Neutron tagging with gadolinium loaded PMMA

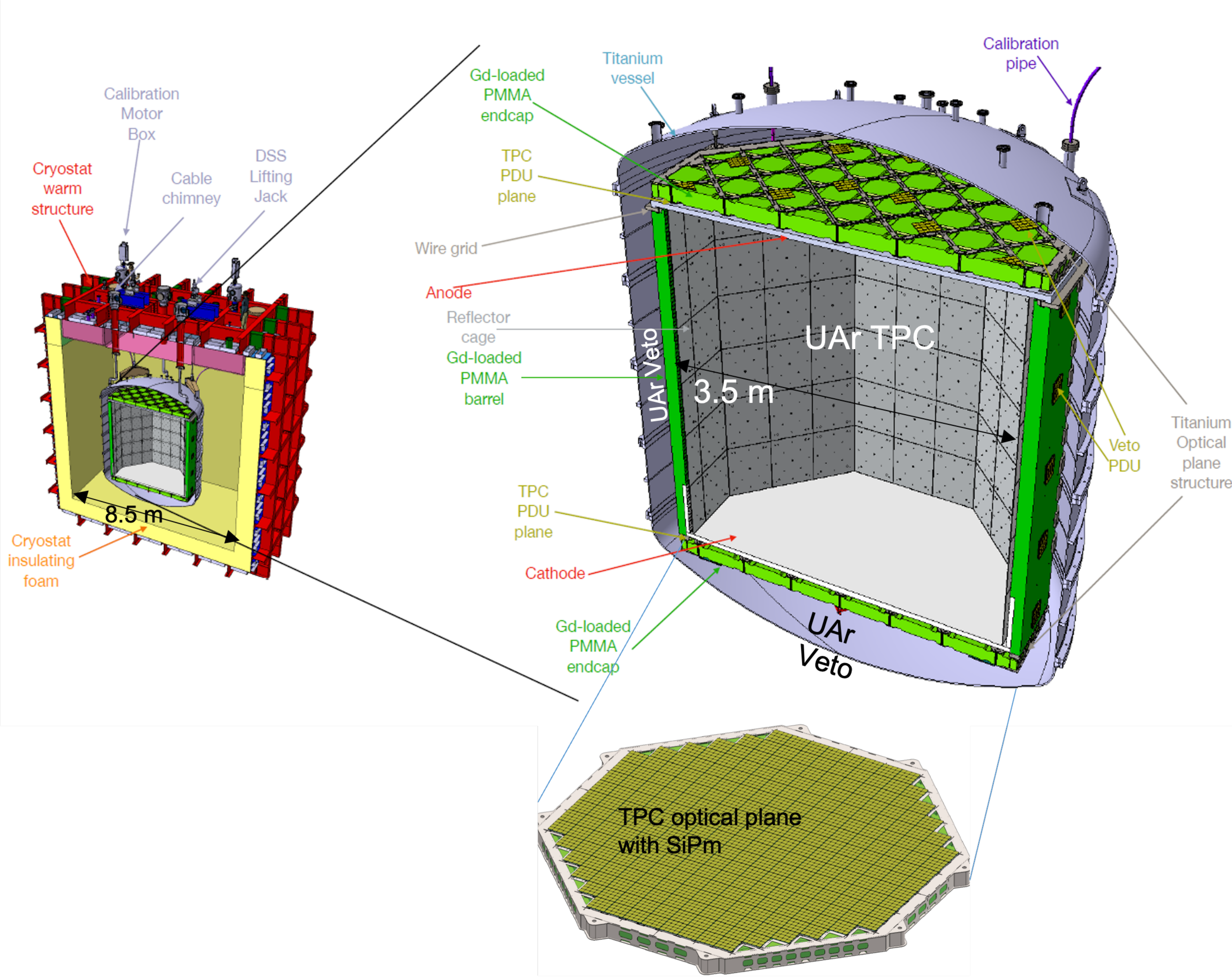
ICRM-LLRMT 2022

LNGS, Assergi (Aq)

A. Marini (INFN-Genova), on behalf of the DarkSide collaboration

DarkSide-20k
Key points

- Direct dark matter search experiment, that looks for Weakly Interacting Massive Particles (WIMPs) events
- The detector will be located… here, at LNGS!
- The detector is based on an ultrapure liquid low radioactivity Argon double-phase Time Projection Chamber (TPC)
- In the TPC the detection of two signals occurs:
    1. Liquid phase → **S1**: produced by the scintillation light due to both the Ar dimers and the recombined fraction of ionized atoms
    2. Gas phase → **S2**: produced by the free electrons escaped from recombination and drifted up to the liquid surface and extracted with an electric field
- Goal: 0.1 nuclear recoil events/200 tons*year.



