Recent activities in ^{212}Pb generator development at PNNL

Matthew J. O'Hara, Lucas P. Boron-Brenner, Hannah R. Hallikainen, Daniel M. Cain*, Jared C. Johnson

Radiochemical Science Team
Nuclear Sciences Division

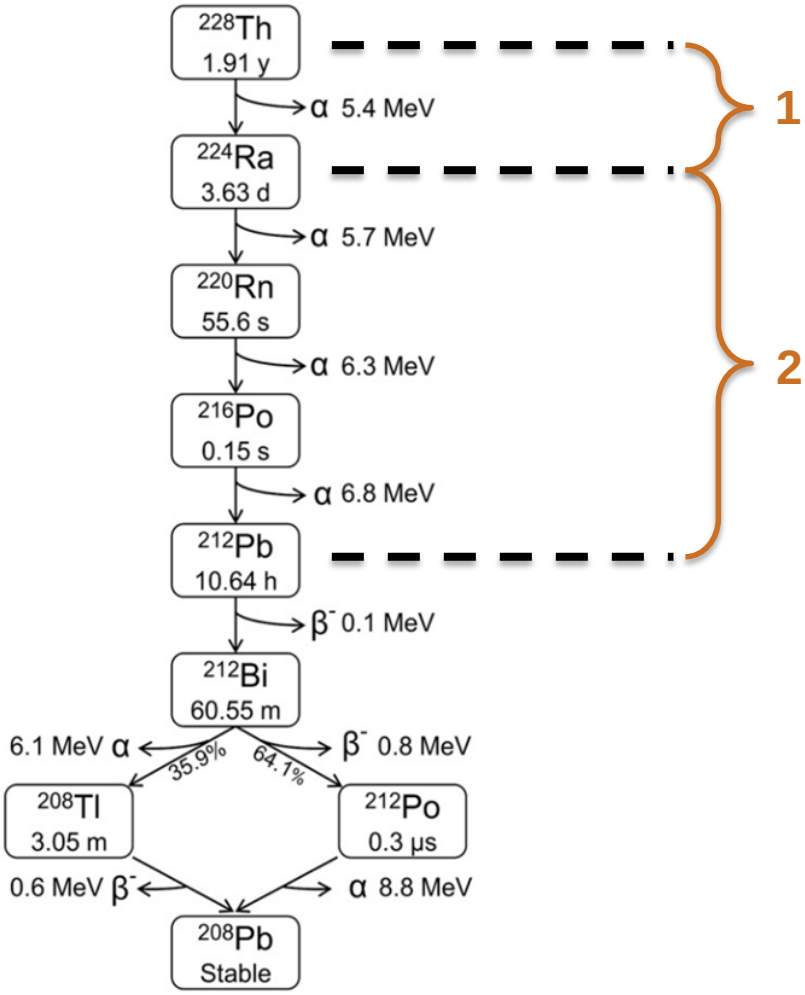
*Advanced Engineering Systems
Operational Systems & Technology Division

