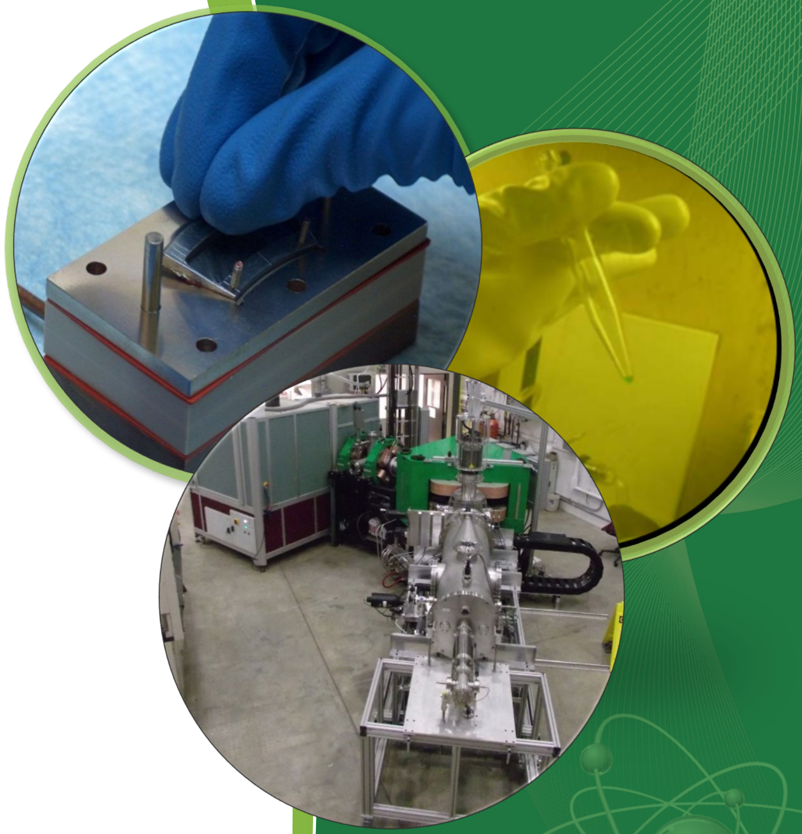
Handling of Ra-226 feedstock at ORNL

DOE Isotope Program: Virtual Roundtable on the Best Practices and Safe Handling of Radium-226

May 30, 2024



Dan Stracener
Oak Ridge National Laboratory

This work is supported by the U.S. Department of Energy Isotope Program, managed by the Office of Science for Isotope R&D and Production.

