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Evaluating Detection Capabilities of Irradiated Methylammonium Lead Iodide Perovskite Crystals

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Motivation

Motivation

- Why this material?
  - High interest in field of radiation detection in recent years
    - Cost-effectiveness compared to traditional detector materials like sodium iodide
- Why put it in a nuclear reactor?
  - Ability to detect radiation is only one of several important qualities to consider when selecting a material
    - Lots of exciting results for highly controlled, short-term exposures to radiation sources
    - Very little data on effects of high-fluence radiation exposure for this material
- Ultimately, we want to answer the questions:
  - *"Is this a worthwhile material to make detectors from?"*, and, if so,
  - *"What kind of lifetime can we expect from these detectors?"*

Motivation (cont.)

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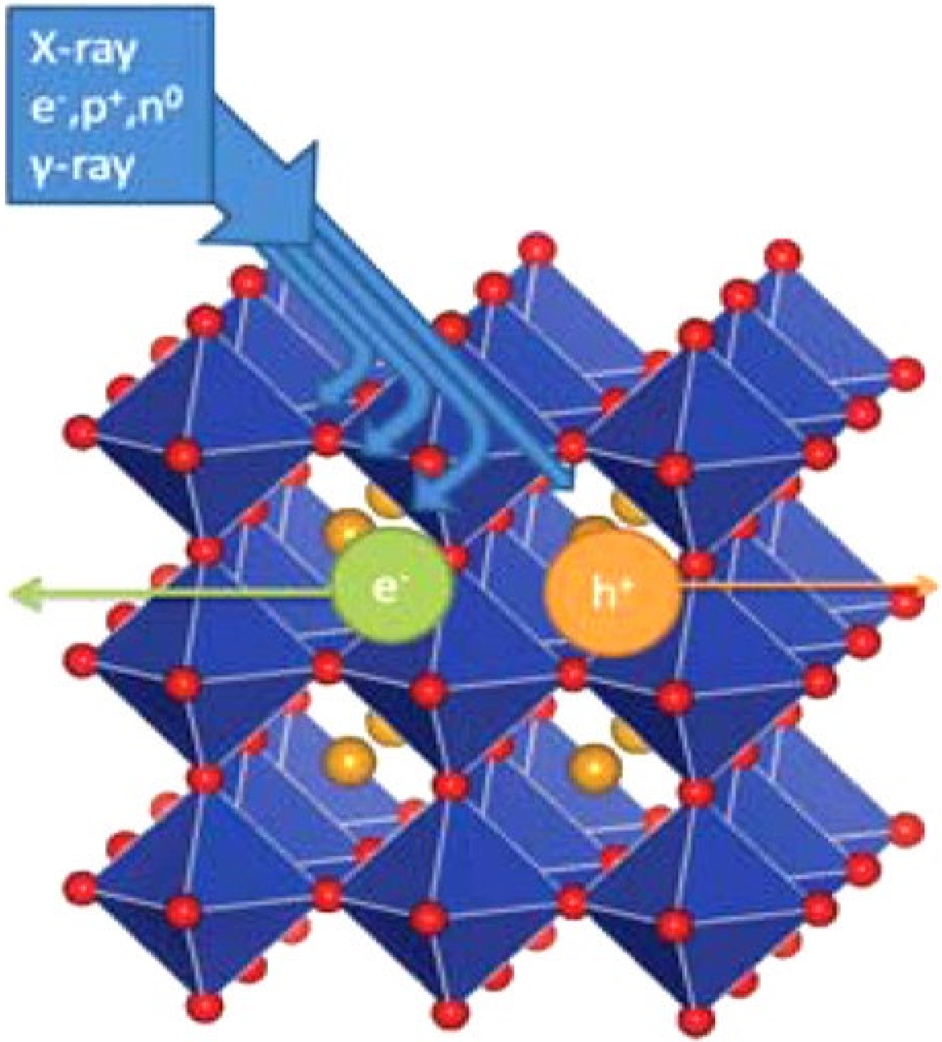
Fig. Methylammonium lead iodide lattice structure with possible radiation particle interactions [1]



Motivation (cont.)

