SAND2021-10388C

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ACCOUNTABLE NUCLEAR MATERIAL PRODUCTION FROM FISSILE ISOTOPE PRECURSORS IN ADVANCED FUEL CYCLES: The Case of Protactinium

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INMM & ESARDA JOINT VIRTUAL MEETING | 25 AUGUST 2021

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SPECIAL THANKS TO…

Sandia National Laboratories

OAK RIDGE National Laboratory

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Nicholas Luciano, ORNL

Natalie McGirl, ORNL

Jeffrey Powers, ORNL

Richard Reed, ORNL

Louise G. Evans, ORNL

WITH LOTS OF GUIDANCE FROM…

Benjamin Betzler, ORNL
Mark Bruhnke, SNL
Ben Cipiti, SNL
Emory Collins, ORNL
Bill Del Cul, ORNL
Zoe Gastelum, SNL
Brandon Grogan, ORNL
Gary Hirsch, ORNL

Donald Kovacic, ORNL
Alan Krichinsky, ORNL
Bradley Patton, ORNL
Barry Spencer, ORNL
Nathan Shoman, SNL
Matthew Sternat, SNL
David Williams, ORNL
Michael Whitaker, ORNL

AND SUPPORT FROM…

Office of International Nuclear Safeguards

NNSA
National Nuclear Security Administration

DIFFERENTIATING FERTILE, FISSILE, AND *FISSILE PRECURSOR* NUCLEAR MATERIALS

Fertile Materials	**"Fissile Precursor" Materials ***	**Fissile Materials**
Neutron capture converts these to fissile material	Spontaneously decays to fissile material; intermediary isotope between fertile and fissile material	Able to undergo nuclear fission with neutrons of all energies
Thorium-232 Uranium-232 Uranium-234 Uranium-238 Plutonium-238 Plutonium-240	Protactinium-233 ($T_{1/2}$ ~27 days) Protactinium-235 ($T_{1/2}$ ~24 mins) Neptunium-239 ($T_{1/2}$ ~2.4 days) Neptunium-241 ($T_{1/2}$ ~14 mins)	Uranium-233 Uranium-235 Plutonium-239 Plutonium-241

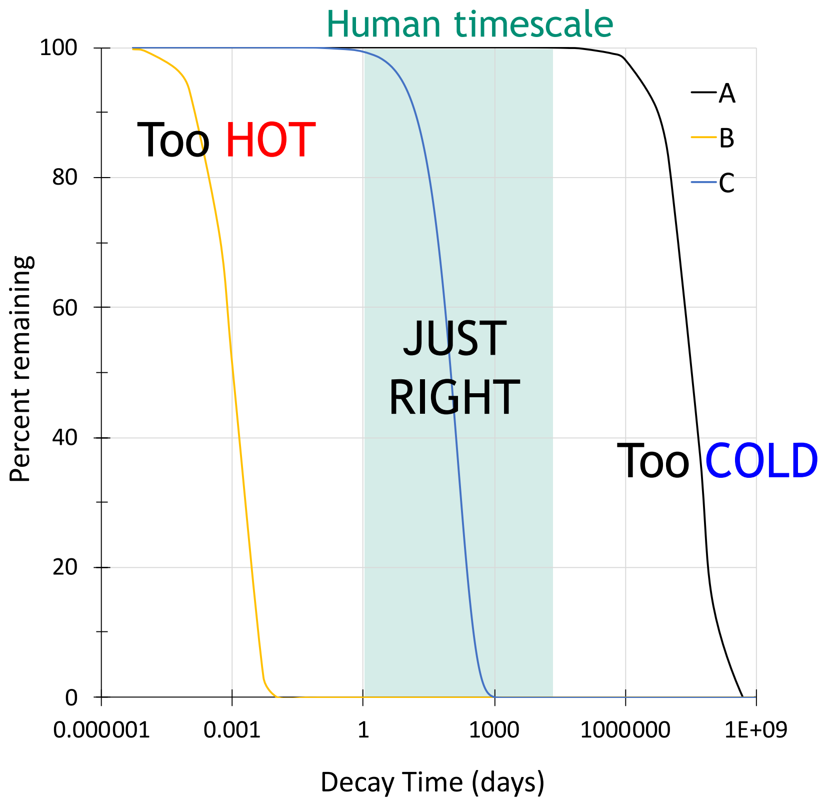
* Not subject to materials accountancy

Thorium-Uranium Cycle: $^{232}Th \xrightarrow{(n,\gamma)} {}^{233}Th \xrightarrow{\beta^-} {}^{233}Pa \xrightarrow{\beta^-} {}^{233}U$

