



中国科学院高能物理研究所

Institute of High Energy Physics Chinese Academy of Sciences



Gd-PMMA

a novel neutron tagging technology for low background detectors

Yi Wang

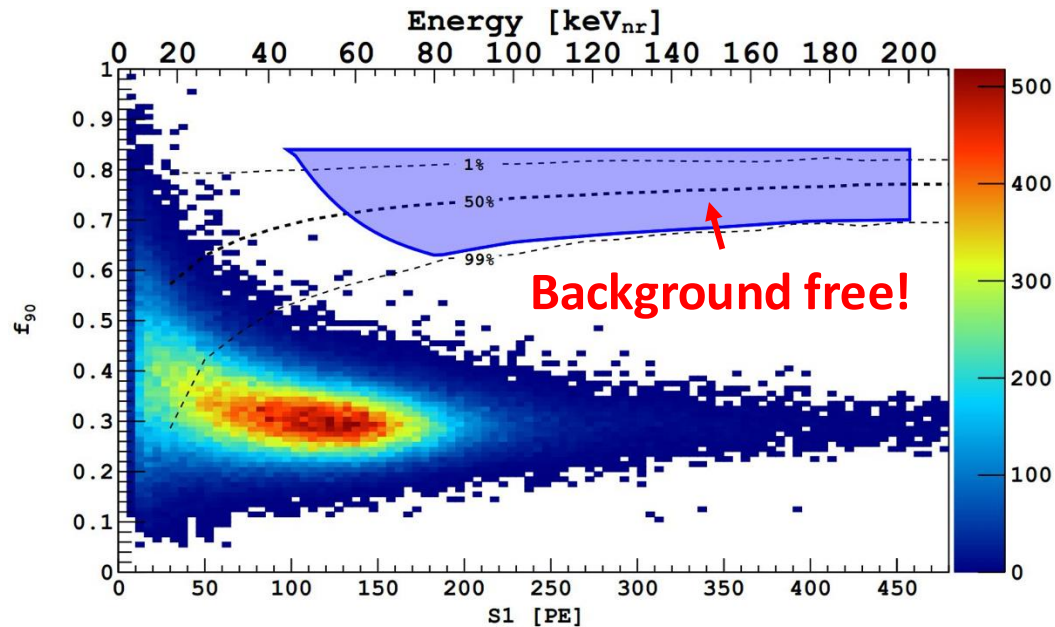
Institute of High Energy Physics, CAS

On behalf of the Gd-PMMA working group

LRT 2024, Kraków, Poland, 10/04/2024

Motivation

- DarkSide-50 has achieved background-free results in the search for WIMPs :
 - S1 Pulse Shape Discrimination (PSD);
 - Water Cherenkov Detector (WCD);
 - Liquid Scintillator Veto (LSV).



532 live-days data [Phys. Rev. D 98, 102006 \(2018\)](#)

