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Cosmogenic Activation Backgrounds

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LRT 2024

NSF/J. Yang

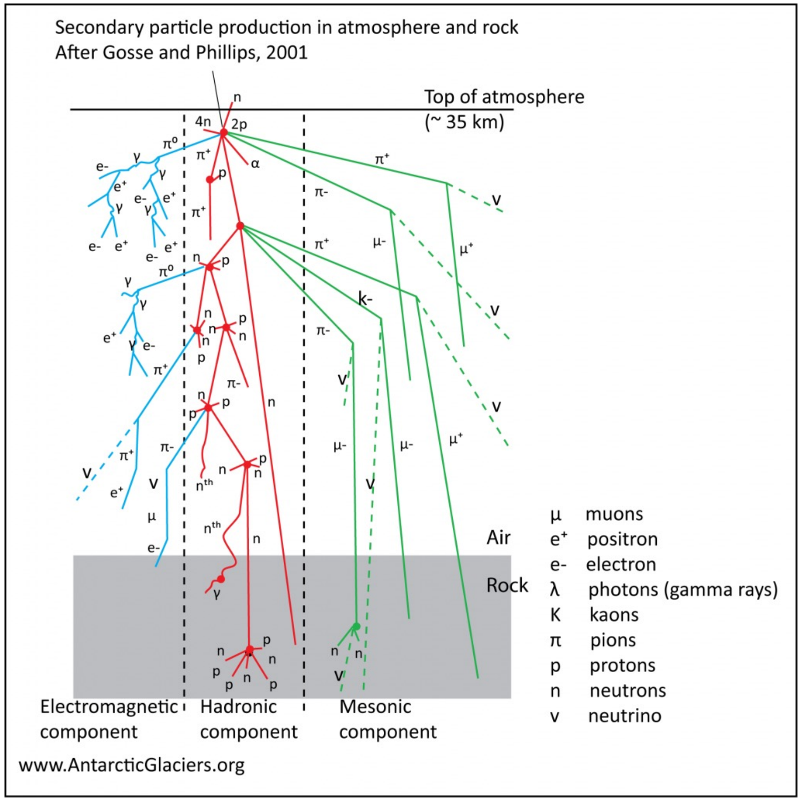
U.S. DEPARTMENT OF ENERGY BATTELLE

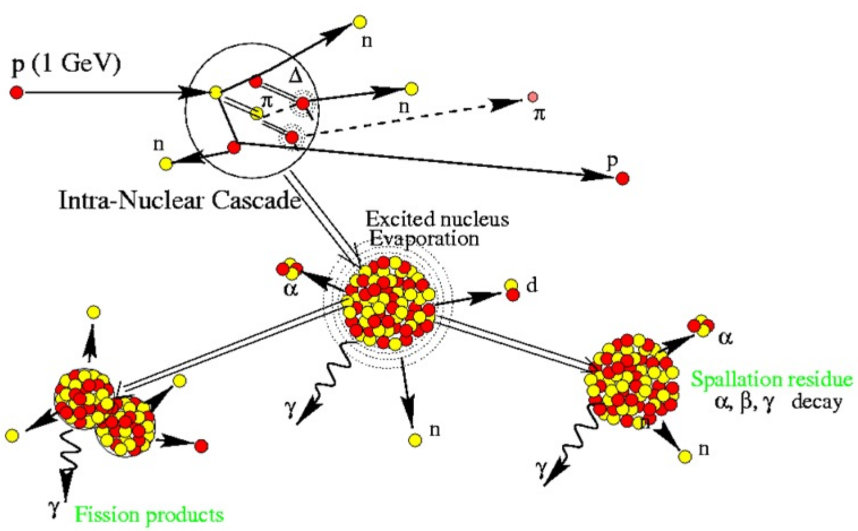
PNNL is operated by Battelle for the U.S. Department of Energy

Cosmogenic Activation

As low-radioactivity experiments get bigger and more sensitive, understanding the production rate of cosmogenic isotopes becomes more and more important

Needed to evaluate the total surface residency time, transportation options, and storage requirements for all low background detector components





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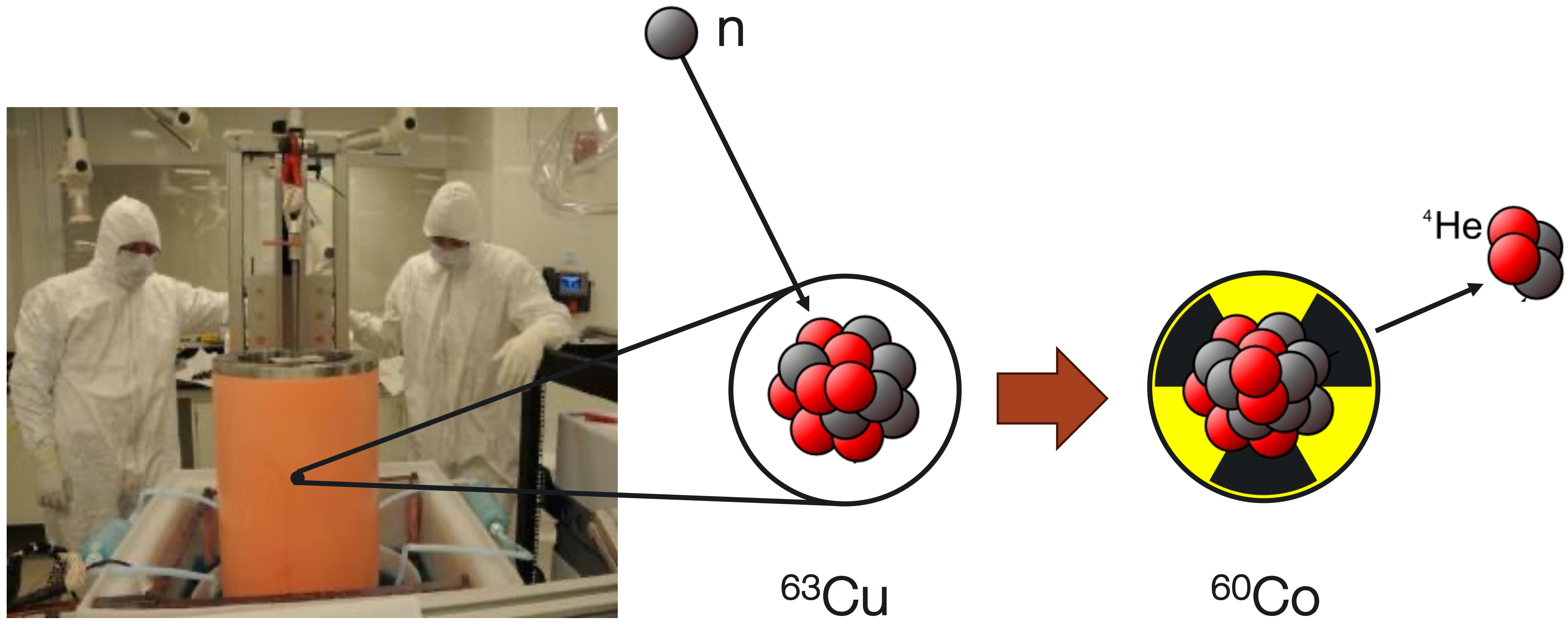
Cosmogenic Activation



Sea-level Production Rate:
30-90 atoms/kg/day

Sea-level Saturation Rate:
0.3-1.0 mBq/kg

Cosmogenic Production Rate

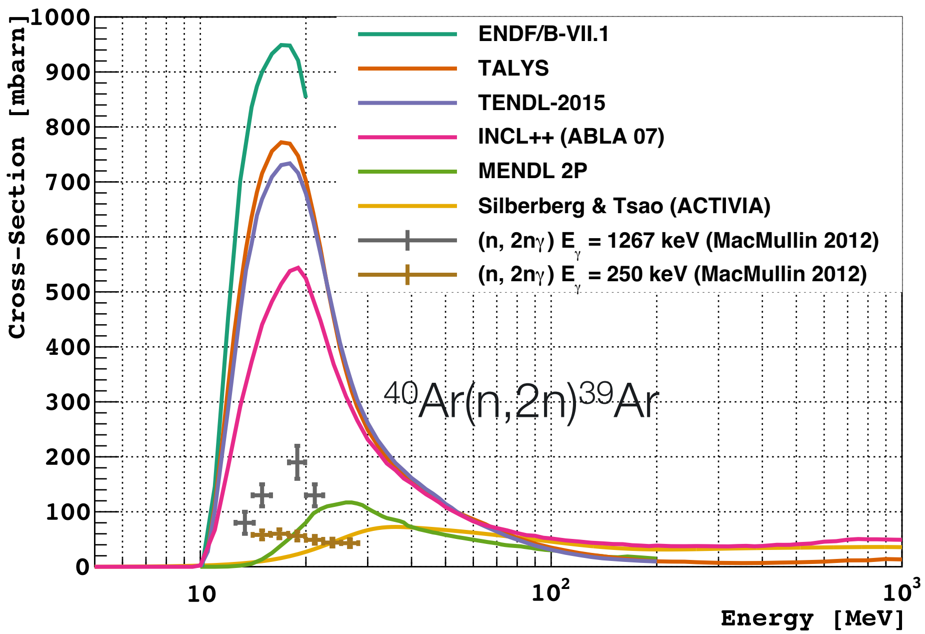
$$P(\vec{r},t) = \int \Phi(E,\vec{r},t)\,\sigma(E)\,n\,dE$$

