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Tritium Production and Release in Particle Accelerator Environments From Calculations to Experiments

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Outline

- Introduction
- Planned tritium diffusion experiment
- Radiation transport model setup
- Residual dose and tritium production calculations with FLUKA.CERN and PHITS
- Tritium diffusion estimates
- Summary

Collaborators

Local “Tritium Team” members (FNAL):

*Will Asztalos (IIT/FNAL), Alajos Makovec, Nisarg Patel,
Quinn Peterson, Don Athula Wickremasinghe*

Athula proposed the tritium diffusion experiment!

Colleagues from other facilities:

FRIB: Tom Ginter, Juan Zamora

ORNL: Yong Joong Lee, Vitaly Pronskikh

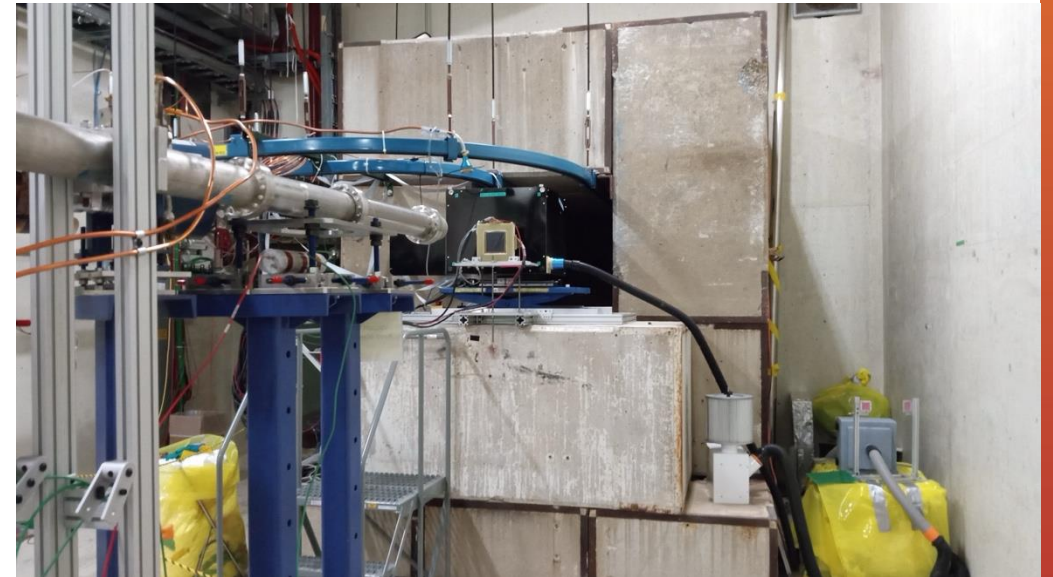
We plan to carry out similar experiments at FRIB and ORNL and possibly other facilities

✚ Tritium Diffusion Experiment Concept

- Target module: target surrounded by material samples
 - Common materials used in accelerator environments
- Irradiate the target at MTA (MeV Test Area) at FNAL
 - Tritium produced in the samples by secondaries
- After the beam is off, remove the samples from the module and submerge them in water so that tritium would be diffused from a sample into water
- Collect tritiated water and send it to the local LSC experts to measure tritium content
 - Can estimate diffusion coefficients in various materials

✚ Facility at Fermilab

- **MTA (MeV Test Area):** beamline enclosure hosting **ITA (Irradiation Test Area)**, a high energy beam facility devoted to studying the effects of radiation on components and materials.
- The ITA provides experimental space and infrastructure at the end of the 400 MeV (up to 7.5×10^{11} pps approved) MTA beamline in which users can place components and materials to study the effects of radiation for future experiments and fabrication.



Photos courtesy of Matt Strait (FNAL)

Experimental Setup: Irradiation in ITA

Courtesy of Quinn Peterson

- **Target module details**

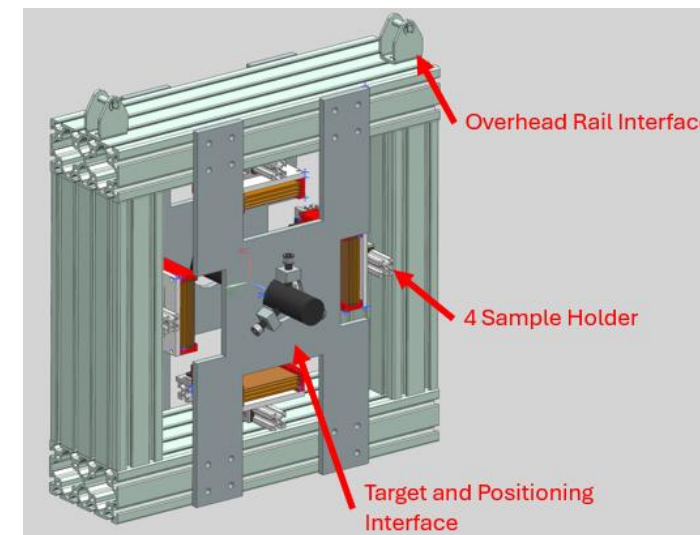
- Graphite target (6 cm $\varnothing$, 40 cm long; density 1.8 g/cc)
- 16 samples: Cu, SS316, Al6061, A36; 10 × 10 cm, 1–4 mm thick
- 4 sample holders, 13.5 cm from target, one material per side

- **Experimental assembly**

- 560 × 560 × 160 mm frame on lift table, interfaces with ITA
- Beam alignment with film + alignment group: ± 0.5 –1 mm
- 3D-printed holders on rails; latch allows remote removal

- **Requirements**

- Uniform water layer on all sample surfaces, constant temperature
- ≥ 10 mL of water, kept small to maximize tritium concentration

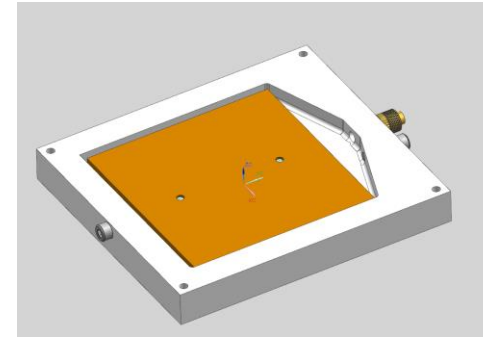




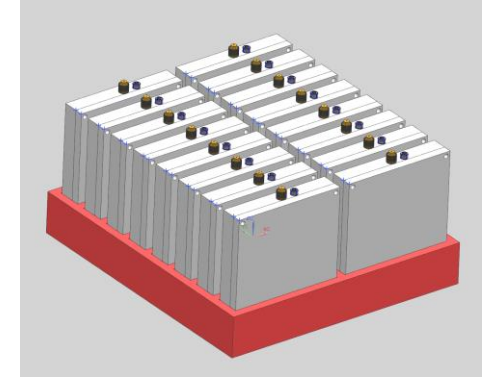
Sample Retrieval and Diffusion Procedure