^{212}Pb Users Meeting
NIDC virtual seminar
July 30, 2020

Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by Battelle *Since 1965*

Two stages to preparation of traditional "wet" $^{224}Ra/^{212}Pb$ generator

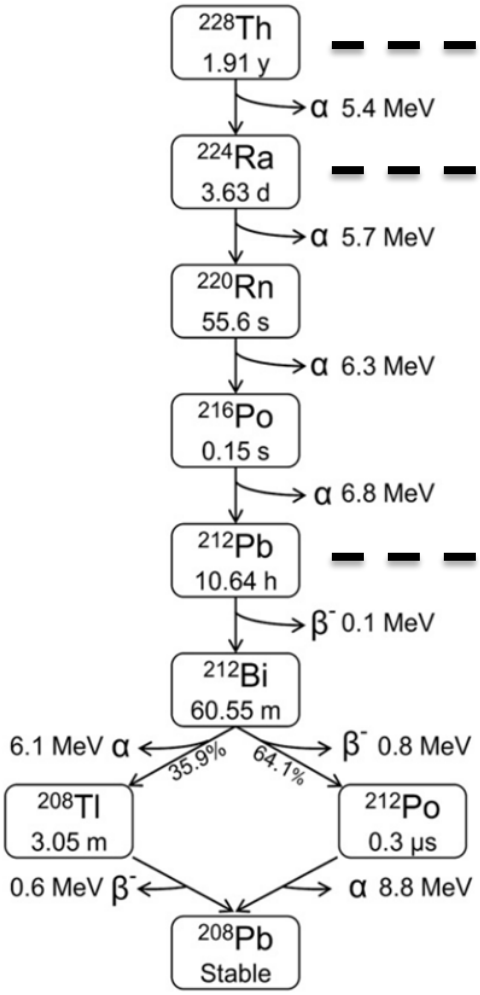


1 Isolation of ^{224}Ra

2 Preparation of ^{224}Ra / ^{212}Pb generator column

Decay scheme is from Westrøm et al., Nucl Med Biol, 2017. 51:1-9

Two stages to preparation of traditional "wet" $^{224}Ra/^{212}Pb$ generator



1 Isolation of ^{224}Ra

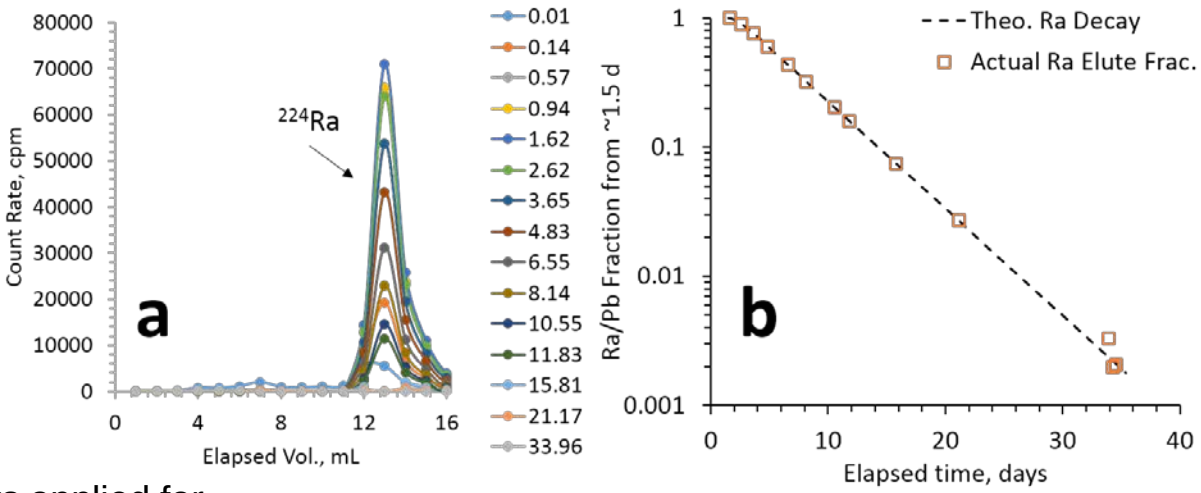
2 Preparation of ^{224}Ra / ^{212}Pb generator column

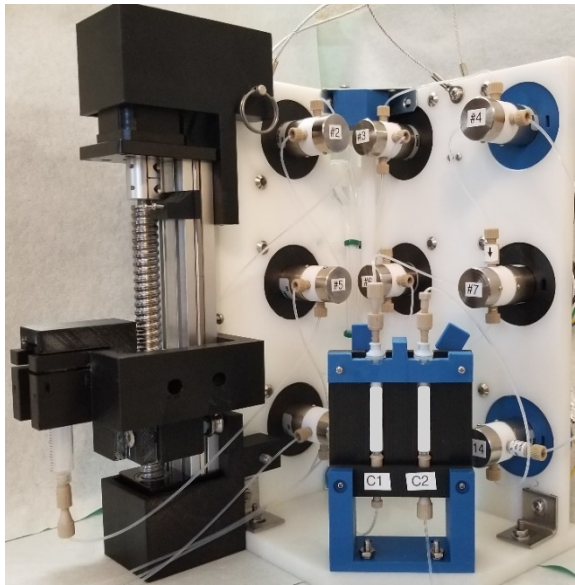
- Traditionally prepared through a series of high-dose manual radiochemical processes
- New methods are being developed to facilitate remote / automated generator production