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- Choice of perovskite, Methylammonium Lead Iodide (MAPbI)
  - Perovskites describe a large family of crystals with general formula ABX_3
    - MAPbI crystals are among the easiest to produce
    - Also has one of the highest theoretical charge collection efficiencies within lead halide group (compared to other candidates like Br) [1]-[3]

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Experimental Setup

Experimental Setup – Crystal Production

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- Crystal purity is vital to charge collection and resistance to radiation-induced damage
  - Production will be done using highest possible purity reagents and precipitation reaction parameters will be continuously monitored

- Production will be divided into 3 batches of 10 crystals
  - First batch – control batch; no prior radiation exposure
  - Second batch – to be irradiated at 900kW for 5 minutes
  - Third batch – to be irradiated at 900kW for 1 hour

Experimental Setup – Crystal Production (cont.)

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- General reaction for production:
  - $CH_3NH_{3(aq)} + 3HI_{(aq)} + PbCH_3COO_{(aq)} \rightarrow CH_3NH_3PbI_{3(s)}$
- Under optimal conditions, crystals can be grown in a lab environment on the order of 10-50 mg in mass
  - Reaction progresses slowly (~2 weeks to synthesize fully-grown crystal)
  - Faster processes exist, but precipitation reaction allows for minimization of oxidation by atmosphere during growth

Experimental Setup – Photocurrent Measurement

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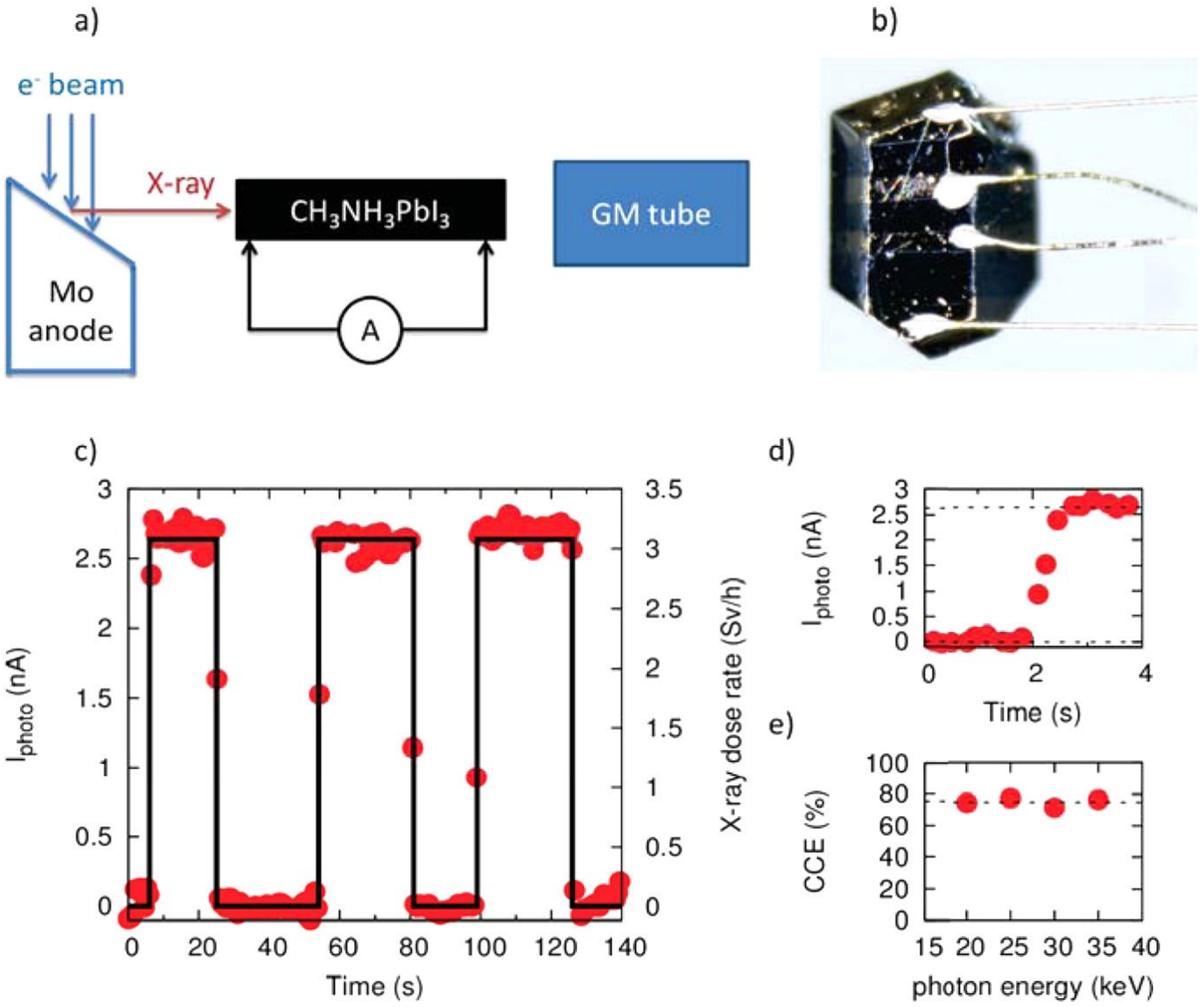
- Crystals will be arranged in setup to obtain several sets of photocurrent data
    - Crystals will be soldered to a low natural resistance wire, such as gold wire, and placed in parallel with a picoammeter
    - Wired crystals will be encapsulated and placed in front of a ^{137}Cs source to obtain count data
    - Calibrated G-M detector will be placed behind wired crystal to obtain attenuation data