ORNL is managed by UT-Battelle
for the US Department of Energy

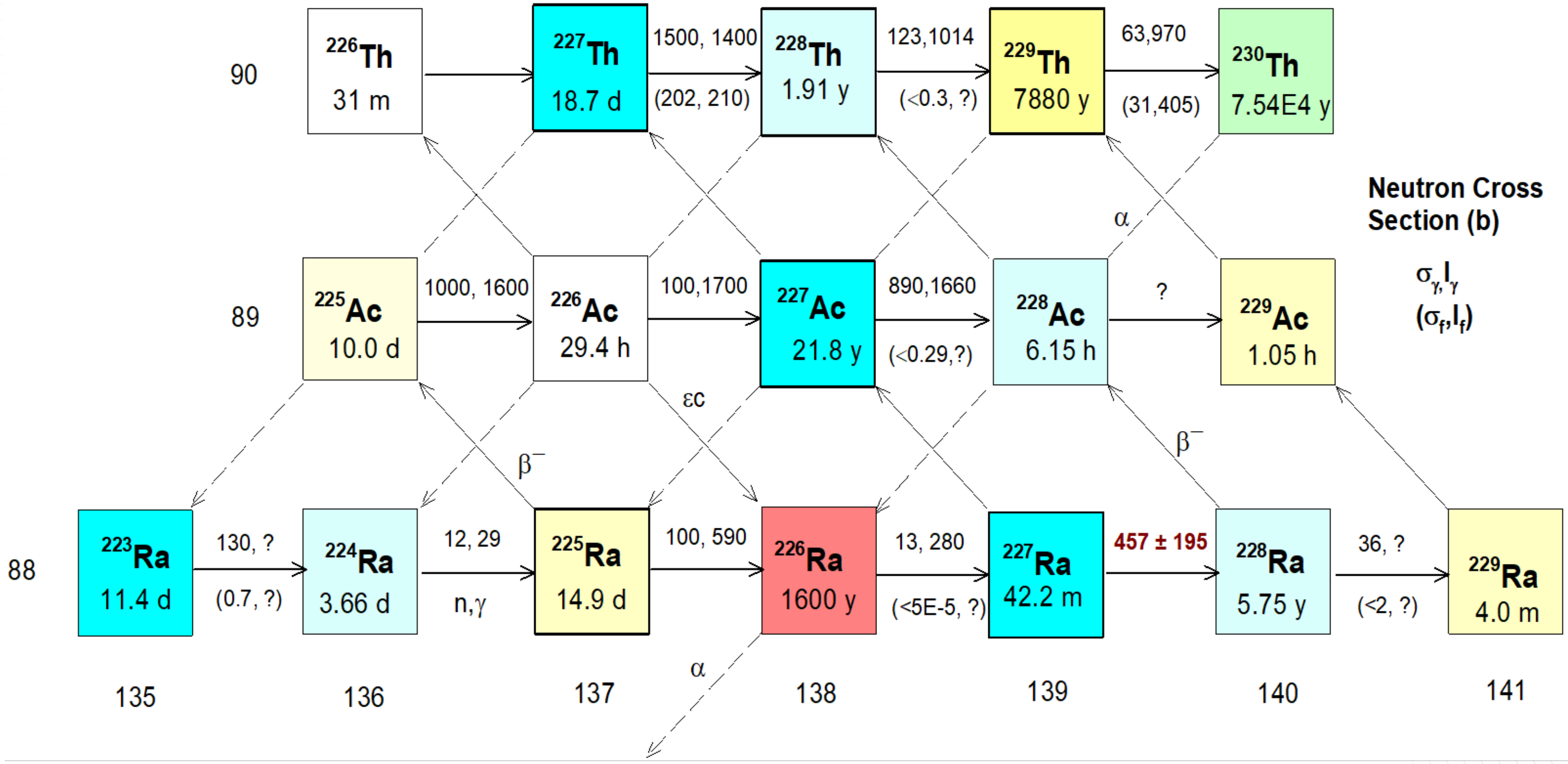
OAK RIDGE
National Laboratory

^{226}Ra handling/processing at ORNL - Overview

- In 2016, ORNL started a focused effort to recover ^{226}Ra from legacy medical devices (waste items)
  - The recovery and purification of ^{226}Ra is performed in hot cells at ORNL
  - This material is used mainly for production of radioisotopes at HFIR (^{228}Th and ^{227}Ac)
- The ORNL team responsible for these activities is quite diverse
  - Includes hot cell technicians, radiochemists, QA staff, analytical lab technicians, radiological control technicians, facility maintenance, transportation, safety, and other support staff (welders, manipulator maintenance, metrology, …)
- My role at ORNL is Project Manager for these activities
  - Trained as a nuclear physicist and was involved with production of radioactive ion beams for most of my career at the Holifield Radioactive Ion Beam Facility at ORNL

^{226}Ra is important for the DOE Isotope Program

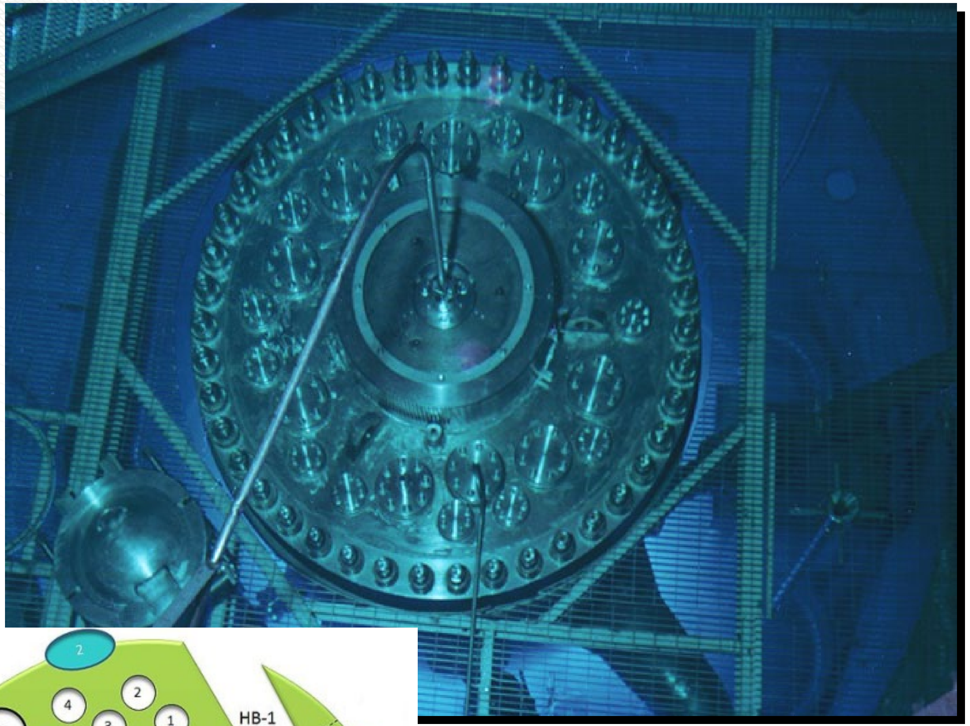
- Isotopes produced at the High Flux Isotope Reactor (HFIR) during irradiation of a ^{226}Ra target
  - A valuable feedstock for production of alpha-emitting radioisotopes
- ORNL is uniquely qualified for large scale processing and irradiation of ^{226}Ra targets
  - HFIR, infrastructure (hot cells, gloveboxes, transportation), safety systems, radiochemistry experience