Decay scheme is from Westrøm et al., Nucl Med Biol, 2017. 51:1-9

Two stages to preparation of $^{224}Ra/^{212}Pb$ generator (Stage 1)

- A new ^{224}Ra isolation **method** was developed to allow for in-line isolation of ^{224}Ra from ^{228}Th stocks
- A new fluidic **system** was developed to perform the new method remotely:
  - Platform has ~10” x 10” footprint
  - All platform components are radiolytically robust
    - Combination of commercial off-the-shelf and 3D printed parts
  - Isolated ^{224}Ra provided in ~1 h

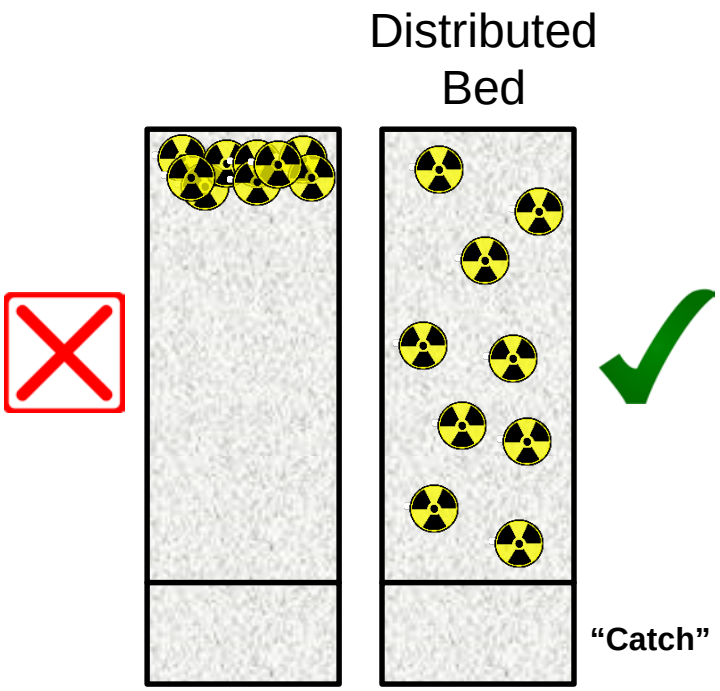


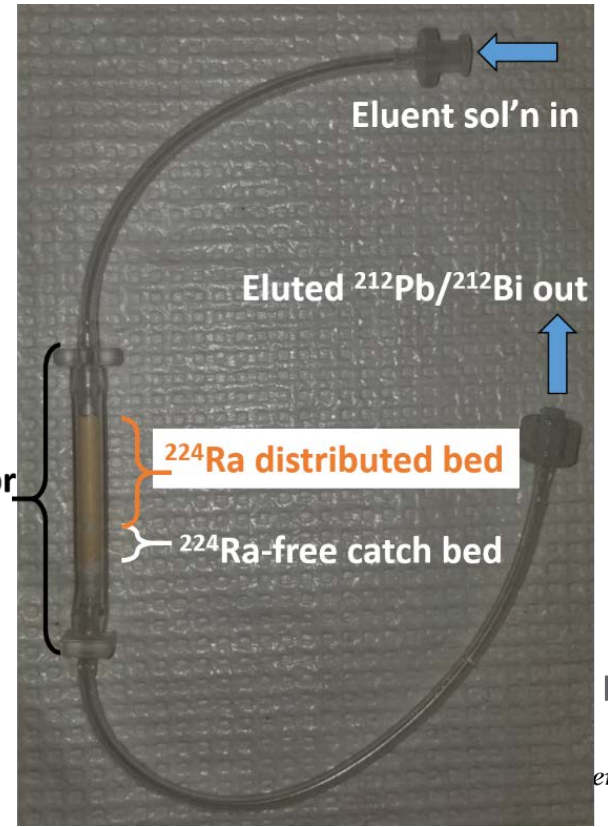


Patents applied for.