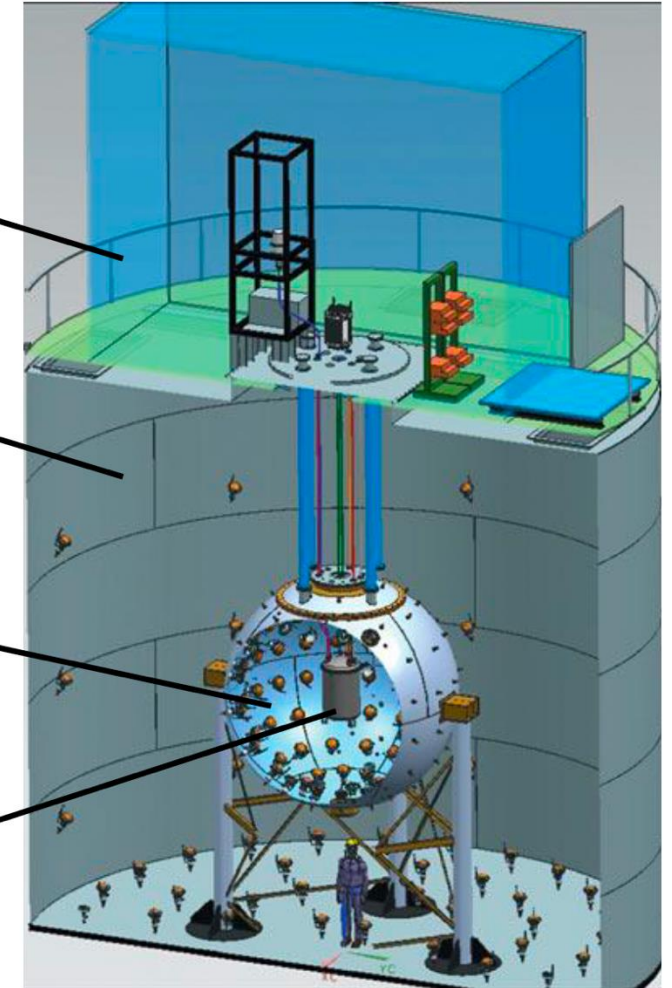
@LNGS Hall C

Radon free clean room

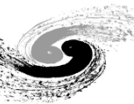
Water cherenkov detector (WCD)

Liquid scintillator veto (LSV)

TPC

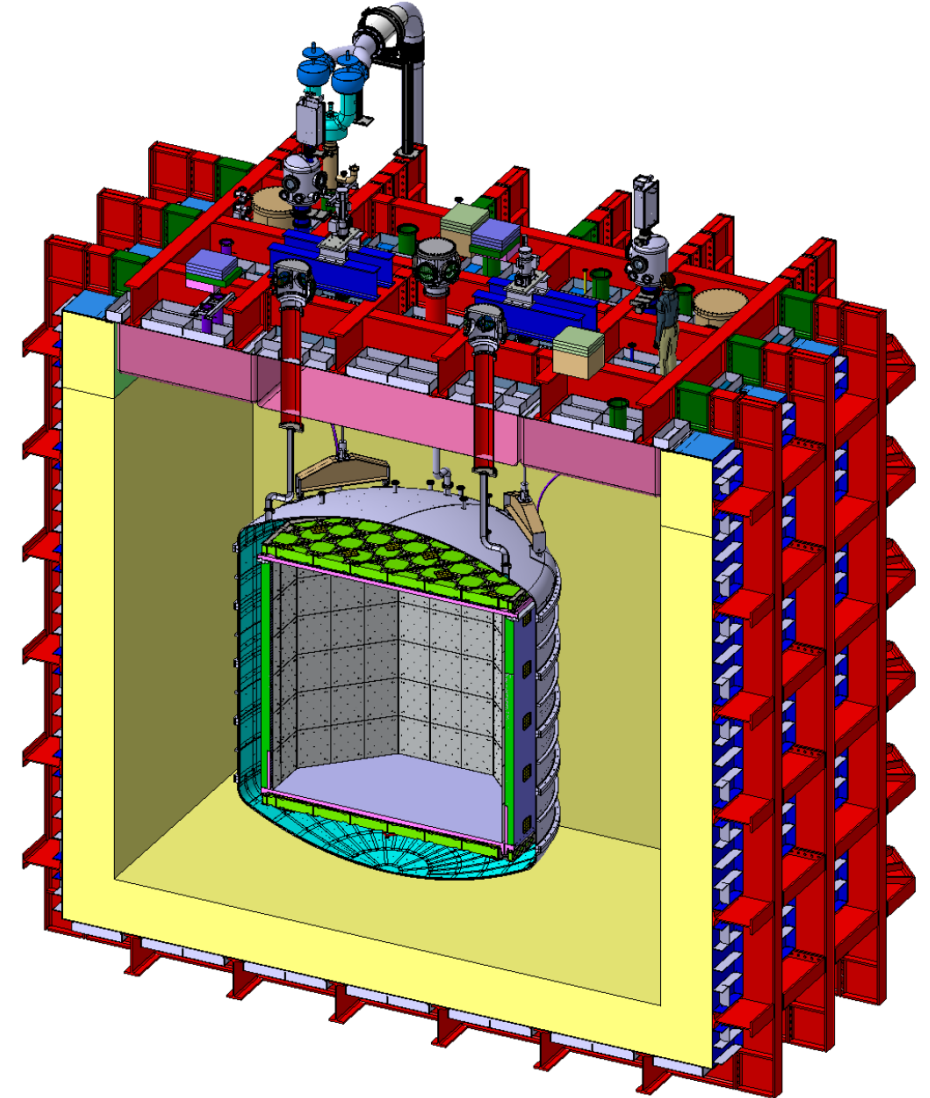


A strategy for rejecting neutrons is needed for the next generation of experiments.