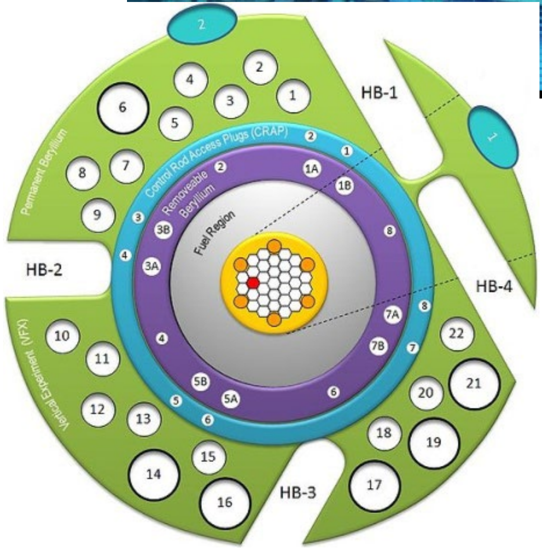


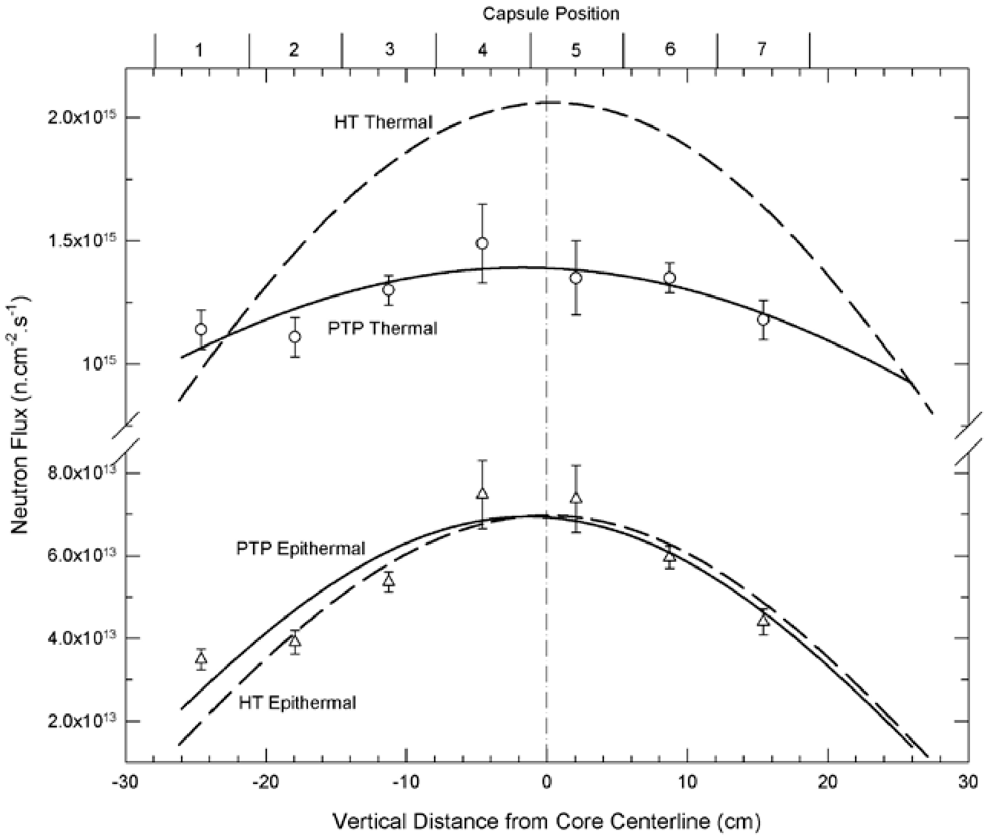
S. Hogle et al., *Reactor Production of Thorium-229*, Appl. Radiat. Isot. 114, 19 (2016)

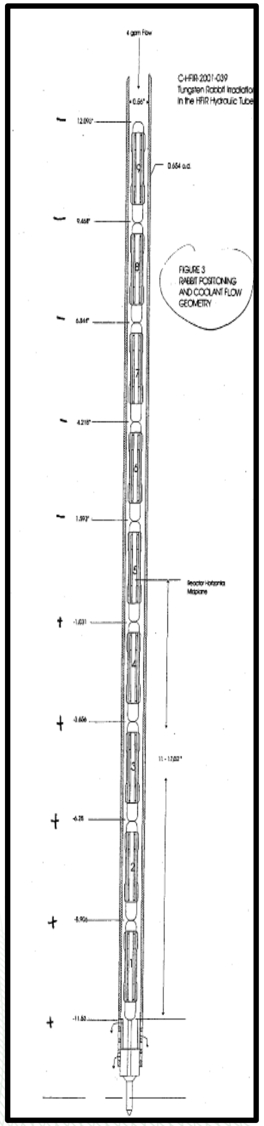
OAK RIDGE National Laboratory

Hydraulic Tube at the 85 MW High Flux Isotope Reactor



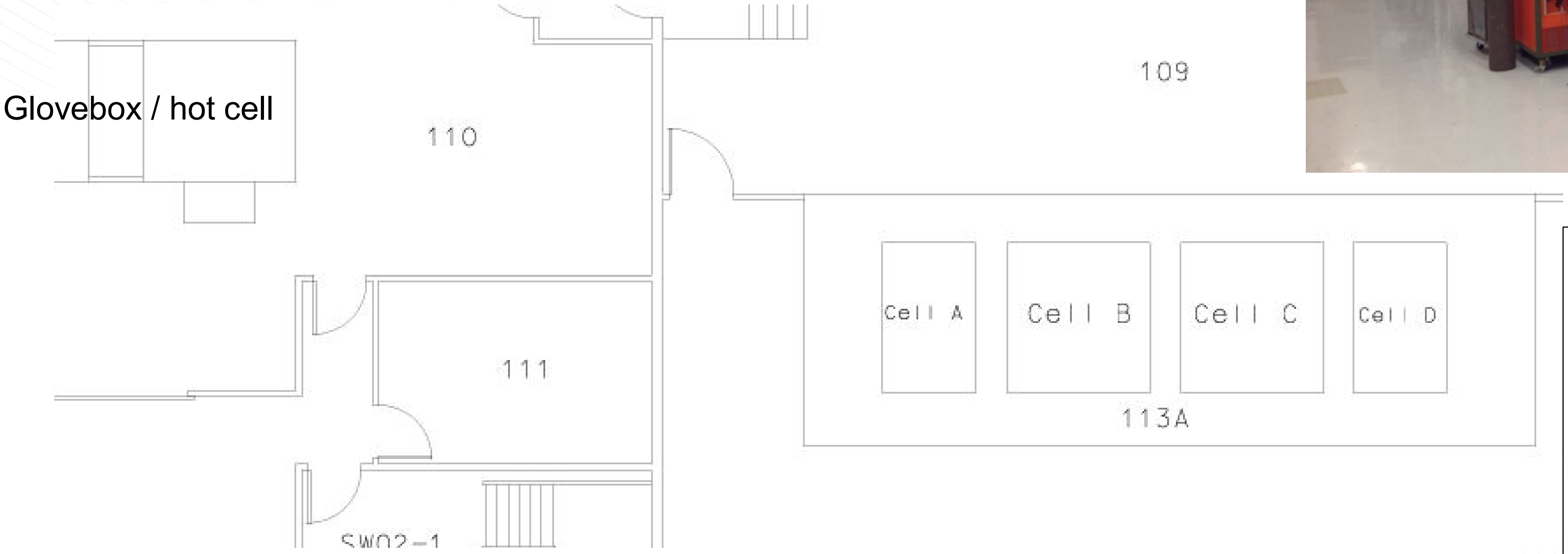






- The HFIR hydraulic tube facility allows for targets to be insert/removed during operation of the reactor
- This makes it possible to optimize production of isotopes, such as ^{227}Ac and ^{228}Th (while minimizing unwanted radioisotopes that contribute to radioactive waste)