Two stages to preparation of $^{224}Ra/^{212}Pb$ generator (Stage 2)

- Purified ^{224}Ra needs to be loaded onto cation exchange resin, which becomes the ^{212}Pb generator column
- A distributed ^{224}Ra-bearing column bed is required to reduce radiolytic degradation of the resin support





Pacific Northwest NATIONAL LABORATORY

erated by Battelle Since 1965

Two stages to preparation of $^{224}Ra/^{212}Pb$ generator (Stage 2)

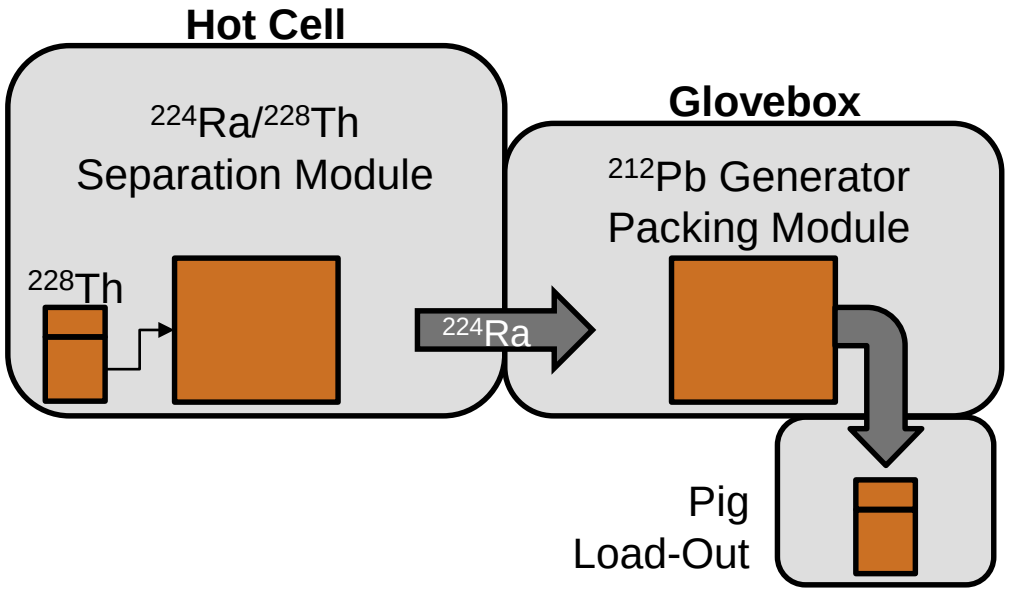
- A fluidic system has been designed to remotely prepare distributed-bed ^{224}Ra-loaded columns for ^{212}Pb generators
  - Receives purified ^{224}Ra directly from the Stage 1 module
  - Fluidic workstation prototype dispenses a pre-determined volume of resin; uniformly contacts it with freshly isolated ^{224}Ra; auto-packs ^{224}Ra-adsorbed resin to a column
  - End-to-end process takes ~1.25 h