Courtesy of Quinn Peterson

- **Dose constraints from radiation analysis**
 - ITA limit: 10 mrem/h at 30 cm (50 mrem/h with approval)
 - 1 h after beam off: module $\sim$ 500 mrem/h, single sample <10 mrem/h
- **Retrieval (no room for extra shielding in ITA)**
 - Sample holders pulled out with long-arm tooling
 - Assembly pushed $\sim$ 3 m into ITA cave on overhead rail to cool down
- **Diffusion**
 - One SS316 container per sample, all surfaces in water
 - Water injected by syringe (Luer lock) for precise volume
 - 16 containers in indexed array in temperature-controlled tank



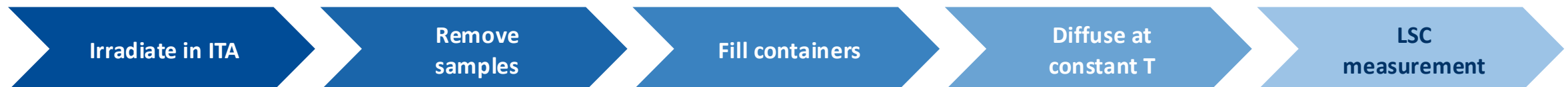
SS316 diffusion container



Container array

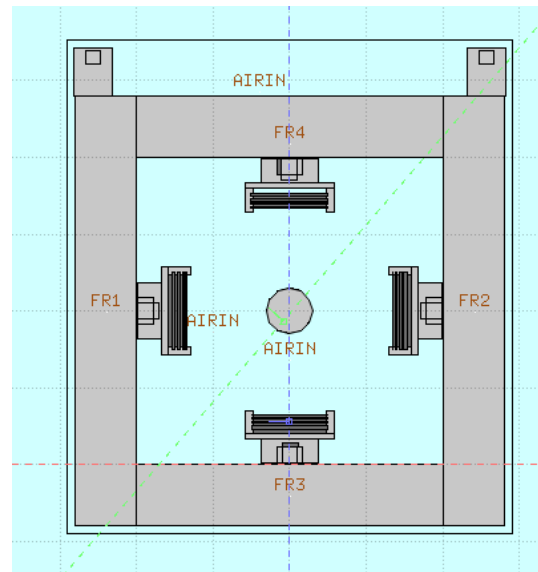
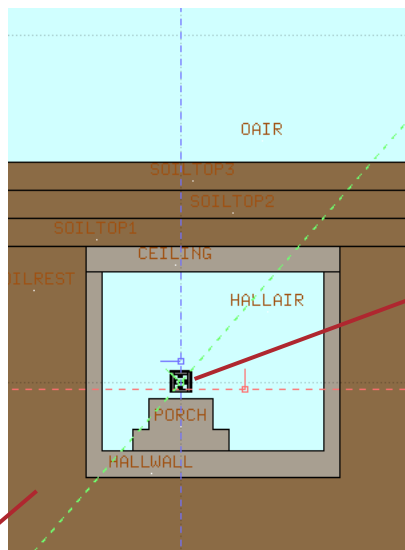


Temperature-controlled tank

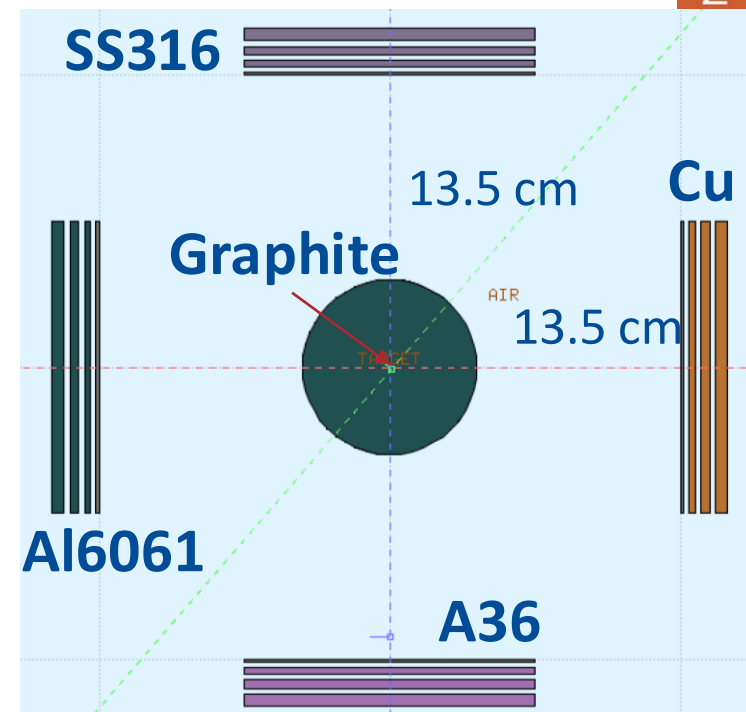
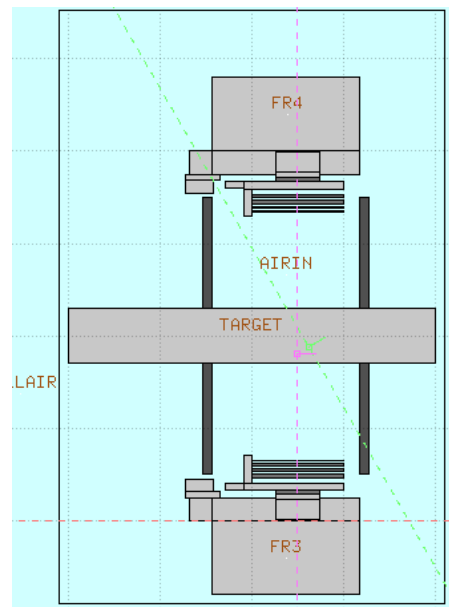
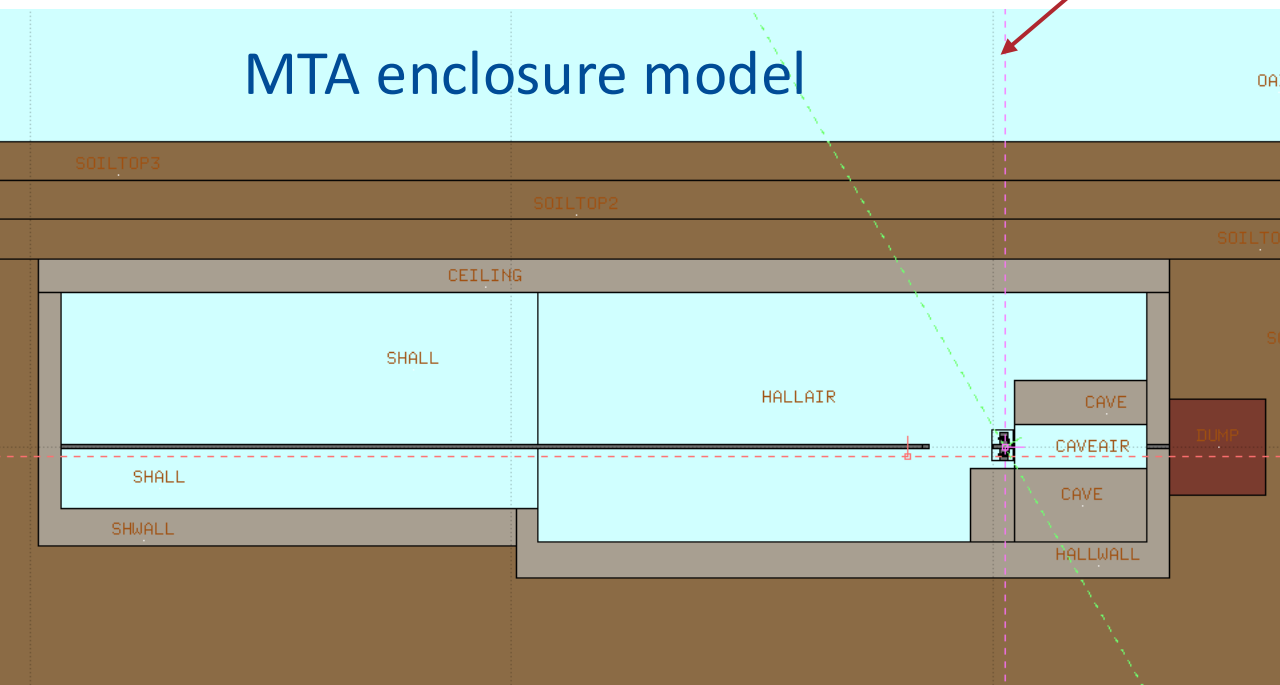