DarkSide-20k scheme

DarkSide-20k
Background types

Background

Induced by — *Induced by*

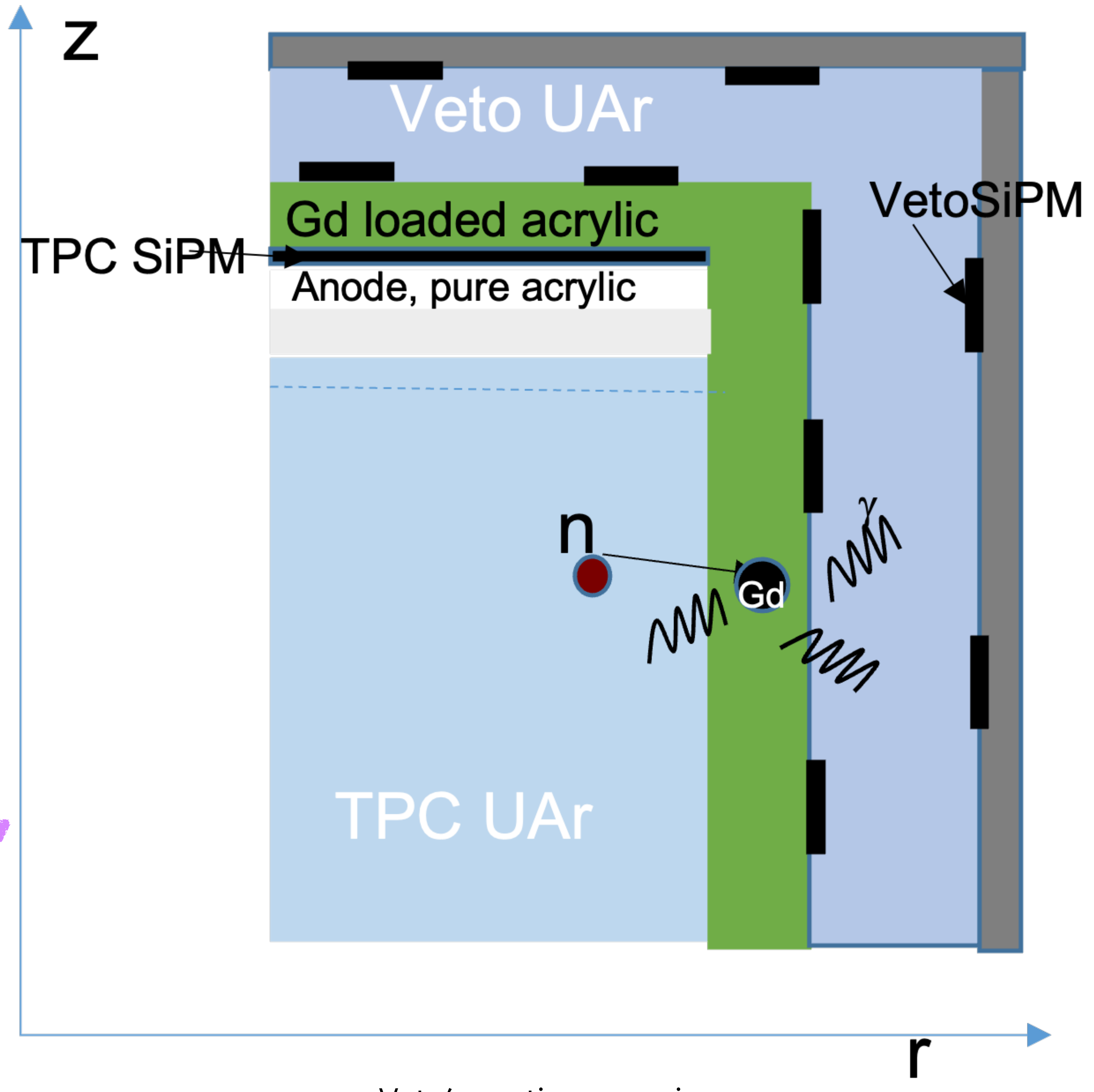
Electron recoil ← PSD → Nuclear recoil

How to overcome? — *How to overcome?*

- Low ^{39}Ar content
- Use of dual-phase TPC
- PSD
- Strict materials selection

- Active Veto detector
- Strict materials selection

Extraction of underground Ar (UAr, ^{39}Ar content equal to $1/1400$ with respect to atmospheric Ar)



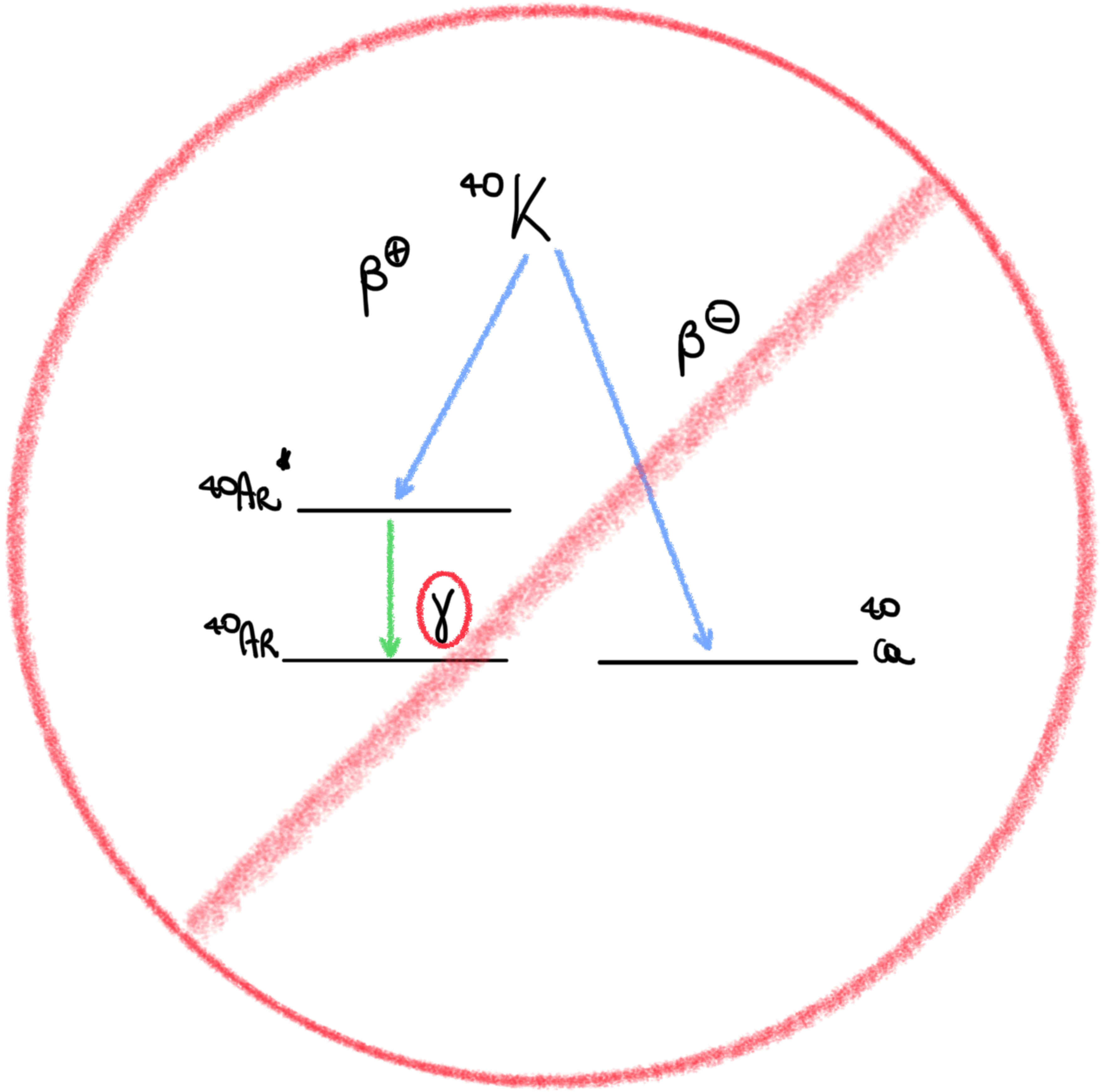
Veto's section overview

DarkSide-20k
Dead time

Probability to tag an event induced by ^{39}Ar decay or a γ interaction as neutron because it happens in coincidence with a true WIMP event within a time window of 800 μs.