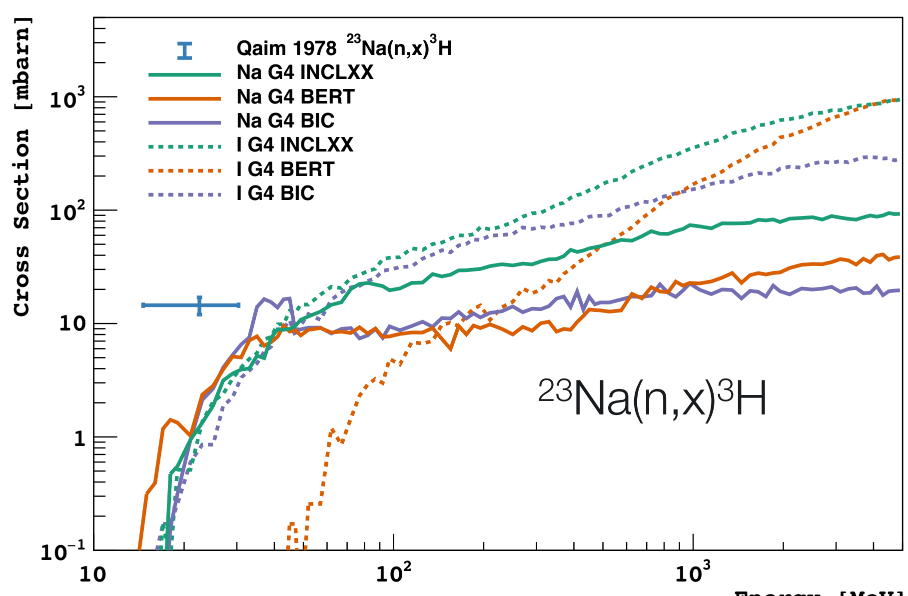
The production rate depends on 3 things

- Cosmogenic particle flux
- Production cross-section
- Target material

Cosmogenic Activation Cross-Sections

There are **very few experimental measurements** of key production cross-sections





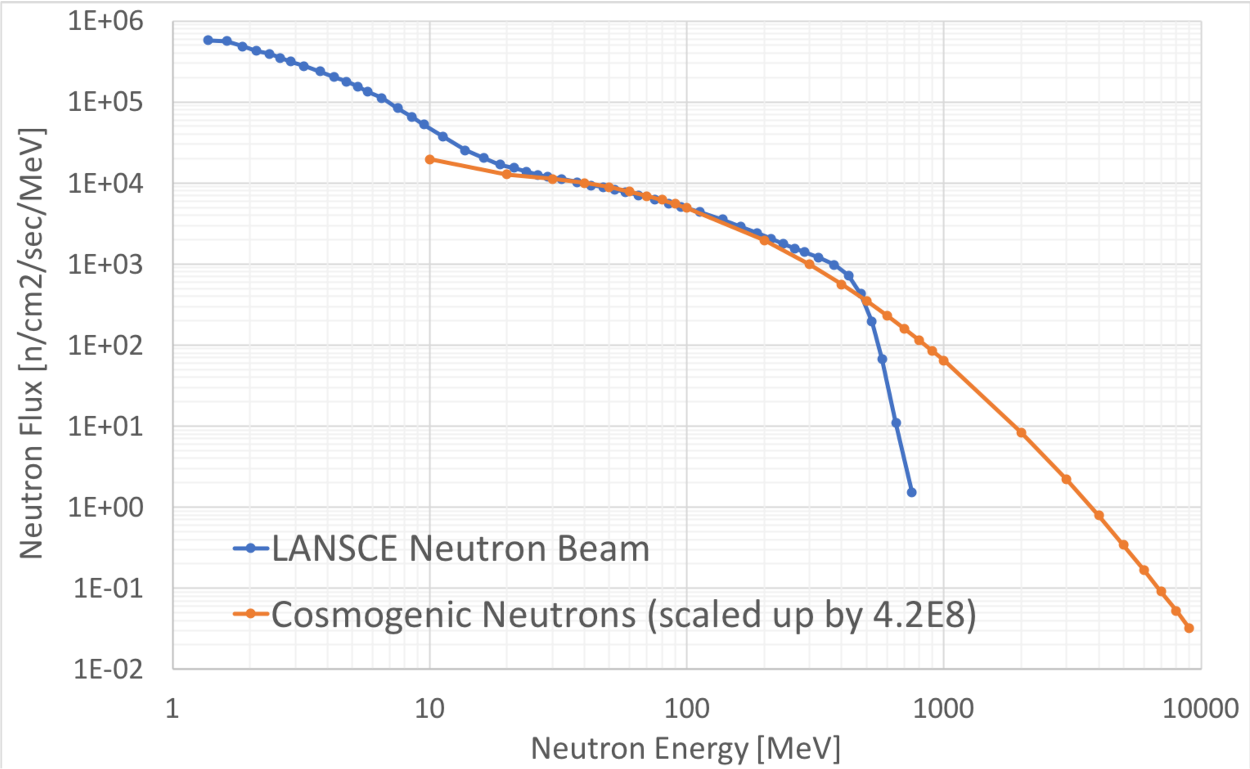
- Difficult to produce monoenergetic high energy neutron beams
- Products of interest to dark matter and other rare event searches can be low energy and not easy to detect

Cross-sections from nuclear data libraries and simulation codes often vary by an order of magnitude or larger

Can we directly measure the production rate through detection of decays?

LANSCE ICE-HOUSE Neutron Beam

Los Alamos Neutron Science Center (LANSCE)
has a neutron beam (4FP30R ICE-HOUSE II) that is
very similar in spectral shape to the cosmic ray spectrum



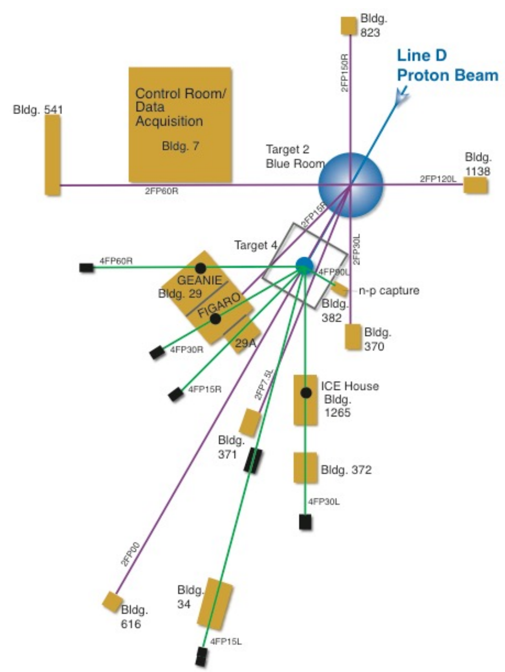
The good agreement in spectral shape from 10–500 MeV allows for low-uncertainty extrapolations to cosmic ray activation rates

The neutron flux is roughly **4.2×10^8 times larger** than the sea-level cosmic neutron flux

1 second on beam
~ 13 years on the surface

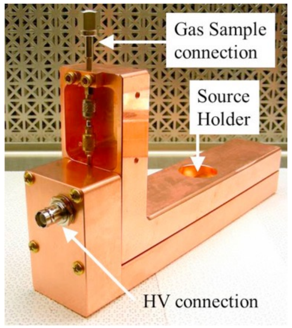
Measurement Technique

Use high intensity neutron beam to increase production rate compared to sea-level cosmic rays



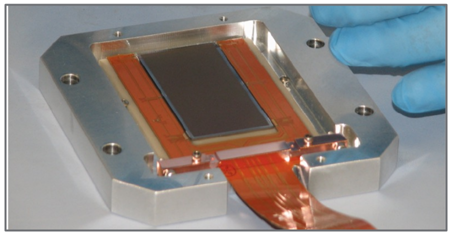
Irradiate material of interest and use radiation detection techniques to directly measure decay of activation products

Argon

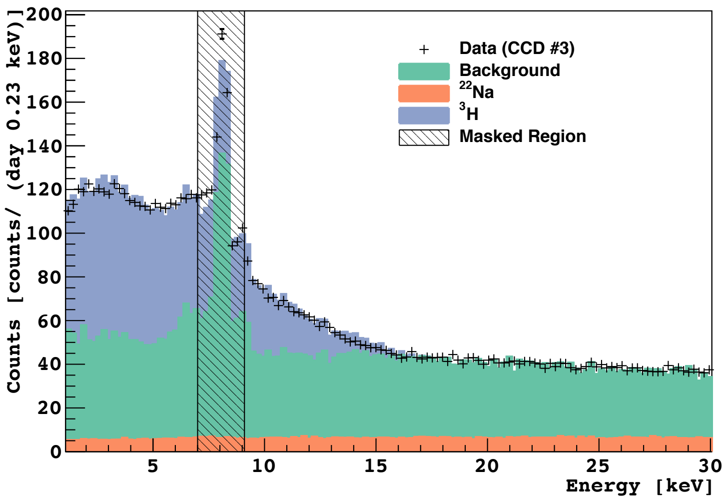


NaI

Si
CCD



Use **known** neutron fluence, and **measured activity** to extrapolate to natural cosmogenic production rate