FLUKA Model Details

- Target module is placed in the beamline in the MTA enclosure
- Calculations are performed with FLUKA.CERN and PHITS

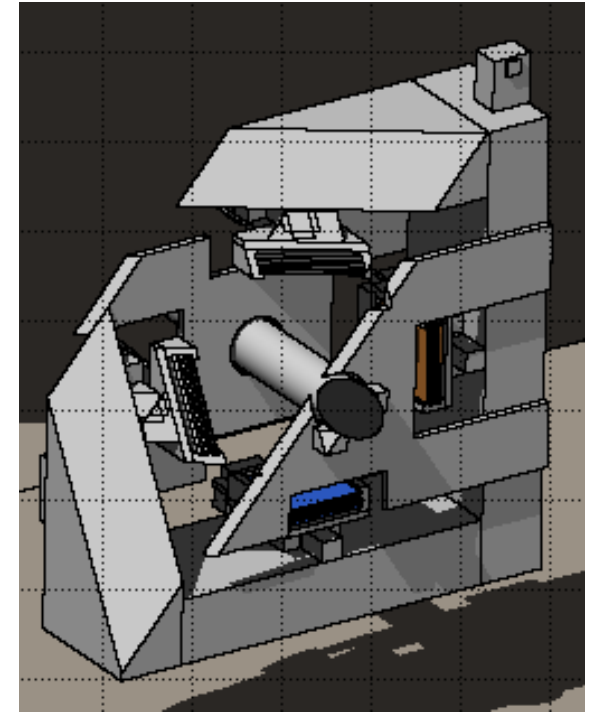


Module containing target & samples



⚙️ Radiation Analysis Input & Output

- Beam parameters
 - 400 MeV protons, 7.5×10^{11} pps, $\sigma_x = \sigma_y = 1$ cm
- Addressing a possible rad safety issue
 - To avoid contamination with irradiated powder, graphite target will be wrapped in 3M™ Aluminum Foil Tape 425 (included in the FLUKA model)
- Calculate tritium production and material activation
 - Optimize for maximum tritium production and minimum activation
 - Samples: minimize volume, maximize surface area (to facilitate diffusion)



⚙️ Residual Dose Maps

Irradiation 1 week, cooling 1 hour

Mesh parameters are:

$x = -300$ to 450 cm; $y = -300$ to 400 cm; $z = -2000$ to 500 cm

$dx = dy = dz = 10$ cm

Cross section

-230 to 400 cm

-100 cm to 80 cm

Numbers in mrem/h

Vertical view

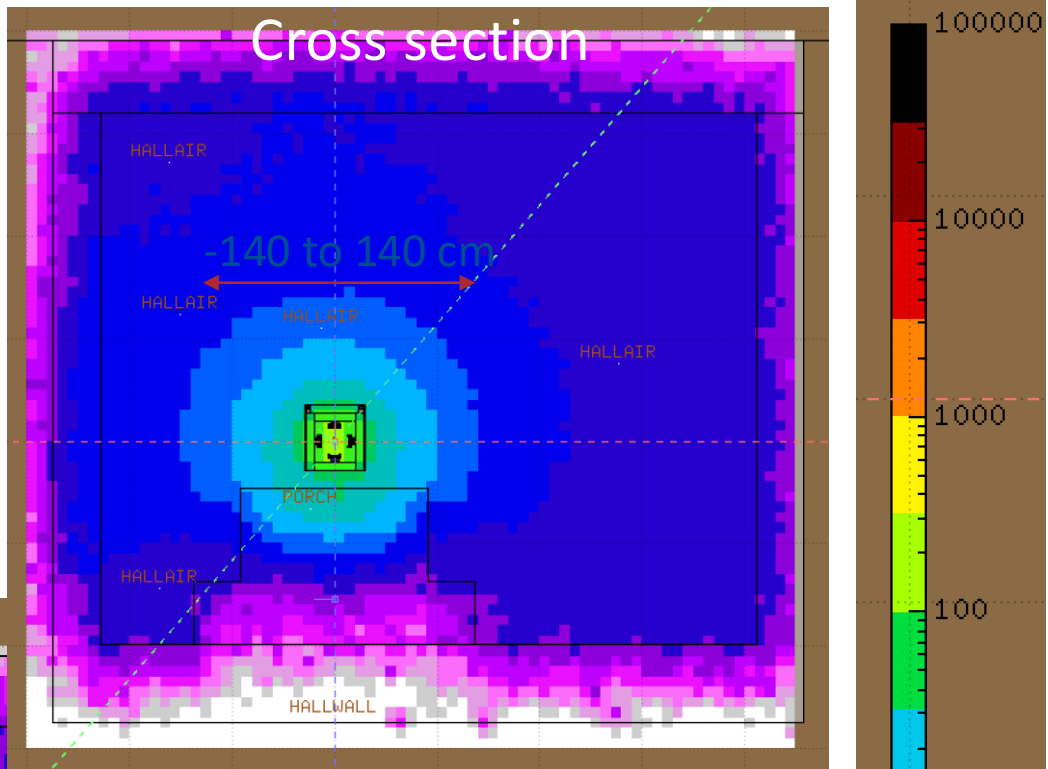
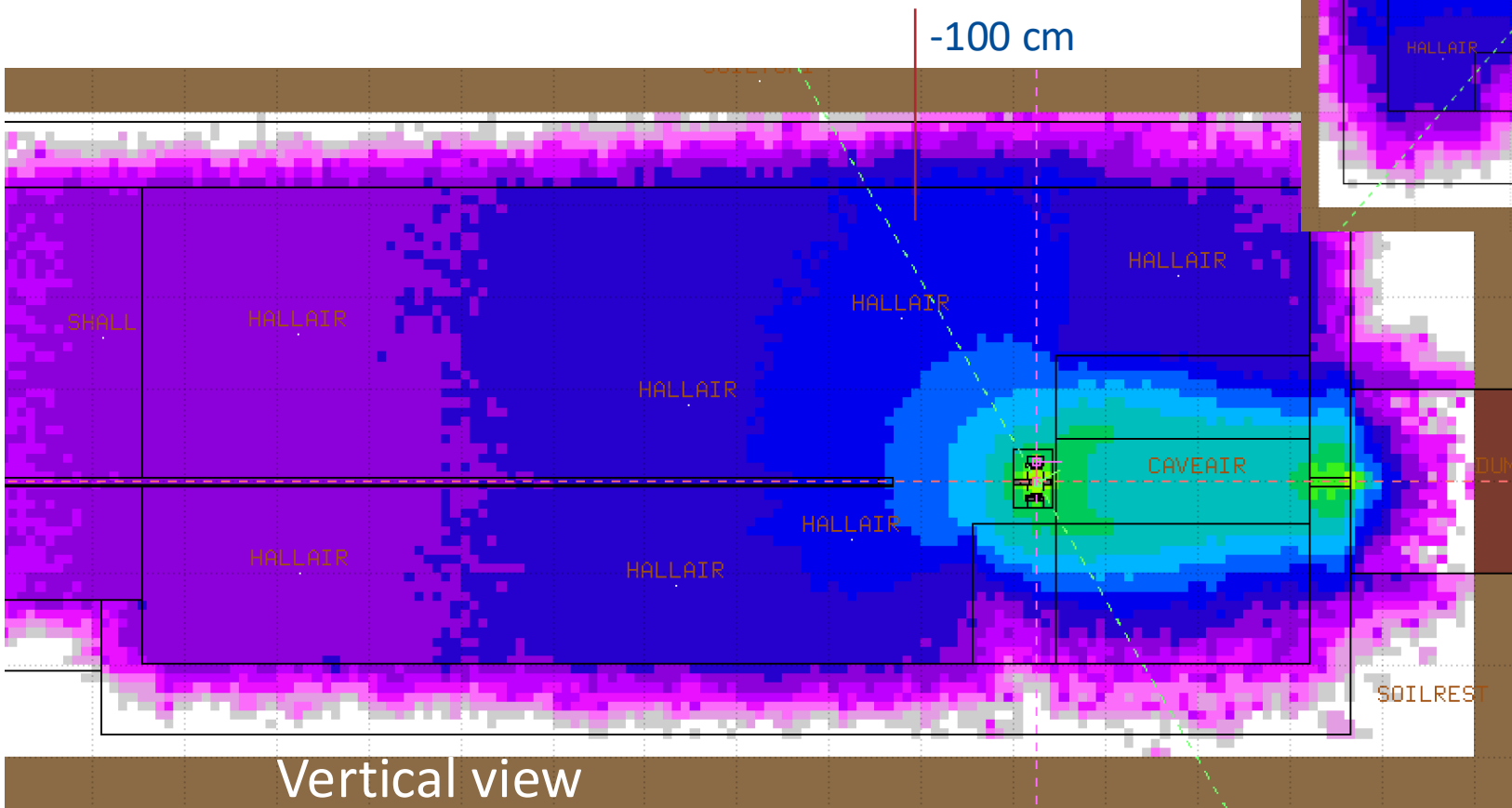
Residual Dose Maps

Irradiation 1 week, cooling 1 day

Mesh parameters are:

$x = -300$ to 450 cm; $y = -300$ to 400 cm; $z = -2000$ to 500 cm

$dx = dy = dz = 10$ cm



Numbers in mrem/h

Residual Dose Summary for the Target Module

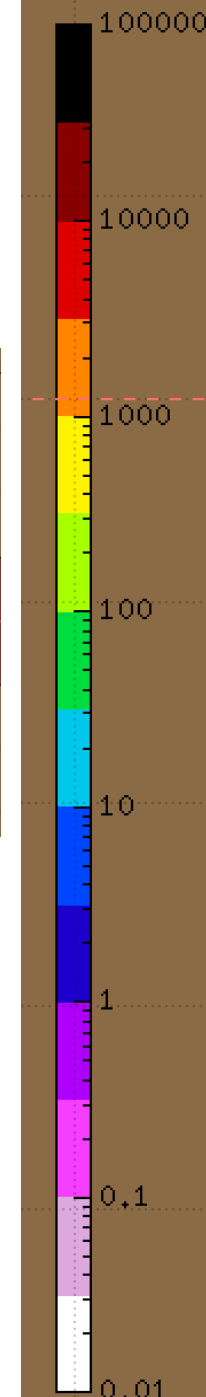
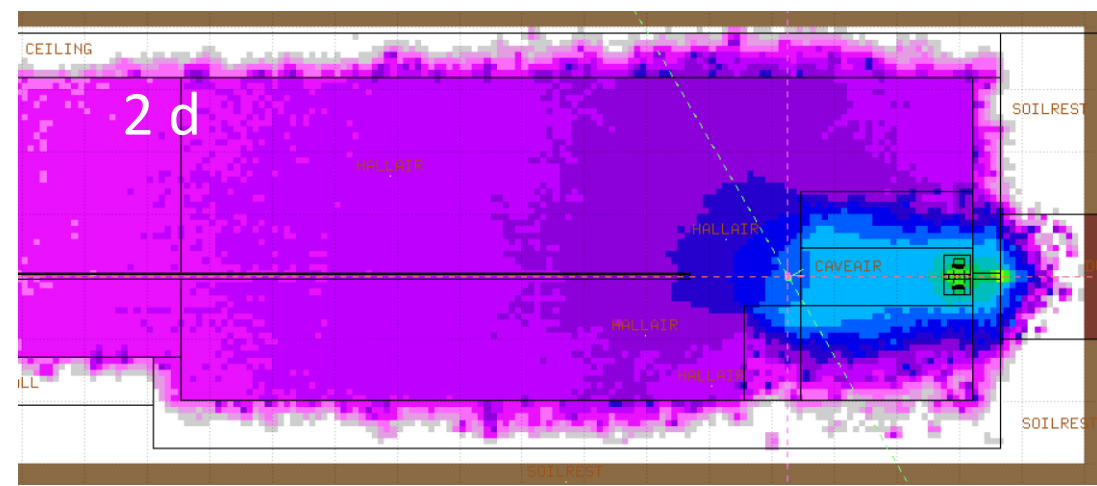
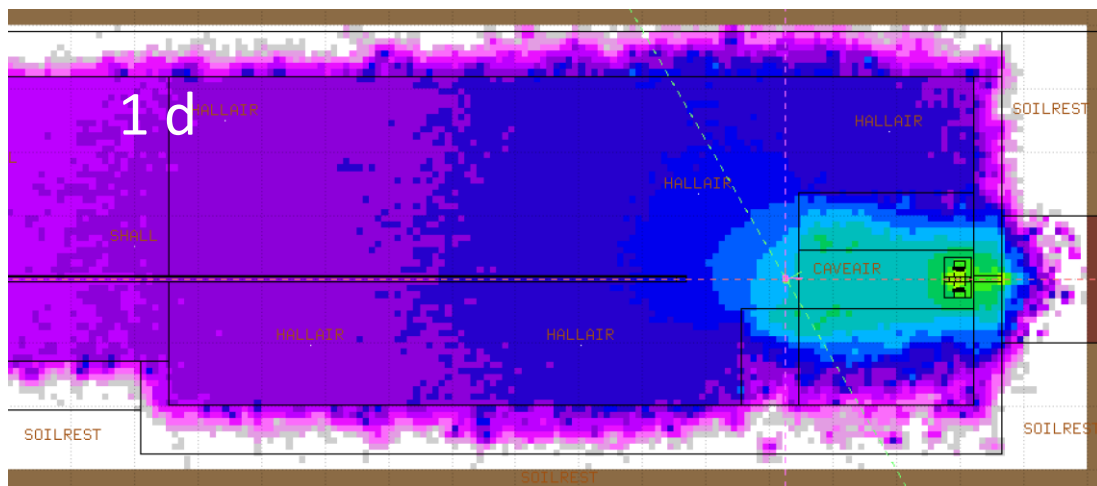
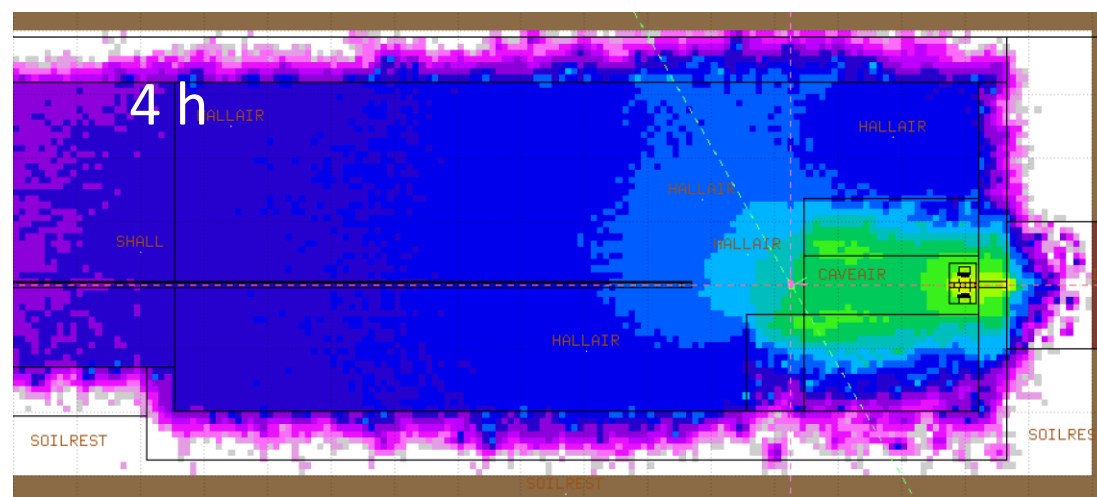
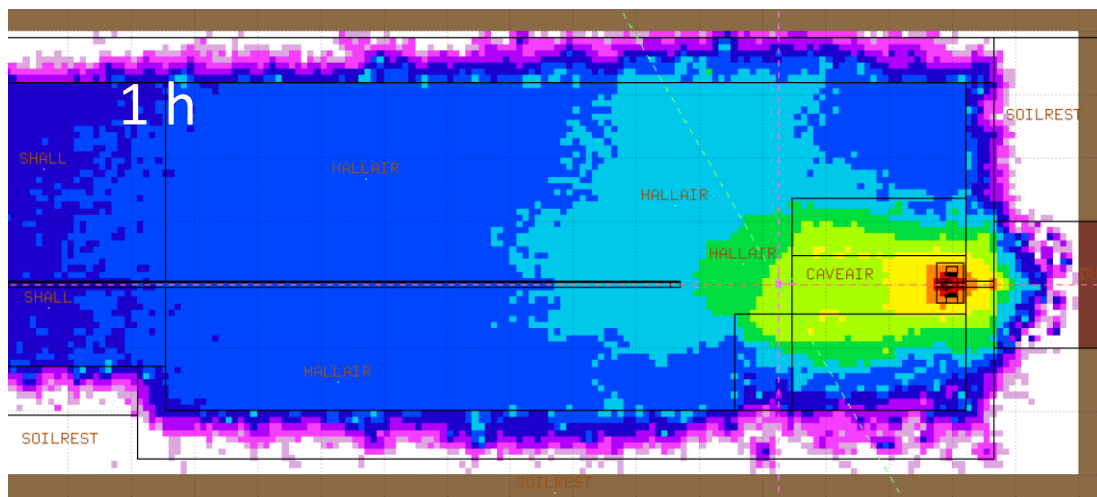
Residual doses at 30 cm from the module, mrem/h