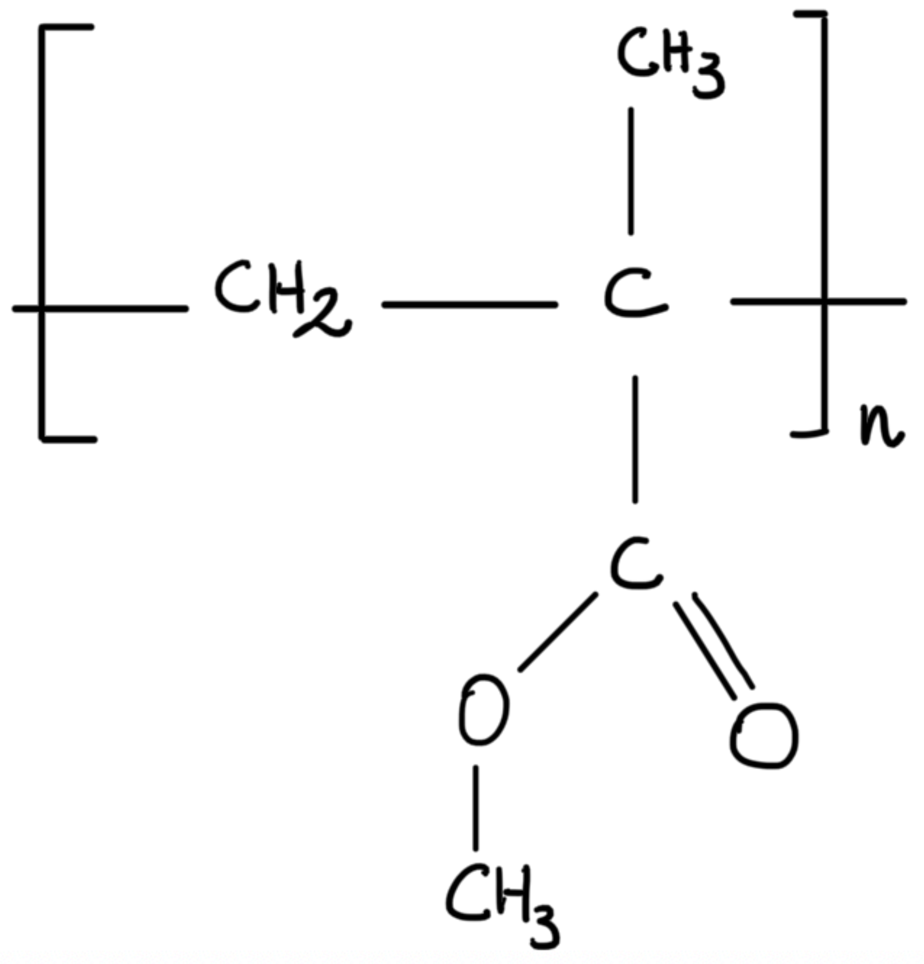
The main sources of random coincidences with WIMP events within the time window Δt are:

- Decays of ^{39}Ar in the inner Veto or TPC;
- Events due to γ-rays depositing energy in the Veto or TPC;
- Pile-up events: two events of the previous categories happening very close in time.



