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Gd-PMMA

a novel neutron tagging technology for low background detectors

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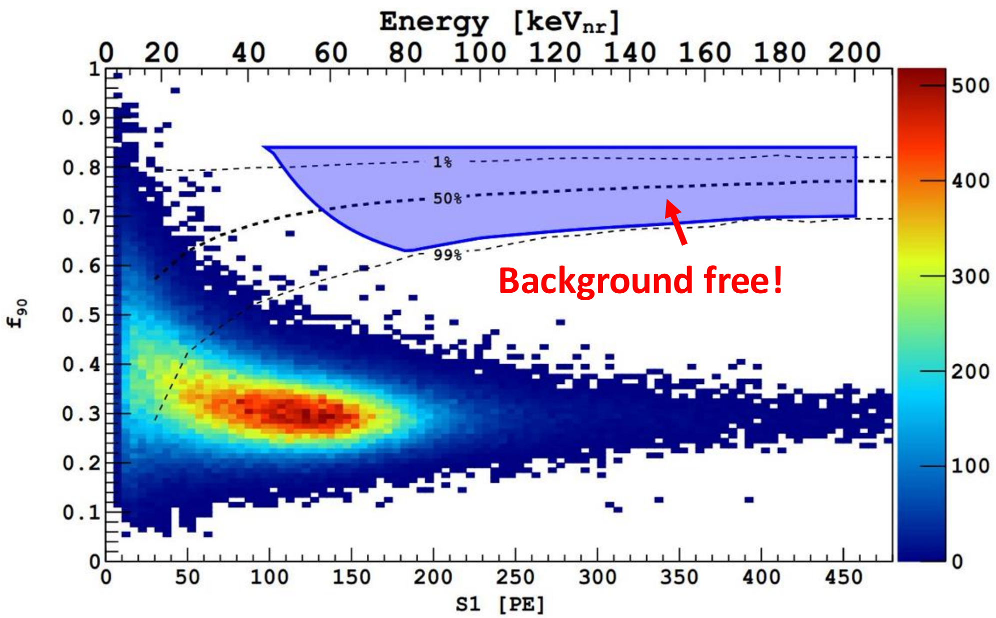
Motivation

- **Nuclear-recoil dark matter searches:**
  Neutron-induced nuclear recoils can mimic WIMP signals.

- **Reactor CEνNS experiments;**
- **Low-energy antineutrino experiments;**
- **Neutrinoless double-beta decay searches.**

DarkSide-50: background-free results

- DAr-TPC with two veto detectors:
  - ➢ Liquid-scintillator veto (LSV)
  - ➢ Water-Cherenkov detector (WCD)



532 live-days data

Phys. Rev. D 98, 102006 (2018)

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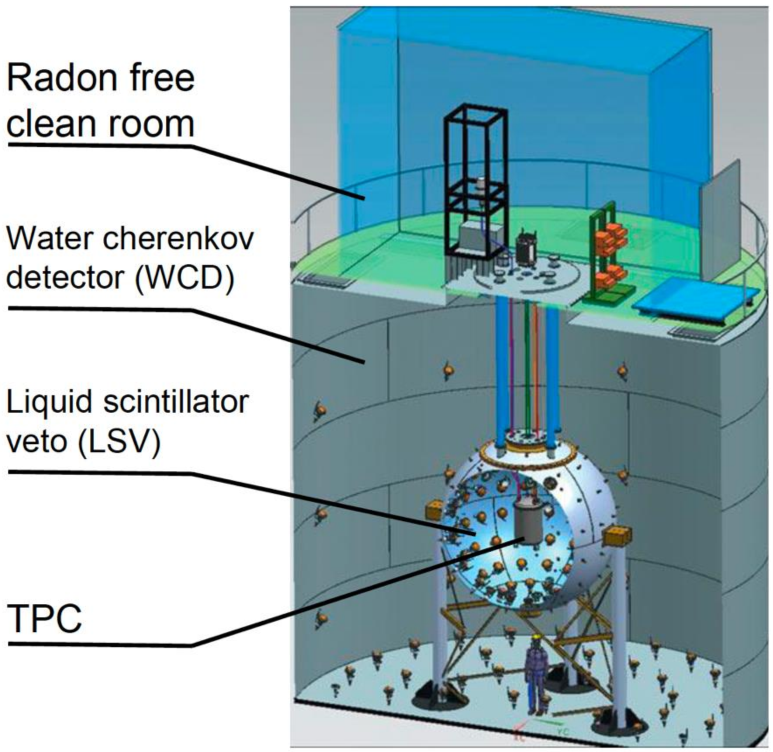
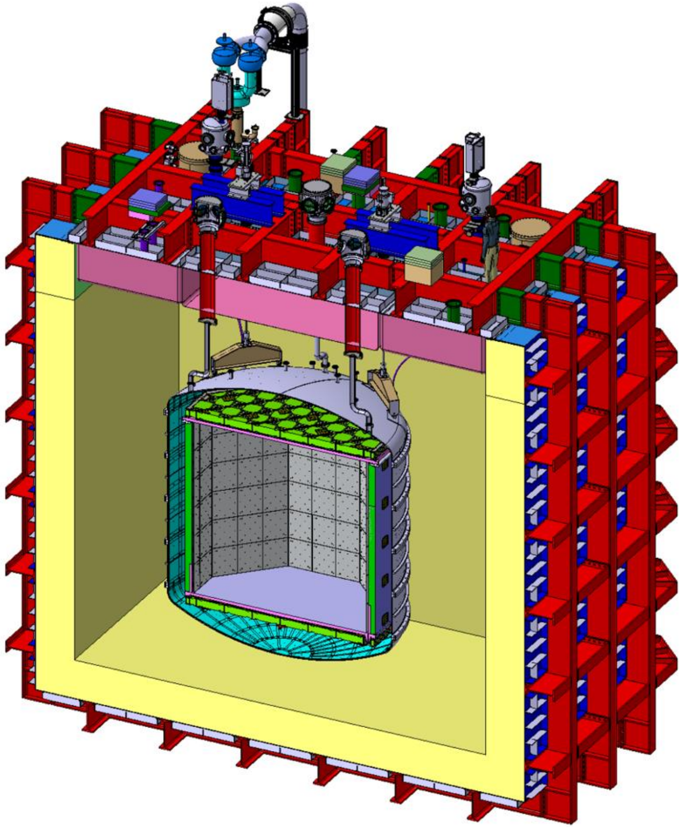