Auto-packed generator columns

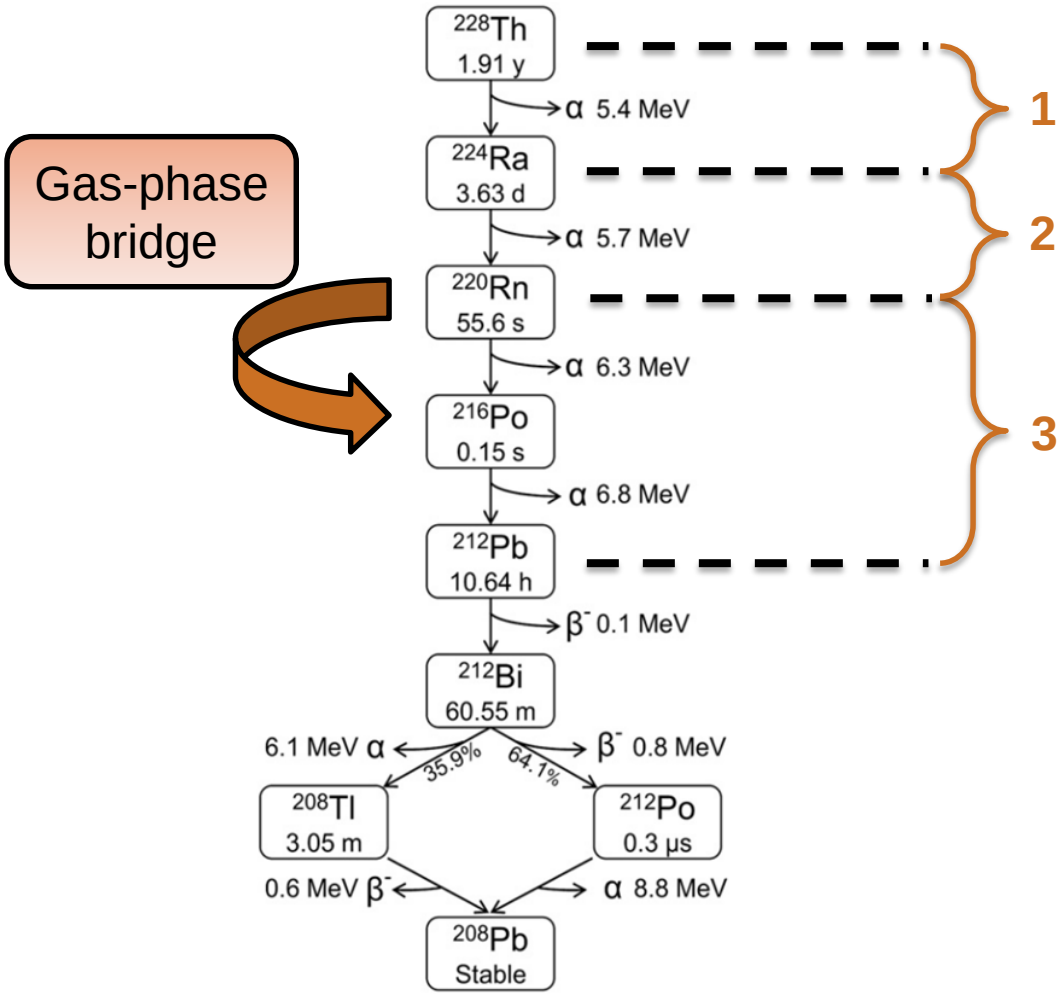
Patents applied for.

Vision for routine $^{224}Ra/^{212}Pb$ generator preparation

- Goal: Reproducible generator preparation with:
  - Reduced personnel dose / production time / production cost
- Scale-up generator testing is planned for FY21
  - Up to 20 mCi and beyond (?)
- Will evaluate ^{212}Pb generator milking performance studies in-house
- Will work with NIDC to make test generators available for testing by end-users
  - If positive response, will evaluate technology integration into routine production