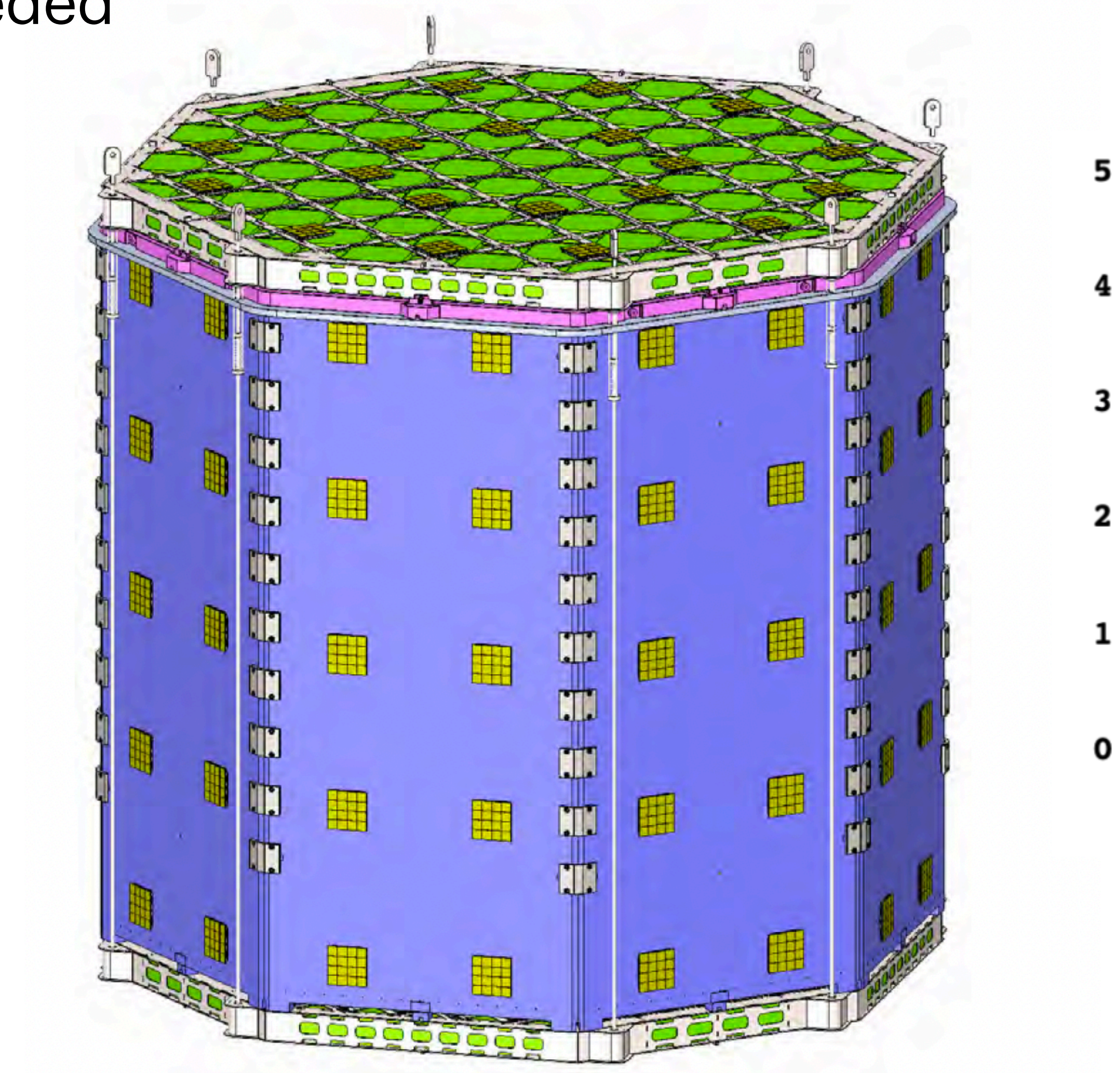
DS-20k n tagging

Veto

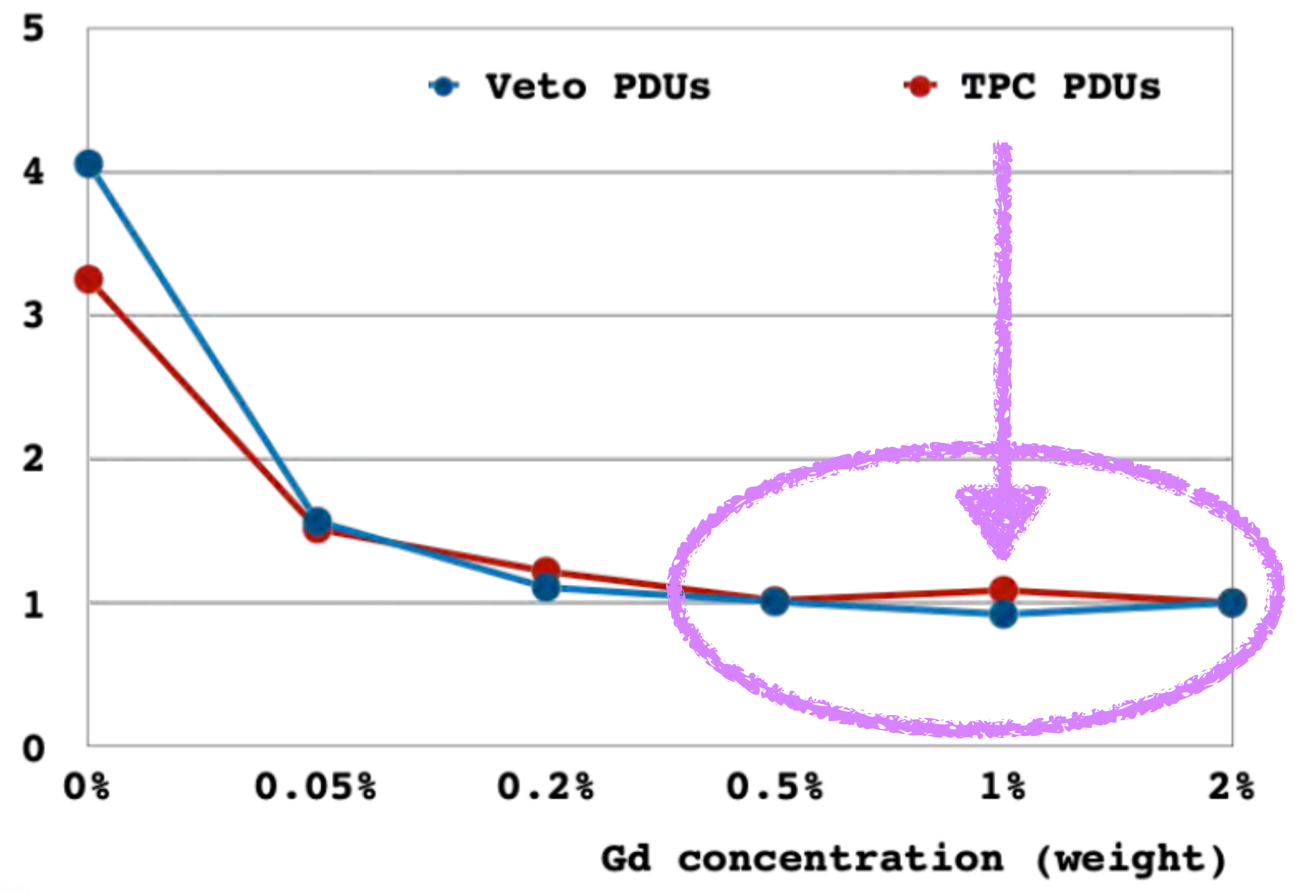


Rich in Hydrogen

- Gd-PMMA is the chosen material for the neutron veto → 11.2 tons needed
- Layers of Gd-PMMA surround the entire TPC volume.
- Gd-PMMA is highly efficient at moderating and then capturing neutrons.
- The capture resulting in the emission of several γs, with energy up to 7.9 MeV.



Focus on Veto walls



Capture relative inefficiency vs Gd content

Gd-loaded acrylic

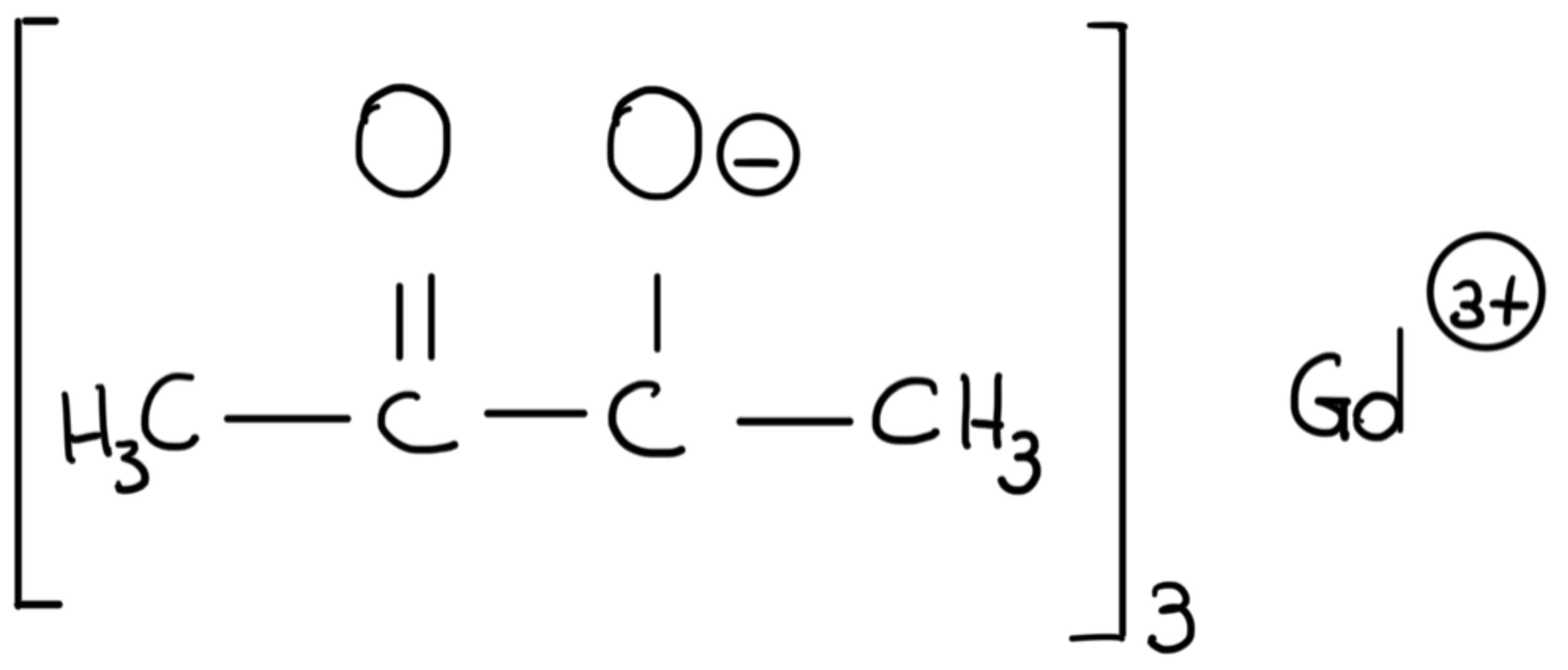
Development of the hybrid material

We performed R&Ds in Italy, Russia and China.