Cooling	Upstream	Downstream	−x side	+x side	Bottom	Top	Max along	Max across
1 h	358	529	381	368	411	268	529	411
4 h	113	268	156	157	284	107	268	284
1 day	47.4	109	63.9	63.7	114	44.0	109	114
2 days	19.5	41.7	24.5	24.4	40.1	17.1	41.7	40.1
3 days	10.1	19.1	11.3	11.3	15.5	8.1	19.1	15.5
4 days	6.8	11.4	7.0	7.0	7.3	5.5	11.4	7.3
5 days	5.6	8.6	5.7	5.6	5.2	4.5	8.6	5.7
6 days	5.1	7.5	5.1	5.1	4.4	4.0	7.5	5.1
1 week	4.7	6.9	4.8	4.8	3.9	3.8	6.9	4.8

- **Cause at each cooling time:** at 1 h the dose is mostly C-11 (20-minute half-life) in the graphite. From about 4 h to 1 day it is mostly Na-24 (15 h) in the aluminum structure. By 1 week what's left is mostly Be-7 in the graphite (about 7 mrem/h), which decays slowly with a 53-day half-life.
- **For handling:** after 1 day one would still see about 100 mrem/h at 30 cm, close to the beam axis downstream or under the module. It falls to about 5–7 mrem/h after a week, and much of the drop happens between days 1 and 3 as the Na-24 decays.

☼ Residual Dose Maps: Target Module in Cave

Activated target module moved into the cave by **275 cm** from its original location.
Irradiation time 1 week; cooling times 1 hour, 4 hours, 1 day, 2 days





Residual Dose Summary: Target Module in Cave

At (0, 0, 0), mrem/h

Cooling	Module	Hall/cave	Sum
1 h	12	87	101
4 h	3.3	72	77
1 day	1.4	29	31
2 days	0.61	9.9	10.7
3 days	0.34	3.7	4.1
4 days	0.25	1.6	1.9
5 days	0.21	0.93	1.1
6 days	0.20	0.68	0.83
1 week	0.19	0.57	0.72

- **Target module moved into cave by 275 cm.** These are residual doses at its original location before moving. Activated concrete walls dominate the doses. Doses are around 10 mrem/h after 2 days of cooling.



Residual Doses for Samples Only

Aiming for ~10 mrem/h or less

Residual dose of the samples on their own, 30 cm from the outer face (mrem/h)

Material / Thickness	1 mm	2 mm	3 mm	4 mm	Sum of 4
1 h after beam-off					
Cu	1.25	2.05	2.58	2.78	8.7
A36	1.50	2.16	2.92	3.31	9.9
SS316LN	1.21	2.84	3.12	3.09	10.3
Al6061	0.78	1.10	1.09	2.01	5.0
4 h after beam-off					
Cu	0.70	1.24	1.56	1.67	5.2
A36	1.03	1.60	2.08	2.37	7.1
SS316LN	0.84	2.03	2.40	2.26	7.5
Al6061	0.65	0.92	0.92	1.67	4.2

Residual dose rates in individual samples even after 1 hour of cooling are very low, always below 10 mrem/h. Max number is around 10 mrem/h for 4 stainless steel samples.

⚙️ Target Module Optimization Examples

- Graphite vs aluminum rod
 - ☐ Aluminum foil tape to avoid contaminated graphite powder
- Graphite target
 - ☐ Material impurities
 - ☐ Power deposition and temperature assessment
 - ☐ DPA calculations
 - ☐ Be-7 production and plating
- Aluminum vs stainless for the support components



Tritium Production in Samples

Comparison of FLUKA & PHITS results led to several % difference

Tritium atoms produced per proton ($\times 10^{-6}$)

Material	1 mm	2 mm	3 mm	4 mm	Total	Rel. error
Cu	5.9	9.6	11.9	13.8	41.2	$\pm 5\%$
A36	4.8	8.8	8.9	10.1	32.6	$\pm 6\%$
SS316LN	4.5	7.3	8.9	10.4	31.1	$\pm 6\%$
Al6061	2.1	3.3	4.7	7.6	17.7	$\pm 8\%$

Tritium activity in the samples at the end of the 1-week irradiation ($\times 10^5$ pCi)

Material	1 mm	2 mm	3 mm	4 mm	Total	Rel. error
Cu	1.29	2.10	2.60	3.01	9.0	$\pm 5\%$
A36	1.05	1.92	1.94	2.21	7.1	$\pm 6\%$
SS316LN	0.98	1.59	1.94	2.27	6.8	$\pm 6\%$
Al6061	0.46	0.72	1.03	1.66	3.9	$\pm 8\%$

Local LSC experts: Good to have $1e2 - 1e4$ pCi/ml in water

Once the beam is off and the samples are immersed in water, these amounts of tritium will be slowly diffused into water over the time, depending on the diffusion coefficients for each material.

Tritium Diffusion Estimates

- Diffusion coefficient: $D = D_0 \cdot \exp(-E_a/RT)$
 - In structural metals (like stainless), $D = 2e-12 \text{ cm}^2/\text{s}$ (increases with T)
 - Drops by 2-3 orders of magnitude due to surface oxide barriers
 - For Cu, $D = 6e-10 \text{ cm}^2/\text{s}$
 - At room T, tritium diffuses ~300 times faster in Cu vs stainless steel
- **Caveat: Diffusion coefficients greatly vary across the literature!**

Diffusion Timescales

Both copper and aluminum alloy release tritium within days

$$D_T = 6e-10 \text{ cm}^2/\text{s}$$

Copper	Time in days	
Sample Thickness, mm	Time to 50% Tritium Release	Time to 90% Tritium Release
1	10	40
2	40	170
3	90	380
4	160	680

$$D_T = 4e-9 \text{ cm}^2/\text{s}$$

Al6061	Time in days	
Sample Thickness, mm	Time to 50% Tritium Release	Time to 90% Tritium Release
1	1.5	6
2	5	23
3	12	52
4	21	92

$$D_T = 2e-12 \text{ cm}^2/\text{s}$$

Stainless steel has a very low diffusion coefficient, and it takes years to release tritium