DarkSide-20k: different n veto strategies

- DarkSide-20k is 3 order magnitude larger than DarkSide-50.

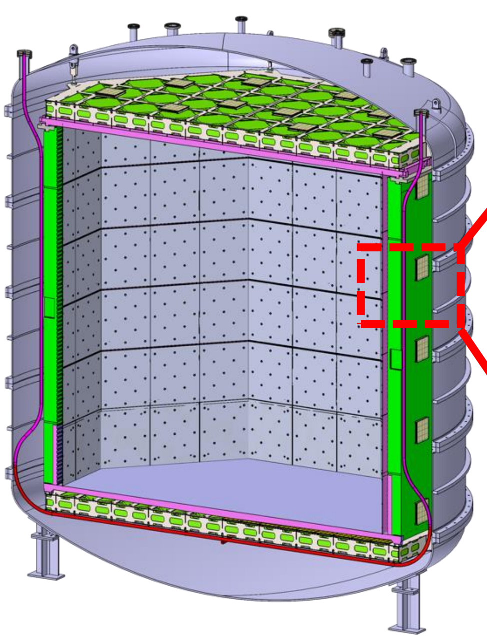


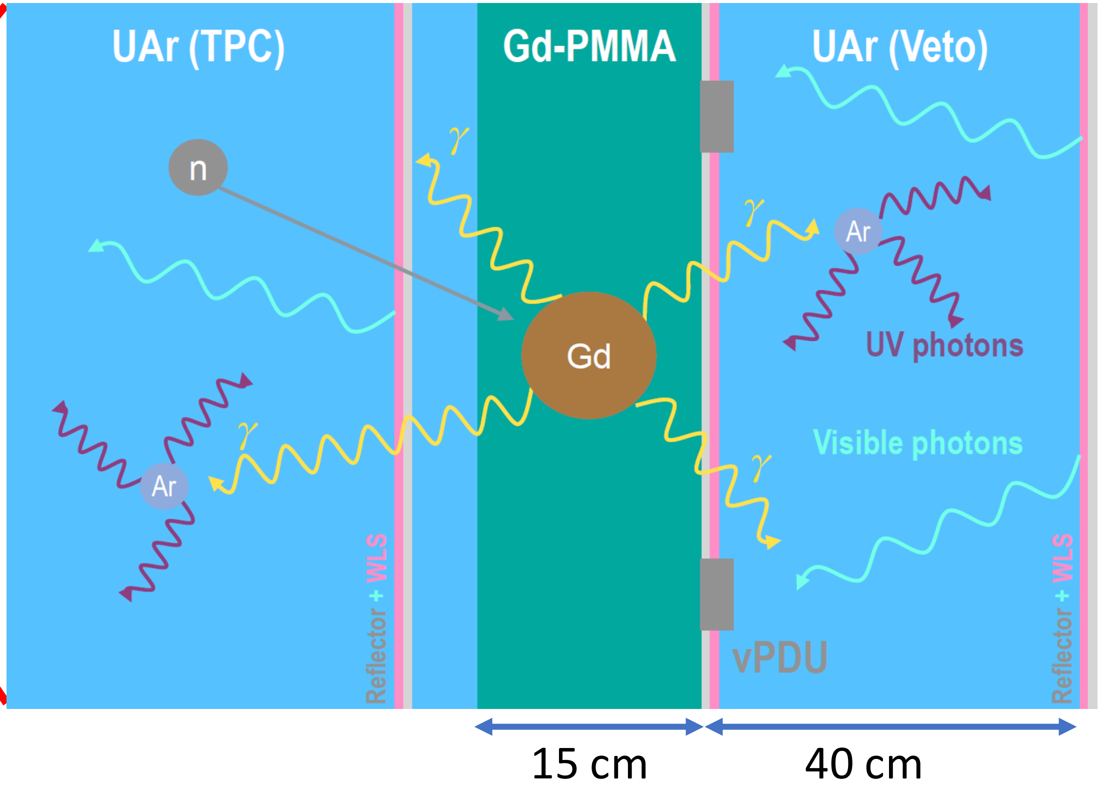
- **CH_4-doped argon**
  Hydrogen-rich, but suppresses argon scintillation light.
- **Gd-PMMA in atmospheric Ar**
  Outer placement was less effective.
- **Gd-PMMA in underground Ar**
  TPC structural material and neutron capture.