- Promising results have been obtained with Gd_2O_3, $Gd(acac)_3$, and $Gd(MAA)_3$.
- All of these compounds are satisfactory from the chemical and radiopurity points of view.

The compounds ready for industrial production are:

- $Gd(MAA)_3$ dispersion in the liquid monomer (MMA).
- Gadolinium oxide nanograins (Gd_2O_3) mechanical dispersion in MMA.

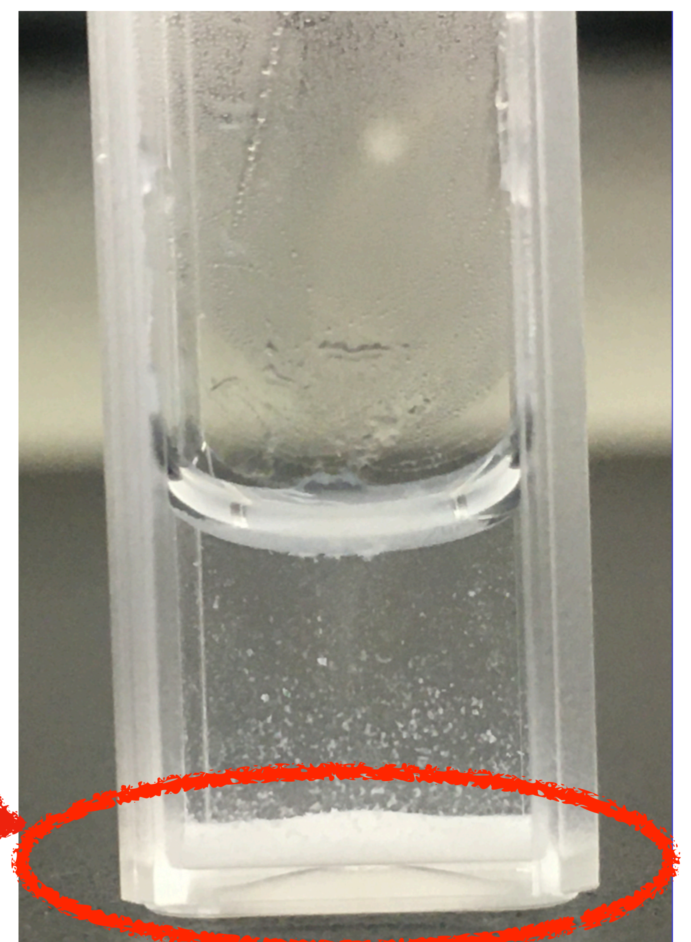


- Radiopure acrylic matrix
- Good machining experience with radio pure plastics.

Proprietary production chain

- Commercially available
- Radiopure

Not soluble



How to overcome?