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Activation Measurements for Dark Matter Experiments

^{39}Ar, ^{37}Ar in Argon

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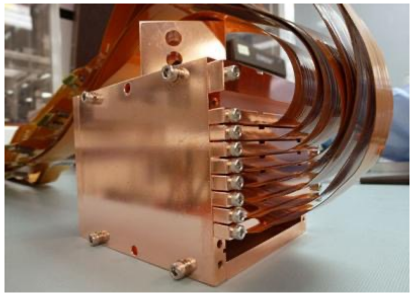
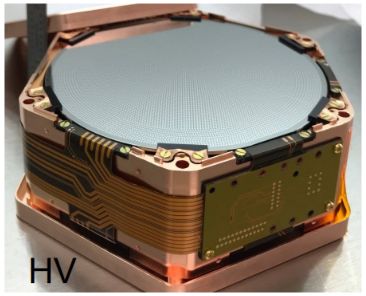
Phys. Rev. C 100, 024608 (2019)
arXiv:1902.09072

^{3}H (Tritium), ^{7}Be, ^{22}Na in Silicon

Pacific Northwest NATIONAL LABORATORY · Los Alamos NATIONAL LABORATORY EST. 1943 · CENPA Center for Experimental Nuclear Physics and Astrophysics · KICP Kavli Institute for Cosmological Physics at The University of Chicago

Phys. Rev. D 102, 102006 (2020)
arXiv:2007.10584



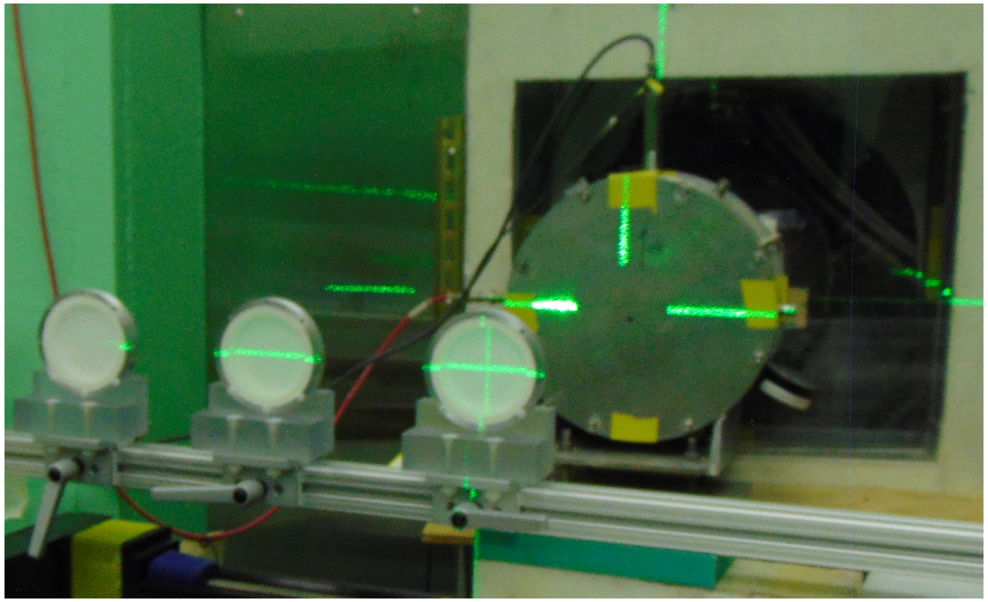


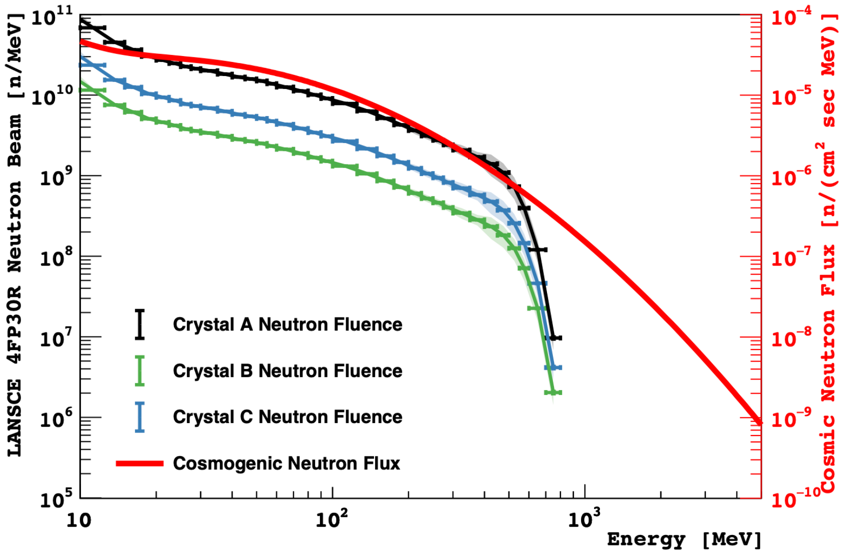
Cosmogenic activation of Sodium Iodide

Pacific Northwest NATIONAL LABORATORY · Los Alamos NATIONAL LABORATORY EST. 1943 · Australian National University · Yale · ILLINOIS UNIVERSITY OF ILLINOIS AT URBANA-CHAMPAIGN