Experimental Setup – Photocurrent Measurement (cont.)

Fig. (a) Example photocurrent measurement experimental setup (b) Picture of MAPbI crystal with gold wire soldering (c-e) sample data collected for x-ray source irradiation [1]



Experimental Setup – In-core Irradiation

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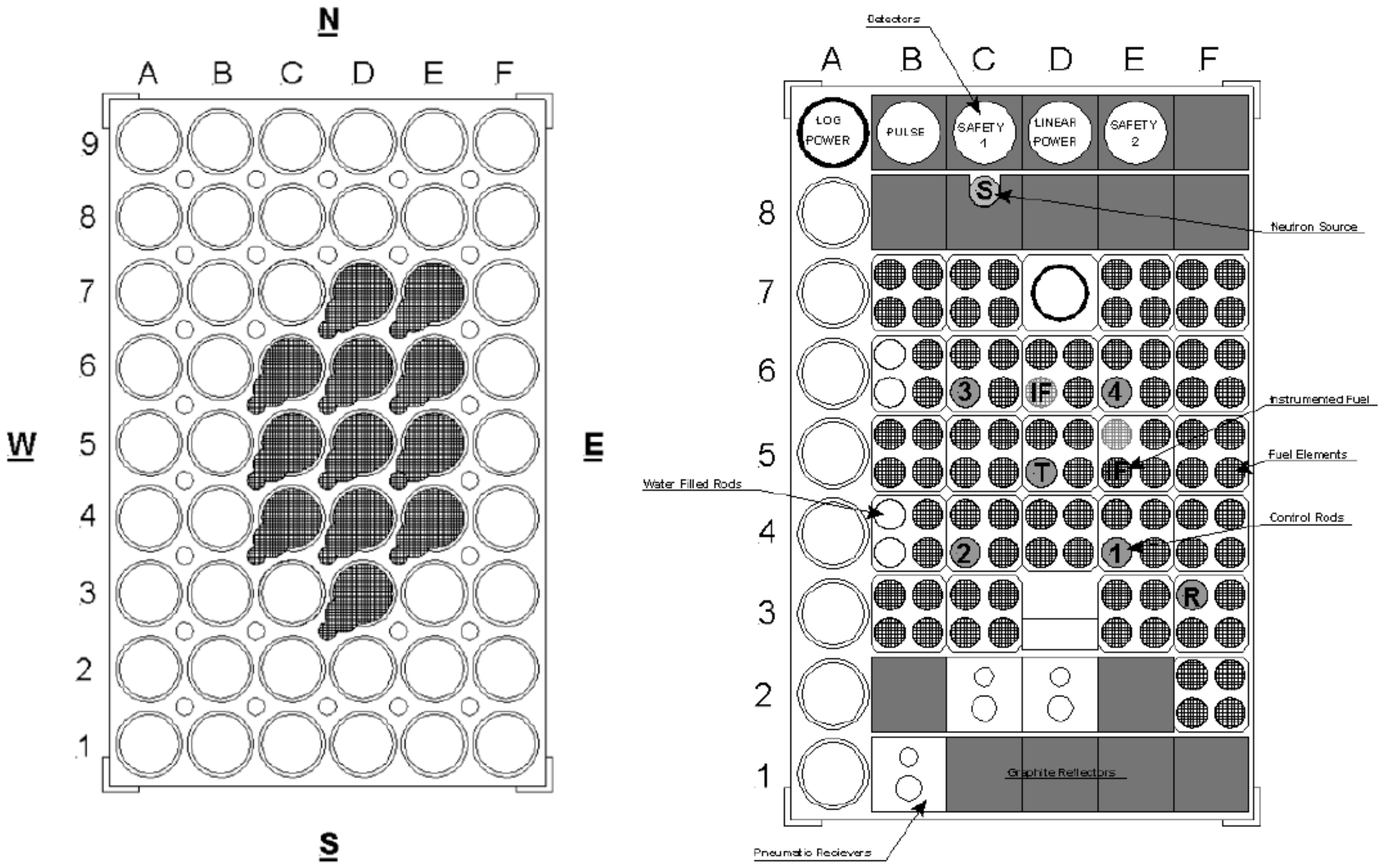
- Crystals will be arranged in an evenly-spaced, radial fashion in sample tubes in the highest flux region of the tube
  - Sample tube will be loaded onto the "A6" Position of the reactor grid plate of the Nuclear Science Center Reactor (NSCR)
  - Within sample tube, sample can loaded with batch of crystals will be placed in highest-flux position inside the tube