Surface treatment

Gd_2O_3 functionalization

Nanograins coating

Nanograins

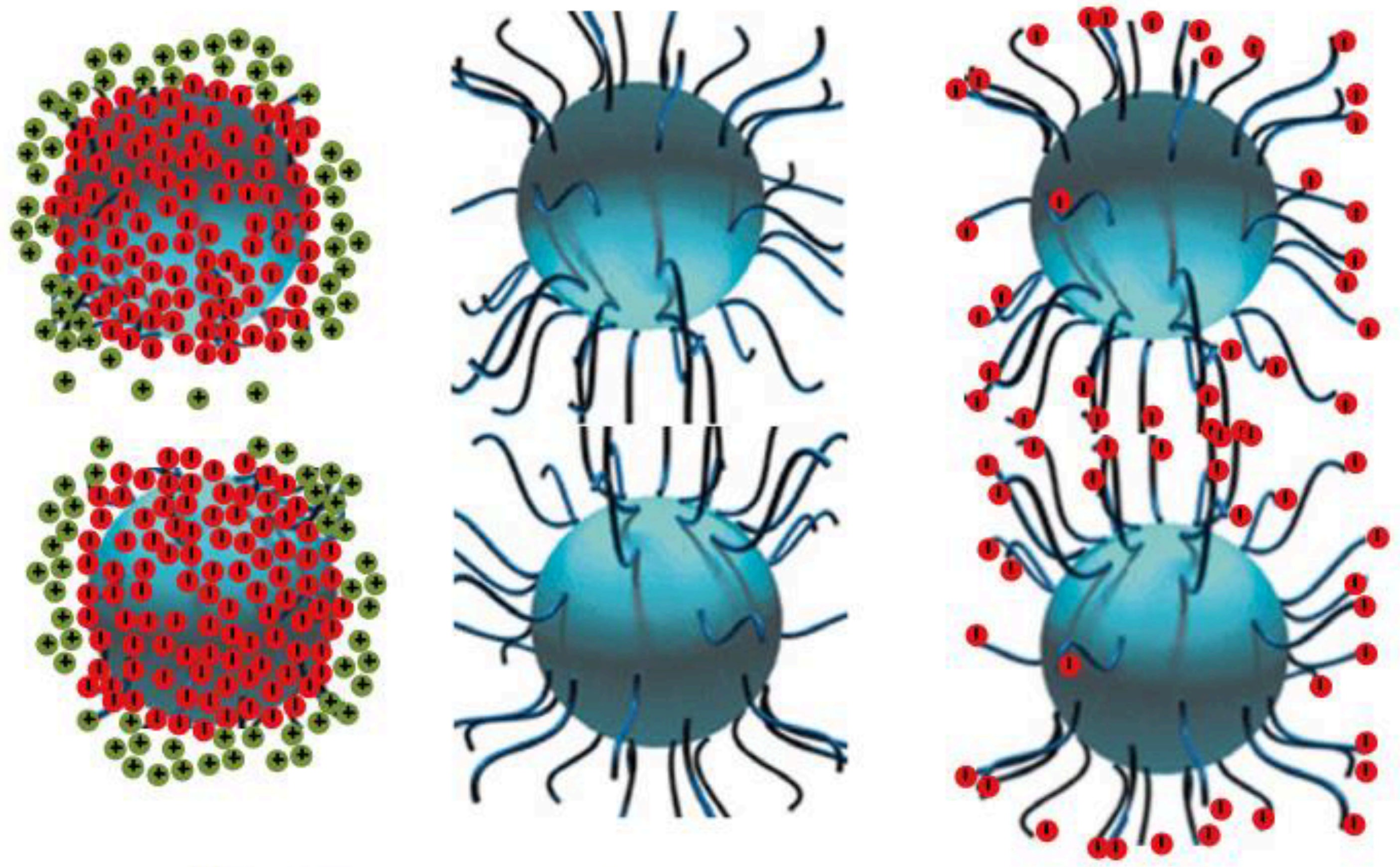
+

Igepal CO-520

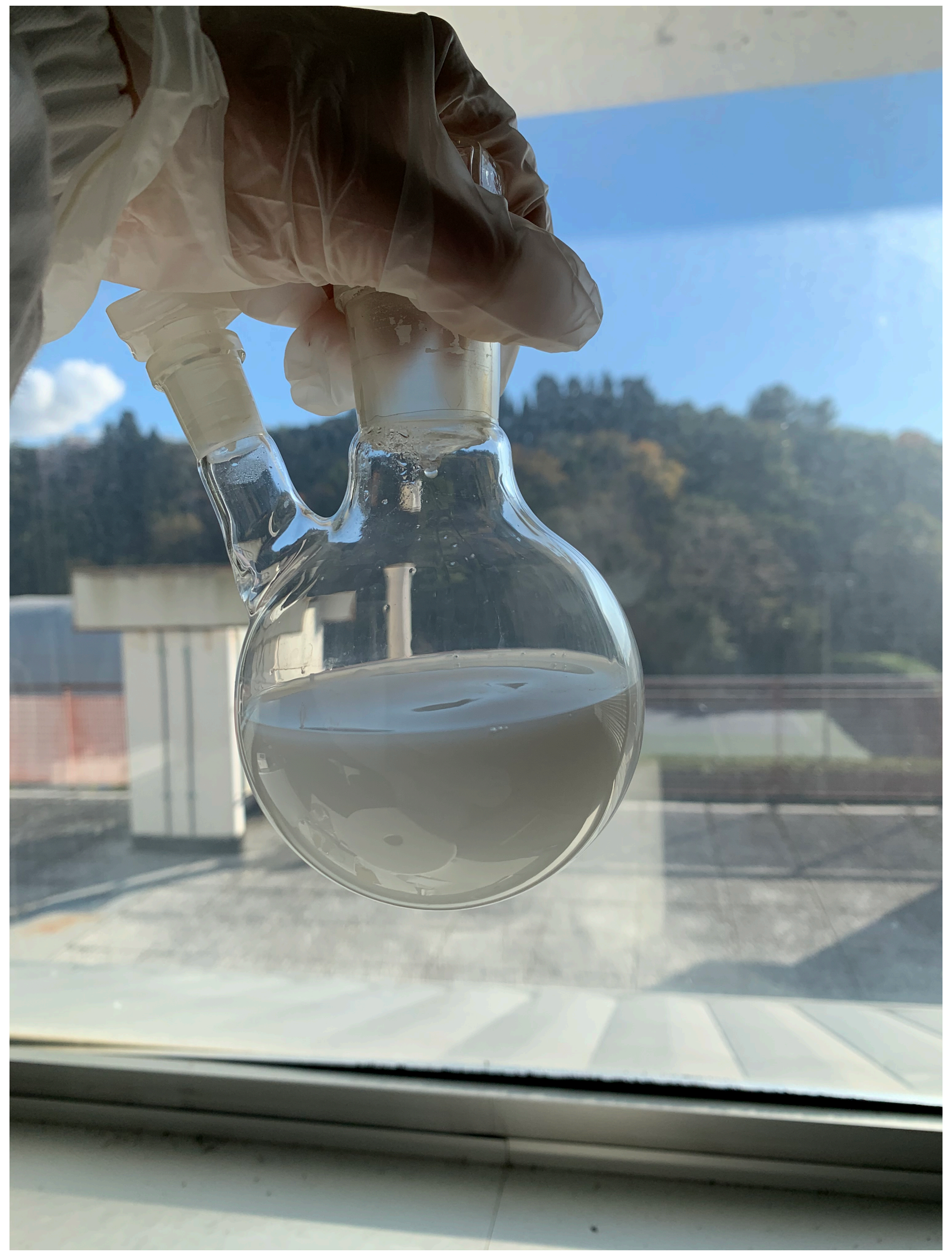
+

Solvent

=



Zhang W. Nanoparticle Aggregation: Principles and Modeling. Advances in Experimental Medicine and Biologu, vol 811. Springer, Dordrech. https://doi.org/10.1007/978-94-017-8739-0_2



But... Is the surfactant radiopure?

	U (ICPMS) [mBq/kg]	Th (ICPMS) [mBq/kg]	^{40}K (HP-Ge) [mBq/kg]
Igepal CO-520	<0.041	<0.12	$3.19*10^4$

LSC

Radiopurity of the components

Purifications & screening

- The radiopurity of industrial oxide is well within requirements
- A strategy was found to reduce the K amount in the surfactant
- An ionic exchange resin was used (H^+ vs K^+)
- Then various ICP-AES and ICP-MS screenings were performed on the samples

Gd_2O_3 nanograins qualification (HPGe)

Sample	^{238}U [mBq/kg]	^{232}Th [mBq/kg]	^{40}K [mBq/kg]
ShinEtsu 1	13.6 ± 3.0	< 27	< 37
ShinEtsu 2	6.6 ± 1.8	< 19	< 23
ShinEtsu 3	2.68 ± 0.47	2.31 ± 0.68	< 13

LSC

Surfactant K contamination after purification (ICP-AES and ICP-MS)

Sample	ICP-AES [mBq/kg]	ICP-MS [mBq/kg]
Sol. 1 (raw Igepal)	$1.87*10^4$	$1.78*10^4$
Sol. 2	$4.80*10^3$	-
Sol. 3	$6.35*10^3$	-
Sol. 4	$4.64*10^3$	-
Sol. 5	$2.48*10^3$	$2.63*10^3$
Sol. 6	$1.04*10^4$	-
Sol. 7	$1.53*10^4$	-
Sol. 9	$5.20*10^4$	$1.89*10^4$
Sol. 10	-	$5.11*10^3$
Sol. 12	$1.83*10^4$	-
Sol. 13	$5.82*10^3$	-
Sol. 14	-	$1.01*10^4$
Sol. 15	-	$6.50*10^3$
Sol. 16	-	$8.67*10^3$
Sol. 17	$3.19*10^2$	-

Genova

LNGS

What does it mean?

Let's see the γ background

- Igepal 0.1% not purified: safe, but we can still have room for improvements
- Igepal 0.1% purified: way below the PMMA contribution, so it's perfectly safe