Hot Cells used for working with ^{226}Ra at ORNL



These hot cells are used for

- Working with large quantities of ^{226}Ra to fabricate targets
- Processing of targets after irradiation at HFIR (high levels of radioactivity)
- Controlling personnel dose and contamination

The hot cells are 8’ x 8’ x 13’ with a 3-foot wall thickness (concrete)

OAK RIDGE National Laboratory

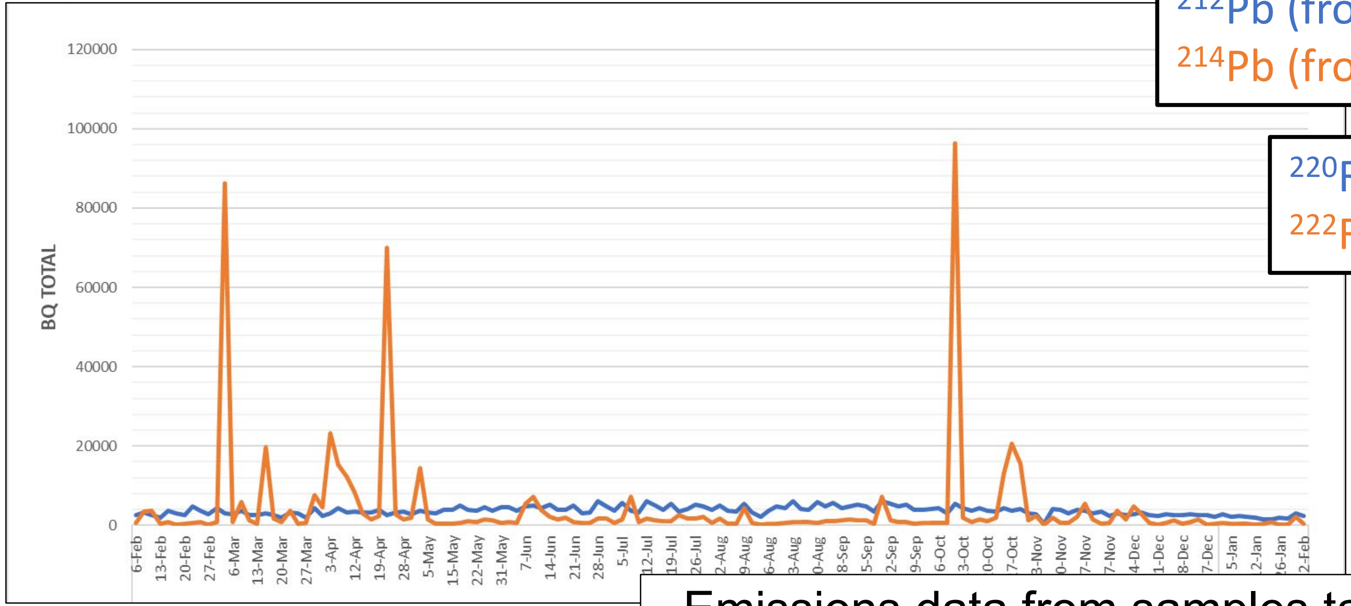
Fabricating ^{226}Ra targets for HFIR irradiation

- Purified ^{226}Ra is pressed into solid pellets for irradiation at HFIR
- Heat is generated in the target during irradiation, and it must be removed efficiently to prevent melting
  - Aluminum powder is added to provide thermal conductivity
  - Tight dimensional tolerances are required to ensure good contact between the pellet and the walls of the target capsule
  - The target is also filled with helium prior to welding to improve heat transfer
- Pellets are pressed in a glovebox (dexterity required)
  - A lead shield reduces personnel whole-body dose
  - Finger ring dosimeters monitor dose to the hands
  - The total amount of ^{226}Ra per pellet is limited to control personnel dose
  - Working with ^{226}Ra outside of the hot cell is limited to a single shift due to the increase in dose from the buildup of daughter activity

Preparation of ^{226}Ra targets in a glovebox

Emissions from the hot cells are monitored

- The hot cells are maintained at negative pressure with respect to work areas
- There are two ventilation systems that exhaust through a bank of HEPA filters and into a monitored stack
  - The main hot cell exhaust is high volume and vents directly to the stack
  - A hot off-gas (HOG) system that is high flow rate, low volume – exhaust gases pass through a caustic scrubber before venting to the stack