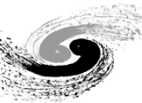
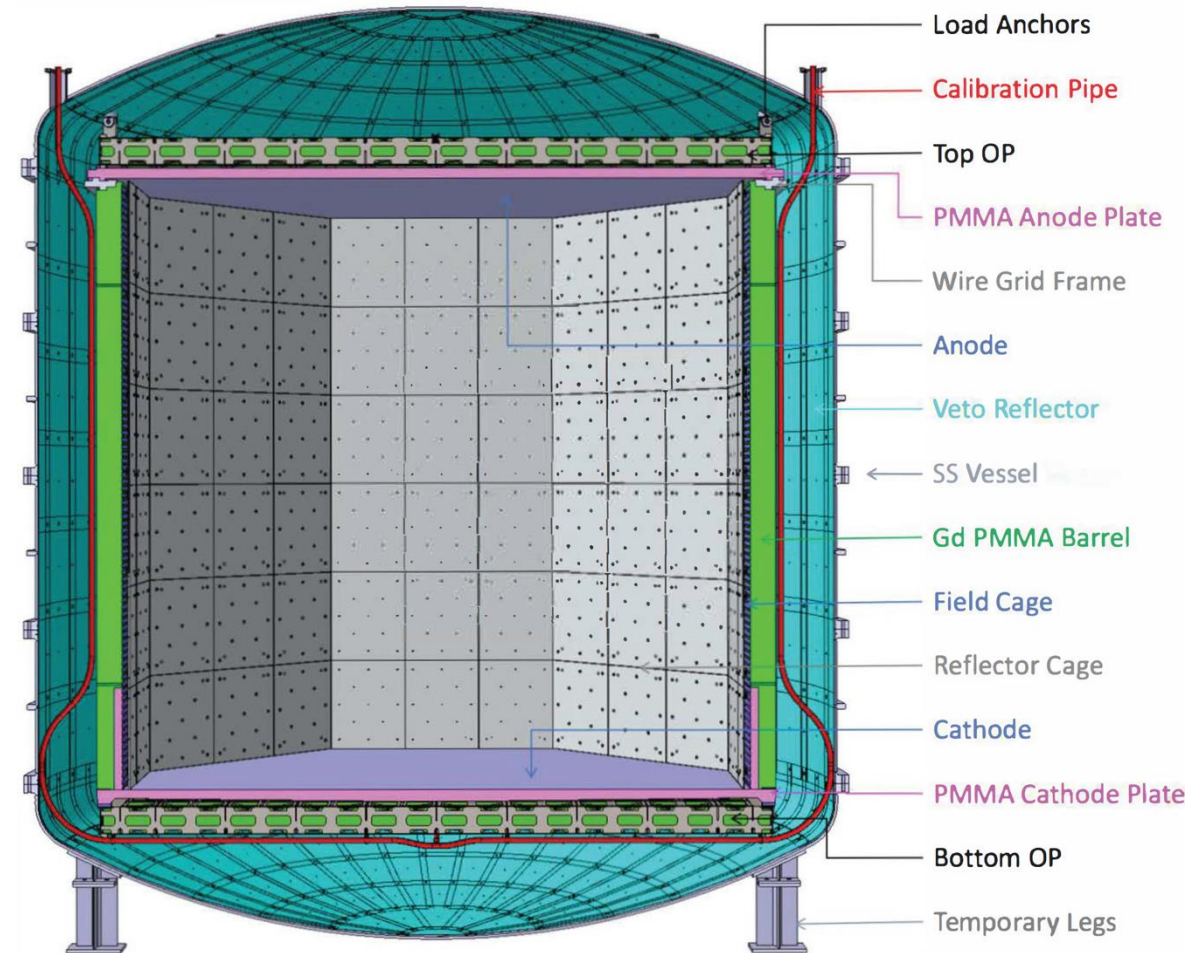
Neutron Veto Strategies Considered

- Several neutron veto strategies has been considered for the next generation WIMP search experiment with argon (DarkSide-20k):
 - Load CH_4 in argon (atmospheric);
 - Gadolinium-doped acrylic in atmospheric argon;
 - **Gadolinium-doped acrylic in underground argon.**