Alternate ^{212}Pb generator concept: ^{220}Rn emanation generator



1 Isolation of ^{224}Ra

2 Preparation of ^{224}Ra / ^{220}Rn emanation source

3 Collection of ultra-high purity ^{220}Rn→^{216}Po →^{212}Pb

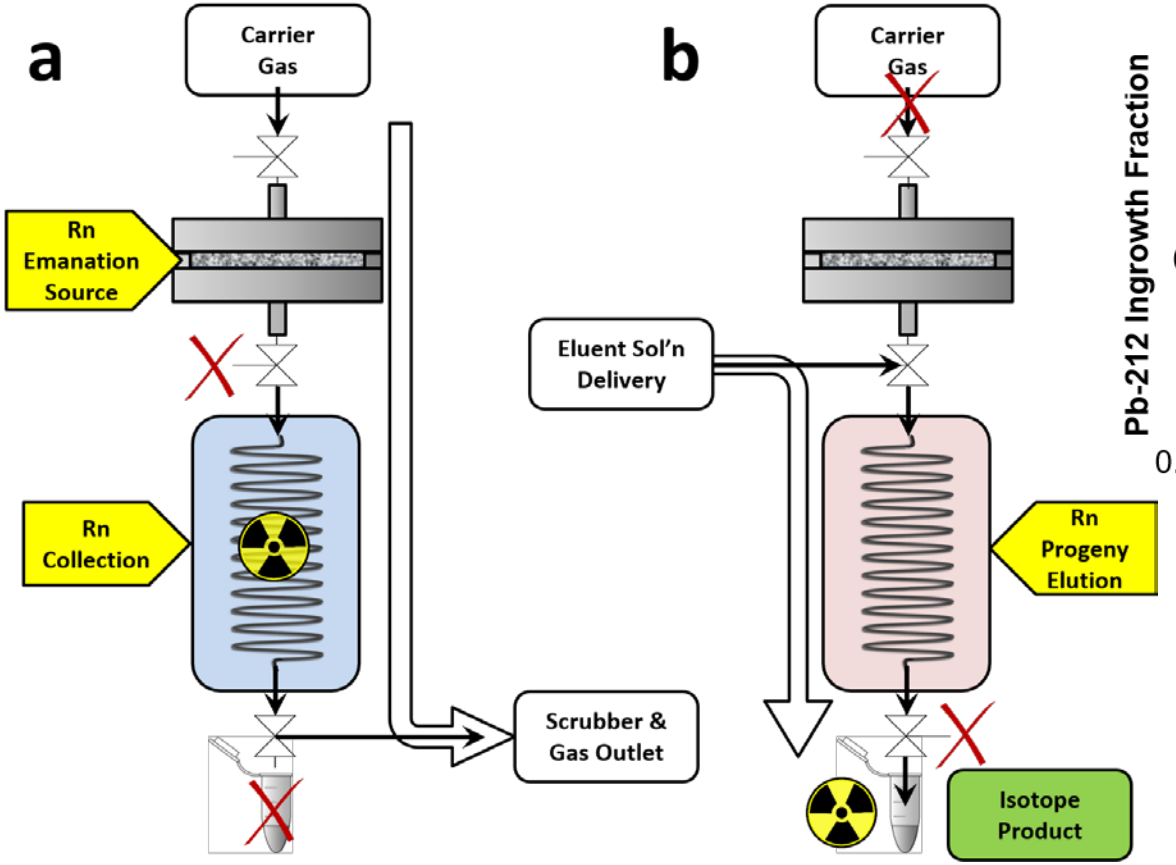
- This new approach has potential to:
  - Reduce possibility of ^{224}Ra breakthrough
  - Reduce presence of metal contaminants
  - Have ^{212}Pb collected in label-ready solution

Decay scheme is from Westrøm et al., Nucl Med Biol, 2017. 51:1-9

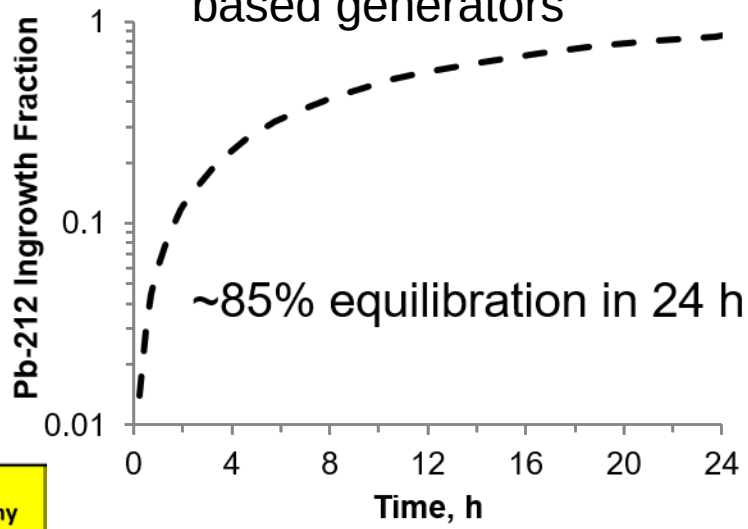
Alternate ^{212}Pb generator concept: ^{220}Rn emanation generator