Uranium-Plutonium Cycle: $^{238}U \xrightarrow{(n,\gamma)} {}^{239}U \xrightarrow{\beta^-} {}^{239}Np \xrightarrow{\beta^-} {}^{239}Pu$

IDENTIFYING *FISSILE PRECURSORS* OF INTEREST

1. Protactinium-233 persists long enough ($T_{1/2}$ ~27 days) to isolate from spent fuel
2. Protactinium-233 decays fast enough to accumulate uranium-233 on IAEA timescales
3. Protactinium-233 may be chemically processed to obtain uranium-233 of high isotopic purity
4. Protactinium-233 is not subject to material accountancy protocols or international safeguards



Material Category	Example	Timeliness Goal
Unirradiated direct-use	Plutonium, uranium-233, high enriched uranium in fresh fuel rods	1 month
Irradiated direct-use	Plutonium, uranium-233, high enriched uranium in spent (irradiated) fuel rods	3 months
Indirect use	Natural or depleted uranium Thorium	12 months

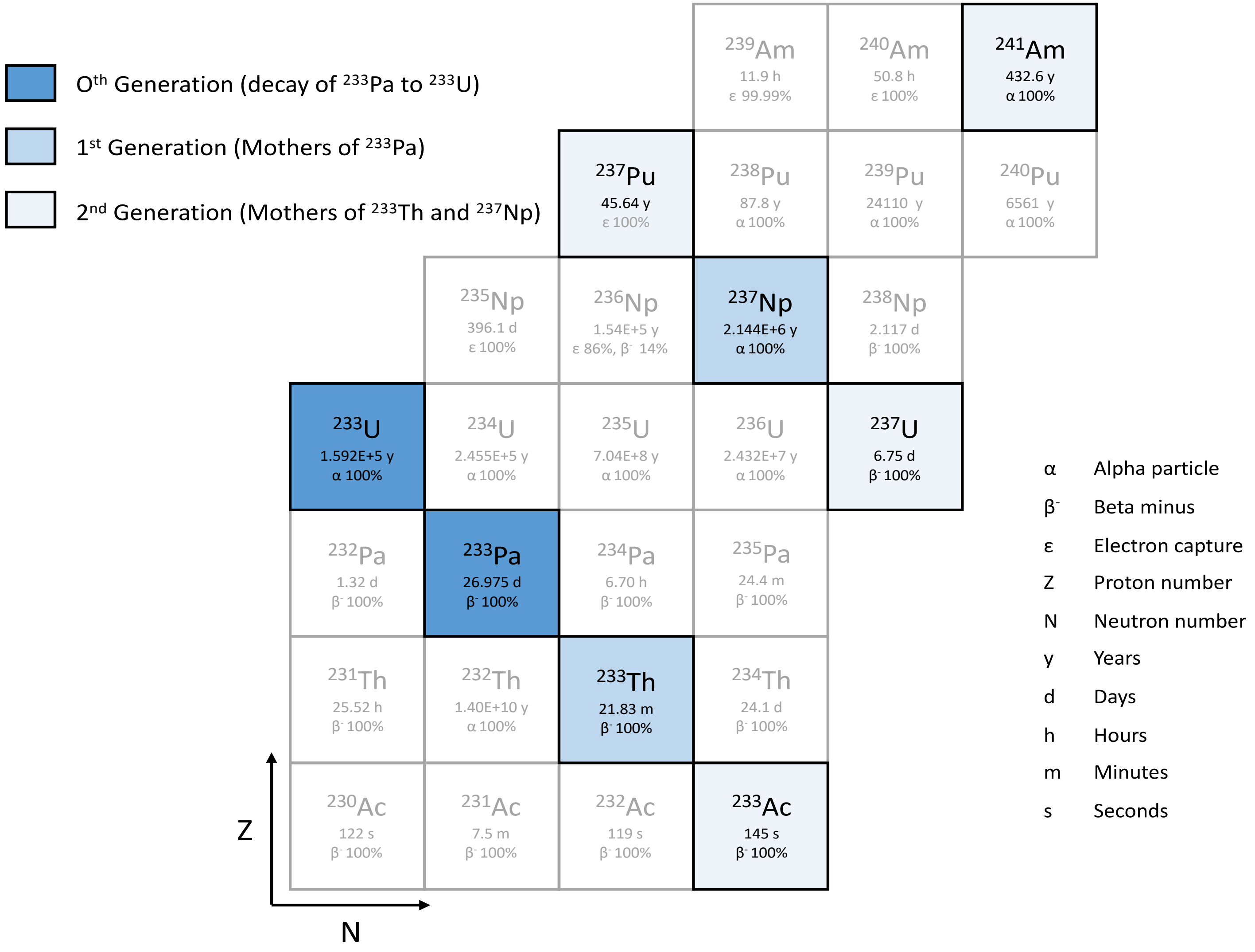
KEY RESEARCH QUESTION

Will technological advances in nuclear fuel cycles enabling short-cooled or online spent fuel partitioning result in the need to monitor fissile isotope precursors, *in order to meet accountancy and timeliness goals for nuclear materials?*

A PROTACTINIUM CASE STUDY:

- How is Pa-233 produced? → Identify leading candidate fuel cycles
- How much Pa-233 is generated, and on what timescales? → Conduct reactor simulations
- Can Pa-233 be isolated on meaningful timescales? → Conduct chemical separations calculations
- How can we monitor and verify Pa-233? → Simulate detector responses

PROTACTINIUM-233 IS GENERATED IN ALL THORIUM FUEL CYCLES



Thorium-Uranium Cycle:

$^{232}Th \xrightarrow{(n,\gamma)} {}^{233}Th \xrightarrow{\beta^-} {}^{233}Pa \xrightarrow{\beta^-} {}^{233}U$