Bring neutron capture closer to the active volume to improve tagging of neutrons, e.g. from photosensors and the stainless-steel vessel.

How Gd-PMMA tags neutrons

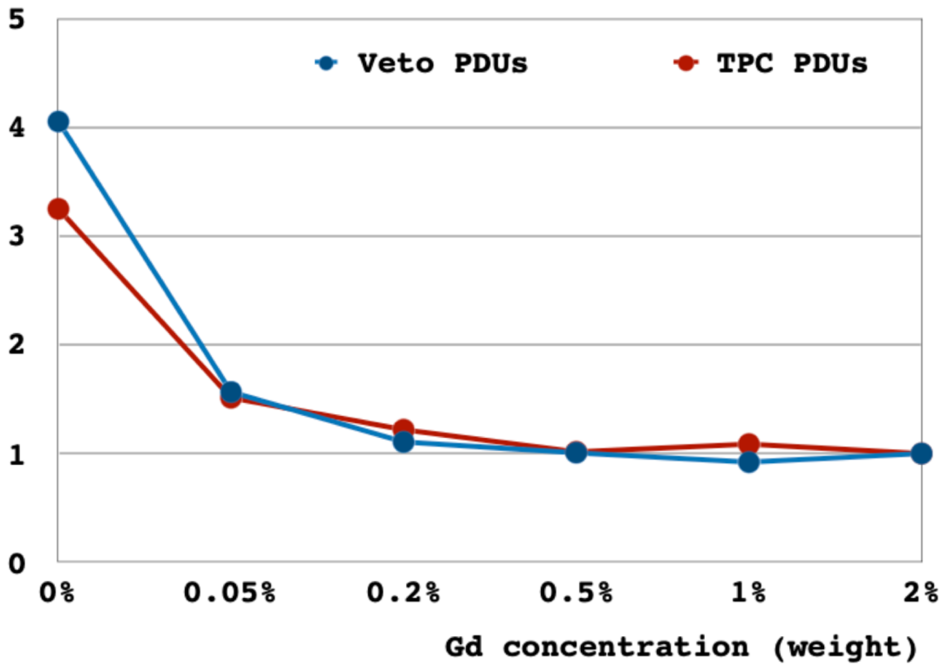
- Gd in the TPC structure captures neutrons, releasing γ rays.
- Capture γ rays can deposit energy in argon on either side.
- Photosensors detect the resulting scintillation light.





Requirements for Gd-PMMA

- Simulation based on the DarkSide-20k geometry indicates that 1 wt% Gd is sufficient for maximizing the neutron tagging efficiency.



Requirement to the R&D:

- Gd concentration ≧ 1 wt%;
- Dimension of the single panel can reach 4 m x 2 m x 15 cm;
- Can be operated in LAr.

Timeline of the Gd-PMMA development

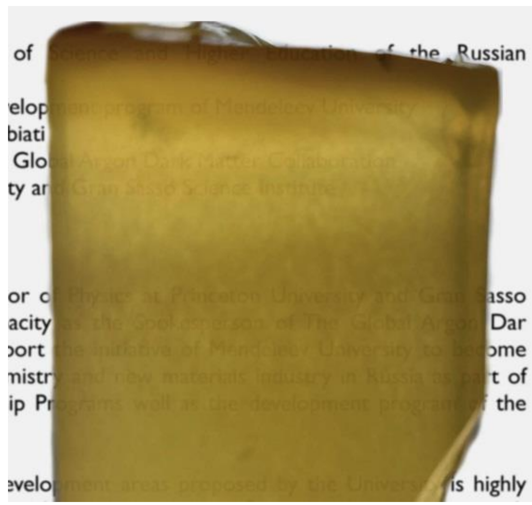
Recipe R&D	Lab-scale production	Industrialization R&D	Pre-production	Production
2020~2022	2023	2023~2024	2024~2025	2025~2026

- **Radiopurity control at every stage**
  Assess the radioactivity introduced at each process step.

- **Residual stress control for liquid-argon operation**
  Control stress and annealing to reduce cracking risk.