Fig. Grid plate and core layout of NSCR [4]



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Experiment Simulation Results

Sample Activity Estimates

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- Radiological concerns for irradiating this compound ($CH_3NH_3PbI_3$):
  - Lead
    - ^{209}Pb
    - 3.3-hour half-life; 0.6 MeV beta decay radiation [5]
    - Precursor ^{208}Pb makes up ~50% of all natural lead samples (can vary from sample to sample)
  - Iodine
    - ^{128}I
    - 25-minute half-life; 2.1 MeV beta decay radiation [5]

Sample Activity Estimates (cont.)

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- Radiological concerns for irradiating this compound ($CH_3NH_3PbI_3$):
  - All other feasible radioisotopes have negligibly low:
    - Parent isotope natural abundance
    - Neutron absorption cross sections
    - Decay constants
    - Branch ratios
    - Some combination of the above

Sample Activity Estimates (cont.)

- Isotopic production estimated and inventory tabulated over time using in-house developed Python executable script
  - Program uses NumPy's *expm* method to solve Bateman equation over a "run time" activation time series immediately followed by a "decay time" wait period
  - Library of constants such as half-life and thermal absorption cross sections taken from IAEA Chart of Nuclides and ENDF-BVII.1
  - Library of flux data for each sample position generated using gold foil and cadmium-covered gold foil data

Sample Activity Estimates (cont.)