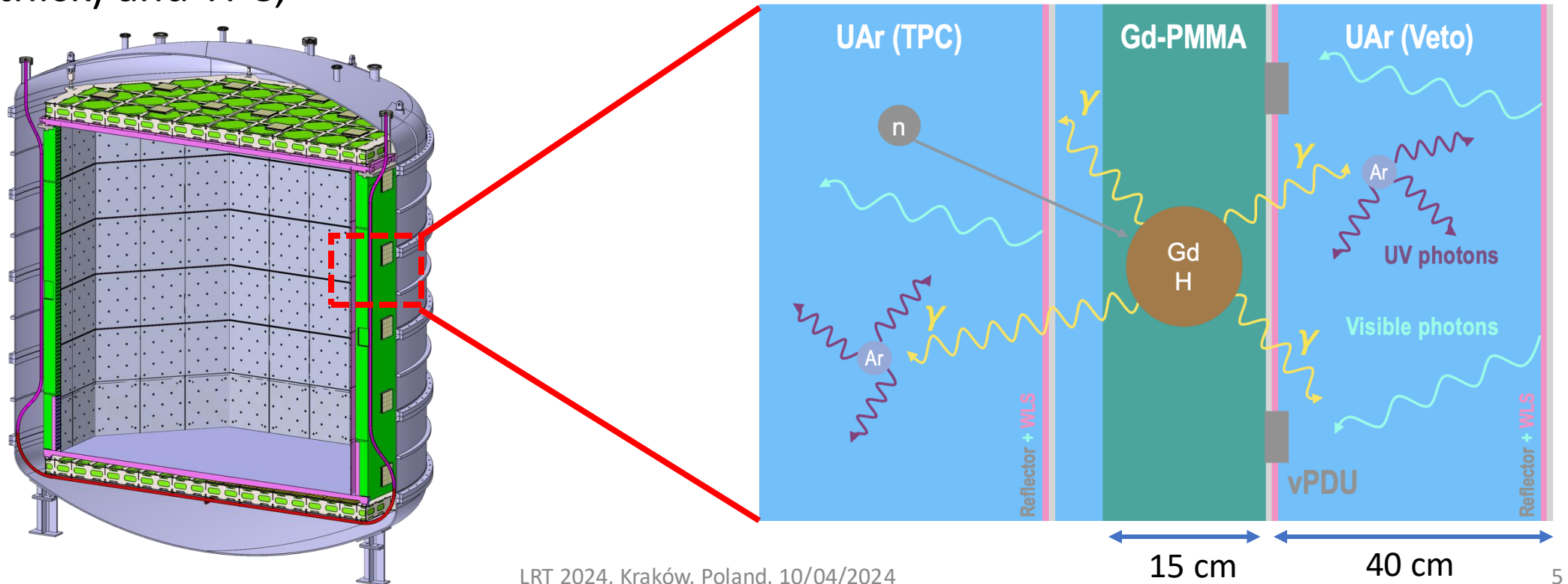
Dual-phase Ar TPC with Neutron Veto

- Gadolinium-doped acrylic is considered a solid material suitable for building the detector.
- Proposed design of a O (50) tonnes dual-phase Ar TPC integrated with a neutron veto.
- SiPM as the photosensor.
- Neutron Veto:
 - Gd-PMMA for TPC main structure, top and bottom endcaps;
 - TPC active volume 4π covered by Gd-PMMA;
 - Ar buffer in the veto volume;
 - Ar in the TPC.