Phys. Rev. D 107, 022006 (2023)
arXiv:2209.14898

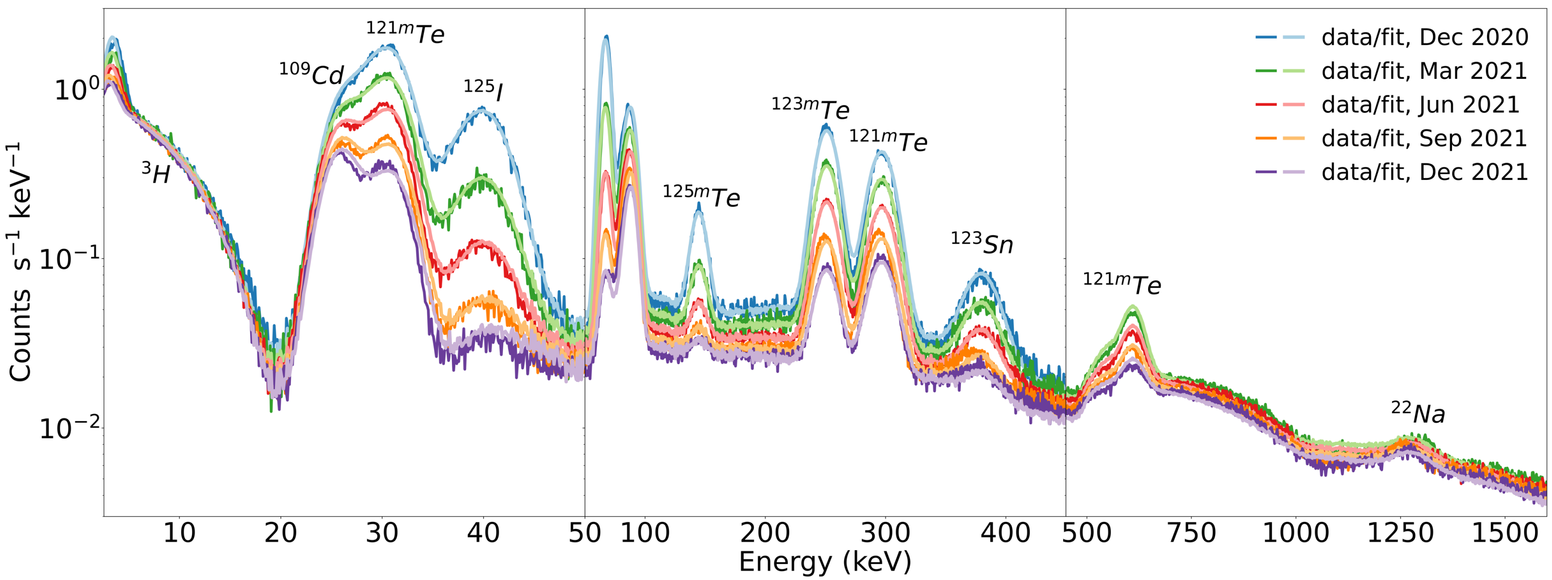
NaI Beam Irradiation

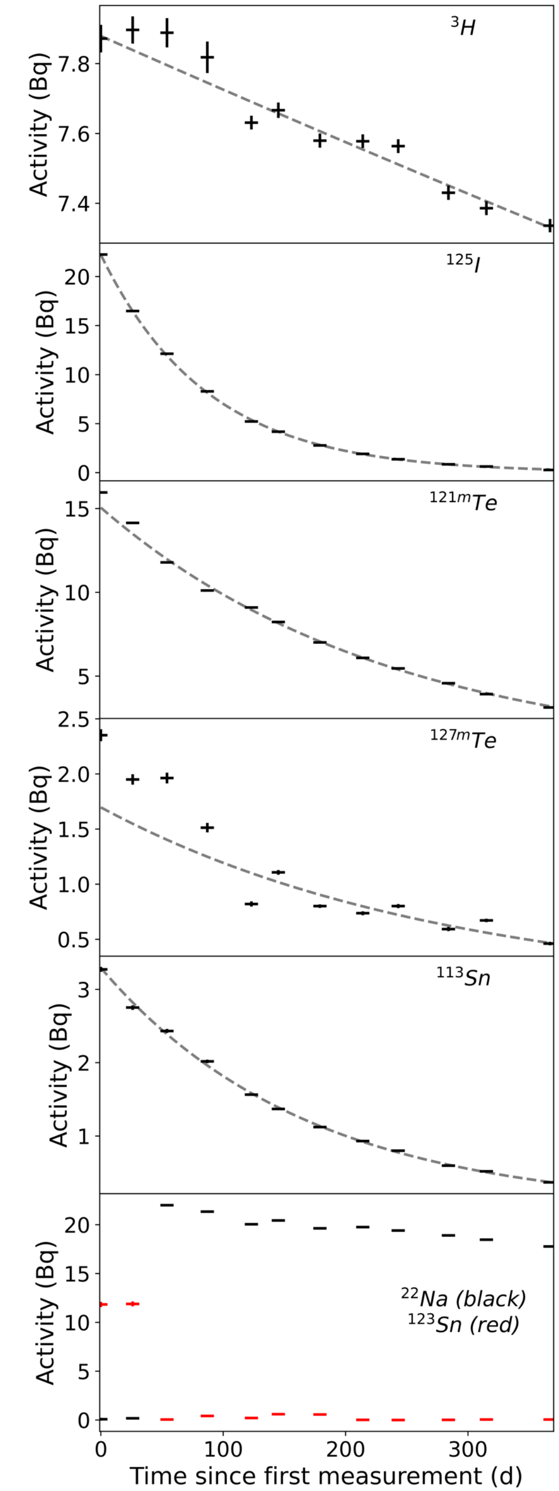




Irradiate NaI crystals on LANSCE beam with different exposures
(concerned about possible radiation damage)

NaI Counting

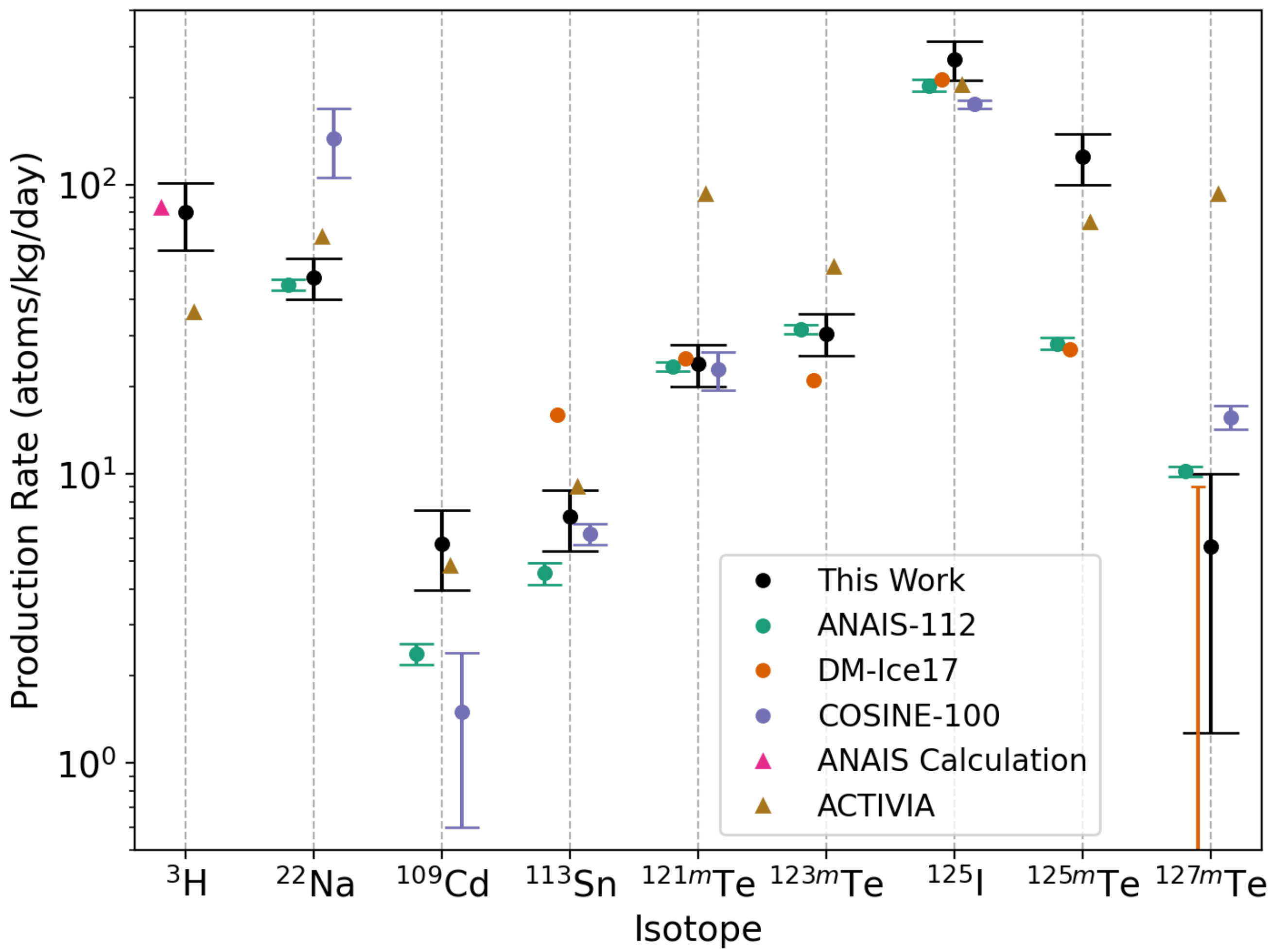




Mounted PMT to crystal and self-counted induced activity

Data was fit in both energy and time
to identify rates of several different isotopes

Production Rate Results



By extrapolating the measured activity to the sea-level cosmic neutron flux (accounting for cross-section shape uncertainties) we measured the production rates of 9 different activation products, including the first experimental measurement of tritium production in NaI

Cosmogenic Production Rate

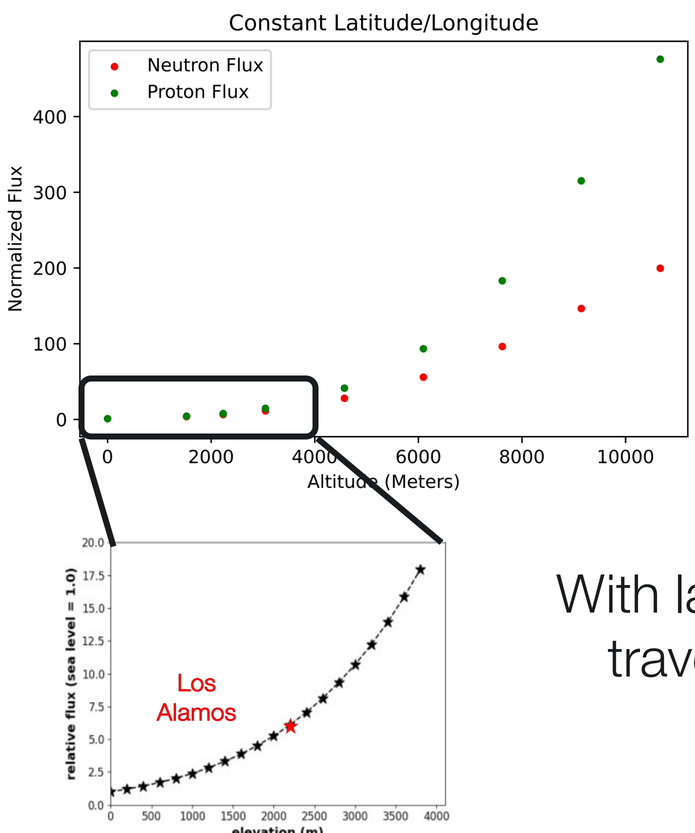
$$P(\vec{r},t) = \int \Phi(E,\vec{r},t)\,\sigma(E)\,n\,dE$$