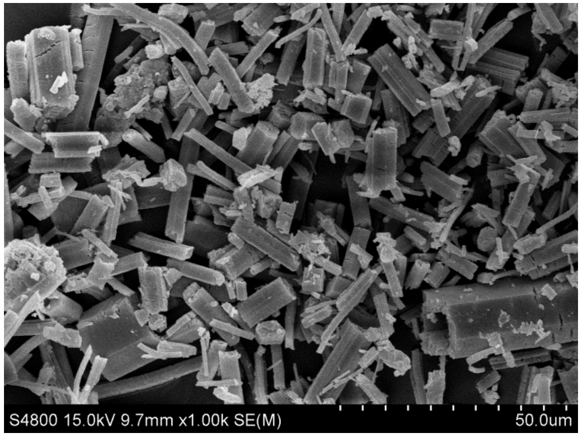
Recipes R&D

- Three candidate routes have been developed by the DarkSide collaboration.

Gd_2O_3	$Gd(acac)_3$	$Gd(MAA)_3$
		
Particle dispersion in MMA	Dissolved complex in MMA	**Dissolved complex in MMA**
JINST 19 (2024) P09021	Materials 14 (2021) 3757	Rare Metals 34(4) (2010) 568–573

Gd(MAA)$_3$ recipe was selected

- A dedicated recipe dissolves Gd(MAA)$_3$ into the MMA precursor.
- The formulation supports a broad range of Gd loadings (upto 10 wt%).
- Good chemical stability due to the molecular connection between Gd(MAA)$_3$ and MMA.



Gadolinium methacrylate

Dissolving recipe developed by Yangzhou University and IHEP

揚州大學
YANGZHOU UNIVERSITY

中國科學院高能物理研究所
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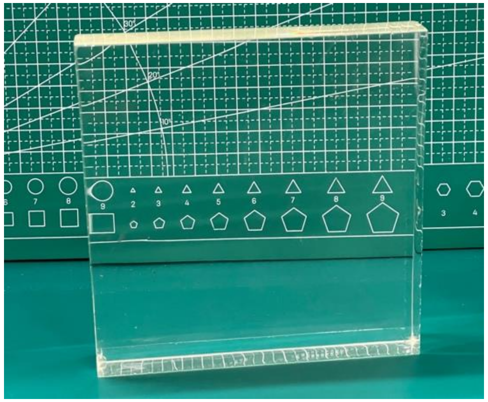
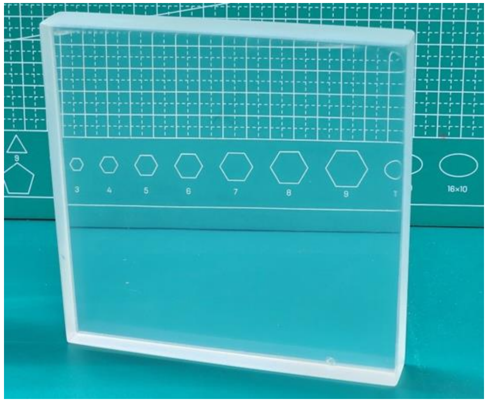
Lab-scale production

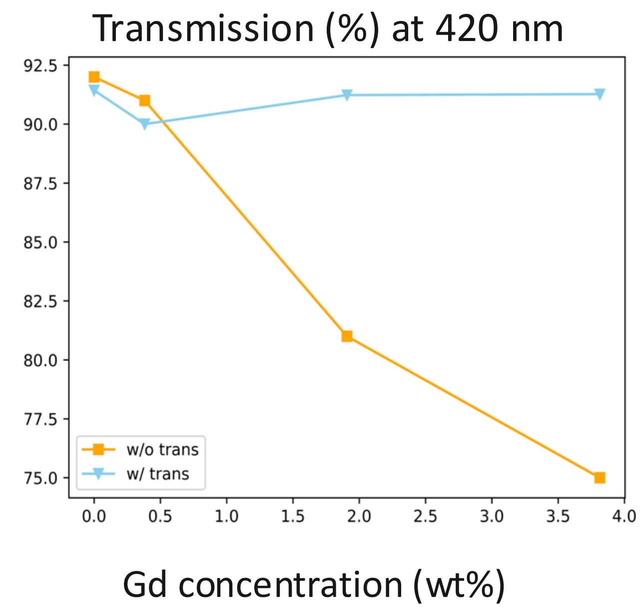
- A dedicated initiator recipe was developed to prevent self-inhibition and implosion during polymerization;
- High optical transparency is achievable.