1. ^{220}Rn collection (^{212}Pb ingrowth) stage

2. ^{212}Pb stripping stage

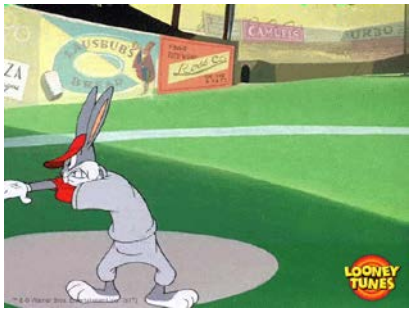


^{212}Pb production rate is the same as with column-based generators

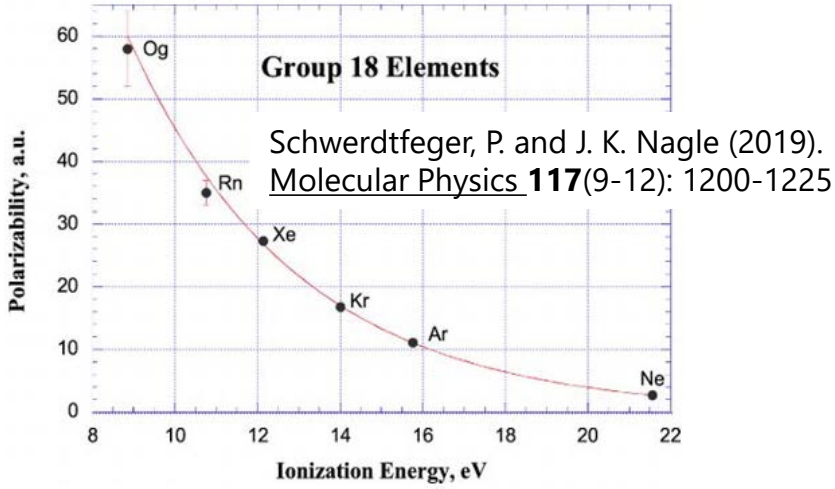


Patents applied for.

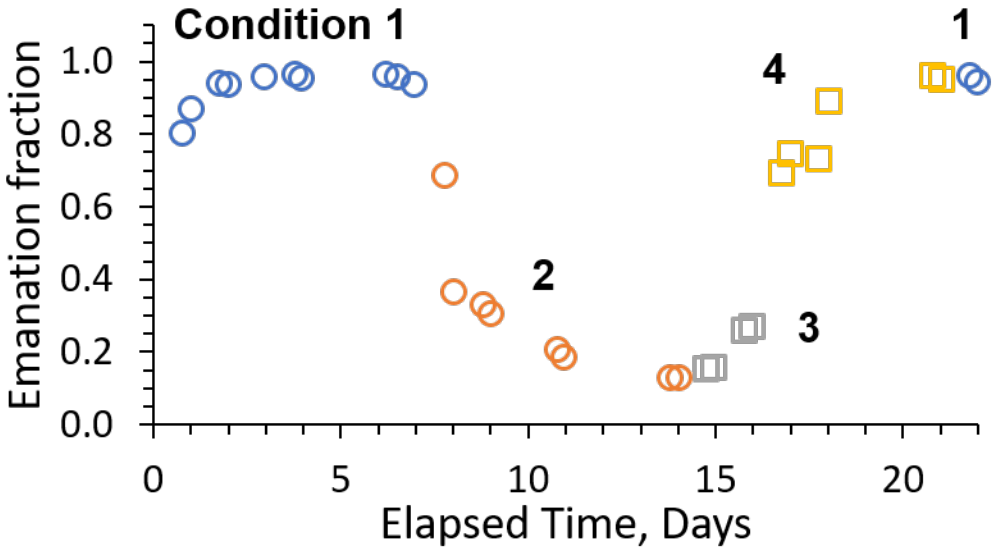
The Rn pitch: Evaluation of emanation source media and conditions



- Emanation-based generator source must allow for efficient ^{224}Ra deposition and ^{220}Rn emanation
  - Carrier gas can be used to transport ^{220}Rn away from source
- Due to increased polarizability of heavy noble gas elements, efficient emanation from many material surfaces is not feasible
  - We are evaluating a class of media that emanates Rn efficiently under certain carrier gas conditions



Schwerdtfeger, P. and J. K. Nagle (2019). Molecular Physics **117**(9-12): 1200-1225