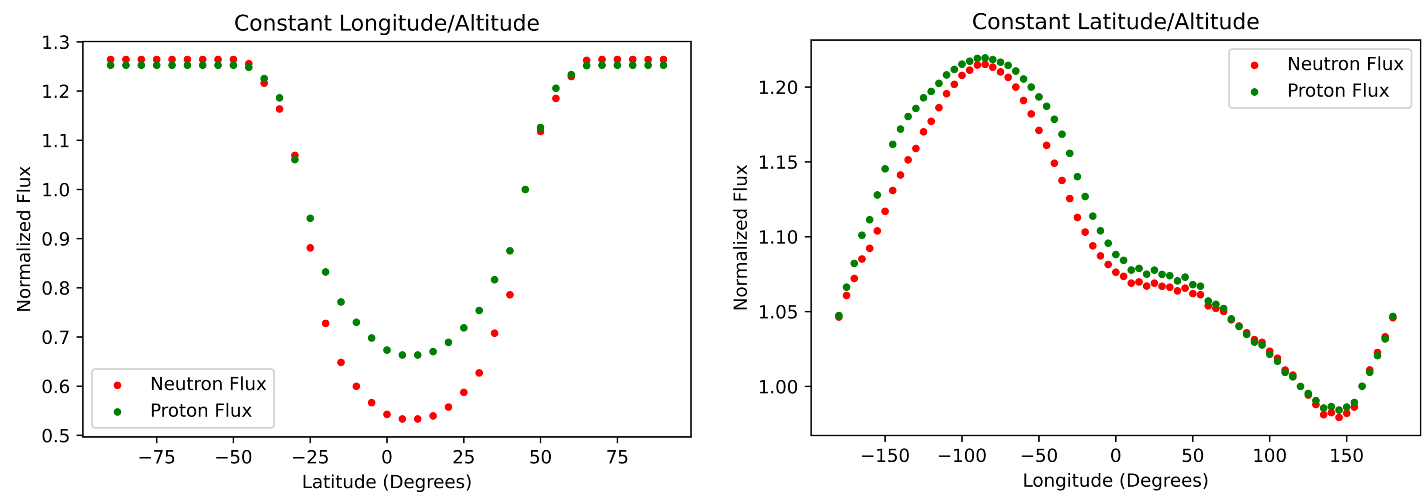
The production rate depends on 3 things

- Cosmogenic particle flux
- Production cross-section
- Target material

Cosmogenic Flux Variation

Flux of cosmogenic neutrons and protons varies by altitude, latitude, and longitude





With large international collaborations, detector components often travel across the globe before finally being assembled in deep underground sites

Careful tracking is needed to calculate the activation of components during transportation and storage

Witness Samples

Witness samples: Specifically chosen materials that can be measured for induced activity to determine the cosmogenic exposure of detector parts

Advantages:

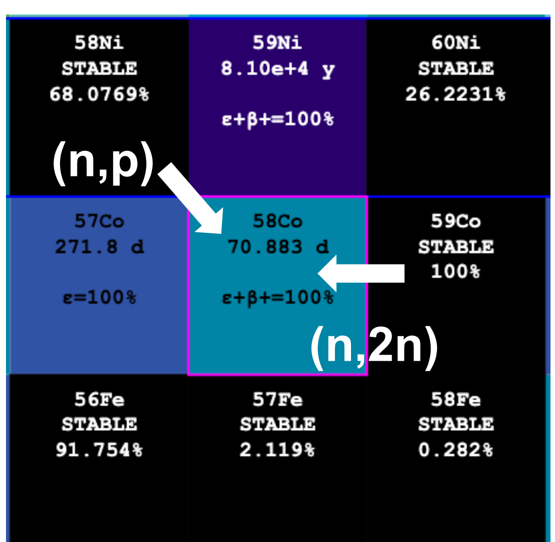
- Direct measurement of the relevant particle flux (compared to muon flux counters)
- Can be tailored to the specific reactions of interest
- Simple, passive system, easily deployed

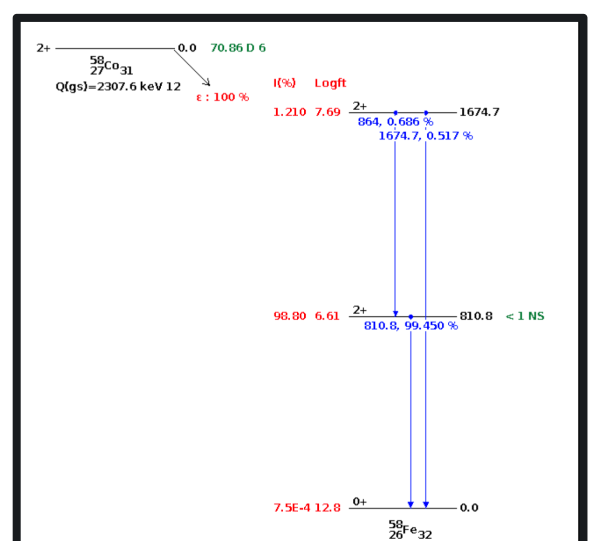
Disadvantages:

- Production rates for cosmogenic-induced reactions are low.
    - Cannot be used as a live monitor. Must be transported to a facility to be analyzed. Exposure and radioactive decay during transport must be taken into account

Identifying Witness Materials

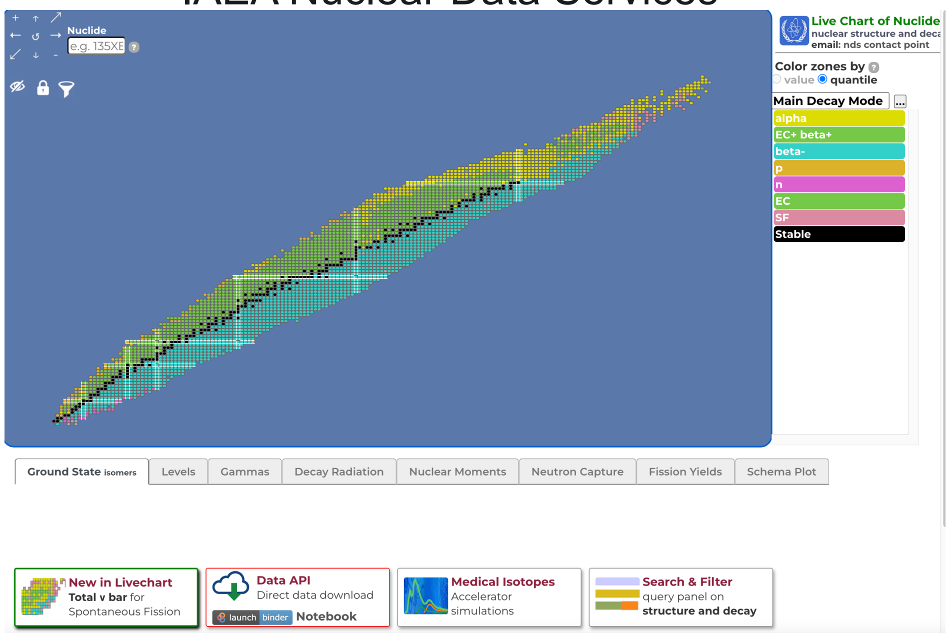
- Should contain a target isotope (with high natural abundance) with a nearby lighter radioactive isotope that
  - Has decay products that can be easily detected (e.g. gamma emitters)
  - Has a production cross-section that allows easy detection of activity (i.e. should not require a dark matter detector to be able to measure it)
  - Should have a relatively short half-life
- Ideally:
  - Reaction will be particle-type specific (e.g. (n,p), or (p,n))
  - Target will have single element composition
  - Target will be non-toxic, relatively cheap, and easy to procure.



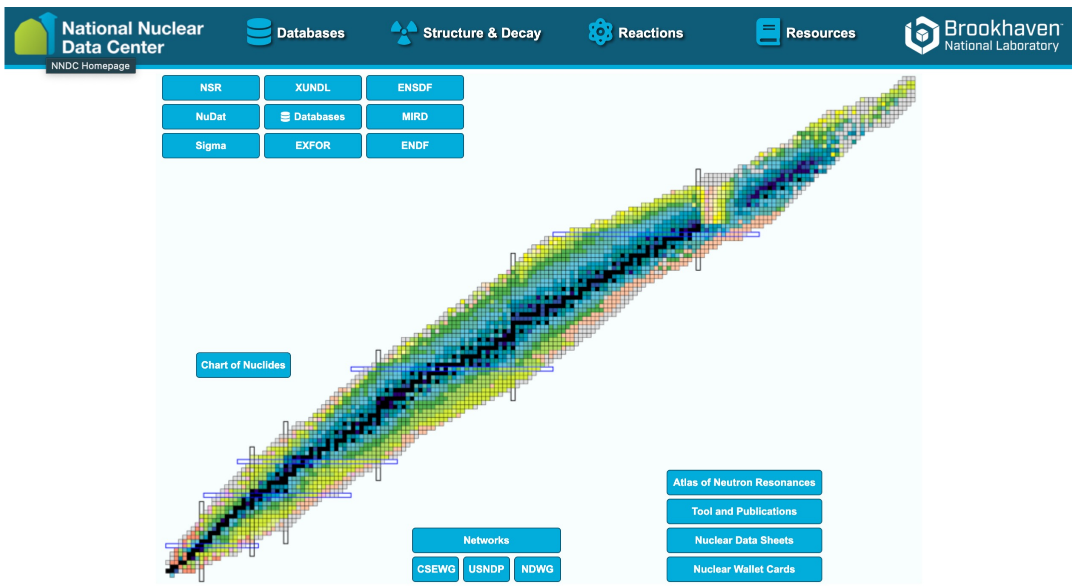


Identifying Witness Materials

IAEA Nuclear Data Services



BNL National Nuclear Data Center



Use existing isotope databases to collect full set of possible isotope for witness materials
Downloaded all relevant isotope data and saved in dataframe for subsequent work