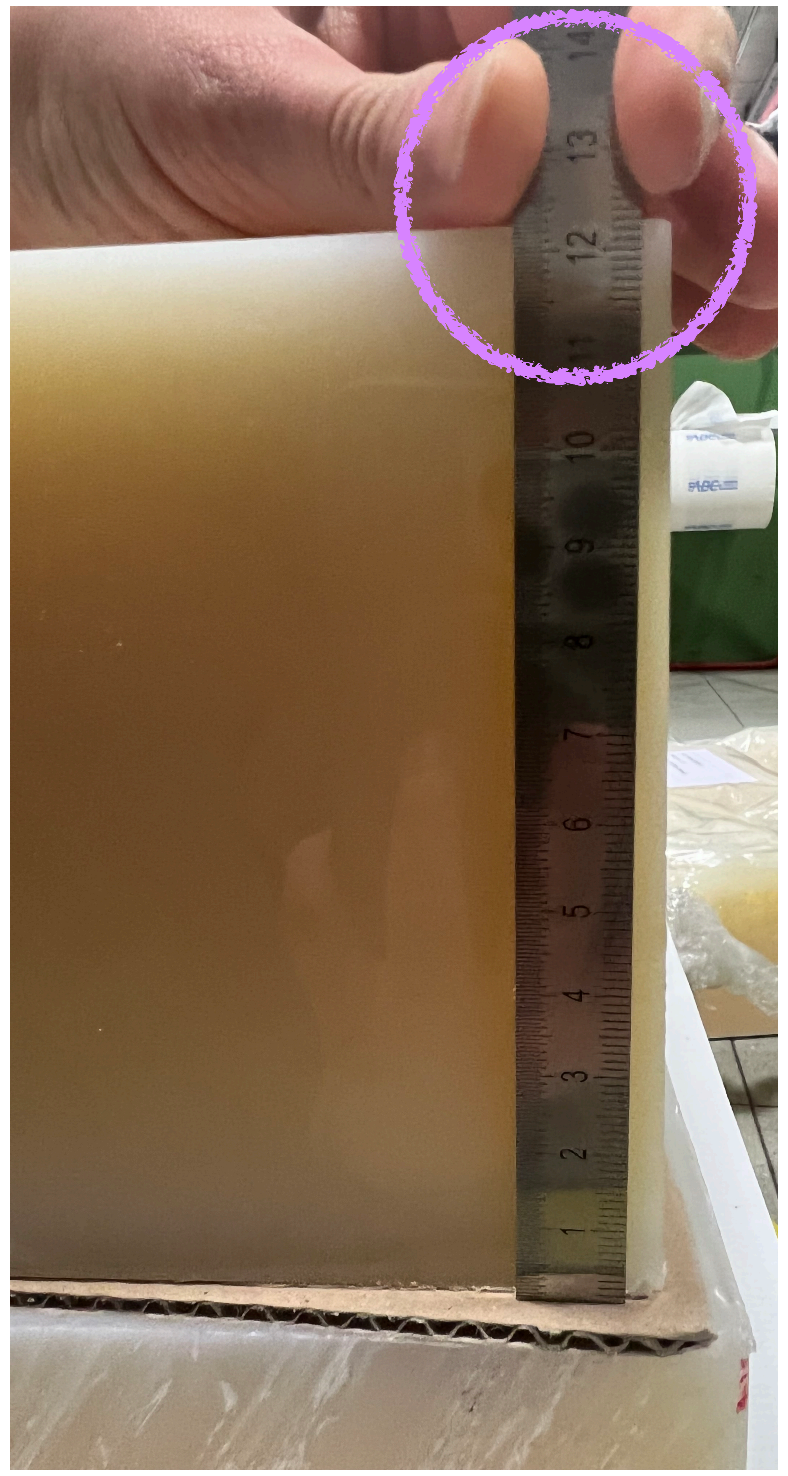
Contributor	Contributions to γ-background [Hz]			
	Igepal $0.1\%_w$ not purified		Igepal $0.1\%_w$ purified	
	TPC	Veto	TPC	Veto
Gd_2O_3	0.4	0.9	0.4	0.9
Igepal	10.2	19.6	0.3	0.5
PMMA	9.7	19.5	9.7	19.5
Total contribution from Gd-PMMA	20.3	40.2	10.1	20.5

The γ bkg is safe

And now?

Next steps for Gd_2O_3

- The R&D is over.
- We are scaling at final mass production capability with an Italian company, Clax s.r.l. (https://www.claxitalia.com)
- In the industrial samples we reached an homogeneity better than 5% with respect to nominal value in a 12 cm thick slab.
- The thickness of these samples has almost reached the DarkSide-20k requirement (17 cm).



Gd-complex solution

$Gd(MAA)_3$

Courtesy of prof. Y. Wang

- Organometallic compound - $Gd(MAA)_3$ produced from Gd_2O_3
- Process developed at Yangzhou University and scaled at DonChamp Chinese company (https://donchampacrylic.en.ecplaza.net)
- Functionalisation of the compound to make it soluble in MMA

$Gd(MAA)_3$ doped acrylic sheet (5 cm thick)

$Gd(MAA)_3$ recipe: radiopurity
Screening

Courtesy of prof. Y. Wang

- The pure acrylic from the company (previous experience with Juno vessel) has already been screened and it's compatible to DarkSide-20k needs
- HPGe screenings on the composite material are still ongoing in China
- $Gd(MAA)_3$

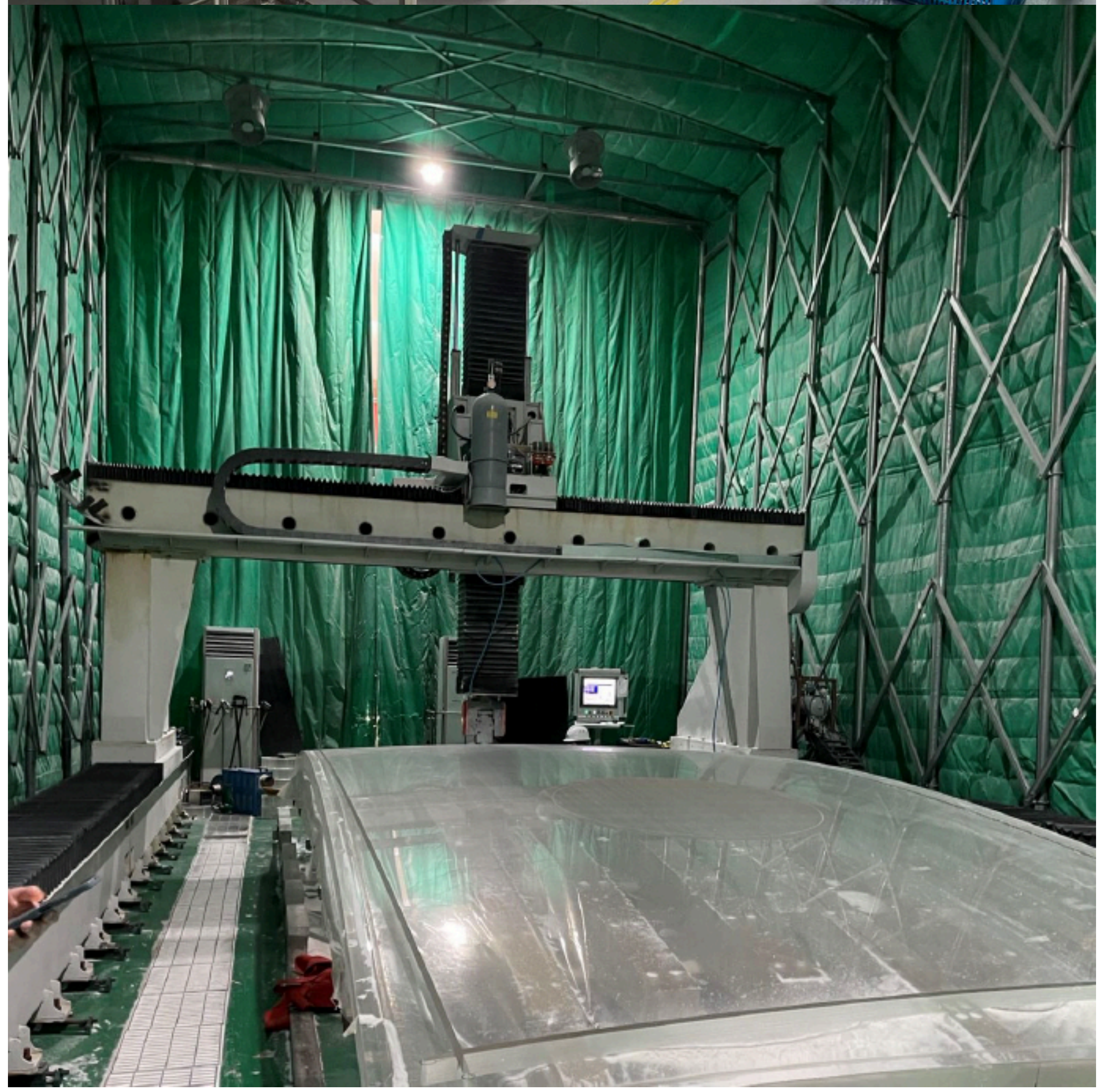
Isotope	[mBq/kg]
^{60}Co	< 2.38
^{137}Cs	< 2.97
^{40}K	125.86 ± 49.96
^{232}Th - ^{228}Ac	23.81 ± 9.64
^{232}Th - ^{228}Th	17.07 ± 6.25
^{235}U	< 3.62
^{238}U - ^{226}Ra	< 29.80
^{238}U - ^{222}Rn	40.48 ± 5.99