SS316	Time in years	
Sample Thickness, mm	Time to 50% Tritium Release	Time to 90% Tritium Release
1	8	34
2	31	135
3	70	303
4	125	538

On the other hand, A36 steel releases most of tritium in a few hours

$$D_T = 2e-7 \text{ cm}^2/\text{s}$$

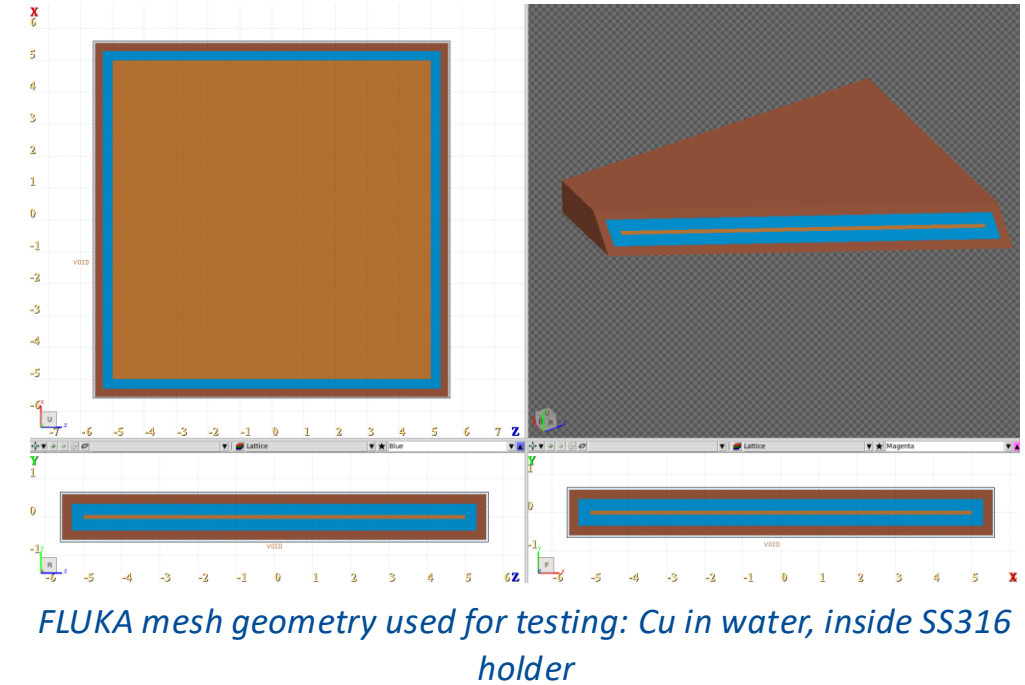
A36	Time in hours	
Sample Thickness, mm	Time to 50% Tritium Release	Time to 90% Tritium Release
1	0.8	3.5
2	3	14
3	7	31
4	13	55



Diffusion Model Builds on Radiation Analysis

Courtesy of Alajos Makovec

- **Input data taken from radiation analysis results**
 - Geometry: FLUKA Cartesian mesh, one material per bin (Cu, water, SS316 holder, air)
 - Tritium in Cu: 1.3×10^{-5} ^3H /primary (between 3 mm and 4 mm plate), 7.5×10^{11} pps, 4 h $\rightarrow C_0 = 1.4 \times 10^{10}$ atoms/cm³
- **3D finite-volume diffusion solver**
 - Bulk diffusion + interface transfer (Cu–water, SS–water, SS–air)
 - Tracks water flushes, air loss, decay; checks mass balance
- **Output: tritium in water (pCi/ml) vs time, 20°C & 50°C**





Diffusion Model: Release and Flush Analysis

Courtesy of Alajos Makovec

- **Simulated Cu sample with periodic water flushes**

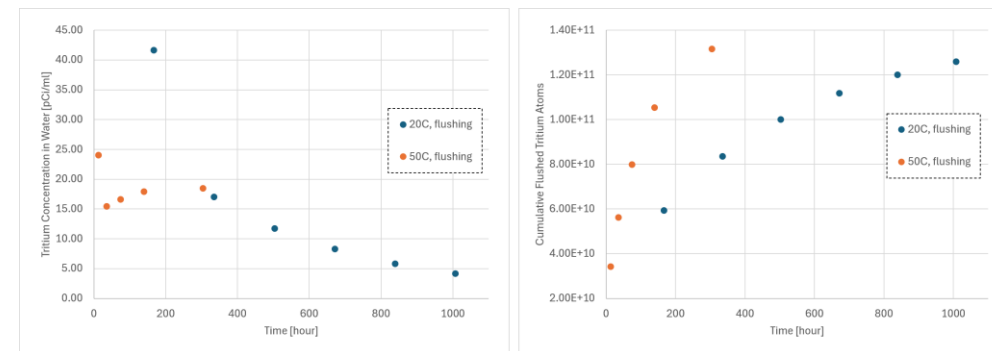
- $D_{\text{Cu}} = 5.8\text{e-}10 \text{ cm}^2/\text{s}$ (20°C), consistent with our estimate (6e-10)
- Predicted water activity: ~5–40 pCi/ml per flush sample
- 50°C ($D \times 4$): more released in < 2 weeks than at 20°C in ~6 weeks

- **Flush analysis, validated on simulation**

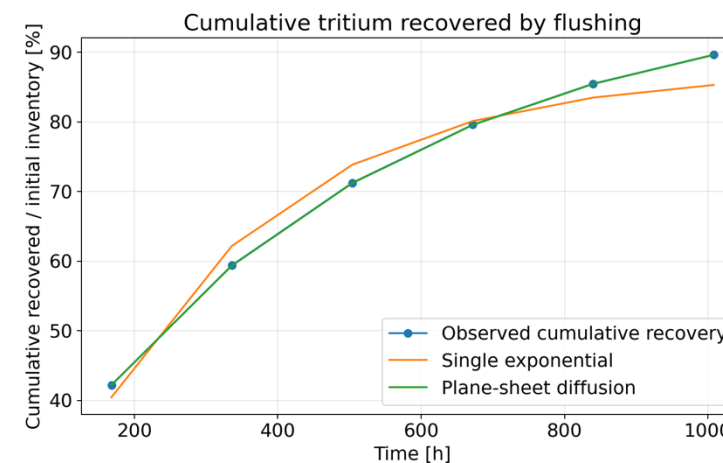
- Plane-sheet diffusion fit recovers $D_{\text{eff}} = 5.84\text{e-}10$ (20°C) and $2.37\text{e-}9 \text{ cm}^2/\text{s}$ (50°C); $R^2 > 0.997$
- 50% / 90% release: 234 h / 1009 h (20°C), 58 h / 249 h (50°C)

- **1–4 mm samples, as in radiation analysis**

- Release time:
 - $\propto L^2$ if bulk diffusion;
 - $\propto L$ if interface-limited



Simulated flush samples: 20°C vs 50°C



Analysis of 20°C simulation: model fits

Summary

This work addresses radiation transport simulations coupled with planned experimental measurements. The Monte Carlo codes CERN-FLUKA and PHITS are being used to model and optimize an experimental configuration in which selected materials are exposed to secondary particles produced by the interaction of a proton beam with a graphite target.

