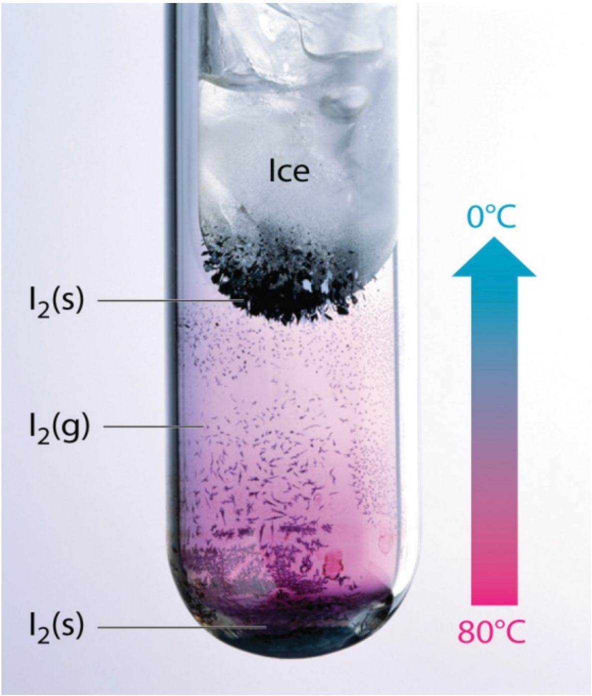
# Operational Sharpness

- Maintaining operational sharpness plays an important role in reducing xenon emissions. Even routine operations must be performed optimally every time for consistency.
- The goal is to minimize off normal events such as breakage that can release volatiles or turning a wrong valve.
- Practice and experience are required for operational sharpness. To achieve this goal, minimal staff turnover and training both play an important roles.

http://www.nordion.com/medical_isotope_update/medical_isotope_photos.asp

# Chemistry

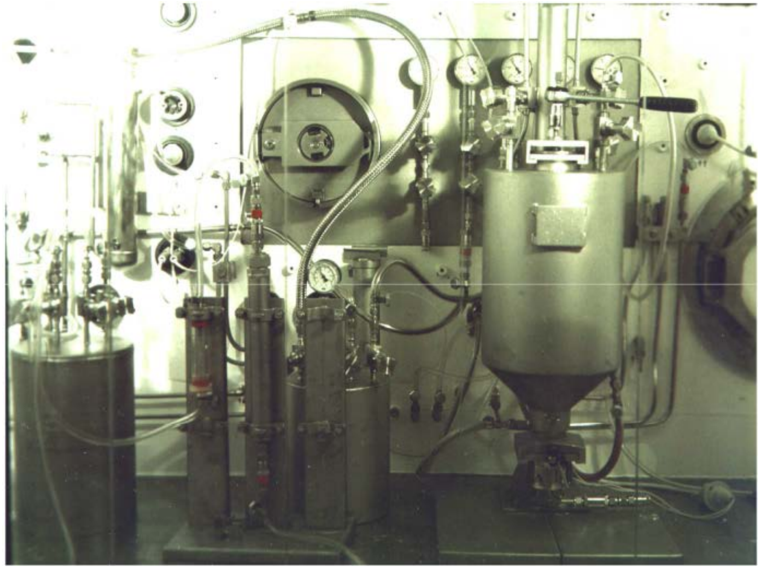
- Xenon is an inert gas and cannot be chemically removed. Engineered barriers are needed to retain radioxenon for it to decay sufficiently before release to the environment.
- Where there is iodine, there is xenon due the decay of radioiodine to xenon. In order to reduce radioxenon emissions iodine abatement must also be considered.
  - $^{133}I \rightarrow ^{133m}Xe \rightarrow ^{133}Xe$
- The chemistry of iodine is complicated making it challenging to control. It can be in solid, aqueous, or gas form depending on the chemistry
  - **Solid** ($I^-$) as salts and organoiodine
  - **Gas** ($I_2$, $CH_2I_2$, $HIO_3$, $IO_2$ dimer, $I_2O_5$)
  - **Aqueous** $I^-$, $I_2$, $I_3^-$, $IO_3^-$, $IO_4^-$, $IO_2$, $I_2O_4$, $I_2O_5$, $HIO_3$
- Solubility of iodine is acid dependent
  - $I_2 (aq) + 2H_2O \leftrightarrow 4I^- + O_2 + 4H^+$
    - $I_2$ is only slightly soluble