High optical transparency

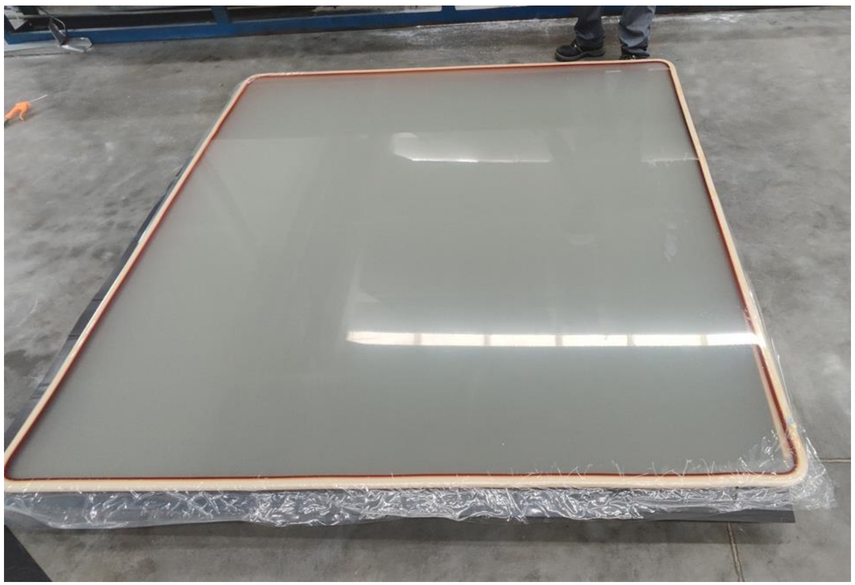


Normal optical transparency



Industrial production: single sheets

- Industrialization is in cooperate with Donchamp.
- Optimize single-sheet dimensions to support high production yield.



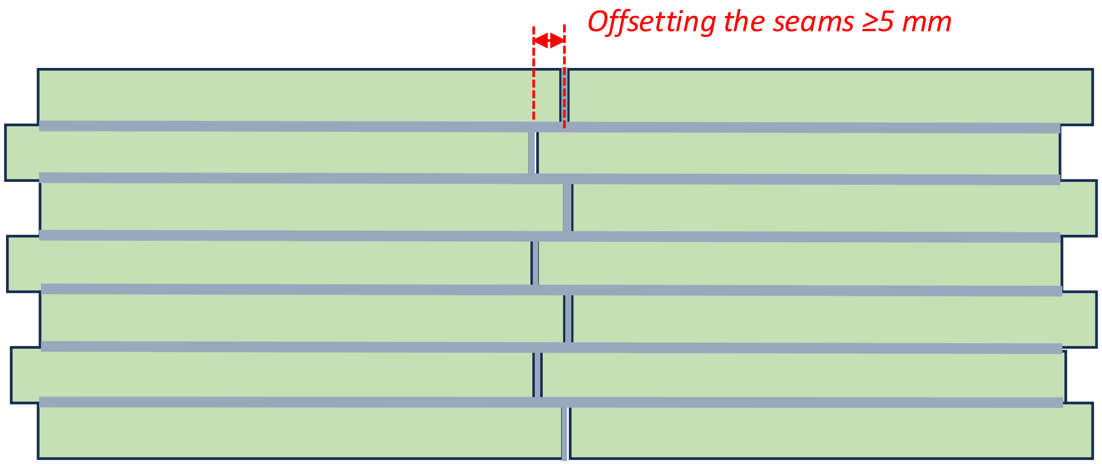
First 2 × 2 m sheet: Donchamp, spring 2024

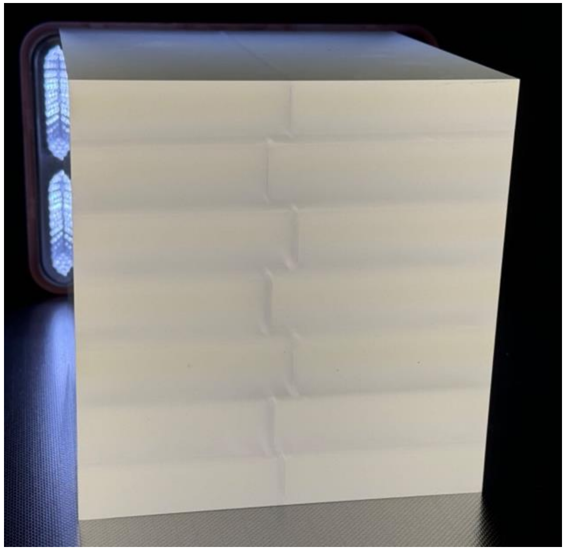
Optimized single-cast sizes:

- High transparency:
  100 × 70 × 4 cm
- Normal transparency:
  200 × 200 × 2 cm

Bonding and lamination

- A dedicated bonding and lamination process has been developed to enable multiple sheets to be assembled into a single panel with the required final dimensions.
- The adhesive uses the same ingredients as Gd-PMMA, specifically $Gd(MAA)_3$ dissolved in MMA.