~ 1600 isotopes

~ 190,000 decay schemes

Identifying Witness Materials

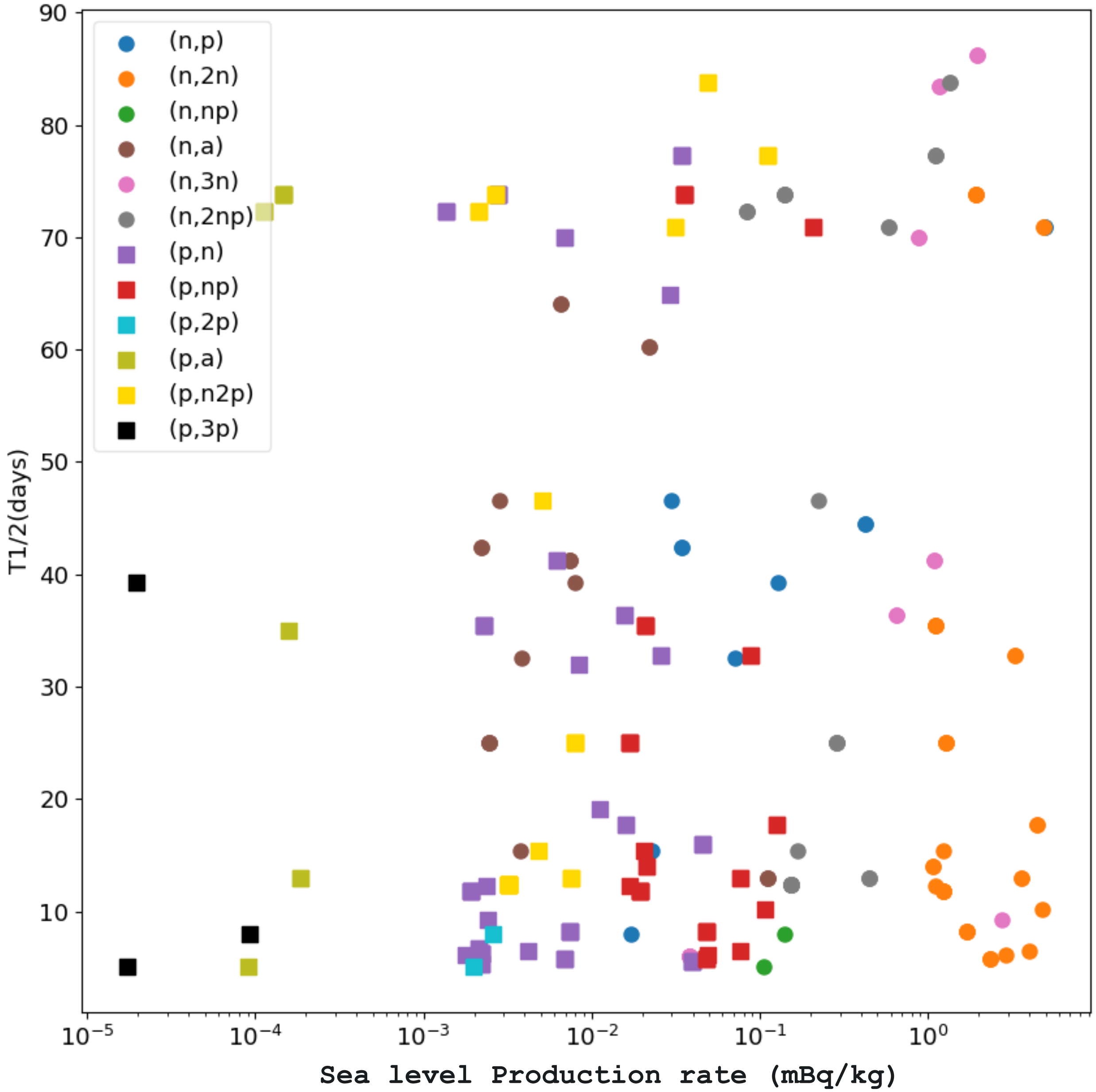
Select radioisotopes with suitable half-lifes

Select isotopes with gamma ray transitions above a threshold energy and intensity

For each reaction select target isotopes above a threshold natural abundance

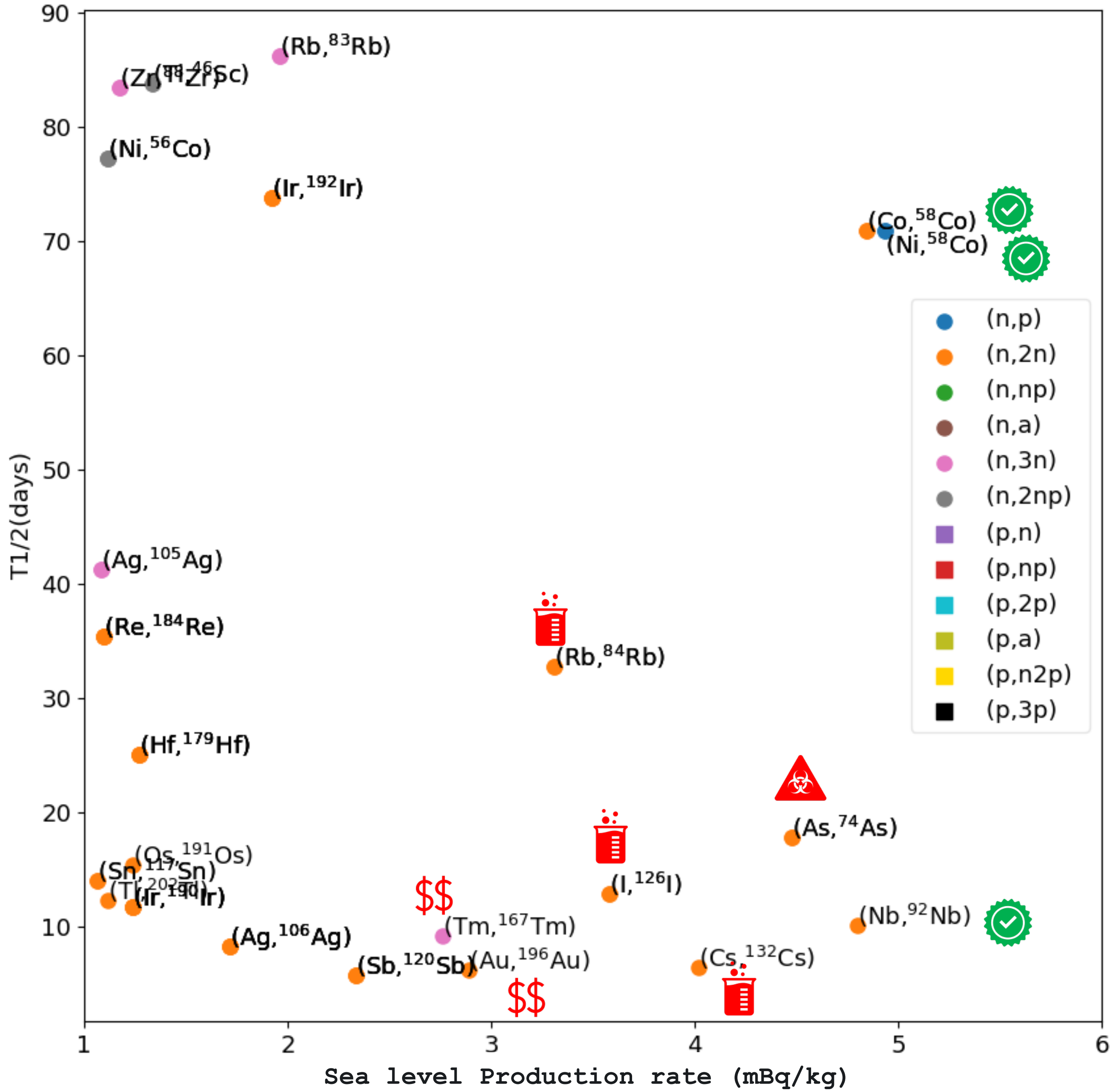
Extract reaction cross-sections from TENDL

Integrate cross-section with particle flux (EXPACS) to calculate production rates



(n,2n) reactions typically have highest production rates
Proton-induced reactions having lower rates due to the lower flux at sea-level

Candidate Witness Materials



Besides production rates and half-lifes, one also needs to take into account toxicity, chemical reactivity, price, etc.