Uranium-Plutonium Cycle:

$^{238}U \xrightarrow{(n,\gamma)} {}^{239}U \xrightarrow{\beta^-} {}^{239}Np \xrightarrow{\beta^-} {}^{239}Pu$

THE LEADING CANDIDATES FOR THORIUM FUEL CYCLES ARE:

Reactor Type/Fuel Cycle	Developer	Focus for this work
Pressurized Water Reactor (PWR) with once through cycle or multiple fuel recycling	ThorEnergy (Norway)	Compare ^{233}Pa production rates for a variety of fuel compositions. Compare ^{233}Pa production rates for fresh vs. recycled fuel.
Advanced Heavy Water Reactor (AHWR) with fuel recycling	BARC (India)	Compare ^{233}Pa production in startup, transition, and equilibrium cores.
Fast Breeder Reactor (FBR) with fuel recycling	BARC (India)	Determine ^{233}Pa concentrations in axial and radial thorium blankets.
Molten Salt Breeder Reactor (MSBR) with continuous reprocessing to remove fission products and protactinium	Flibe Energy (USA), CAS (China)	Quantify ^{233}Pa in fuel salt and in online reprocessing system.

WHAT IS A SIGNIFICANT QUANTITY OF PROTACTINIUM-233?

Material		Significant Quantity	Applies to…	Select Precursors (half-life, decay mode)
Direct use	Plutonium[a]	8 kg	Total element	Neptunium-239 (2.356 days, β^-)
	Uranium-233	8 kg	^{233}U	**Protactinium-233 (26.975 days, β^-)** Neptunium-237 (2.14E+06 yrs, α)
	High enriched uranium ($^{235}U \geq 20\%$)	25 kg	^{235}U	Protactinium-235 (24.44 mins, β^-) Plutonium-239 (24110 yrs, α)

[a] For plutonium containing less than 80% plutonium-238

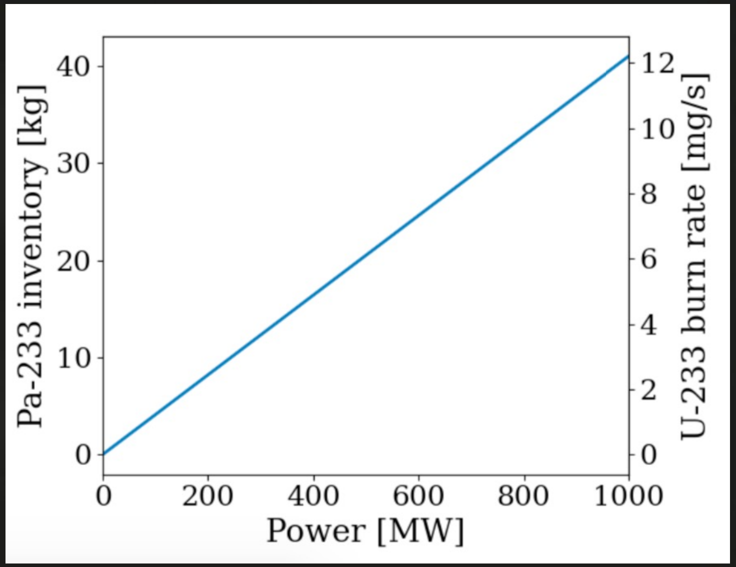
A significant quantity of protactinium-233 will decay spontaneously to a significant quantity of uranium-233 within several half-lives (~few months).