Bonded and laminated test cube at IHEP

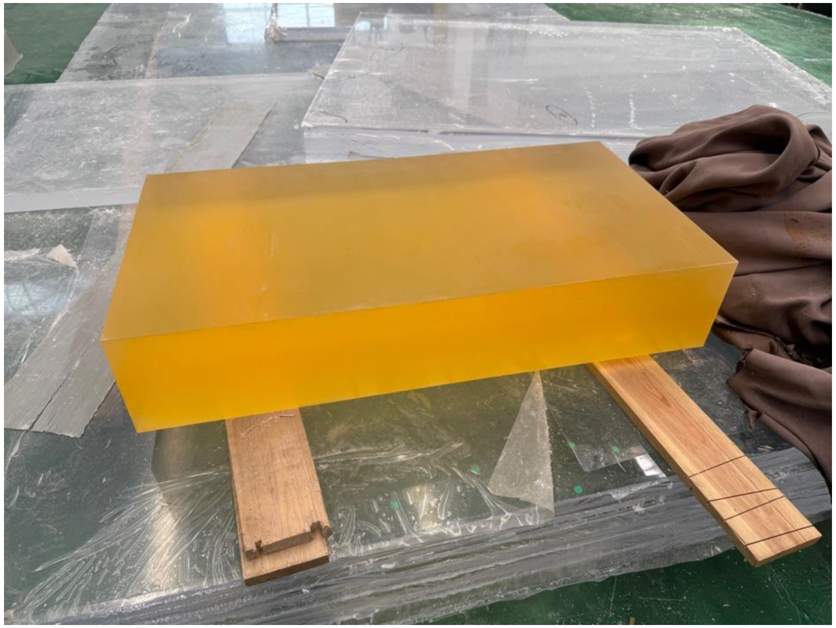
Pre-Production

- Outcome of the pre-production.
- Bonded and laminated panel: 80 × 80 × 16.4 cm.
- Delivered to the University of Alberta for further testing.

Mass production for DarkSide-20k

Production schedule

Period	Stage
Q3 2025	Gd_2O_3 procurement
Q4 2025	$Gd(MAA)_3$ production
Q1–Q3 2026	Sheet casting, bonding, lamination and annealing
Q4 2026	Machining



Gd-PMMA panel mass production

- The entire production process takes ~ 1 year.
- All Gd-PMMA parts for DarkSide-20k will be delivered to LNGS in mid-2027.

Radiopurity assays

- Joint assays with Boulby Underground Laboratory, CJPL, LNGS and LSC.

Samples prepared for radiopurity assays

ICP-MS results

Isotope	Activity (mBq/kg)
^{232}Th	0.08
^{238}U (top)	0.19

HPGe Results

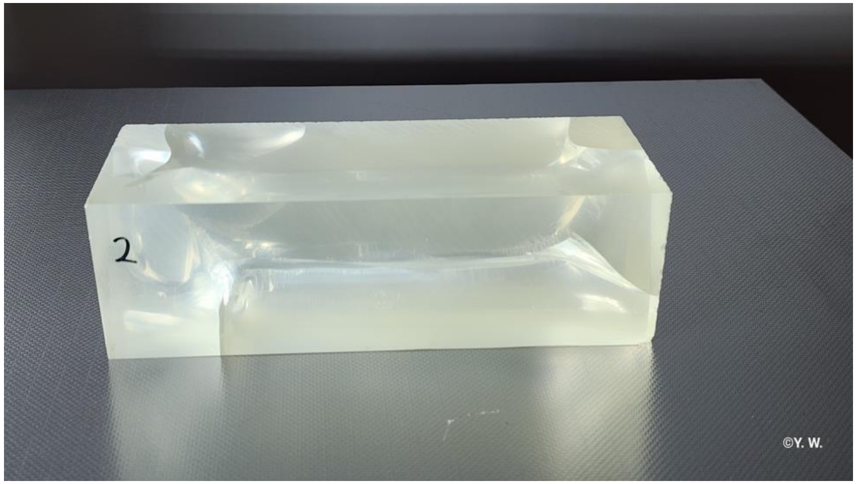
Activities in mBq/kg

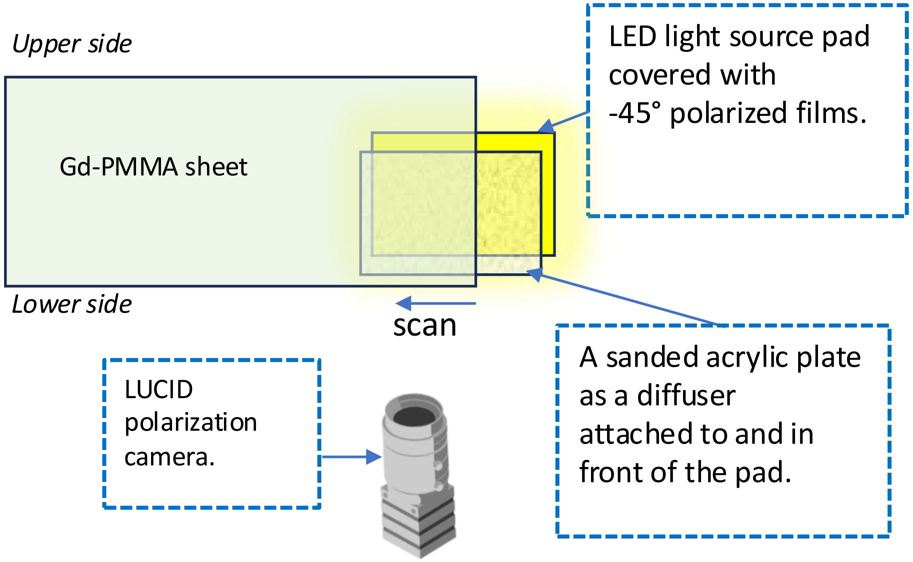
Isotope / chain	JUNO PMMA	Shin-Etsu 5N Gd_2O_3	Gd-PMMA
^{228}Ra (^{232}Th chain)	< 0.14	< 0.5	(0.21 ± 0.09)
^{228}Th (^{232}Th chain)	< 0.078	0.4±0.1	(0.10 ± 0.04)
^{226}Ra (^{238}U chain)	0.05±0.02	0.5±0.1	(0.20 ± 0.05)
^{234}Th (^{238}U chain)	< 2.1	< 33	< 23
^{234m}Pa (^{238}U chain)	< 1.8	< 7.4	< 1.8
^{235}U	< 0.07	< 0.31	< 0.13
^{40}K	< 0.41	4±1	(3.7 ± 0.9)
^{137}Cs	< 0.025	< 0.079	< 0.055