After irradiation, the materials will be immersed in water for a defined period, and the resulting tritium concentration in the water will be measured. The measurements will provide experimental estimates of tritium release from irradiated materials and enable evaluation and improvement of models describing tritium transport in these materials.

The combined computational and experimental approach will contribute to improved tritium management and safety assessments in particle accelerator facilities.

 **Thank You!**



Questions?

Comments?

Backup Slides

⚙️ Target Module Optimization (Target Cylinder)

- Comparison of residual dose rates for Al6061 vs graphite target cylinder (residual dose rates in mrem/h, at 30 cm from target)

Target	1 h	4 h	1 day	3 days	1 week	1 month
1-day irradiation						
Aluminum	1,580	1,190	436	51	4.3	2.3
Graphite	678	2.7	1.2	1.16	1.1	0.8
1-week irradiation						
Aluminum	2,220	1,750	674	98	24.5	15.9
Graphite	685	9.6	8.0	7.8	7.4	5.5

- Graphite could produce irradiated powder:
graphite rod will be wrapped in 3M™ Aluminum Foil Tape 425
(included in the FLUKA model)

Target Module Optimization

- Graphite impurity effects on the residual doses

Residual dose increase over pure graphite, 1-week irradiation (target alone, 30 cm)

Graphite	1 h	4 h	1 day	3 days	1 week
A. Unpurified (ash ~800 ppm)	0.2%	8.8%	7.5%	5.6%	3.9%
B. Thermally purified	0.0%	1.2%	0.9%	0.6%	0.4%
C. Nuclear grade (ash < 10 ppm)	0.0%	0.1%	0.1%	0.1%	0.0%
Pure graphite, mrem/h	714	10.8	9.1	8.9	8.4

- ☐ **Purified graphite** (thermally or halogen-purified): impurities add at most about 1%, so they don't need to be modeled.
- ☐ **Unpurified graphite**: a few percent difference; at most about 9% around 4 hours, falling to about 4% at 1 week.
- ☐ Iron and nickel are the elements that matter most.

Power Deposition into Target Module

Power deposited during the irradiation (7.5×10^{11} p/s, 400 MeV, 48 W beam)

Component	Power, W	Rel. error
Graphite target	24.1	0.1%
Tape (Al foil + acrylic adhesive)	0.042	0.3%
Samples Cu (4)	0.062	1.5%
Samples A36 (4)	0.059	1.4%
Samples SS316LN (4)	0.059	1.1%
Samples Al6061 (4)	0.026	1.3%
All 16 samples	0.21	
Aluminium structure	1.27	0.3%
Whole module	25.6	

- Beam power: 48 W (7.5×10^{11} p/s $\times$ 400 MeV).
- Graphite target absorbs half the beam power; most of the rest leaves the module in the forward direction. Individual samples get 3–19 mW.

Component Heating, DPA

- Target temperature might reach 77 ° C in the worst case.
 - Graphite oxidation in air begins around 400–450 ° C; tape adhesive: 3M 425 is rated –54 ° C to 149 ° C.
- Temperature not an issue.
- Radiation dose into tape adhesive: ~1.5 MGy in a week. Inspect after irradiation.
 - Sample heating: negligible. At most 19 mW per sample; the samples stay at room temperature.
 - Aluminum structure: no concern. 1.3 W spread over the frame, plates and holders is negligible.
 - Displacement damage in graphite is negligible.

Displacements per atom in the graphite after the 1-week run ($\times 10^{-6}$ dpa)

Region	DPA-SCO	DPA-NRT
Peak (beam axis, first 5–10 cm)	8	14
Beam core ($r < 3$ mm)	6	9
Whole-rod average	1.7	2.6

DPA orders of magnitude below the damage levels at which graphite would noticeably change its dimensions or thermal conductivity.

Be-7 Production

Be-7 production (1-week irradiation at 7.5×10^{11} p/s; activity at the end of the irradiation)

Where	Be-7 per proton	Bq	pCi	Share
Graphite target	4.2×10^{-2}	2.7×10^9	7.4×10^{10}	85%
Hall and cave concrete	7.3×10^{-3}	4.8×10^8	1.3×10^{10}	15%
Air (cave + hall + inside the module)	9.0×10^{-5}	5.9×10^6	1.6×10^8	0.2%
Aluminium structure	6.8×10^{-5}	4.5×10^6	1.2×10^8	0.14%
Tape (adhesive + foil)	1.7×10^{-5}	1.1×10^6	3.0×10^7	0.03%
All 16 samples	2.6×10^{-6} ($\pm 20\%$)	1.7×10^5	4.6×10^6	0.01%
Total	4.9×10^{-2}	3.2×10^9	8.7×10^{10}	

- Be-7 has a 53-day half-life, so these activities change little over the cooling times we've been using. The Be-7 in the graphite is also what keeps the rod at about 7 mrem/h at 30 cm after a week.

⚙ Be-7 Production

- **Be-7 made in the air: about $6e6$ Bq (0.2% of the total).** This is the plate-out source. Not accounting for ventilation → all of it ends up on surfaces.
 - **Be-7 recoiling out of the graphite: none reaches the outside.** Recoils stop in the tape (thicker than the recoil range of $30\text{ }\mu\text{m}$) → Wrapping the rod contains its Be-7.
 - **Recoils from exposed surfaces (outer foil, plates, samples):** Negligible.
- Almost all Be-7 stays locked inside the graphite and the concrete. What can plate out is the $6e6$ Bq made in the air, about 0.2% of the total, mostly in the cave.
- The samples hold about $1.7e5$ Bq of Be-7 in all, roughly $1e4$ Bq each. That is comparable to their tritium (about $1e5$ Bq total), so it's worth keeping in mind for the water-leaching measurements. At room temperature Be-7 stays fixed inside the metal and doesn't diffuse like tritium, so only Be-7 near the sample surfaces could get into the water. The 478 keV gammas also make it easy to identify if it does show up.



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