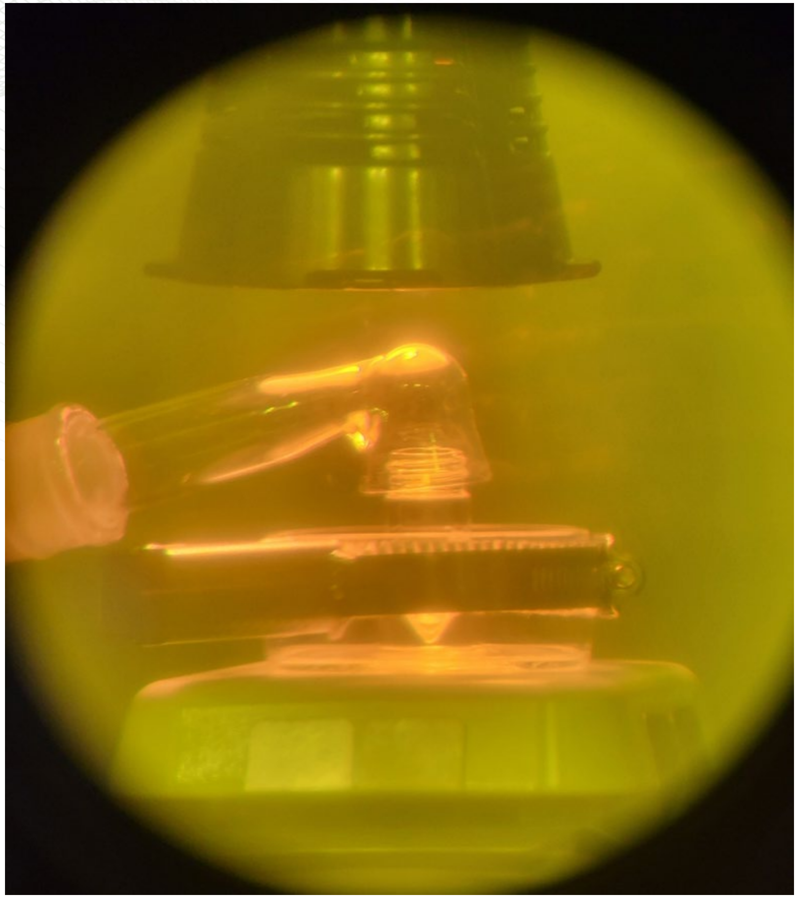
^{212}Pb (from ^{228}Th decay)
^{214}Pb (from ^{226}Ra decay)

^{220}Rn ($T_{1/2}$ = 55.6 sec)
^{222}Rn ($T_{1/2}$ = 3.8 days)

Emissions data from samples taken in the exhaust stack

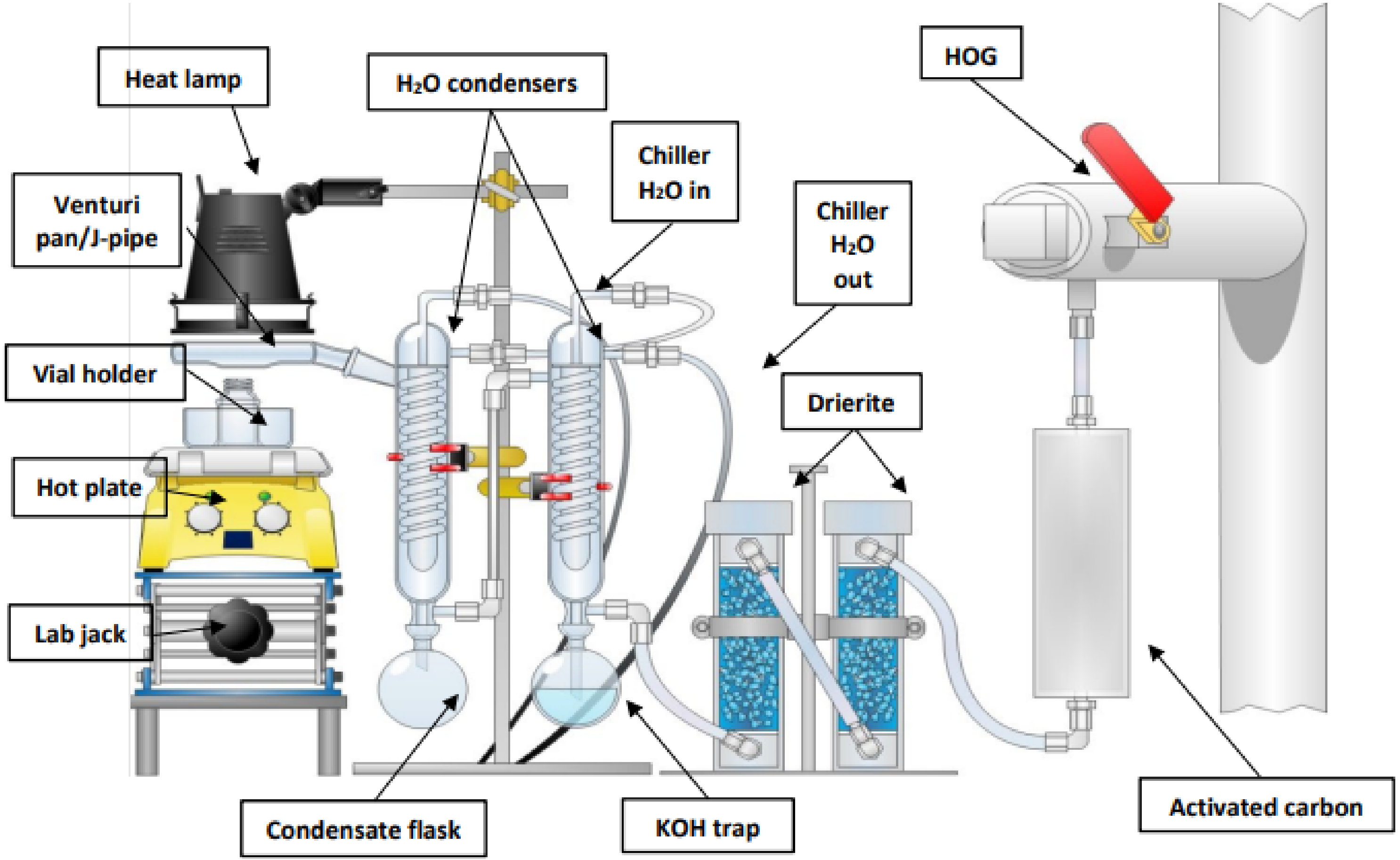
OAK RIDGE National Laboratory

Reducing environmental emissions with in-cell controls



Evaporation set-up in the hot cell with heat lamp and venturi collector that is connected to the HOG system