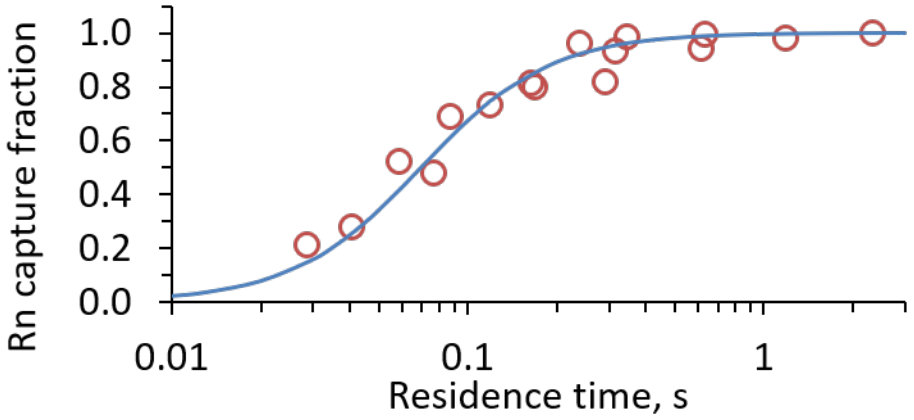


Patents applied for.

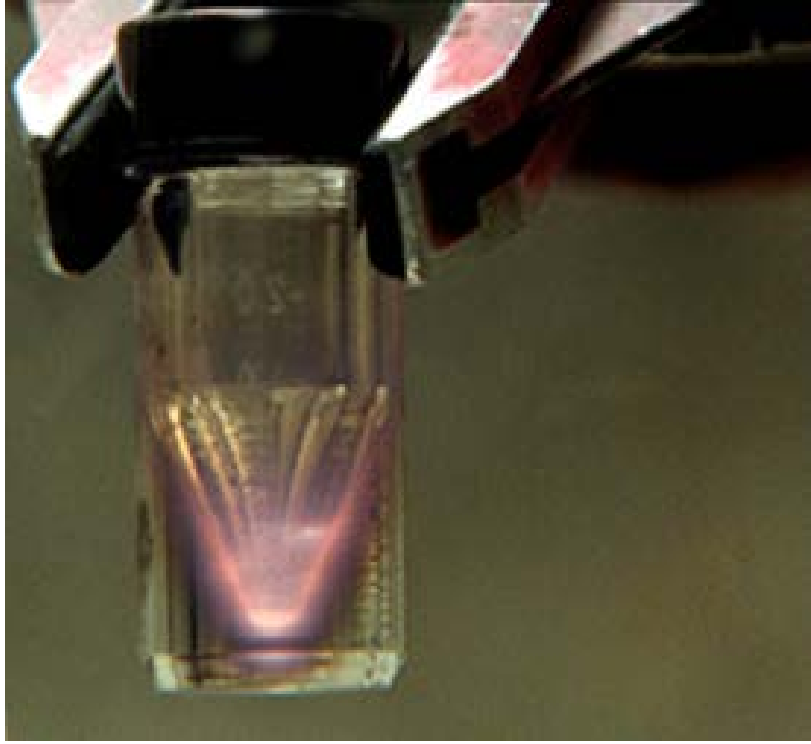
The Rn catch: Evaluation of ^{220}Rn capture agents

- Emanated ^{220}Rn (from source) must be captured from carrier gas stream
- In-line Rn absorption options include cryo-based deposition and nanoporous media
  - Currently evaluating a class of metal-free nanocage materials capable of:
    - Room temperature Rn capture
    - Dissolution for subsequent ^{212}Pb isolation
    - ^{212}Pb product supplied in label-ready buffer sol'n
  - Total ^{212}Pb yield is presently measured at ~85%
  - Final ^{212}Pb product in acetate buffer is conducive to biomolecule labeling

Patents applied for.



Thank You!



Contact Info:
Matt O'Hara
509-375-5579
Matthew.OHara@pnnl.gov

Acknowledgement:
This research is supported by the U.S. Department of Energy Isotope Program, managed by the Office of Science.

Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by Battelle *Since 1965*