IAEA Safeguards Glossary 2001 Edition, International Atomic Energy Agency (Vienna, 2001)

ESTIMATING PROTACTINIUM PRODUCTION

Assuming equilibrium condition

- Rate of Pa-233 Decay = Rate of U-233 Fission
- Constant power, all driven by U-233 Fission
- Minimal loss of Pa-233 to neutron absorption



Starting from:

$$\lambda_{Pa} = \frac{\ln(2)}{T_{1/2}} \qquad R = \frac{PM}{EN_a} \qquad I_{Pa} = \frac{R}{\lambda_{Pa}}$$

Gives us an upper bound on I_{Pa}:

$$I_{Pa} = \frac{MT_{1/2}}{EN_a \ln(2)} * P \approx \text{41 kg / GWth}$$

R = fission rate for U-233
I_{Pa} = Pa-233 inventory
P = reactor power
M = molar mass of U-233
$T_{1/2}$ = half-life of Pa-233
λ_{Pa} = decay constant for Pa-233
E = average energy per fission
N_a = Avogadro's number

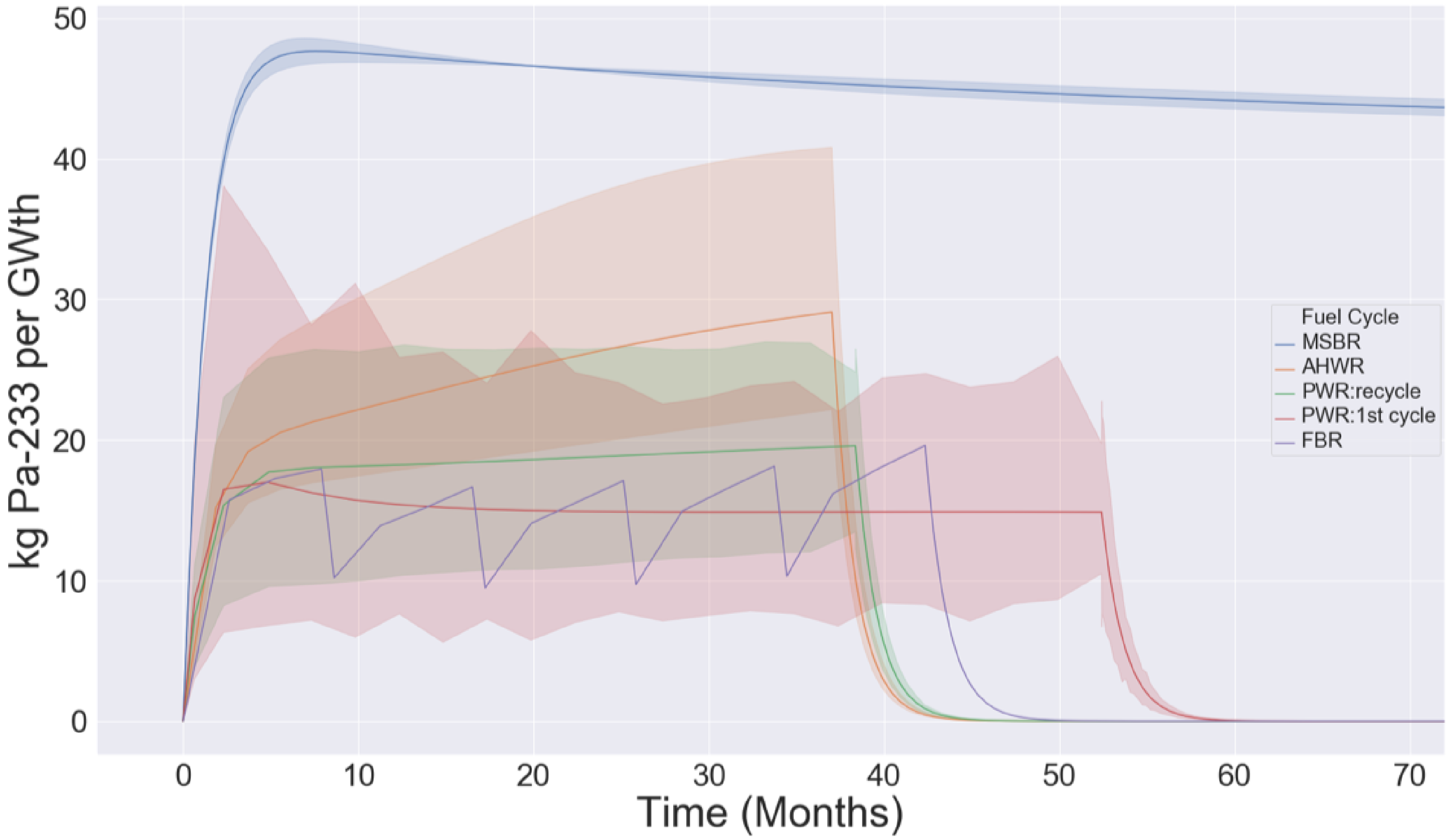
HOW MUCH PROTACTINIUM-233 IS THERE IN A FUEL CYCLE?