http://images.flatworldknowledge.com/averillfwk/averillfwk-fig11_018.jpg

# Chemistry and Abatement

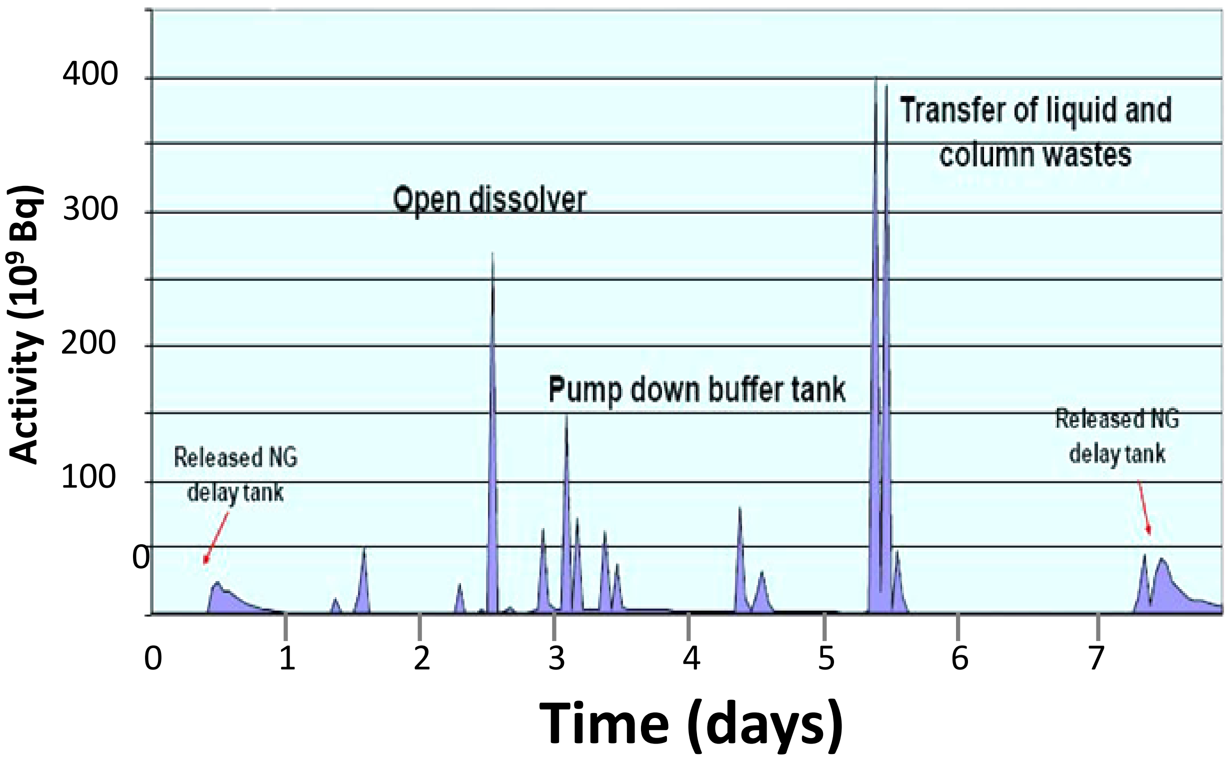
- The largest radioxenon releases occur during dissolution of targets.
- Dissolution chemistry – acidic or alkaline – will determine whether iodine is maintained in the aqueous state or volatilized. Acidic dissolution will favor formation of volatile iodine while alkaline solutions will tend to maintain iodine in solution.
- Designing chemistry for low emissions is important
  - Capture all xenon from dissolution
  - Remove iodine early to avoid xenon releases from downstream processing steps due to iodine decay.
  - Utilize chemical steps that are automatable in sealed/closed systems
    - Any chemistry step that requires a vessel to be opened will cause releases
    - Use material and valves that keep the Xe contained
  - Develop waste management procedures that can be performed in closed/isolated systems
    - Prevents chronic low emissions



**Dissolution and chemical processing hot cell.**
Sameh, WOSMIP 2013

# Stack Monitoring

- Stack monitoring is also an important tool for optimizing radioxenon abatement. Stack monitoring can be used to diagnose radioxenon releases during operational procedures. By assigning xenon spikes to their associated process, potential leaks can be determined and repaired.




**Plot of $^{133}$Xe and associated operating procedures**. Assignment of peaks from stack monitoring are valuable in troubleshooting abatement. Figure from ANSTO presentation, Hoffmann, E., ANSTO, WOSMIP 2009

# Emissions Capture

- Investigation of emission capture technologies for xenon abatment from Mo-99 production facilities
  - Task 1 - Evaluation of Adsorbent Materials (continuation of FY18 support)
    - Continued evaluation of new materials, such as silver mordenite and metal organic frameworks (MOFs) to make a compact design
    - Produce preliminary flow sheets
      - Designed to keep emission below NRC requirements
  - Task 2 - Detailed Exploratory Testing to Verify Emissions Levels
    - Computational models and experiments may be used to test
      - Different adsorbents
      - Cooled abatement traps
      - Trapped gas management and long term storage
      - Trap regeneration
      - Trap design

# Summary

- PNNL has expertise that can be applied to MIP emissions reduction
- Currently working on NNSA's Office of Defense Nuclear Nonproliferation funded emissions reduction project to support xenon abatement
- PNNL will be open and transparent and provide all of our R&D on an equal basis to any producers interested in those results