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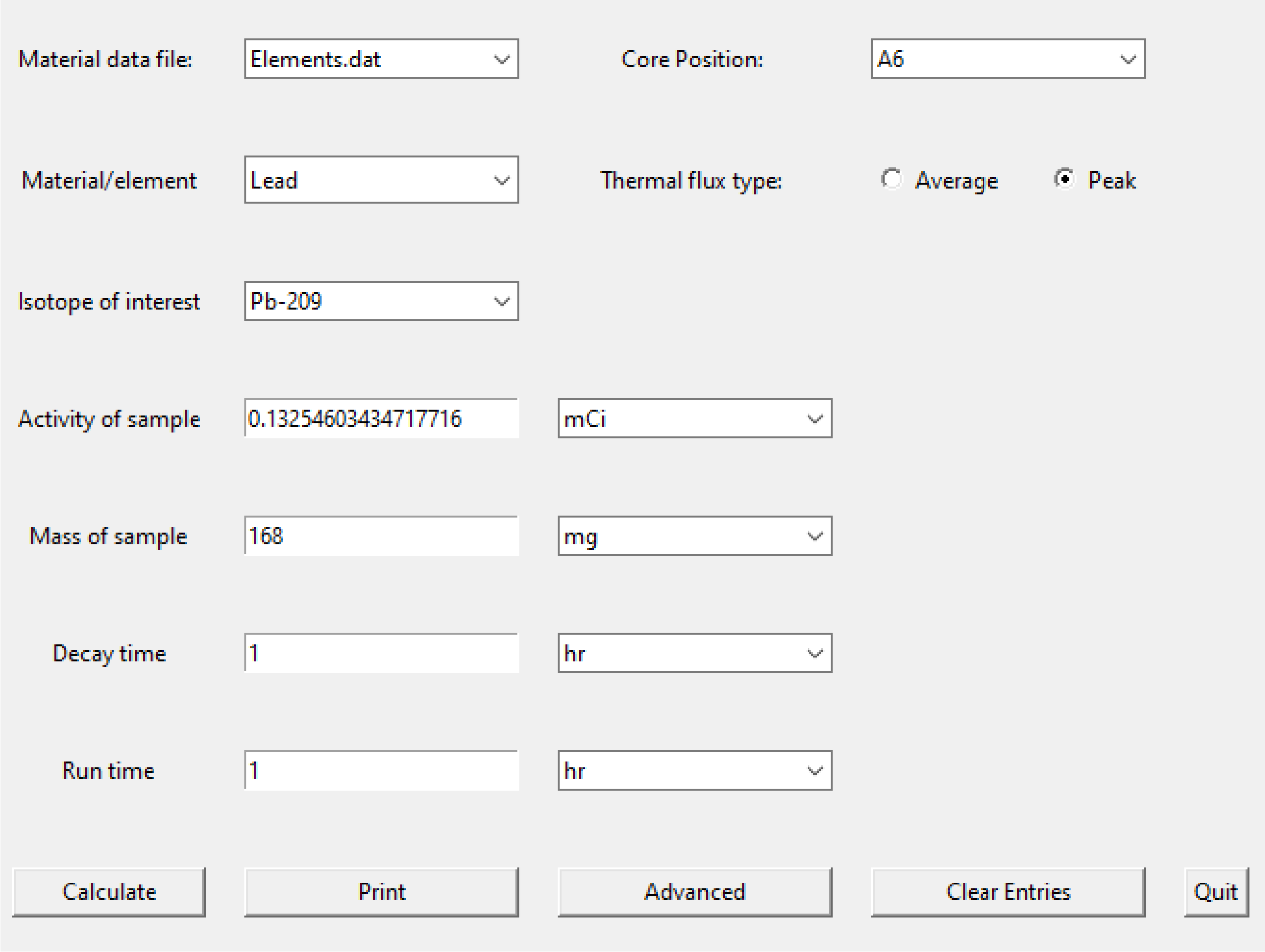
Fig. GUI display for in-house developed code for estimating sample activities for isotope production



Sample Activity Estimates – ^{209}Pb

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	Activity (mCi) after a decay time of:		
	1 minute	1 hour	1 day
5-minute Run	0.015	0.012	8.79E-05
1-hour Run	0.164	0.132	9.58E-04

- ^{209}Pb has a larger cross section, but is also longer-lived than ^{128}I once activated
- Radioactivity won’t fully diminish until 1 day of decay in storage
  - However low enough that a faster extraction could be reasonably achieved with reasonable time, distance, and shielding