Schematic of in-cell gas trapping apparatus



Personnel protection

- Access to the hot cells is possible either through a large door in the back of the hot cell or through an access port at either end of the hot cell bank
- Radon is released from the hot cells whenever the doors are opened or when material is removed using the side ports
- Full PPE, including a powered air-purifying respirator (PAPR), is required for staff during these operations
- Local continuous air monitors (CAM) are used to assess the radon levels around the hot cell bank – alarms during normal operations result in evacuation of the area (local or facility)
- Monitoring of radiological personnel dose includes
  - Personal dosimeters worn during radiological work (electronic with audible alarms)
  - An annual whole-body / lung count with NaI and HPGe detectors and routine urinalyses are used to monitor potential uptake of radiological material

Hot Cell Tech and Radiological Control Tech prepared to remove samples from the hot cell

Waste generation and disposal

- This process generates radioactive waste
  – Waste streams include low-level radioactive solids, mixed waste, liquid waste
  – No TRU waste is generated in this process
- All liquid wastes are sampled and analyzed before disposition
- All solid wastes are characterized using a portable HPGe detector
  – Includes the smaller in-cell containers and the larger overpacks (mainly 55-gallon drums and B-25 boxes)
- In some cases, high activity levels in the waste make it more efficient and cost-effective to use a Type B container

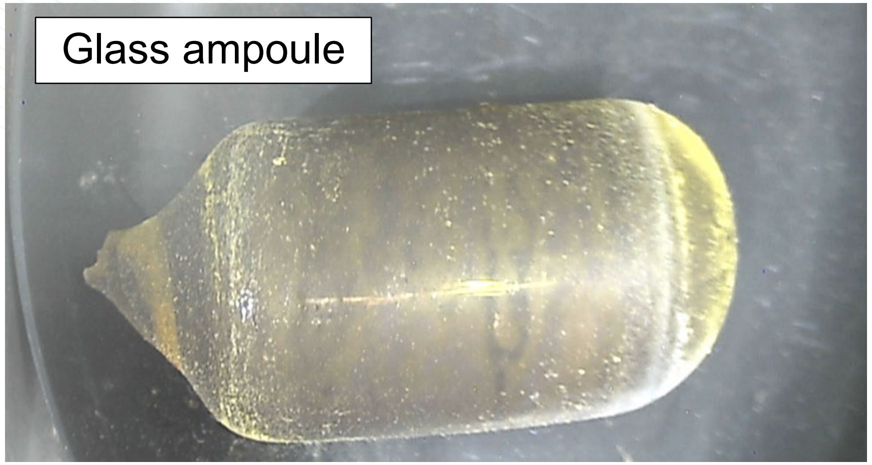
Empty B-25 boxes

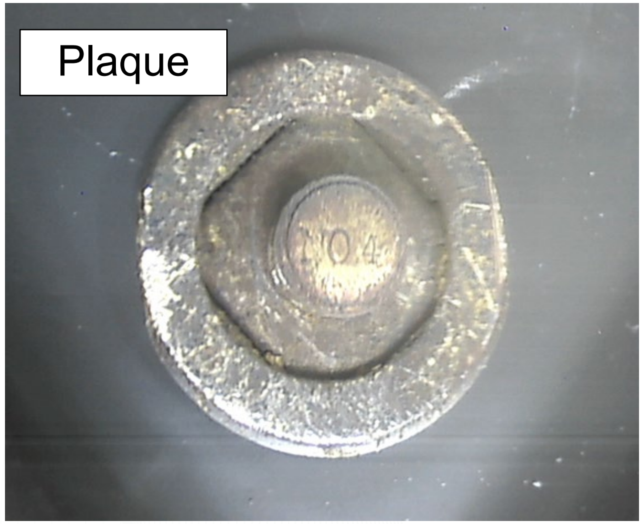
Type A containers used for liquid waste disposal

Type B container with waste

Examples of legacy medical devices containing ^{226}Ra

- Legacy ^{226}Ra devices are stored at ORNL in welded Special Form Capsules (SFC) in shielded 55-gallon drums
- Inside these SFC are a variety of devices