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Deploying Witness Materials

Target	Reaction	Product	Product Half-life [days]	Time to 95% Saturation [days]	PNNL Sat. Activity [mBq/kg]	LANL Sat. Activity [mBq/kg]
Ni	(n,p), (n,2np), (n,3np)	^{58}Co	70.9	307	5.5	31.5
Co	(n,2n), (p,np)	^{58}Co	70.9	307	5.6	32.4
Nb	(n,2n)	^{92m}Nb	10.2	44.1	1.93	11.2
Ti	(p,n)	^{48}V	16.0	69.2	0.057	0.39

Aim to target locations and paths that explore largest flux variations

- Sea-level, High-altitude ground
- Low, High Latitude
- Cross-Pacific/Atlantic Flight

Witness materials will be packaged with GPS tracker to evaluate activation during transportation

Goals

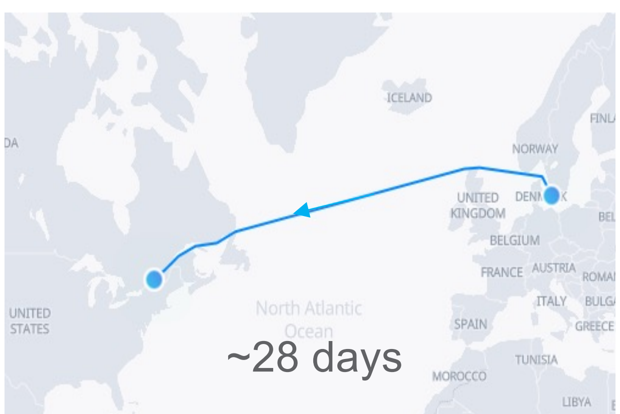
- Establish feasibility of using witness materials to measure high energy (> 10 MeV) particle flux
- Develop software for community to track activation based on location and materials

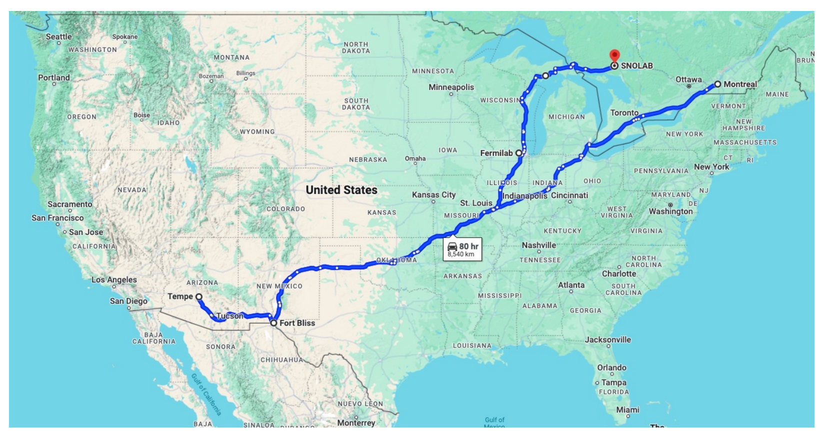
Cosmogenic Tritium

Sea-level production rate of tritium was measured to be
124 ± 25 atoms/kg/day

Corresponds to a background rate of
~ **1.8 mDRU per day of sea-level exposure** in the 0-2 keV energy range

Target Background Goal for Oscura CCD Dark Matter Experiment:
25 mDRU





Extremely short window of time to transport silicon from Europe to North America and fabricate the CCDs,
even accounting for shielded transportation and underground packaging.

Denmark (Ingot) → Arizona, US (Fabrication) → Fermilab, US (Packaging) → SNOLAB, Canada (Assembly)

Is there another solution?

*DRU: events/kg/keV/day

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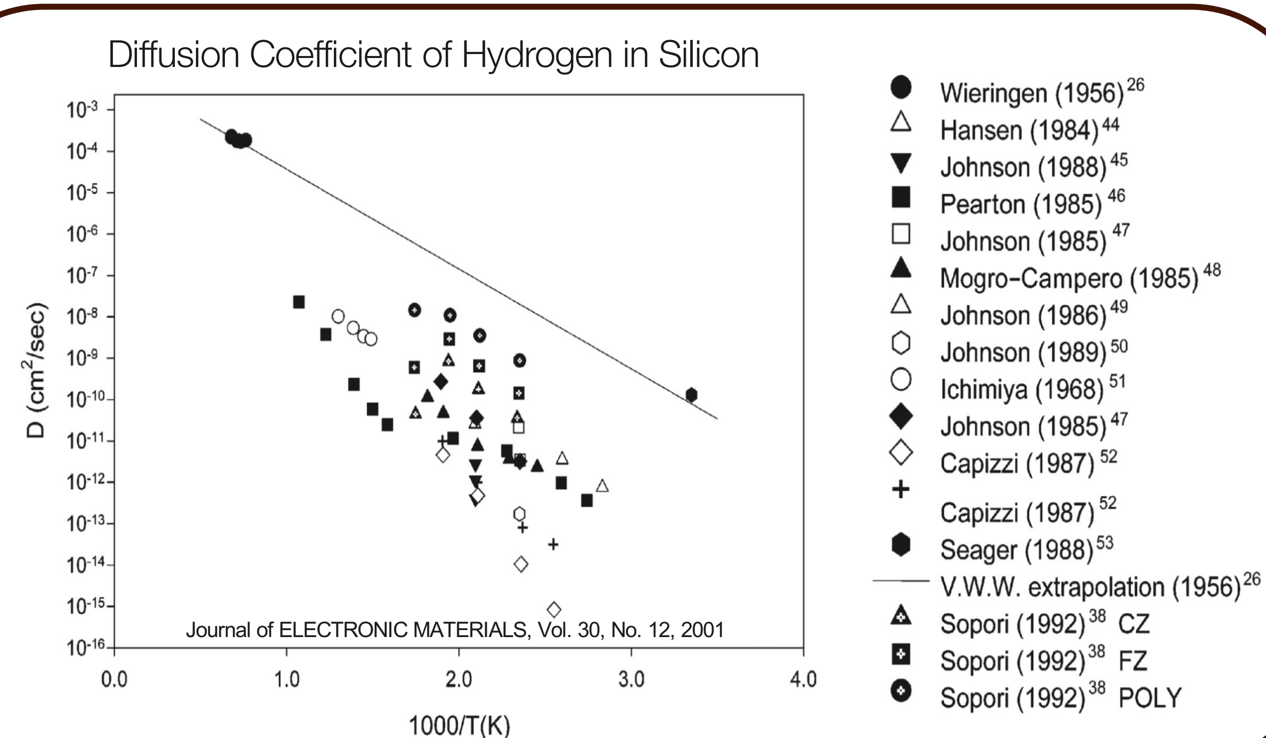
Tritium Bakeout

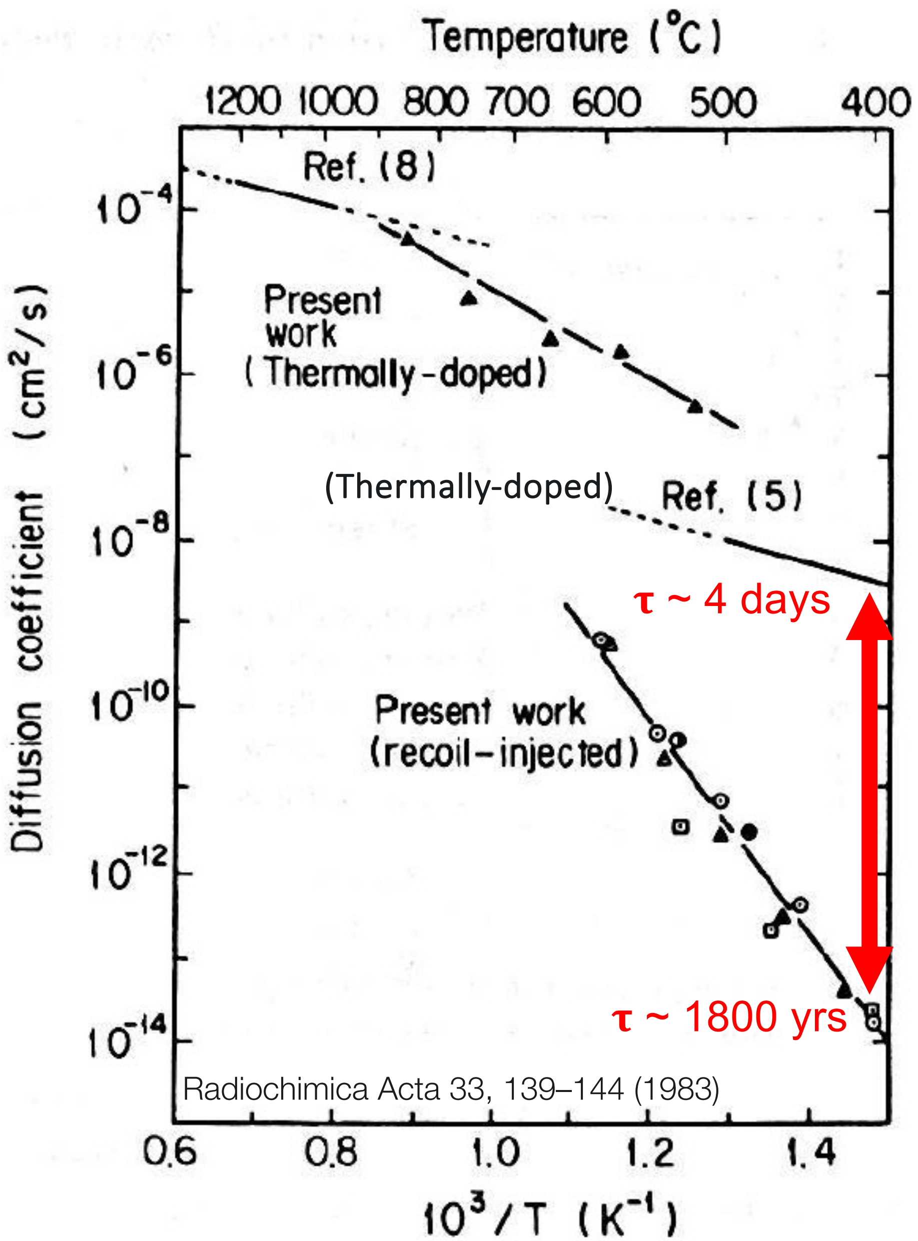
Hydrogen is typically mobile as an interstitial species in the silicon lattice

Diffusion of hydrogen in silicon is commonly used in semiconductor industry to reduce recombination and improve device characteristics.