Example Plotted Results – ^{209}Pb

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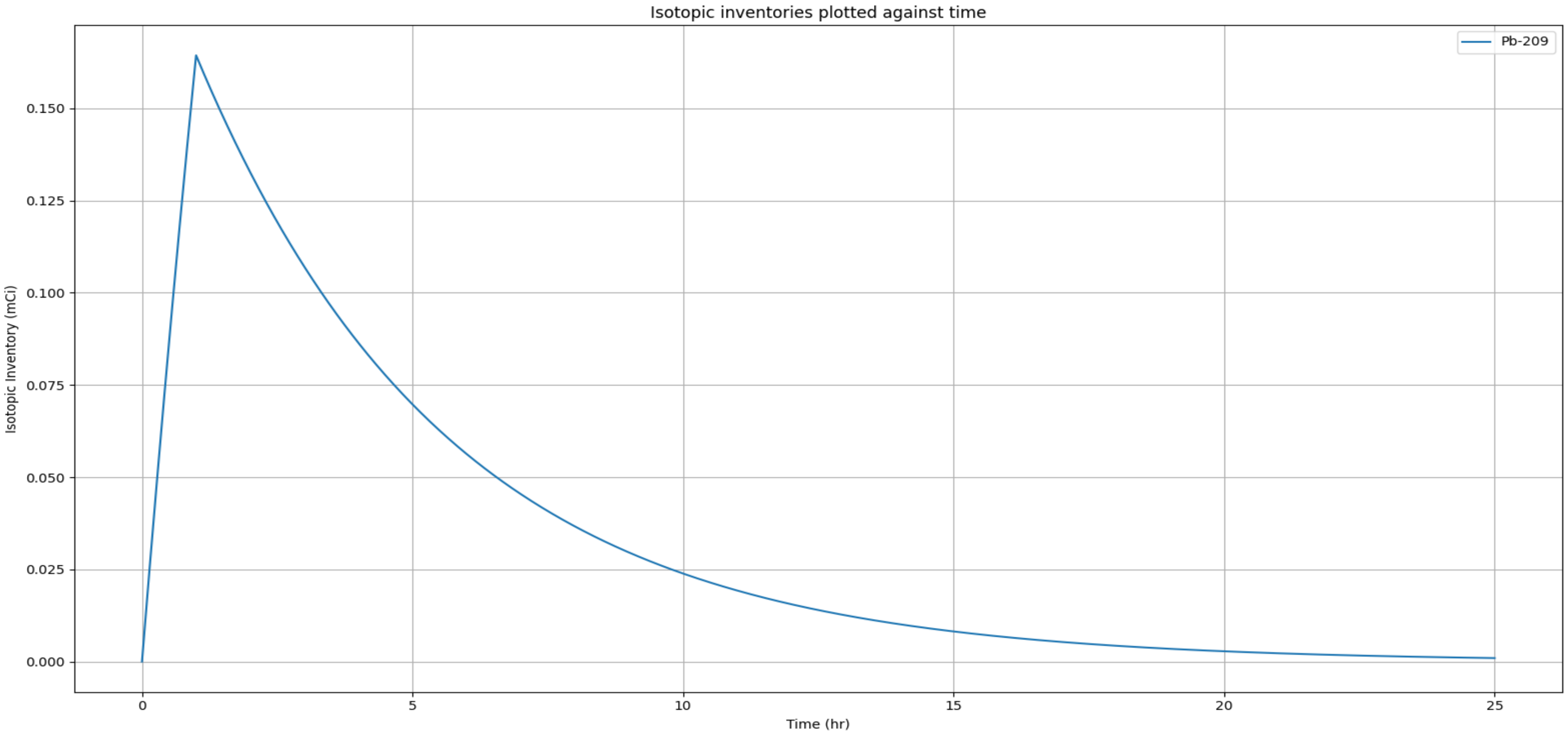

Fig. ^{209}Pb production and decay curve for a 1-hour run of 500mg of MAPbI followed by 1 day of decay in inventory

Sample Activity Estimates – ^{128}I

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	Activity (mCi) after a decay time of:		
	1 minute	**1 hour**	**1 day**
5-minute Run	240.307	46.776	1.11E-15
1-hour Run	1505.122	292.975	6.96E-15

- ^{128}I has a much shorter half-life and higher precursor capture cross section
- ^{127}I also has a higher atom density in MAPbI
- This makes immediate extraction much less feasible
  - However, the 25-minute half-life of ^{128}I means that the isotope has decayed to a negligible amount after ~5 hours