Glass ampoule

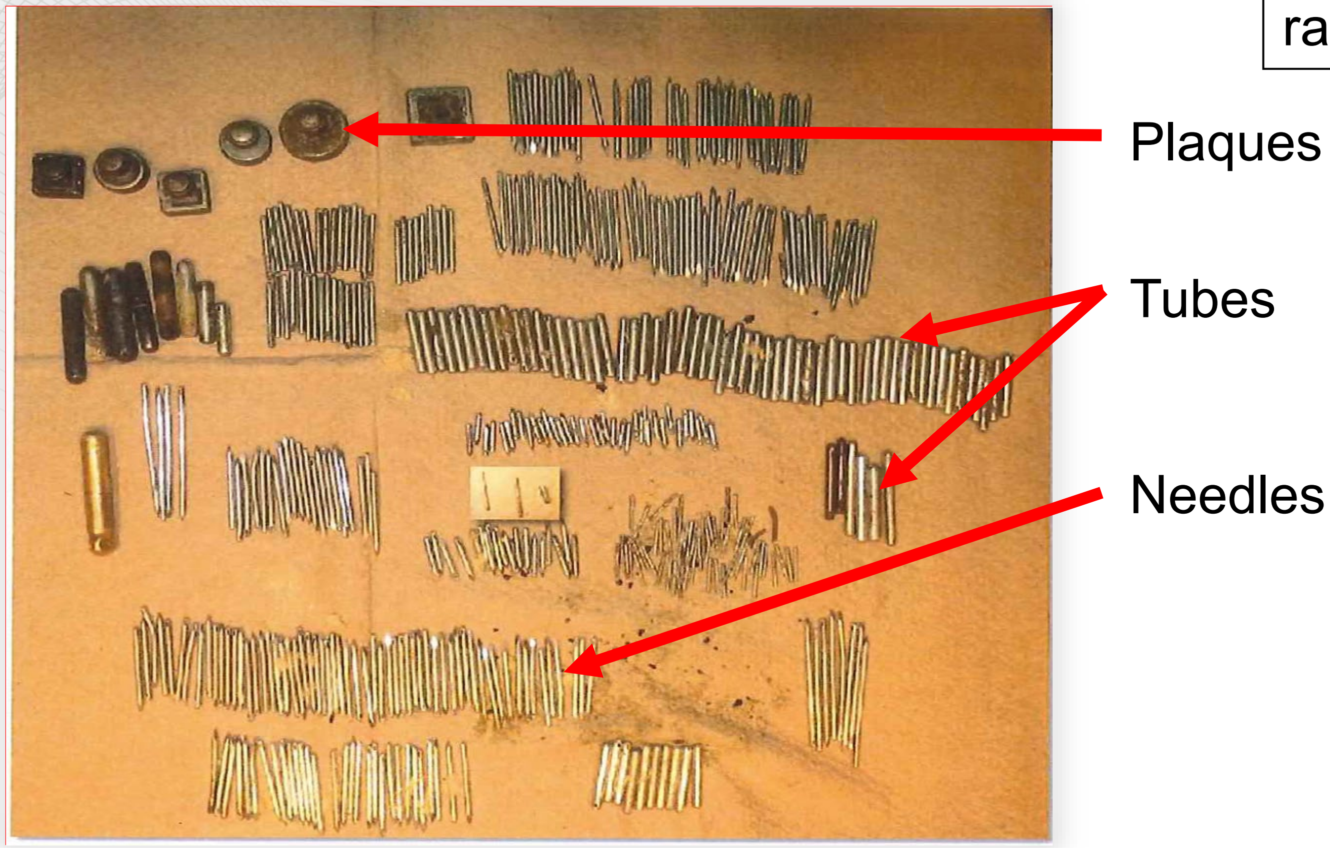

Plaque


Variety of needles, tubes and cylinders in the SFC

More examples of legacy medical devices containing ^{226}Ra

Recovery from Legacy Radium Medical Devices

Devices are mechanically disassembled, and radium is chemically dissolved from the fragments



Individual sources contain various amounts of ^{226}Ra

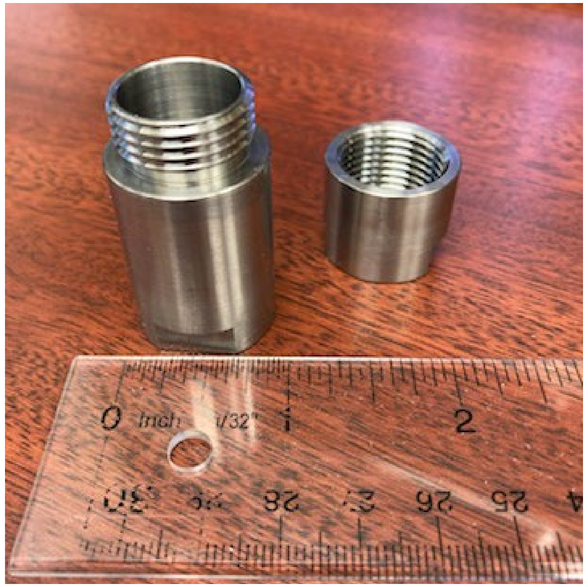
Dissolving and purifying ^{226}Ra

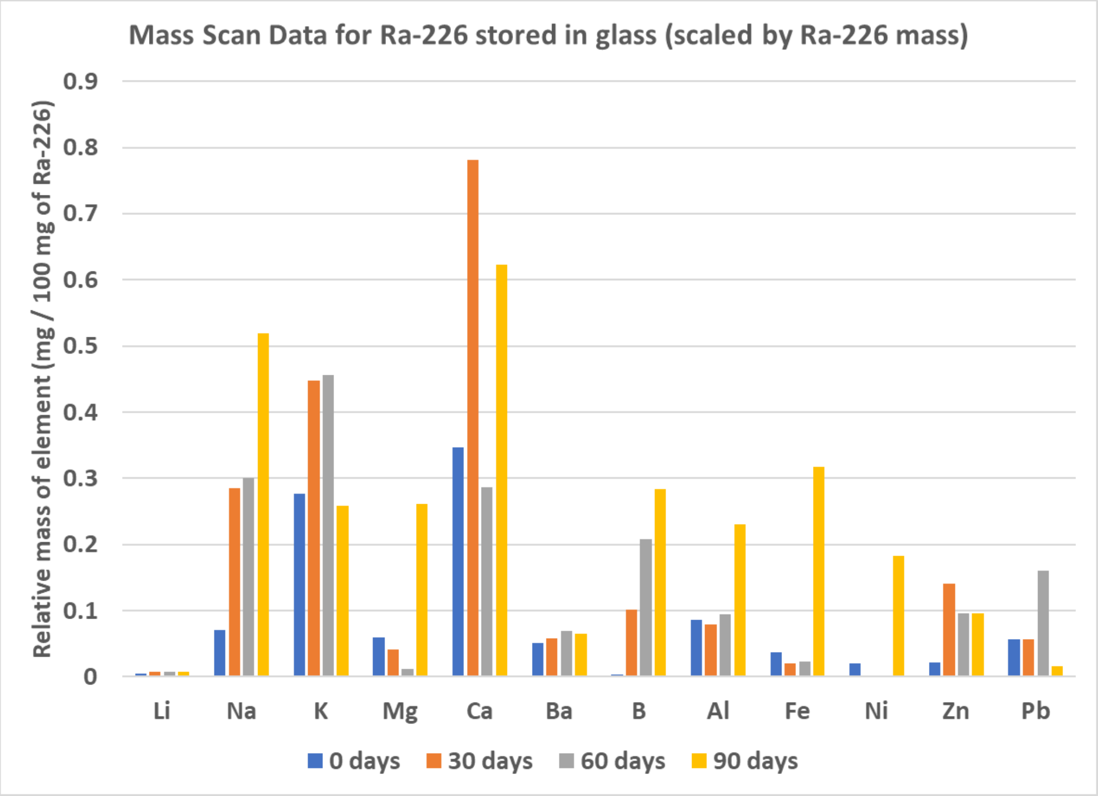
- The chemistry to purify the ^{226}Ra material is not trivial
- The 'filler' material is quite varied, including
  - Group 2 elements (Ca, Ba, …)
  - Transition metals (Cu, Ni, Ag, …)
  - Pb and Bi
  - Pt and Ir from the devices
- The process must be optimized to reduce the volume of waste generated

