Gadolinium in Water Cherenkov Detectors

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2nd EU Workshop on Water Cherenkov Experiments for Precision Physics (WCD-2025)

Kraków, Poland September 17, 2025

Water Cherenkov Technology: Some Points to Remember

1) Gammas are not seen directly. They must Compton scatter off electrons, and the resulting Cherenkov light is seen.

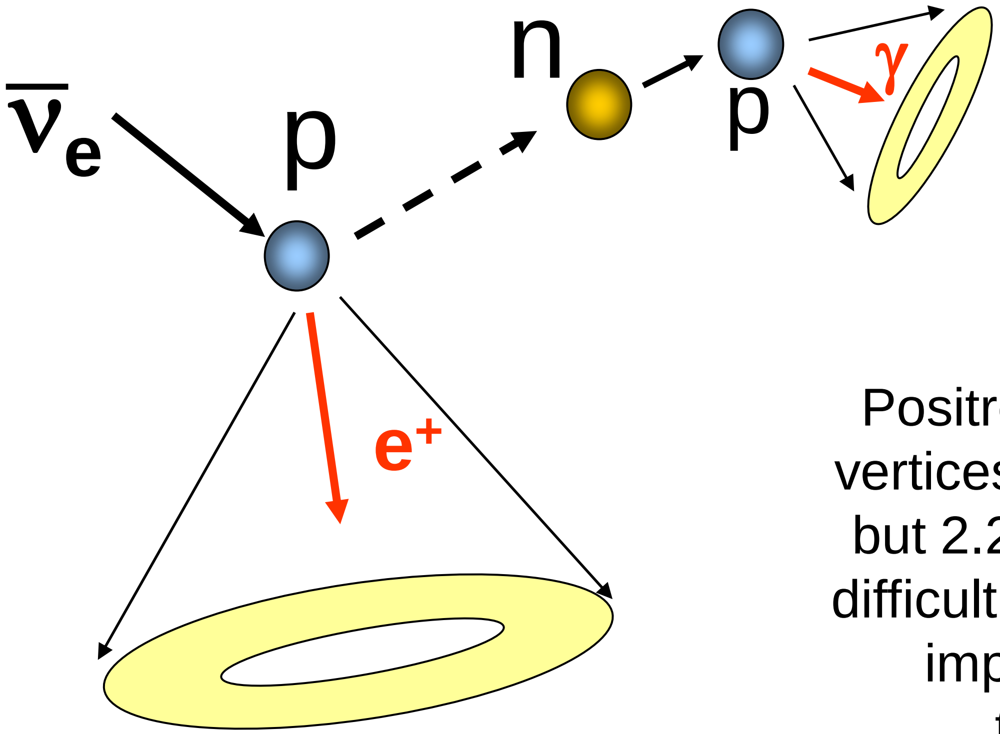
These detectors typically record about six photoelectrons per MeV of kinetic energy above Cherenkov threshold imparted to the electrons.

2) It follows from #1 that all gammas below about 1 MeV are invisible in water.

3) This means positron annihilation photons cannot be seen.

4) Above 1 MeV, the cross section for inverse beta decay [IBD] is two orders of magnitude greater than that for elastic scattering, so IBD is how low energy antineutrinos usually interact.

Inverse Beta Decay



Positron and gamma ray vertices are within ~50 cm, but 2.2 MeV gammas are difficult to detect and, more importantly, difficult to distinguish from backgrounds.

All of the events in the traditional Super-Kamiokande low energy (i.e., below 100 MeV) analyses are <u>singles</u> in time and space.

And this rate is actually very low…

<u>just three ν-like events per cubic meter per *year.*</u>

"Everyone complains about the (supernova neutrino) weather,
but no one *does* anything about it…"

So, after one of the sessions at Neutrino 2002 in Munich, theorist John Beacom and I spent hours sitting in a subway station, brainstorming ideas. But we did NOT start with gadolinium!

Date: Tue, 30 Jul 2002 03:45:09 -0700 (PDT)
From: Mark Vagins <vagins@danka.ps.uci.edu>
To: John Beacom <beacom@fnal.gov>
Subject: The briny, briny deep

Hey John,

I just spoke with Yoichiro Suzuki, who is also attending this meeting in Amsterdam. I brought up our scheme for making SK 1% salty.

He liked the idea a lot, and in fact said that salting SK was one of the future options he had been musing about. Naturally, he did say that we needed to carefully model a salty detector and get a feel for the true numbers.

He went on to say that the necessary water system modifications were possible, and that in the near future

"we must do something to get the new physics."

He also felt that 500 tons of salt was reasonable, saying to me,

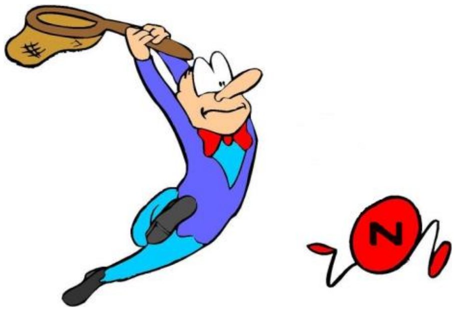
"It's just 50 truckloads - you can shovel it yourself!"

This is a very positive thing indeed (other than the part about my shoveling half a kiloton of salt myself).

So, things sound pretty promising on this end. I wonder if it's worth putting out a phenomenological paper outlining how this will-salt-for-relics could work... it probably is, especially if there is a non-zero likelihood that SK will actually do something about it!

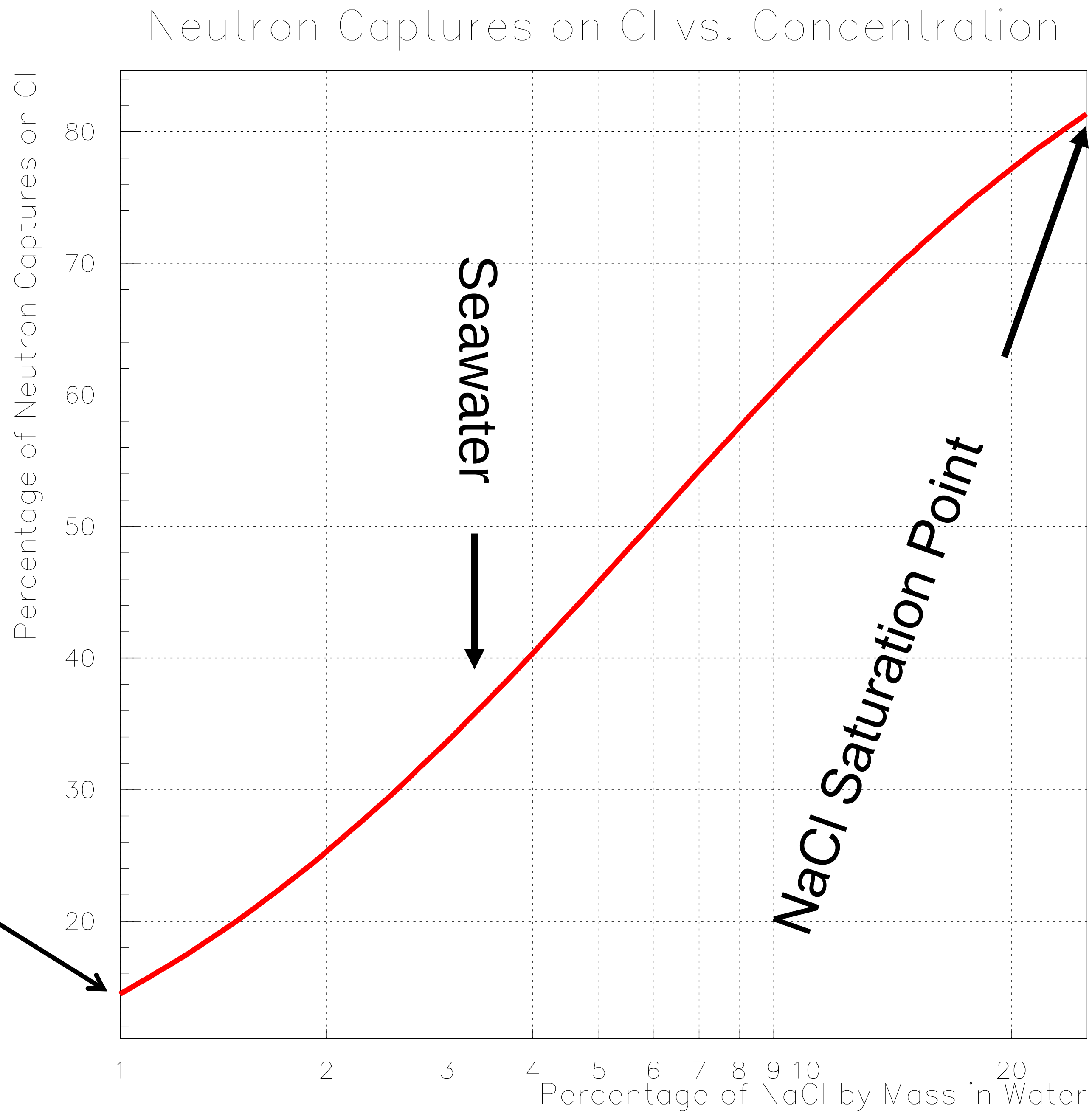
We must do something to get the new physics,
-Mark

Why is so much regular salt needed? Because in light water (unlike in D_2O) we're going to have to compete with hydrogen in capturing the neutrons.



Consequently, plain old NaCl simply won't work well in SK…

We'd need to add 3 kilotons of salt to SK just to get 50% of the neutrons to capture on the chlorine!



500 tons of NaCl in 50 ktons of (light) water → ~14% capture on Cl

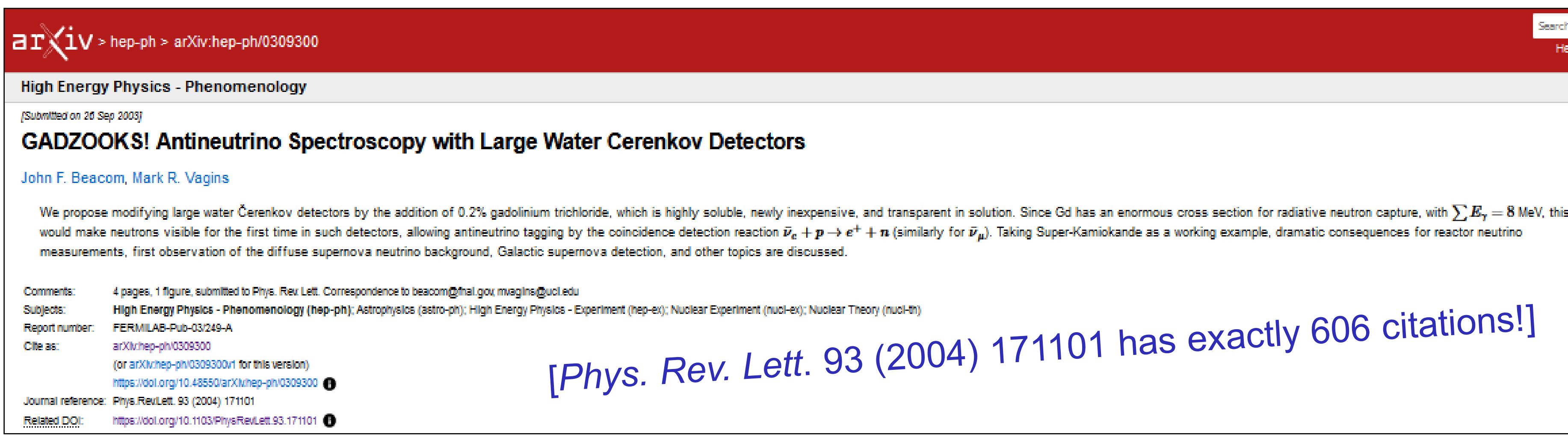
We eventually turned to the best neutron capture nucleus known – gadolinium.

גדוליניום

"Gadol" = Great!