Example Plotted Results – ^{128}I

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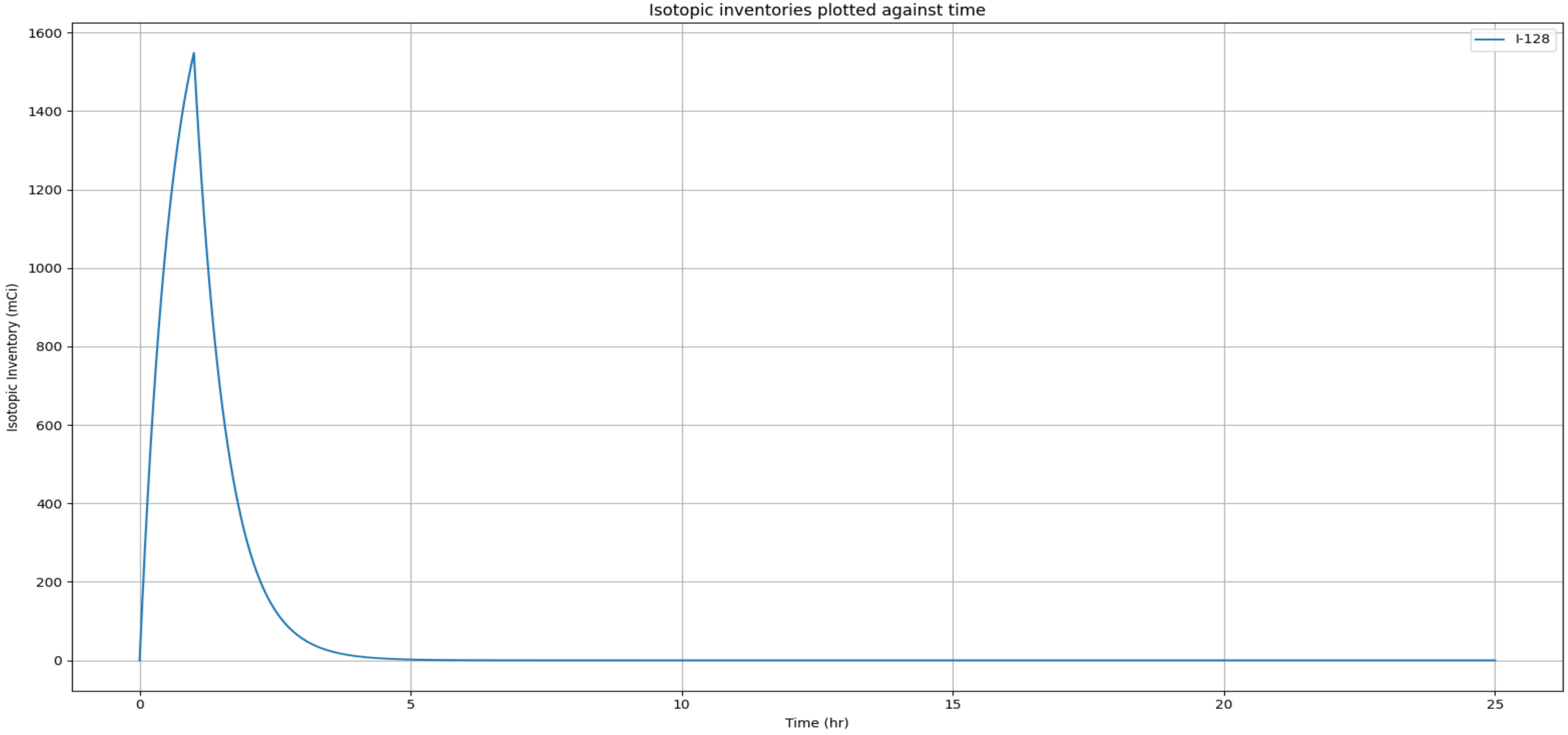

Fig. ^{128}I production and decay curve for a 1-hour run of 500mg of MAPbI followed by 1 day of decay in inventory

Core Reactivity Change Simulation

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- Evaluation of potential changes of core physics prior to running is essential to any experiment performed with a research reactor
  - Tech Spec Requirements at NSCR (T.S. 3.6.1)
    - Absolute value of experiment activity shall be < $1.00 [6]
  - Evaluation performed using a modification of an existing MCNP criticality search for the NSCR core at cold and hot critical

Core Reactivity Change Simulation (cont.)

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- 3 cases evaluated in MCNP to establish bounds
  1. NSCR at cold clean critical; no long tubes loaded in any experimental positions
  2. Sample tube loaded in A6 position with cylinder equal in density (~4.1 g/cc) to batch of crystals [7]
  3. Sample tube loaded in A6 position with cylinder equal to 2 times density of batch of crystals

Core Reactivity Change Simulation - Results (cont.)

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Trial	Final k_{eff}	ρ ($/100)	Δρ ($/100)
No Sample Tube	0.99208	-1.140461023	0
Loaded Sample Tube	1.00673	0.955001412	2.095462
Loaded; Double Density	1.00612	0.868967632	2.009429

- Conclusion: Sample loaded into core will be well under T.S. limits, even with sample tube loaded beyond expected quantity

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Future Work

Future Work

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- Higher precision modeling
  - Run fully modeled samples using ORIGEN for better isotopic buildup estimate; Use full geometry model for MCNP runs
- Design photocurrent measurement circuitry
- Gamma source irradiation setup
- MAPbI crystal production and sample preparation
- Low-power physics testing with setup
  - After low power testing, further MCNP simulations with hot core conditions will also be run prior to testing

References

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5. Livechart - Table of Nuclides - Nuclear Structure and Decay Data, IAEA Nuclear Data Section, 2024, www-nds.iaea.org/relnsd/vcharthtml/VChartHTML.html.
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Questions?