Oak Ridge National Laboratory

Storing ^{226}Ra

- Purified ^{226}Ra is stored in a low-molarity nitric acid solution in a borosilicate glass flask until needed
- For longer-term storage, aliquots of this solution are dispensed into either glass vials or Zircaloy vials and evaporated to dryness as solid radium nitrate

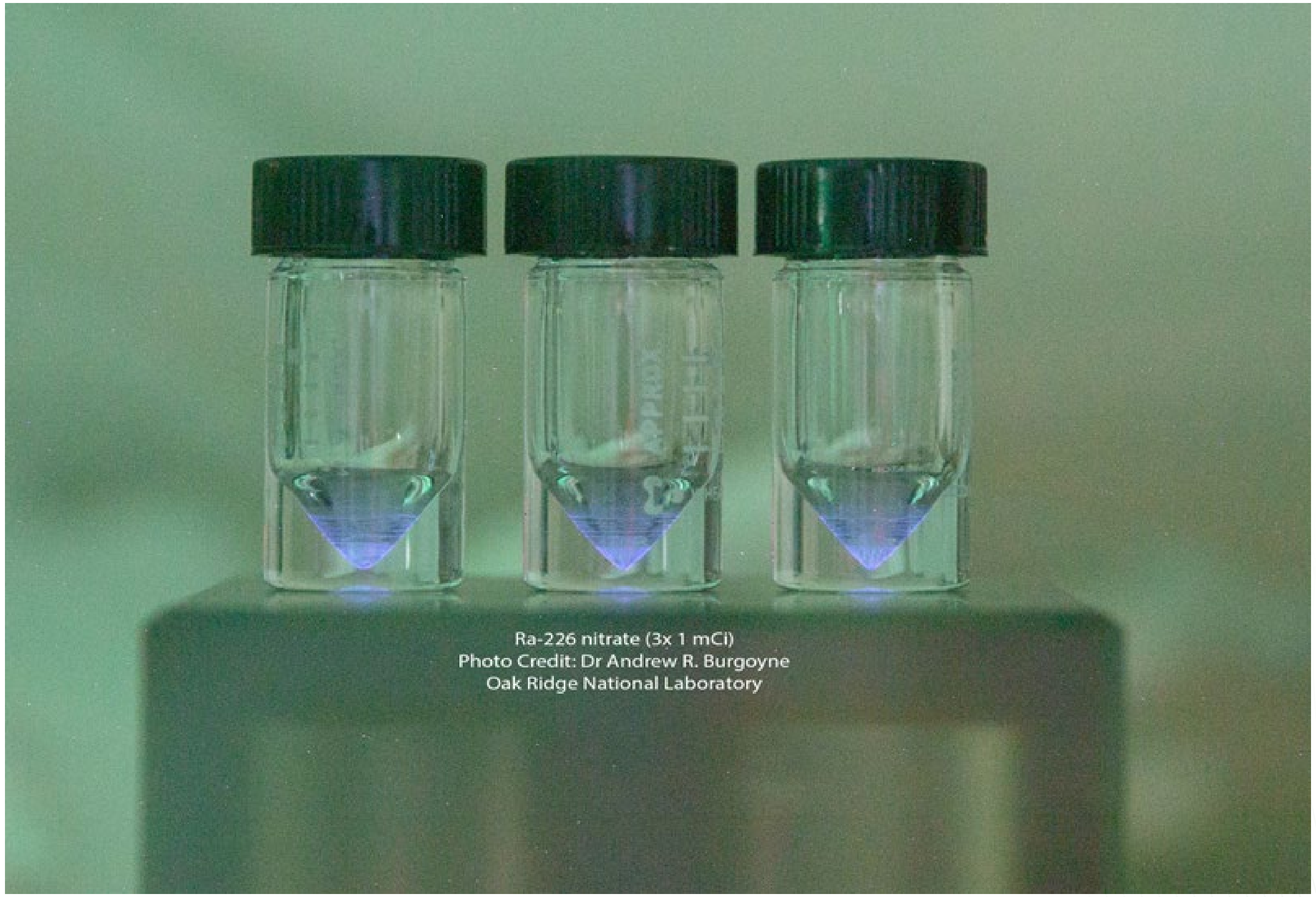




^{226}Ra dissolved in low-molarity nitric acid

OAK RIDGE National Laboratory

Thanks for your attention



Cherenkov radiation from dispensed ^{226}Ra in glass vials (1 mCi each)