- $GdCl_3$ and $Gd_2(SO_4)_3$, unlike metallic Gd, are highly water soluble
- Neutron capture on Gd emits a 8.0 MeV γ cascade
- 100 tons of $GdCl_3$ or $Gd_2(SO_4)_3$ in SK (0.2% by mass) would yield >90% neutron captures on Gd
- Plus, they are easy to handle and store.

As everyone now knows, we did indeed eventually put out that "phenomenological paper."



arXiv > hep-ph > arXiv:hep-ph/0309300

Search... Help

High Energy Physics - Phenomenology

[Submitted on 26 Sep 2003]

GADZOOKS! Antineutrino Spectroscopy with Large Water Cerenkov Detectors

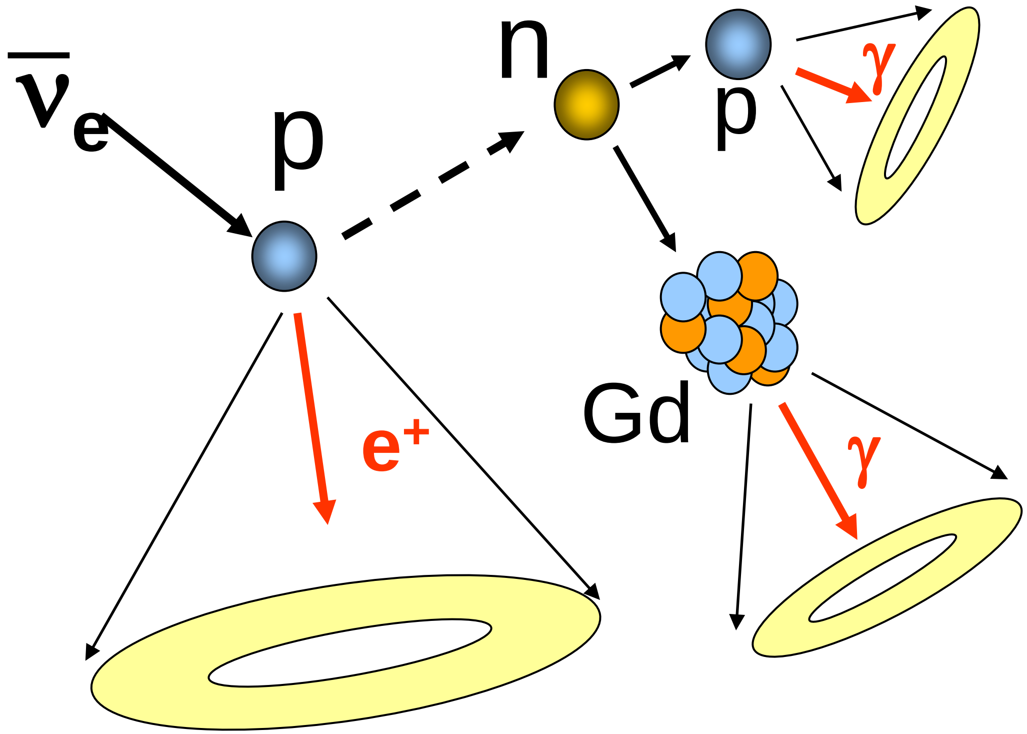
John F. Beacom, Mark R. Vagins

We propose modifying large water Čerenkov detectors by the addition of 0.2% gadolinium trichloride, which is highly soluble, newly inexpensive, and transparent in solution. Since Gd has an enormous cross section for radiative neutron capture, with $\sum E_\gamma = 8$ MeV, this would make neutrons visible for the first time in such detectors, allowing antineutrino tagging by the coincidence detection reaction $\bar{\nu}_e + p \to e^+ + n$ (similarly for $\bar{\nu}_\mu$). Taking Super-Kamiokande as a working example, dramatic consequences for reactor neutrino measurements, first observation of the diffuse supernova neutrino background, Galactic supernova detection, and other topics are discussed.

Comments:	4 pages, 1 figure, submitted to Phys. Rev. Lett. Correspondence to beacom@fnal.gov, mvagins@uci.edu
Subjects:	**High Energy Physics - Phenomenology (hep-ph)**; Astrophysics (astro-ph); High Energy Physics - Experiment (hep-ex); Nuclear Experiment (nucl-ex); Nuclear Theory (nucl-th)
Report number:	FERMILAB-Pub-03/249-A
Cite as:	arXiv:hep-ph/0309300
	(or arXiv:hep-ph/0309300v1 for this version)
	https://doi.org/10.48550/arXiv.hep-ph/0309300
Journal reference:	Phys.Rev.Lett. 93 (2004) 171101
Related DOI:	https://doi.org/10.1103/PhysRevLett.93.171101

[*Phys. Rev. Lett.* 93 (2004) 171101 has exactly 606 citations!]

In addition to first introducing the term "DSNB", basically we said, "Let's add 0.1% gadolinium - using a water soluble gadolinium compound - to Super-K!"



Positron and gamma ray vertices within ~50 cm.

Possibility 1: 10% or less

n+p→d + γ

2.2 MeV γ-ray

Possibility 2: 90% or more

n+Gd →~8 MeV γ

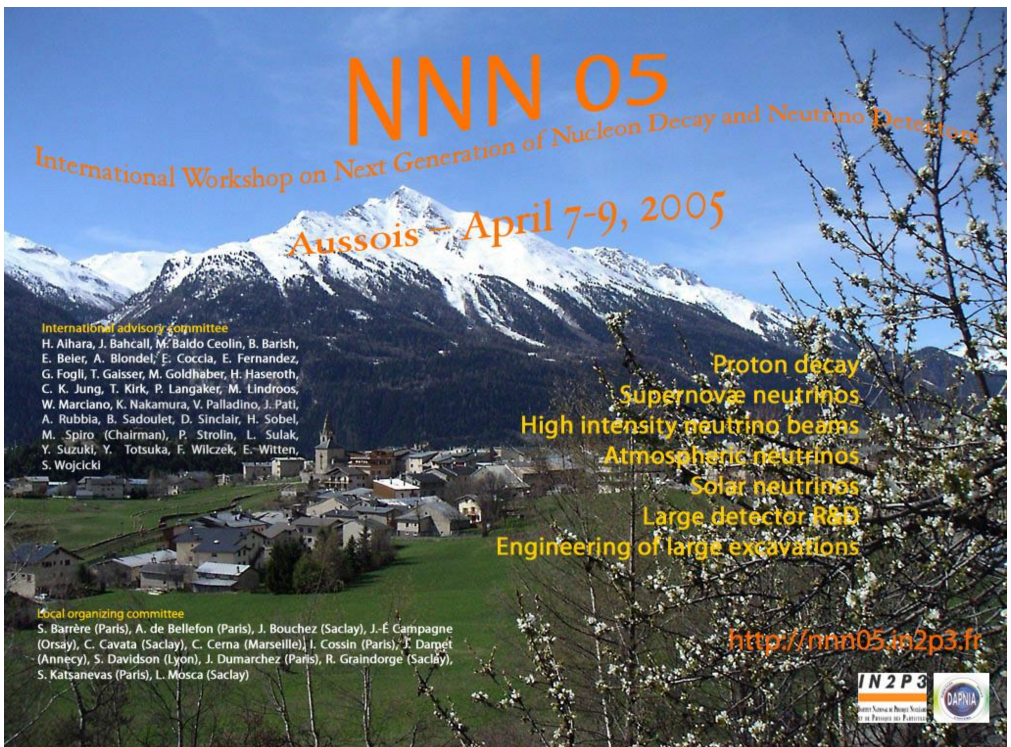
ΔT = ~30 μsec

$\bar{\nu}_e$ can be individually identified by delayed coincidence: "Gd heartbeat"

→ n-tags greatly reduce (10^{-4}) backgrounds to DSNB, etc

Our GADZOOKS! proposal quickly starting getting a lot of attention, especially in the theory community:

At NNN05, <u>before I had even given my talk</u>, John Ellis suddenly stood up and demanded of the senior SK people in attendance:



Why haven't you guys put gadolinium in Super-K yet?

As I told him, studies are under way…

Now, John Beacom and I never wanted to merely propose a new WC technique – we wanted to make it work!

[Snowbird photo by A. Kusenko]

Suggesting a major modification of one of the world's leading neutrino detectors is indeed <u>not</u> the easiest route…

…so began many years of experimental and theoretical studies.

To make Gd loading work, we must:

Dissolve a gadolinium salt in the water

→ Easy and fast (if right Gd compound is selected)

Remove the gadolinium efficiently and completely when desired

→ Straightforward but currently cumbersome
(need about six times as much cation exchange resin
as compared to the mass of the dissolved Gd compound)

Keep pure water transparent, yet retain gadolinium in solution

→ The tricky part; need a selective Gd filtration system

Be sure that the Gd compound is low in radioactive impurities

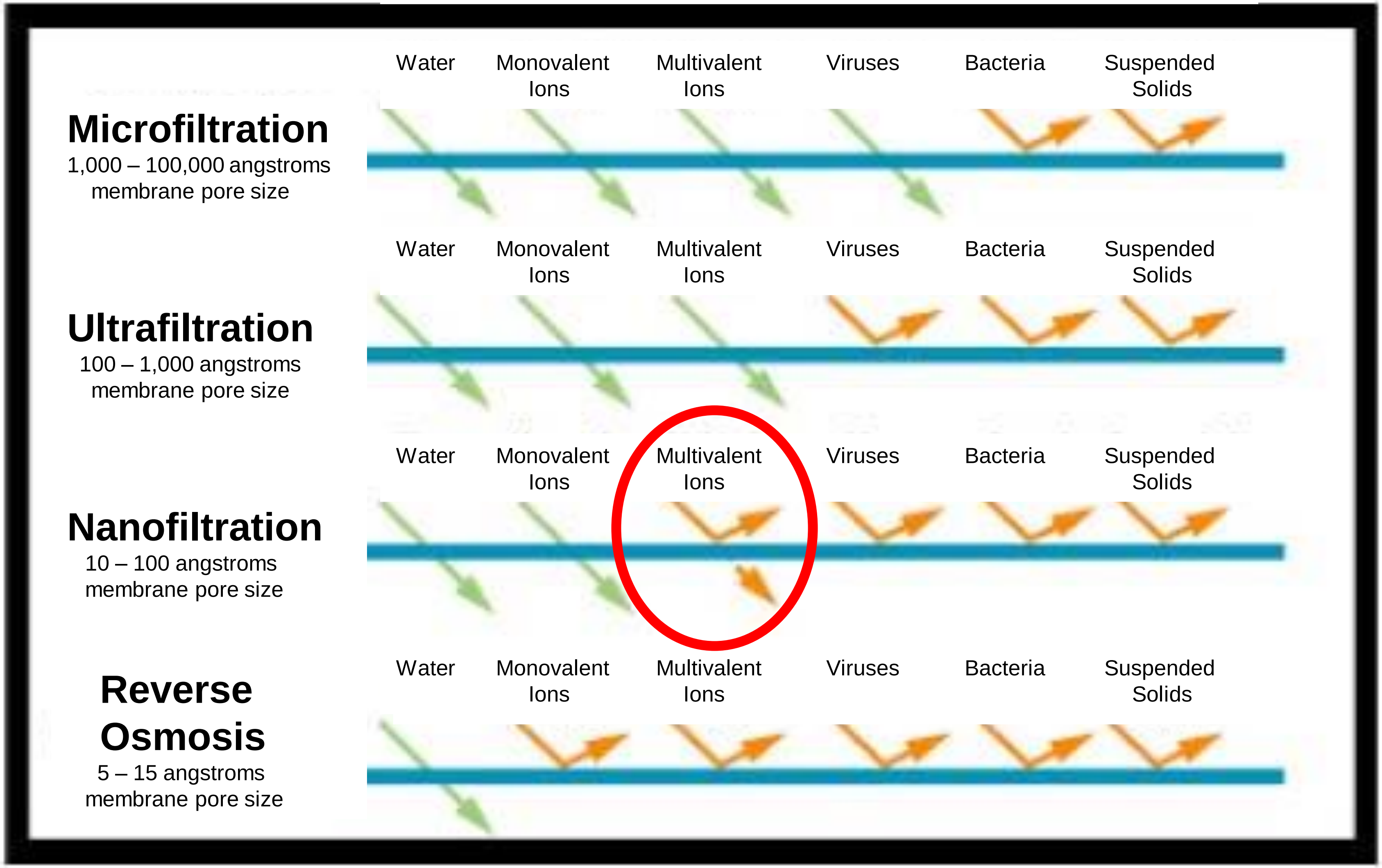
→ Another challenge; partnership with supplier(s) needed