Can we remove cosmogenic tritium from silicon by baking at elevated temperatures?

Tritium Bakeout



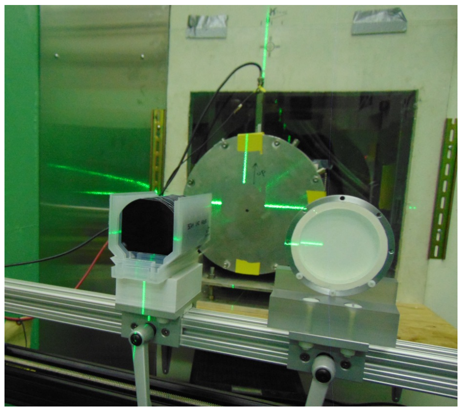
For tritium in silicon, the diffusion coefficient seems to strongly depend on whether the tritium was thermally diffused or implanted through nuclear recoils

This large difference is attributed to possible damage of the silicon lattice during recoil implantation, leading to trapping of tritium on defects

Since cosmogenic tritium is produced through spallation of a silicon nucleus, it is likely to also produce damage and defects in the silicon lattice

Tritium Removal Experiment

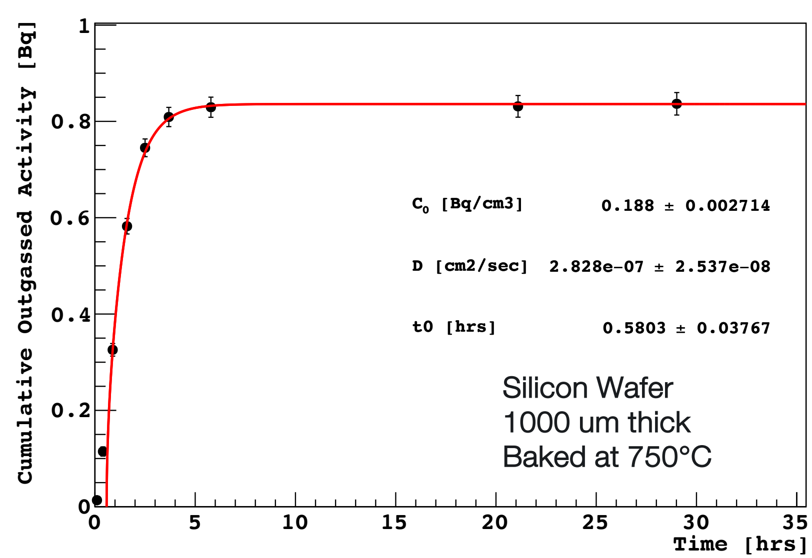
Silicon wafers on LANSCE neutron beam



RADDEC Pyrolyser-Trio Furnace System

Quantulus Ultra Low Background Liquid Scintillation Counter

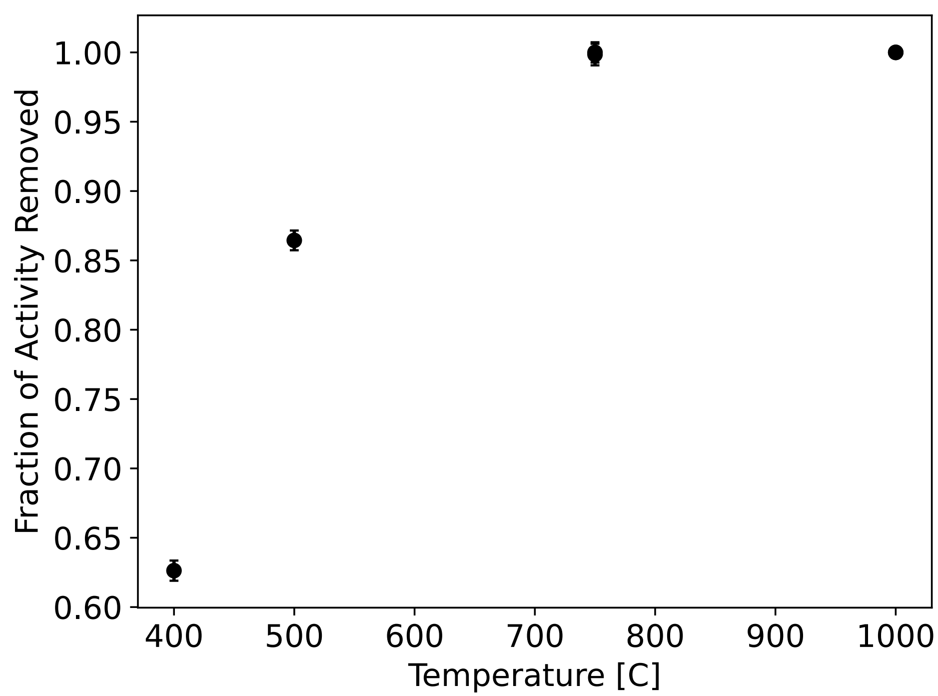
Irradiate on neutron beam → Produce tritium through spallation → Bake silicon at elevated temperatures → Collect removed tritium as a function of time → Count tritium activity in liquid scintillator → Evaluate diffusion and removal fraction

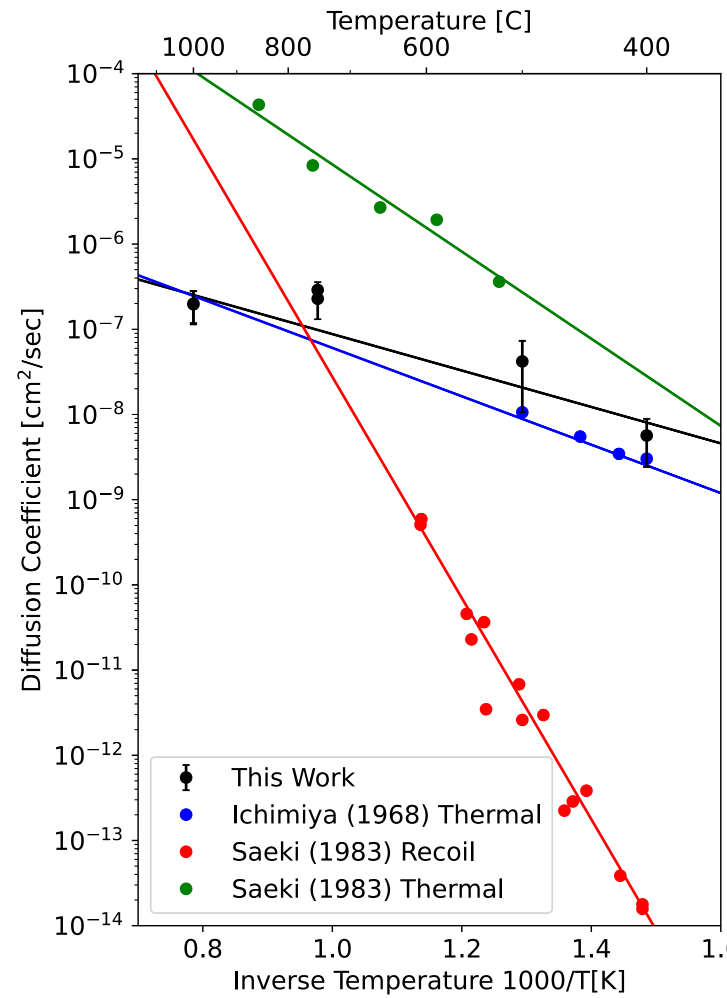


Tritium Removal Experiment

PRELIMINARY

Spallation-induced tritium can be completely removed from high-purity silicon with baking above 750°C for a few hours and diffusion coefficients are consistent with thermally diffused measurements





Results imply that cosmogenic tritium created prior to device fabrication can be removed by a high temperature bake, greatly reducing the constraints on exposure and shielding

Summary

- Understanding the production rates of cosmogenic backgrounds is critical for next generation rare event searches
- Production rates can be measured using the neutron beam at LANSCE and other locations. Measurements have been in argon, silicon, and sodium iodide
- High energy particle flux can be measured using activation of carefully selected witness materials
- In certain cases, removal techniques may be possible.
We have demonstrated the ability to remove spallation-induced tritium from silicon using elevated temperatures.

Thank You

Si Activation

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NaI Activation

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Tritium Removal

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Witness Samples

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