- Pa-233 at shutdown varies from 3 to 42 kg/GWth

MSBR	~40 kg / GWth
AHWR	20 – 40 kg / GWth
PWR	~3 – 30 kg / GWth
FBR	~20 kg/GWth

- All fuel cycles can produce at least a "significant quantity" of protactinium-233

How does potential isolation of protactinium impact safeguards technical objectives?



ISOLATED PROTACTINIUM INVENTORY DEPENDS ON FUEL CYCLE

Power from ^{233}U Fission (% of 1 GWth)	**Quantity ^{233}Pa (kg/GWth)**			
	At reactor equilibrium	**+3 months cooling**	**+6 months cooling**	**+12 months cooling**
100 MW (10%)	4	0.38	0.036	3.4×10^{-4}
200 MW (20%)	8	0.76	0.073	6.7×10^{-4}
500 MW (50%)	20	1.9	0.18	1.7×10^{-3}
1000 MW (100%)	40	3.8	0.36	3.4×10^{-3}

PROTACTINIUM MONITORING TIMESCALES

Reactor startup, transition, & equilibrium

Fuel cycling

Closed fuel cycles with short-cooled partitioning (< 6 months)

Closed fuel cycles with long-cooled partitioning (3-5 years)

Open fuel cycles
Interim & final waste storage (5+ years)

SAFEGUARDS APPROACHES FOR FUEL CYCLE CASE STUDIES

Solid fuel, no reprocessing

- Verify no reprocessing has occurred (continuity of knowledge)
- Verify ^{233}U content using burnup codes, gamma confirmatory measurement
- Item-based safeguards

Solid fuel, long-cooled reprocessing

- Verify no short-cooled reprocessing has occurred (continuity of knowledge)
- ^{233}Pa is sufficiently dilute in THOREX processing streams to allow termination of safeguards on these streams of ^{233}U
- Bulk material accountancy for ^{233}U

Solid fuel, short-cooled reprocessing

- ^{233}U safeguards may require:
  - Verification of ^{233}Pa inventory in multiple process streams
  - Detection of loss or diversion of 8 kg ^{233}Pa in 1 month
  - Monitoring loss of protactinium to aqueous raffinate in THOREX processes
- Inventory measurements must compare to total ^{233}Pa + ^{233}U from burnup codes
- Not cost-effective for commercial purposes

Molten salt fuel, continuous reprocessing

- Fission products and potentially ^{233}Pa removed continuously
- ^{233}Pa held outside of the neutron flux to decay to ^{233}U, which is fed back into core
- ^{233}U safeguards require:
  - Verification of ^{233}Pa inventory in multiple process streams
  - Detection of loss or diversion of 8 kg ^{233}Pa in 1 month

CONCLUSIONS & RECOMMENDATIONS

- If technology advances to allow spent fuel partitioning at short or no cooling, material accountancy approaches for fissile isotope precursors may be needed
- Definition for "short-cooled" fuels depends on the precursor.
  - "Short-cooled" is less than 6 months for Pa-233
- Concepts for material balance with fissile precursors:
  1. Aggregate accounting for fissile isotope and fissile precursor isotope pairs
  2. Material balance "in future"
  3. Flowsheet verification (e.g., similar for Am-241 and Np-237)
- Future work needed:
  - Diversion pathway analysis & diversion indicators for specific fuel cycles
  - More precise nuclear material inventory modeling, especially for molten salt reactors
  - Development of accountancy methods for priority areas

Questions?

Thank you

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