To select the best gadolinium compound we must balance optical and mechanical effects:

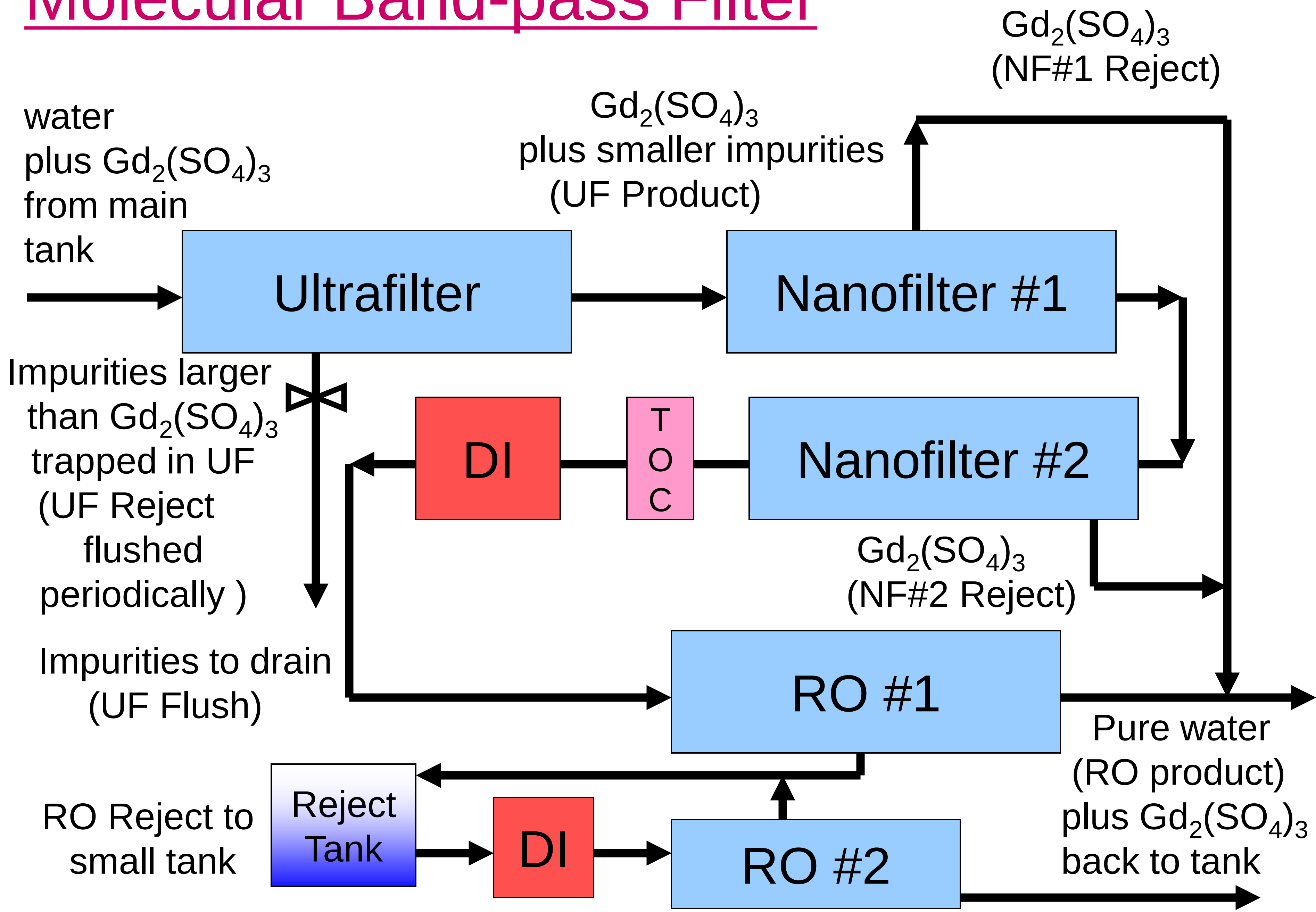
Name	Formula	Pros	Cons
Gadolinium Chloride	$GdCl_3$	Low Cost High Solubility Safety Transparency	Corrosion
Gadolinium Nitrate	$Gd(NO_3)_3$	Low Cost High Solubility Low Corrosion	Absorbs UV
Gadolinium Sulfate	$Gd_2(SO_4)_3$	Transparency Low Corrosion	Low pH Lower Solubility

Membrane-based Filtering Technologies

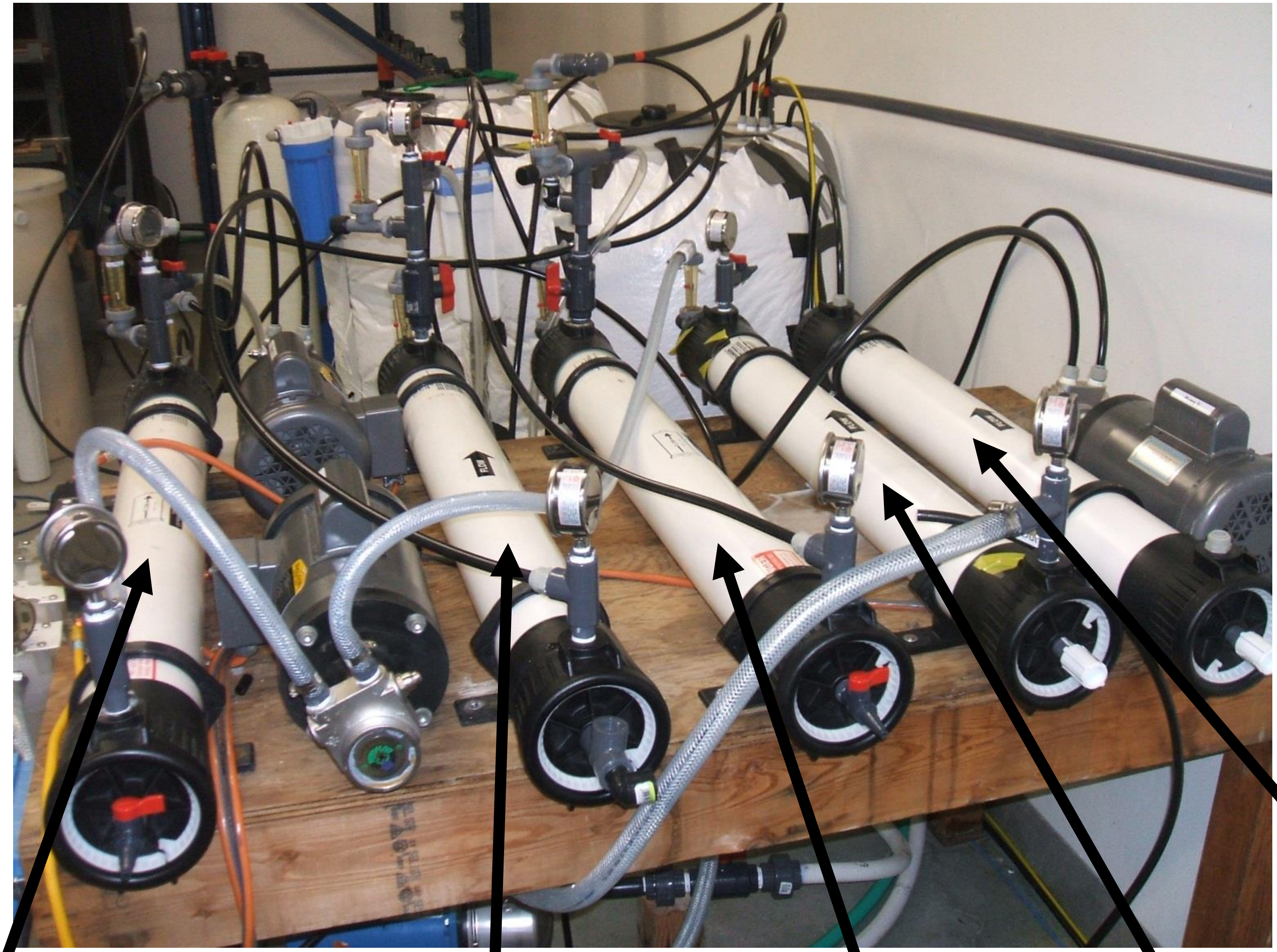
$$Gd_2(SO_4)_3 \rightarrow 2\ Gd^{3+} + 3\ (SO_4)^{2-}$$