- PMMA is essentially pure, thanks to the radiopure PMMA production line developed by Donchamp for the JUNO experiment;
- 5N Gd_2O_3 from ShinEtsu is selected for Gd-PMMA production for low background.

Residual stress

- Control residual stress to limit cracking during cryogenic cooling.
- A residual stress measurement facility is being developed jointly by IHEP and the University of Alberta.

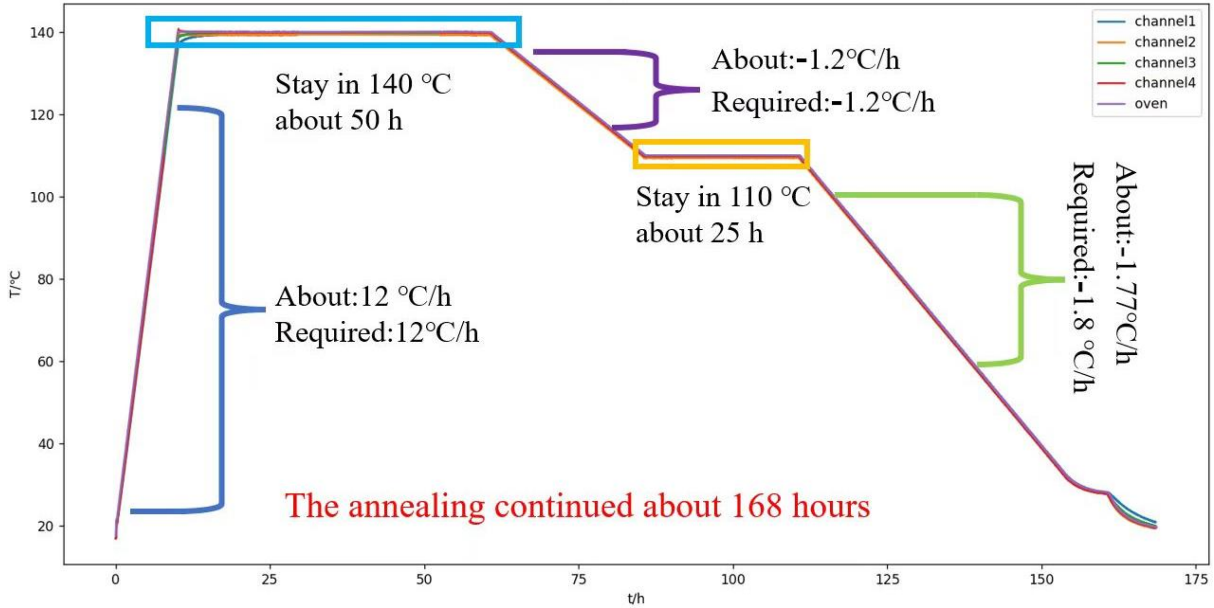




Segmented scan of large sheets

Stress mitigation: Annealing

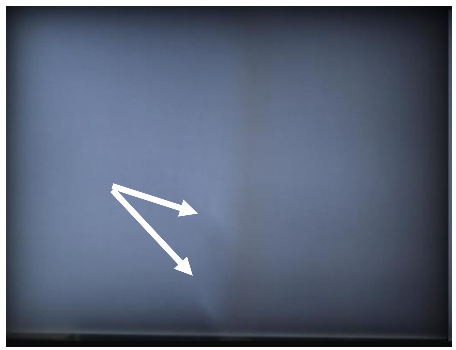
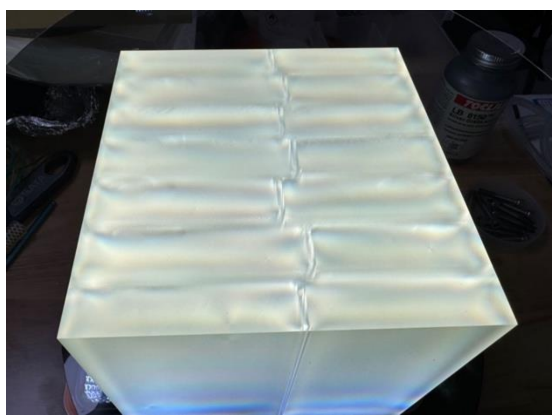
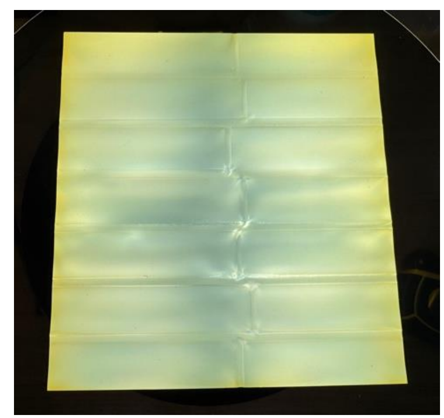
- A three-step annealing strategy has been developed:
  - ➢ After single-sheet casting;
  - ➢ After bonding and lamination (high temperature and longest time);
  - ➢ After machining.