HPGe results of $Gd(MAA)_3$

And now?

Next steps for $Gd(MAA)_3$

- Transport of radiopure MMA from the company to the university labs.
- Dissolve $Gd(MAA)_3$ into MMA $10\%_w$
- Transport Gd-doped MMA back to the company infrastructure
- Dilute with pure MMA to get $1\%_w$
- Polymerisation



R&Ds conclusions

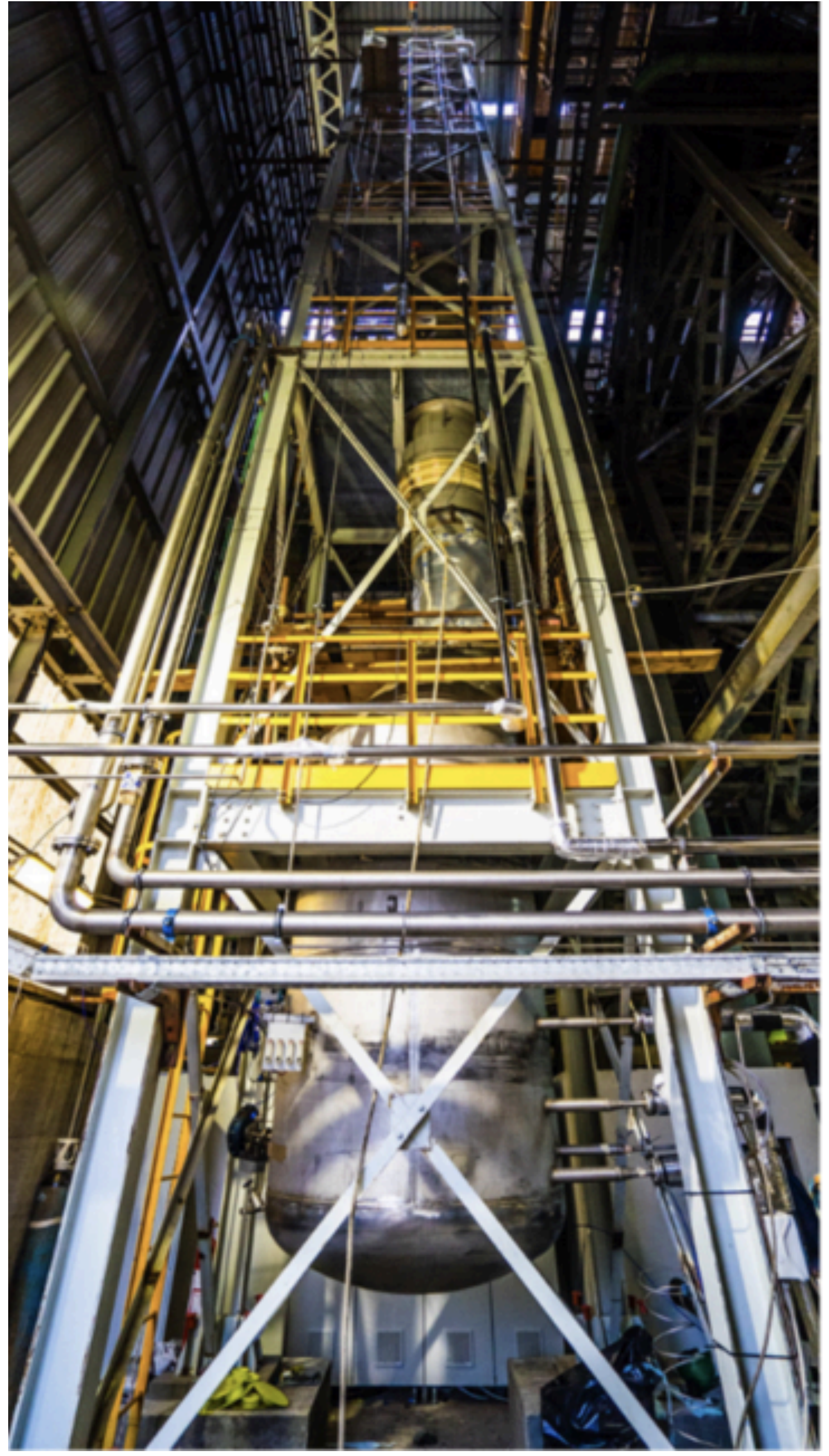
- The need of the DarkSide-20k experiment of tagging neutrons has led to the development of a hybrid material.
- Different strategies have been investigated
- The background requirements of the experiment apply stringent limits on the material radiopurity
- Basing on these requirements, the strategy included severe contamination screenings in the course of the R&D
- Two techniques have been scaled to industrial level production and they both represent a potential solution for the DarkSide-20k experiment

Thank you for your attention!

For further informations: anna.marini@ge.infn.it

DS-20k - the project
UAr and Aria

- The DarkSide-20k TPC will be filled with liquid underground ^{40}Ar
- Atmospheric Ar contains ^{39}Ar, which emits β. Underground Argon (UAr) has a ^{39}Ar content that is 1400 times less than in atmospheric!



Seruci-0, Sardinia, Italy

Extract UAr → Urania: Argon extraction and purification (Colorado) → Aria: final purification plant (Sardinia, Italy)