Molecular Band-pass Filter



Prototype Selective Filtration Setup @ UCI



Membrane Pre-Flush

Nanofilter #1

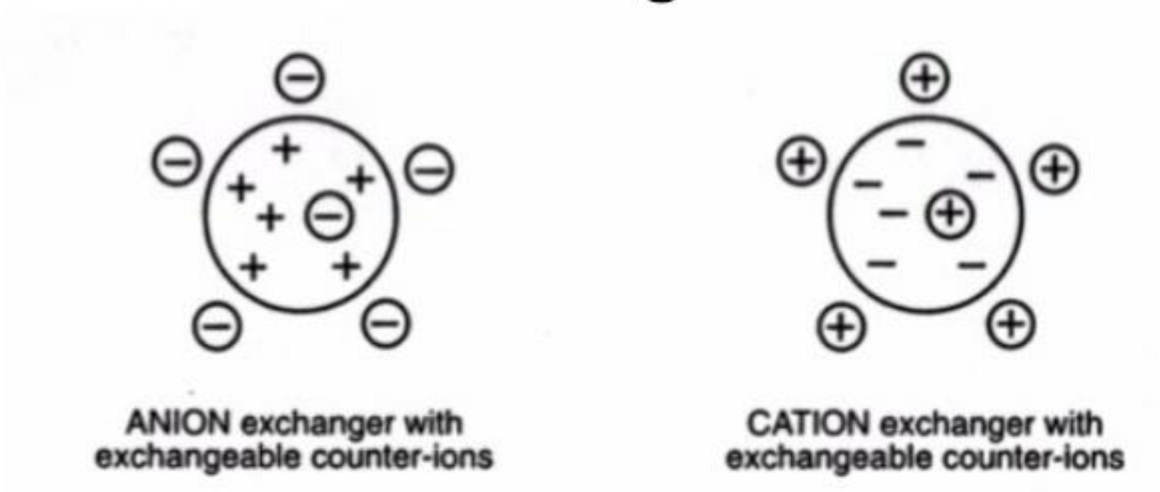
Nanofilter #2

Reverse Osmosis

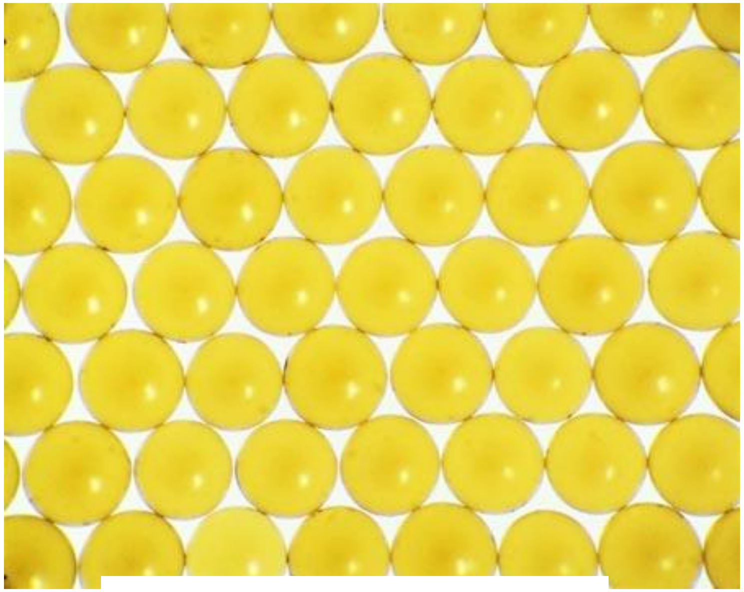
Ultrafilter

Resin-based Filtering Technologies

Ion Exchange Resins

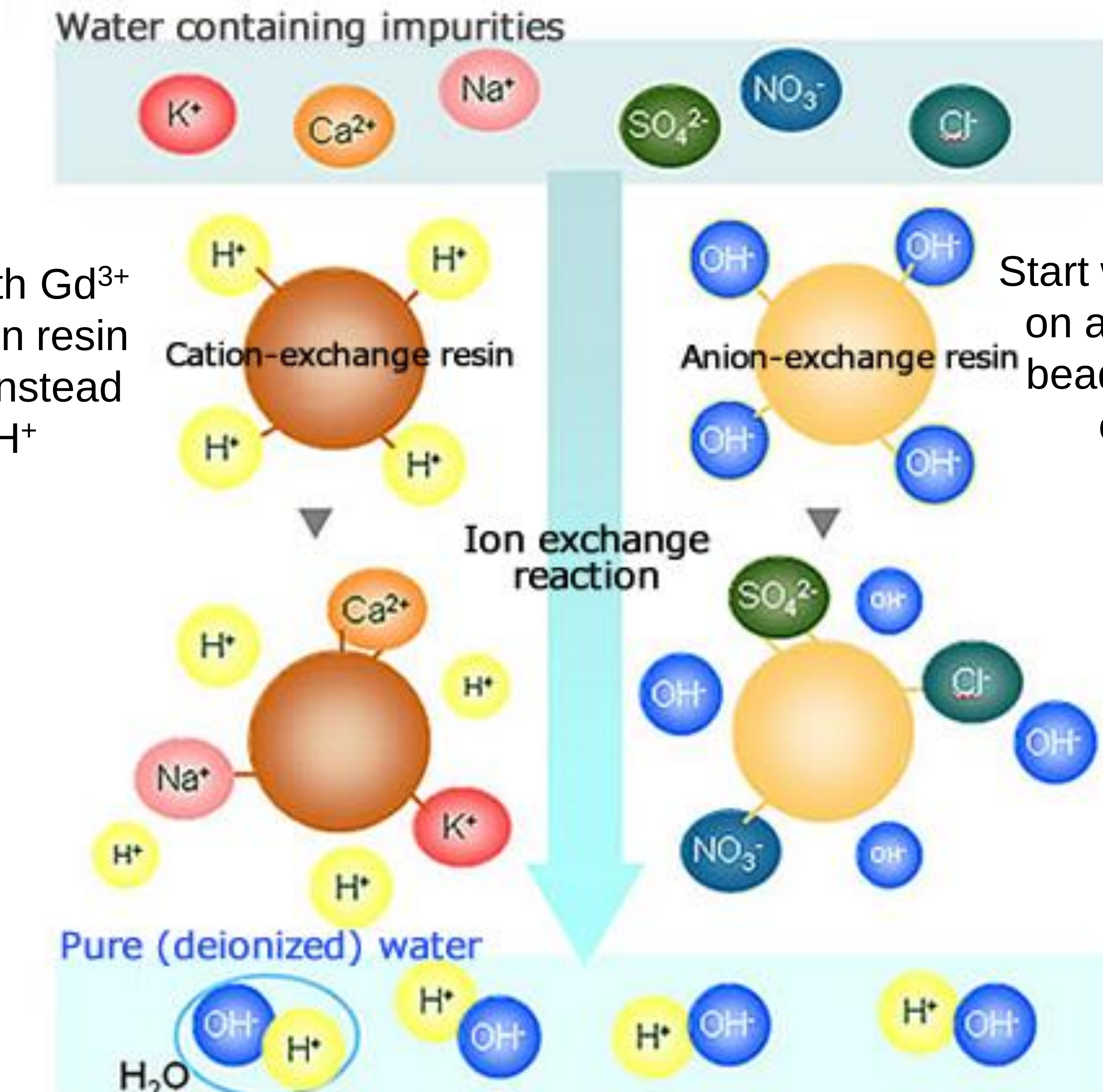


Ion exchange resin



Individual resin beads

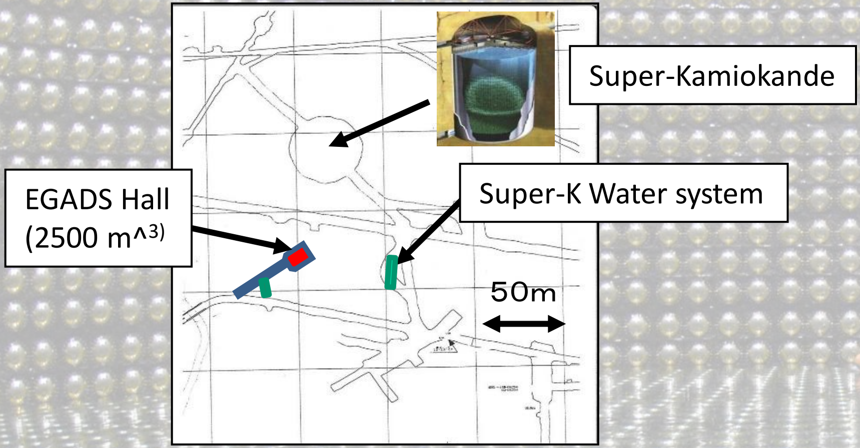
Start with Gd^{3+} on cation resin beads instead of H^+



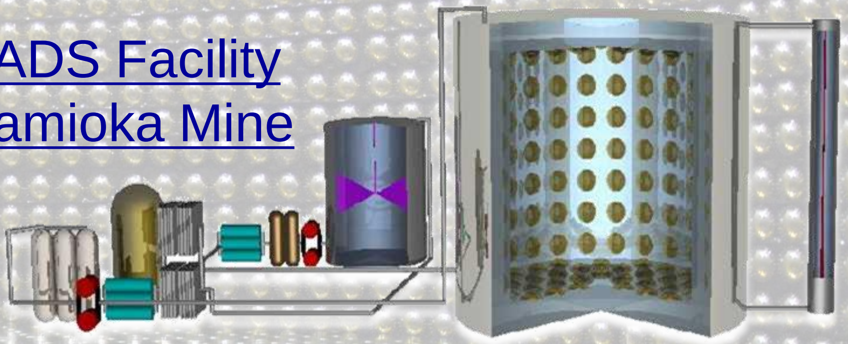
Start with $(SO_4)^{2-}$ on anion resin beads instead of OH^-

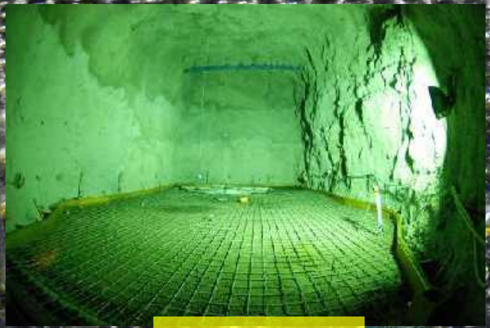
EGADS, the Gd-in-Water Pathfinder

IPMU/ICRR built **EGADS** (Evaluating Gadolinium's Action on Detector Systems), as a dedicated Gd testbed which includes a working 200-ton scale model of SK.