Measured cycle for a 15 cm-thick sample: approximately one week

Stress mitigation after annealing

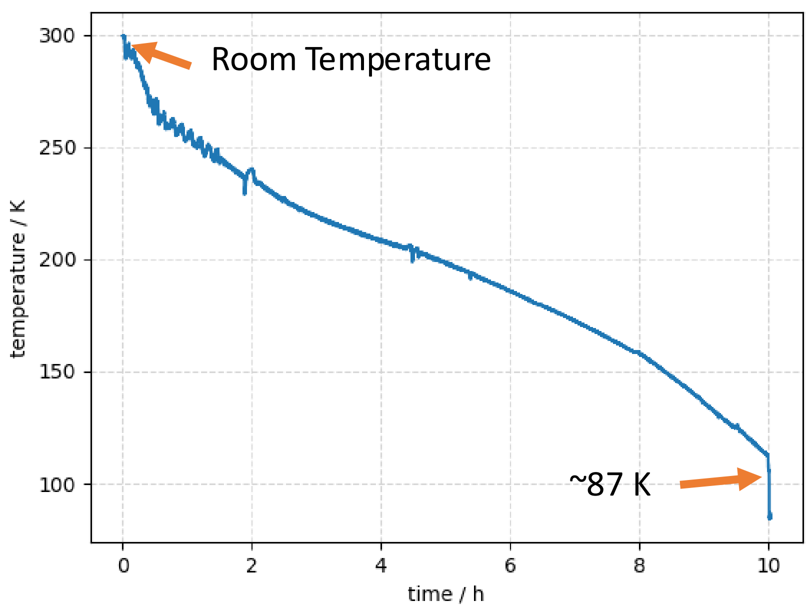
- Polarization images show reduced stress patterns after annealing.

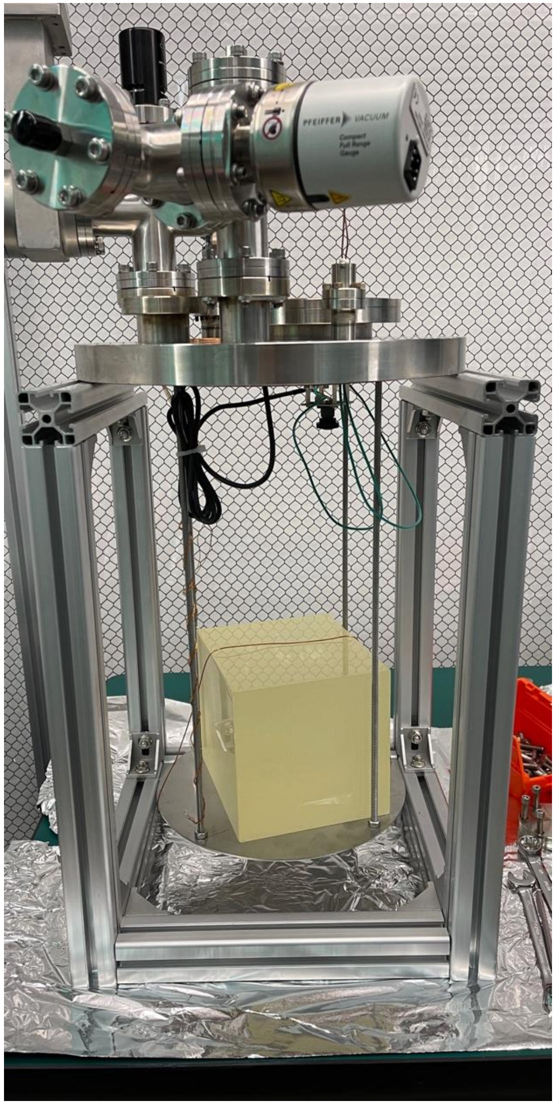
	Before annealing	After annealing
Single sheet		
15 cm-thick sample		

A cooling test

Annealed full thickness sample

- The average cooling rate was ~12 K/hr, while the fastest cooling rate of the cryogenic system is ~20 K/hr.
- No defects have been observed.





Summary

- Gd-PMMA has been developed to meet the stringent background requirements of WIMP dark matter searches.
- $Gd(MAA)_3$-based Gd-PMMA has entered industrial-scale production, with delivery for DarkSide-20k expected in 2027.
- Pre-production Gd-PMMA samples show good radiopurity with respect to ^{238}U and ^{232}Th.
- A dedicated annealing procedure has been developed to meet the requirements for use in noble-liquid detectors.