EGADS Facility
in Kamioka Mine

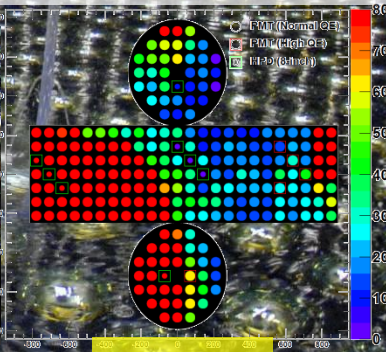




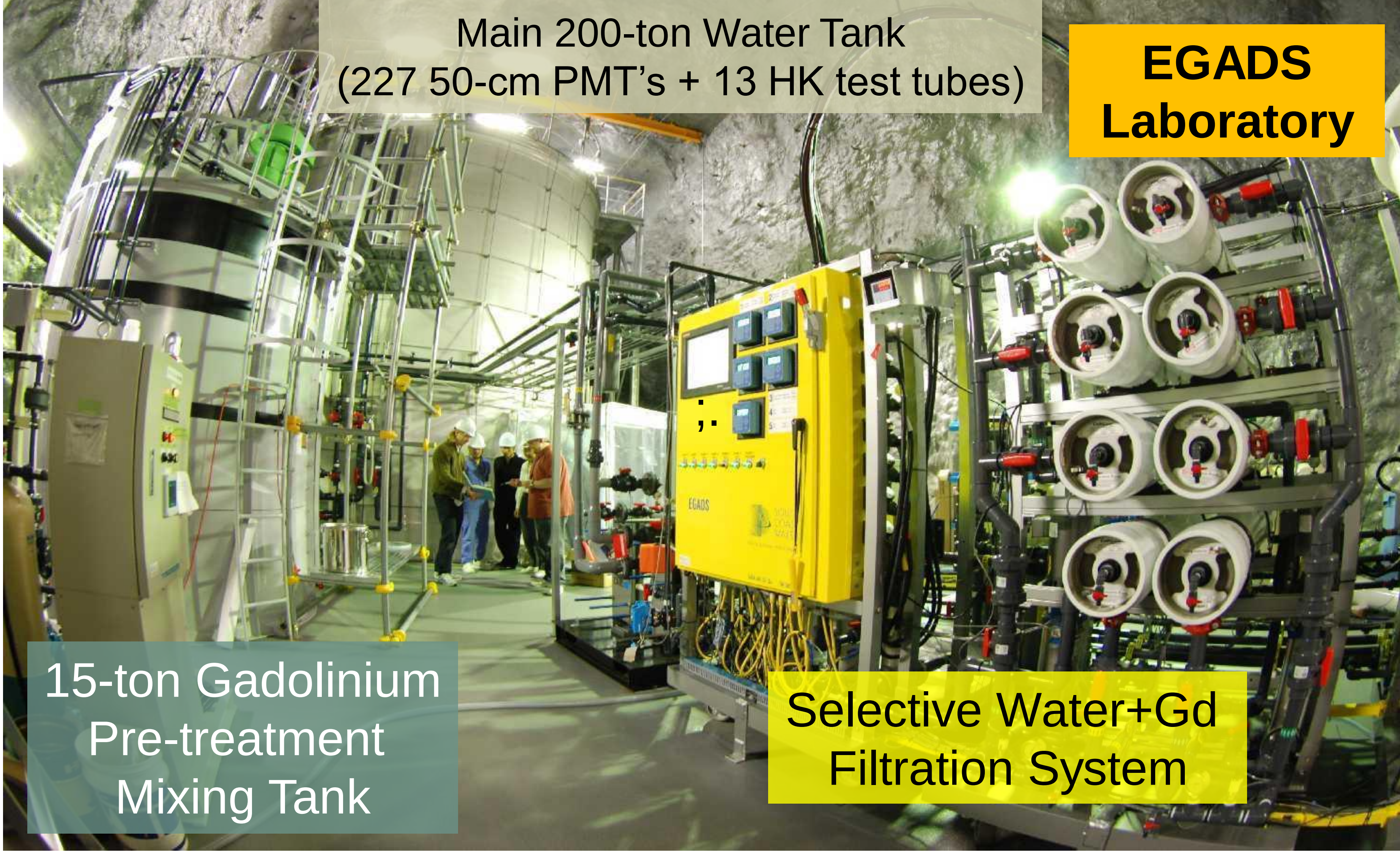
12/2009

11/2011

8/2013



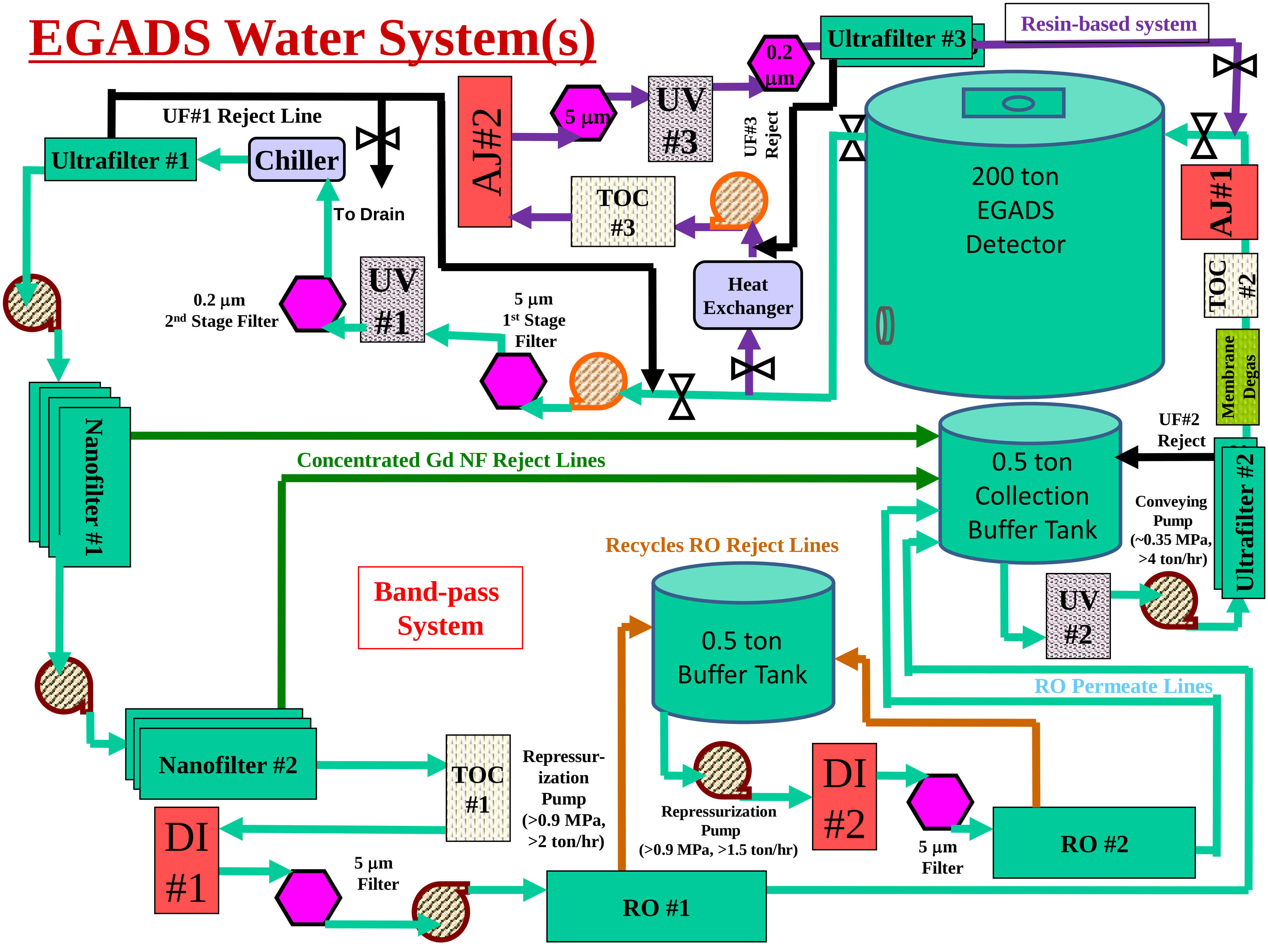
6/2015



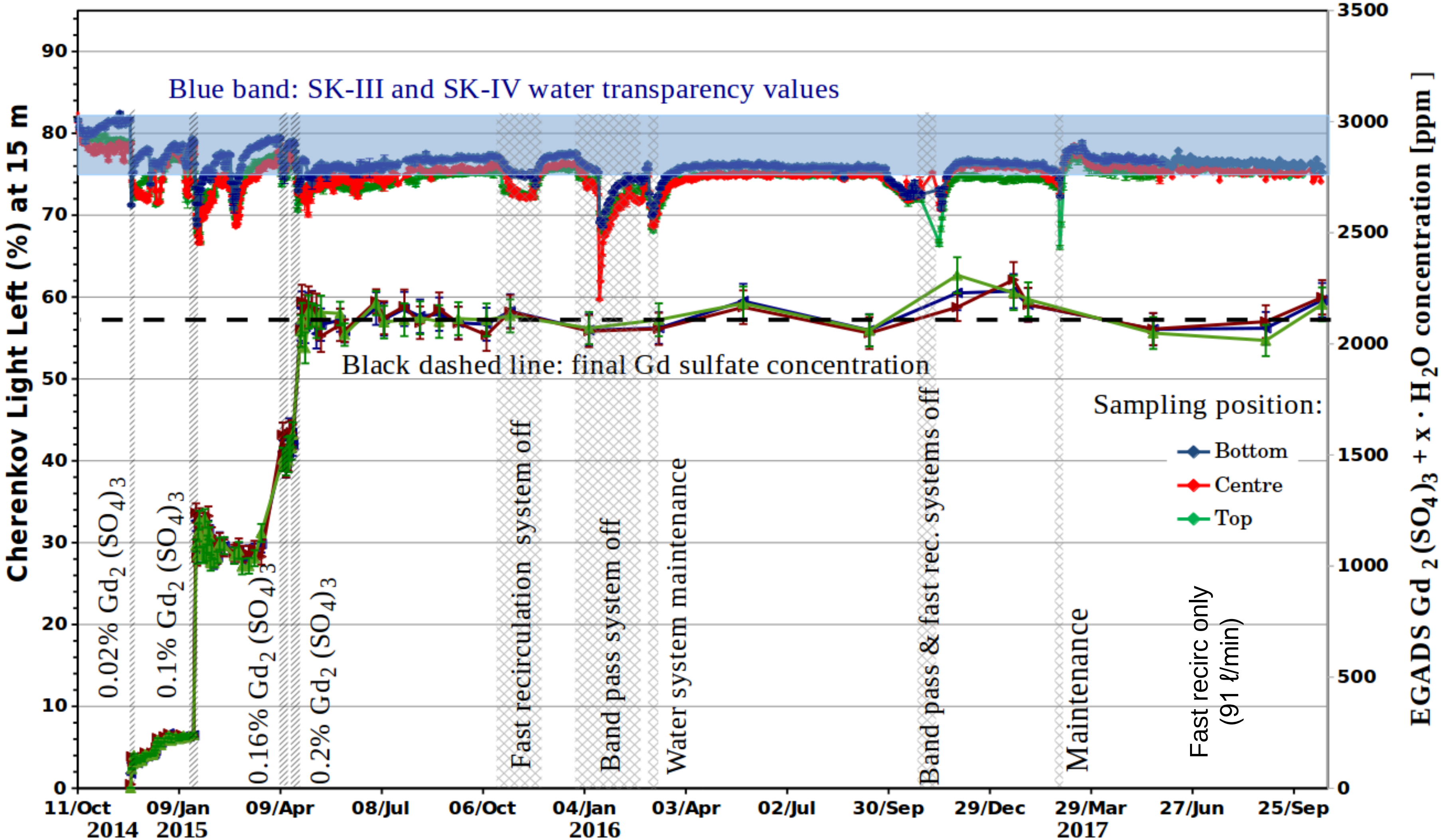
Well over $10,000,000 (36M zł) - not counting salaries - has been spent developing and proving the viability of the Gd-in-water concept.

[“Evaluation of gadolinium’s action on water Cherenkov detector systems with EGADS,” Ll. Marti et al., *Nucl.Instrum.Meth.* **A959** 163549 (2020)]

EGADS Water System(s)



Light @ 15 meters and Gd conc. in the 200-ton EGADS tank



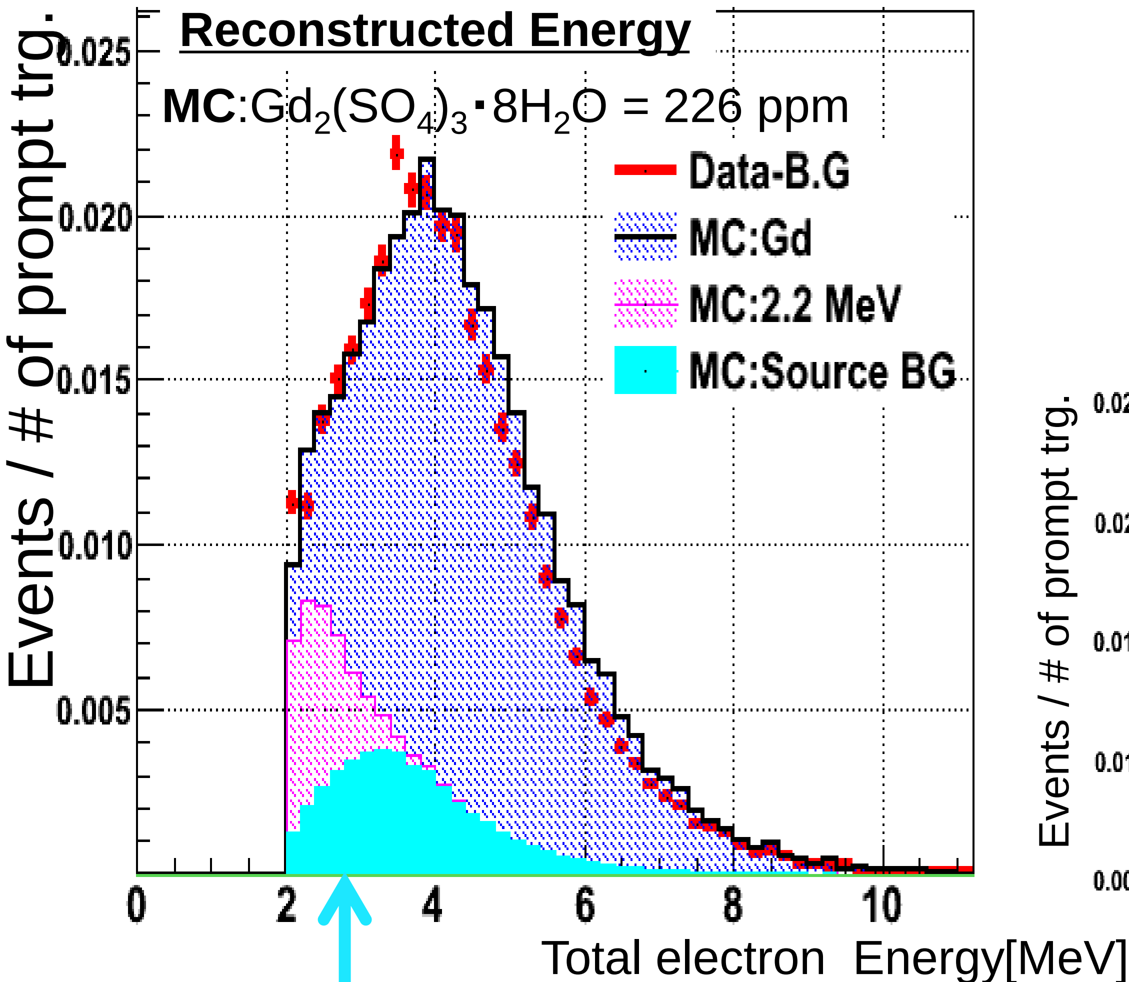
After two and a half years at full (0.2%) Gd loading, during stable operations EGADS water transparency remained within the SK ultrapure range.

→ No detectable loss of Gd after more than 650 complete turnovers. ←

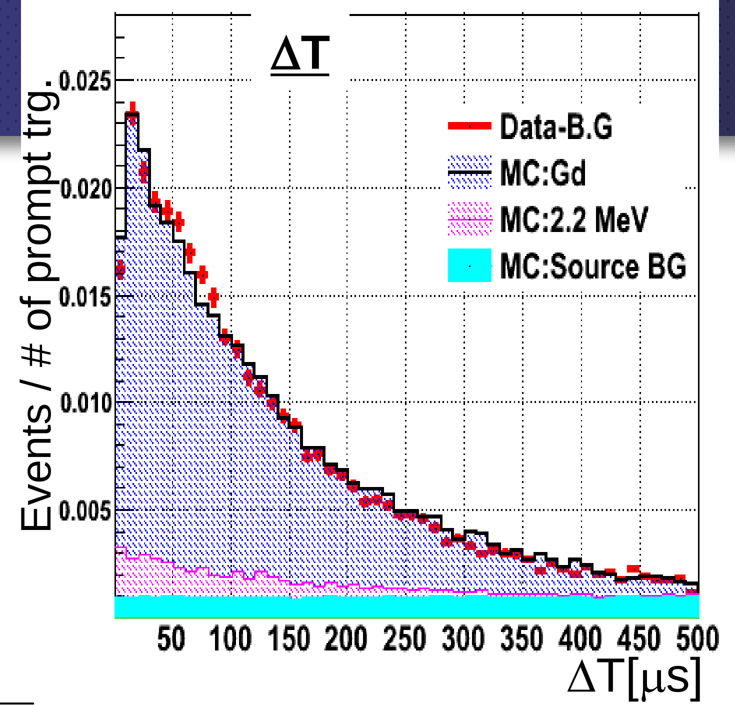
Gd Signals in EGADS

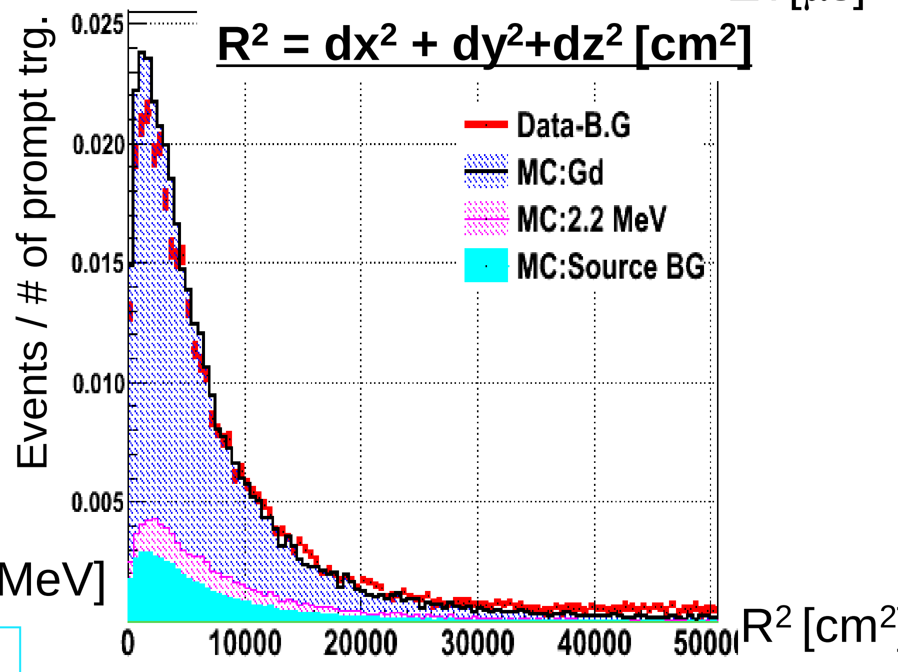
Am/Be source

$\alpha + {}^{9}\text{Be} \rightarrow {}^{12}\text{C}^{*} + n \;\; ; \;\; {}^{12}\text{C}^{*} \rightarrow {}^{12}\text{C} + \gamma$



Assuming 120 Bq Am/Be source intensity





Developing special low-background $Gd_2(SO_4)_3$

Radioactive chain	Part of the chain	mBq/kg
^{238}U	^{238}U	50
	^{226}Ra	5
^{232}Th	^{228}Ra	10
	^{228}Th	100
^{235}U	^{235}U	32
	^{227}Ac / ^{227}Th	300

Radio isotopes in "typical" off-the-shelf gadolinium sulfate

Radioactive chain	Part of the chain	SRN (mBq/kg)	Solar ν (mBq/kg)
^{238}U	^{238}U	< 5	-
	^{226}Ra	-	< 0.5
^{232}Th	^{228}Ra	-	< 0.05
	^{228}Th	-	< 0.05
^{235}U	^{235}U	-	< 3
	^{227}Ac / ^{227}Th	-	< 3

We need from 1-4 orders of magnitude reduction in RI

↑

What the physics requires

Low background Ge counters at Canfranc underground laboratory (Spain)

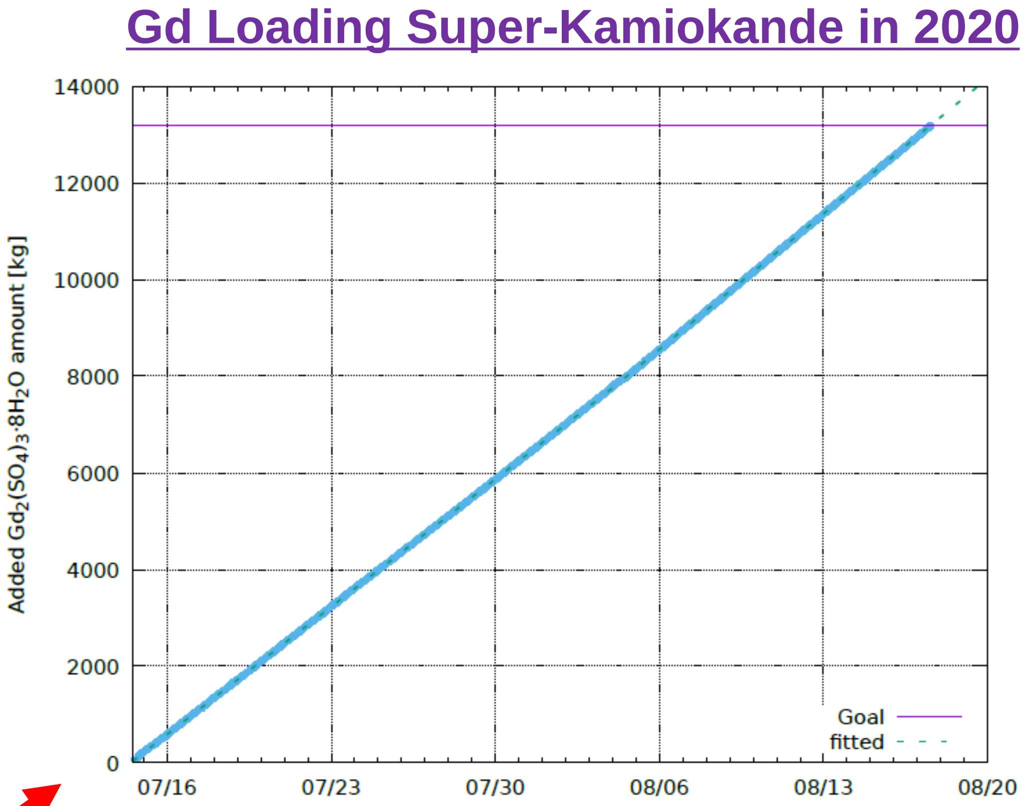
“Development of ultra-pure gadolinium sulfate for the Super-Kamiokande gadolinium project”,
K. Hosokawa et al., *PTEP* **2023** 1, 013H01 (2023)

Kamioka Ge counter (Japan)

Boulby Underground Germanium Suite (UK)

Loading Super-Kamiokande with Gadolinium (First Step)

After nearly 20 years of R&D, planning, and preparation, culminating with a major detector refurbishment in 2018/9, Super-K was finally loaded with 0.01% gadolinium (meaning 13.2 tons of $Gd_2(SO_4)_3 {*} 8H_2O$) in July/August 2020 → SK-VI



"First Gadolinium Loading to Super-Kamiokande", Super-K Collaboration, *Nuclear Inst. And Methods in Physics Research*, **A 1027** (2022) 166248

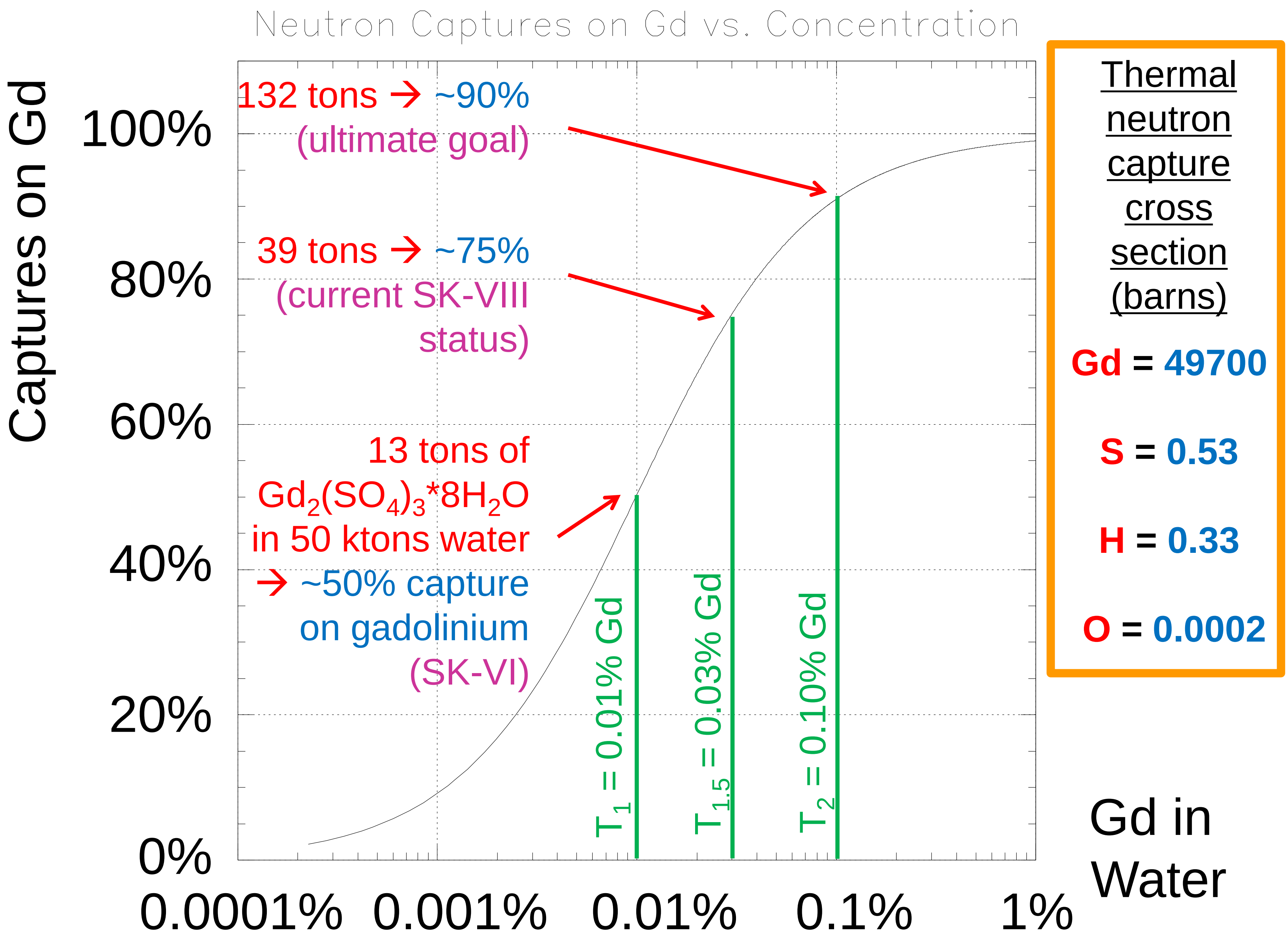
Loading Super-Kamiokande with Gadolinium (Next Step)

To bring the loading up to 0.03% Gd, in the summer of 2022 we added another 26 tons of $Gd_2(SO_4)_3$*$8H_2O$ to Super-K → SK-VII

These two lots of gadolinium sulfate for SK, at 13 and 26 tons, are by far the largest orders of gadolinium in human history… and ultra-pure, too!

About 75% of the neutrons in SK are now being (visibly) captured on Gd.

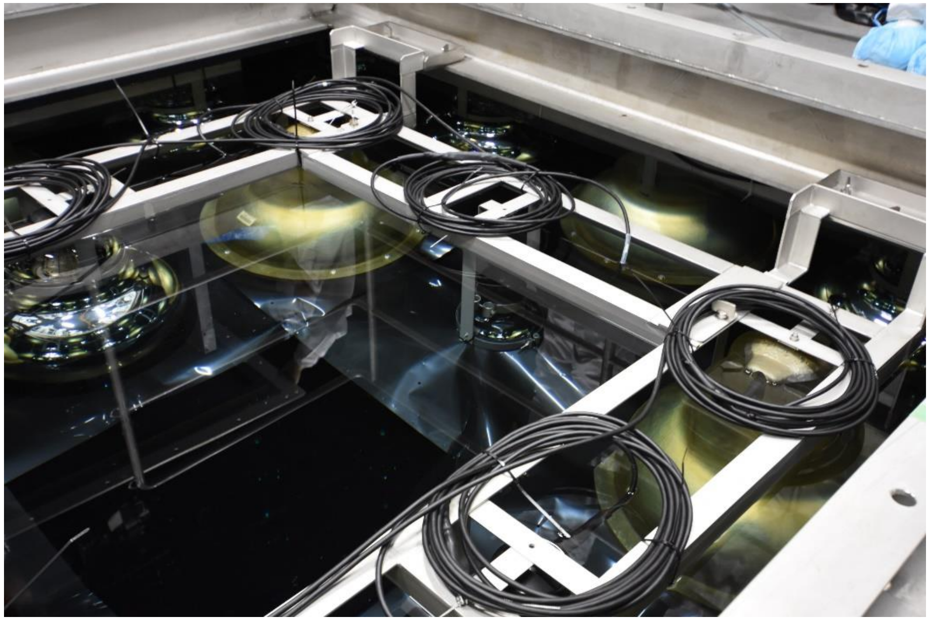
"Second Gadolinium Loading to Super-Kamiokande", Super-K Collaboration, *Nuclear Inst. And Methods in Physics Research*, **A 1065** (2024) 169480



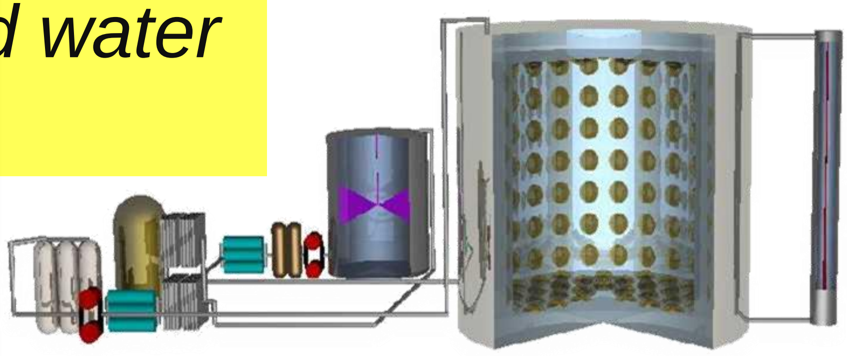
Neutron Captures on Gd vs. Concentration
Captures on Gd
100%
80%
60%
40%
20%
0%
0.0001%
0.001%
0.01%
0.1%
1%
Gd in Water
132 tons → ~90%
(ultimate goal)
39 tons → ~75%
(current SK-VIII status)
13 tons of $Gd_2(SO_4)_3*8H_2O$ in 50 ktons water → ~50% capture on gadolinium (SK-VI)
T_1 = 0.01% Gd
$T_{1.5}$ = 0.03% Gd
T_2 = 0.10% Gd
Thermal neutron capture cross section (barns)
Gd = 49700
S = 0.53
H = 0.33
O = 0.0002

Evaluating Gadolinium's Action on Detector Systems (EGADS) @ Kamioka

[Gd R&D, galactic supernova watch]



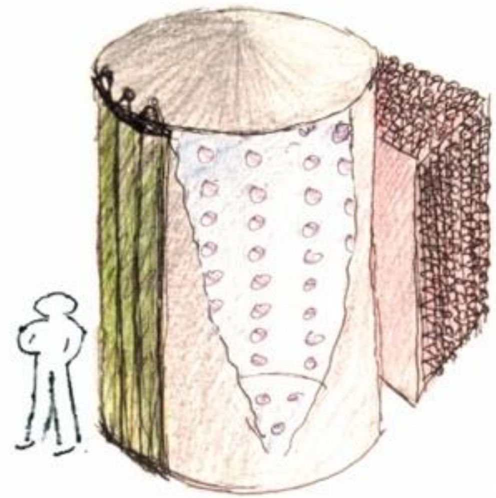
200 tons of $Gd_2(SO_4)_3$-loaded water operating since 2013

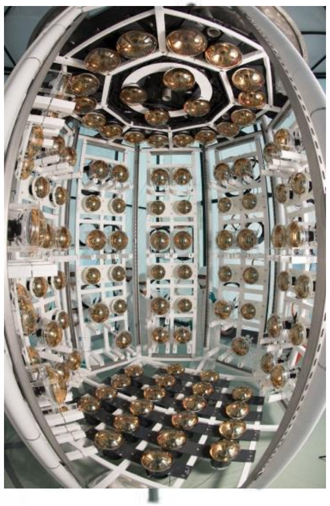


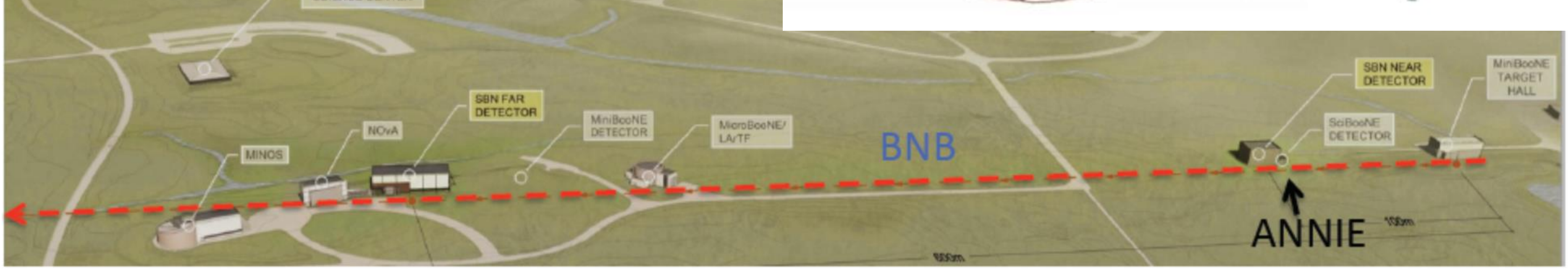
Accelerator Neutrino Neutron Interaction Experiment (ANNIE) @ Fermilab

[ATM-like ν interactions]

[see Amanda Weinstein's talk]







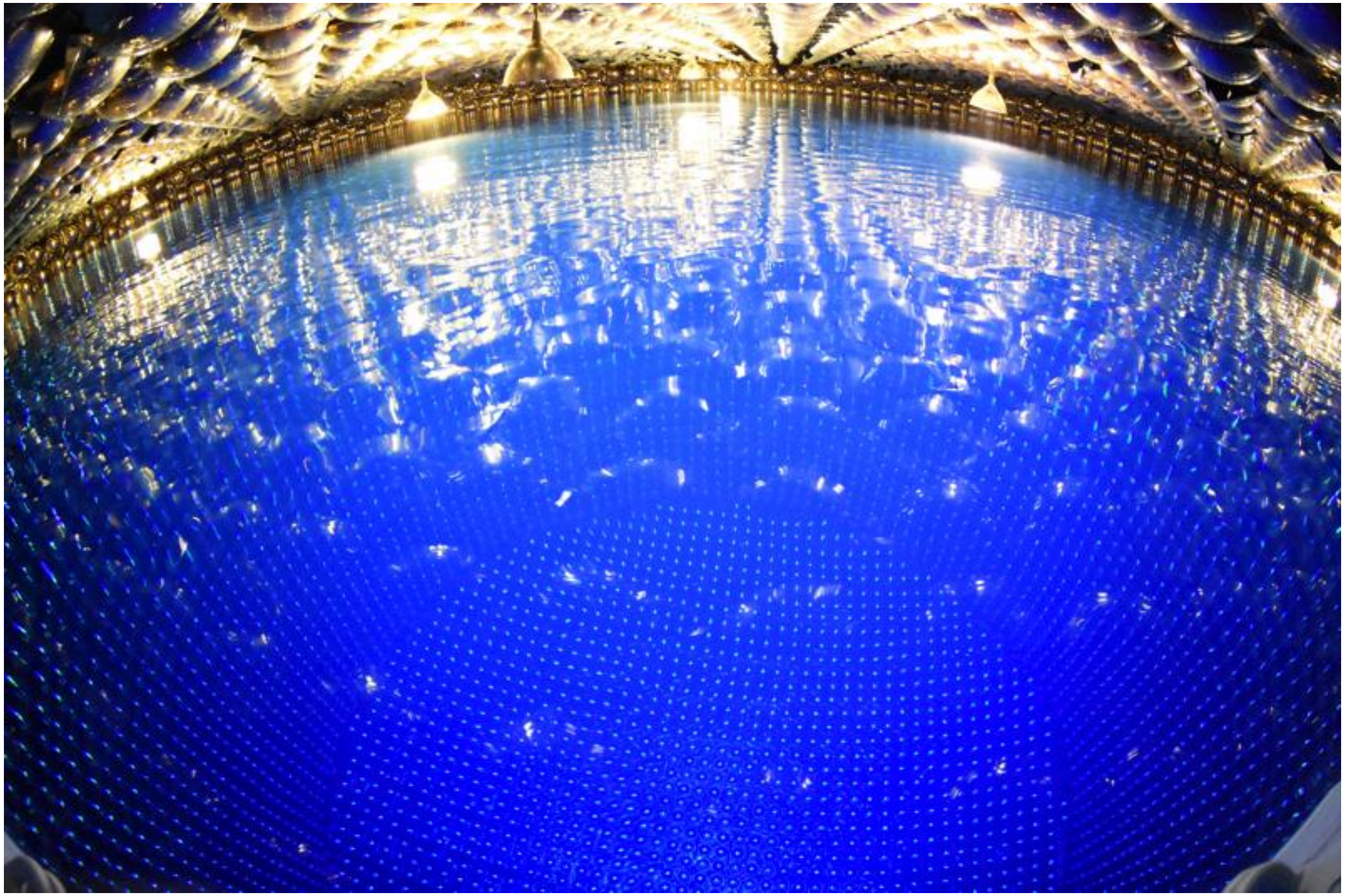
Main target = 26 tons of $Gd_2(SO_4)_3$-loaded water

Fully Gd-loaded in 2019, and taking beam as of 2020

Super-Kamiokande with Gadolinium (SK-Gd) @ Kamioka

[ν astrophysics, ν oscillations, PDK, etc]

[see Hiroyuki Sekiya's talk]



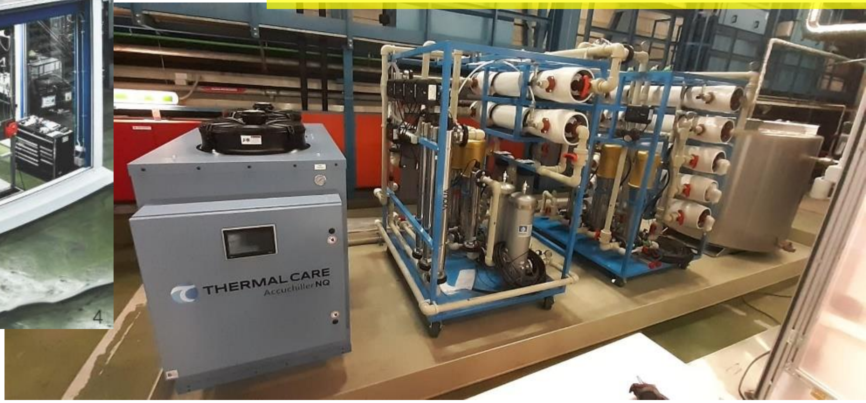
50000 tons of $Gd_2(SO_4)_3$-loaded water

Initial Gd-loading completed on August 17th, 2020

XENONnT @ Gran Sasso

[dark matter search, SN]

Veto shield's Gd water system's arrival; June 2021

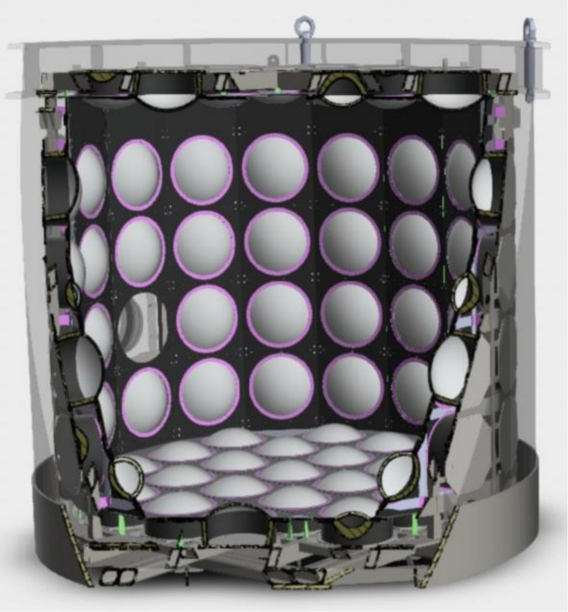


Neutron veto = 700 tons of $Gd_2(SO_4)_3$-loaded water

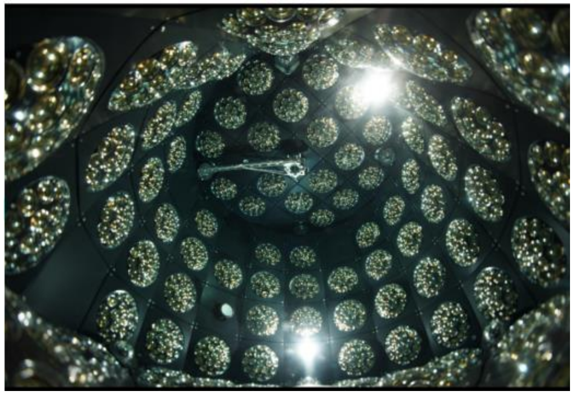
Gd-loaded since 2023

Water Cherenkov Test Experiment [WCTE] @ CERN

[detector/analysis R&D in a test beam]



WCTE
3.6 m tall
3.8 m diameter
~100 mPMT modules
41 tons total volume
2 t/hr flow



[see Lauren Anthony's talk]

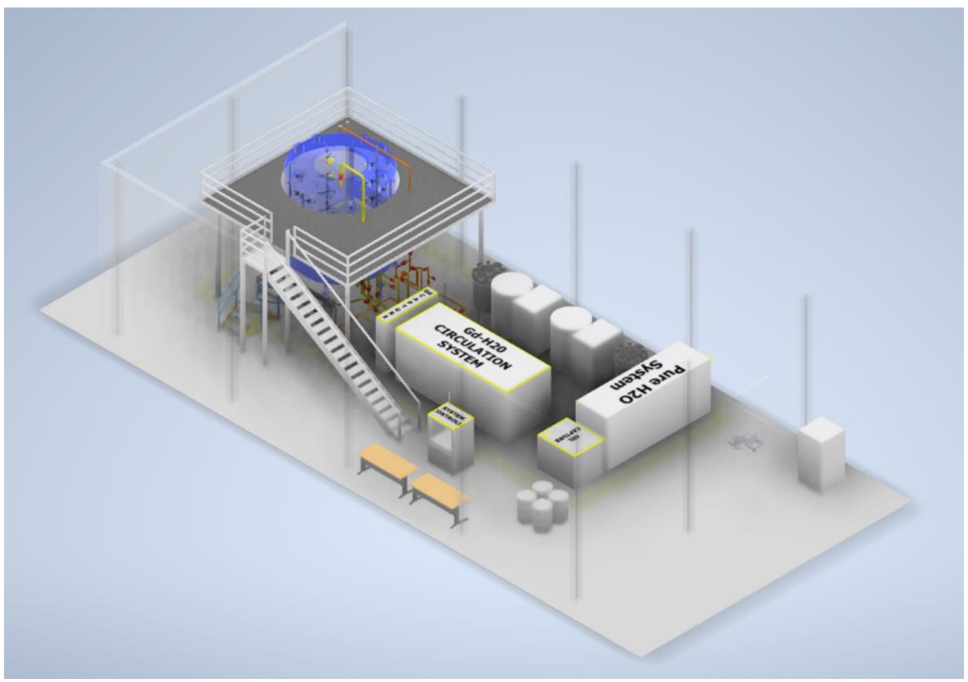
In its final week of data-taking before being de-commissioned, the 41-ton WCTE was loaded with 0.03% Gd – the same as in SK, on May 23rd, 2025.

30-ton Tank @ BNL

[nuclear nonproliferation detector technology R&D]

Back in the US, EGADS's full band-pass filtration technology has been replicated for exploring how to recirculate gadolinium loaded water-based liquid scintillator (WbLS).

$H_2O \rightarrow Gd_2(SO_4)_3+H_2O \rightarrow Gd+WbLS$

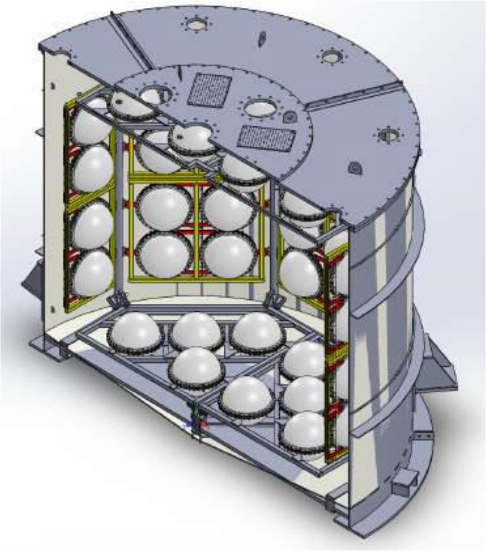


30-ton test facility at Brookhaven National Laboratory in New York

EGADS-style band-pass system installed at BNL; Gd goes in over the next few months!

BUTTON @ Boulby

[technology demonstrator in an underground UK lab]



BUTTON
3.2 m tall
3.6 m diameter
96 PMT modules
30 tons total volume

Like the BNL test tank, the Boulby Underground Technology Testbed for Observing Neutrinos [BUTTON] will evaluate the performance and handling of various fill materials: H20, H2O+Gd, and WbLS. Tests are beginning now, with Gd soon!

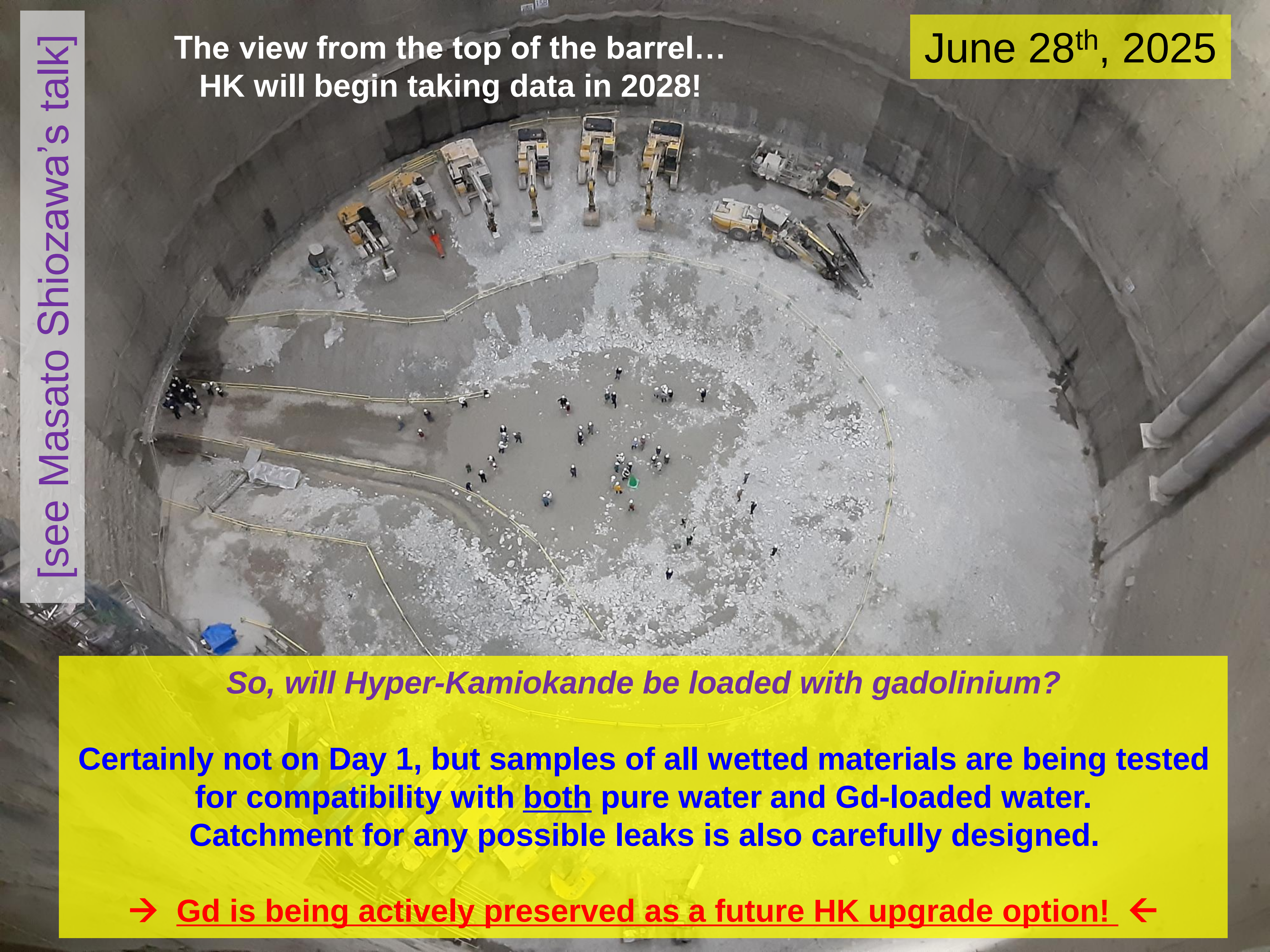
Gd-H_2O: Everybody's Doing It, Man…

Name	Location	Main Goal	Water Volume	$Gd_2(SO_4)_3$ Loaded
EGADS	Kamioka	Gd R&D, SN Watch	200 tons	Since 2013
ANNIE	Fermilab	High-E Neutron Multiplicity	26 tons	Since 2019
Super-K-VI/VII	Kamioka	DSNB, SN Burst, PDK, ATM/Sol/LB ν	50 ktons	Since 2020/2
XENONnT Water Shield	Gran Sasso	Dark Matter Detection	700 tons	Since 2023
WCTE	CERN	IWCD/mPMT Demonstrator	50 tons	May 2025 (completed)
30-ton Test Tank	BNL	Nuclear Non-Proliferation Demonstrator	30 tons	2025/6
BUTTON	Boulby	Underground Demonstrator	30 tons	2026
Hyper-K-II(?)	Kamioka	DSNB, SN Burst, PDK, ATM/Sol/LB ν	258 ktons	203X(?)

[see Masato Shiozawa's talk]

The view from the top of the barrel…
HK will begin taking data in 2028!

June 28th, 2025



So, will Hyper-Kamiokande be loaded with gadolinium?

Certainly not on Day 1, but samples of all wetted materials are being tested for compatibility with <u>both</u> pure water and Gd-loaded water.
Catchment for any possible leaks is also carefully designed.

→ <u>Gd is being actively preserved as a future HK upgrade option!</u> ←