Active Neutron Veto

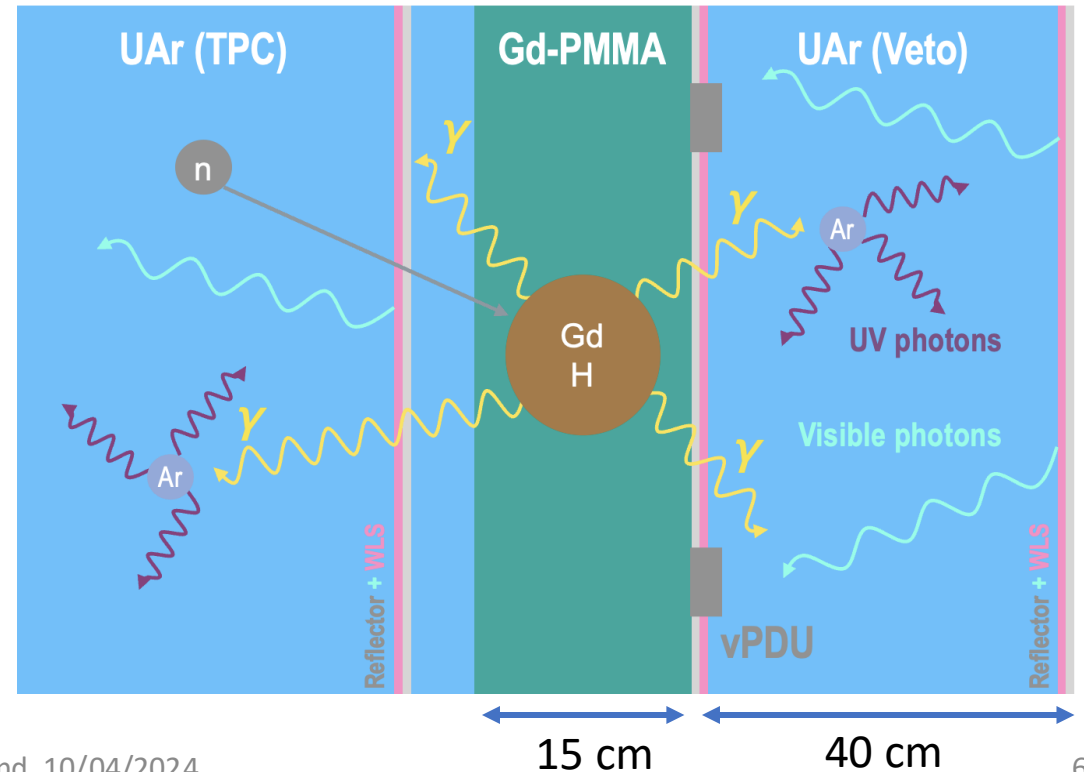
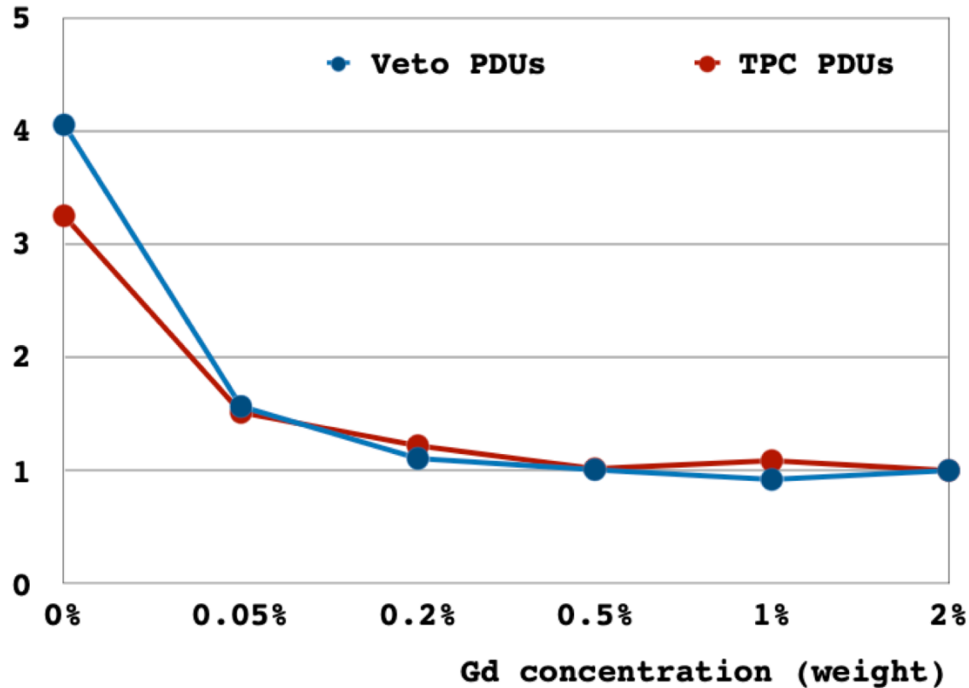
- Gd-PMMA -> Hydrogen + Gadolinium:
 - Hydrogen -> single γ ~ 2.2 MeV;
 - Gadolinium -> multiple γ upto 8 MeV.
- Produced γ rays interact in Ar in both veto buffer (40 cm thick) and TPC;



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- Light yield assumptions:
 - TPC: 10 p.e./keVee;
 - Veto: 2 p.e./keVee.
- Gd concentration: 1 wt%;
- Thickness of Gd-PMMA: ≥ 15 cm;
 - Neutron tagging inefficiency $\sim 5\%$.



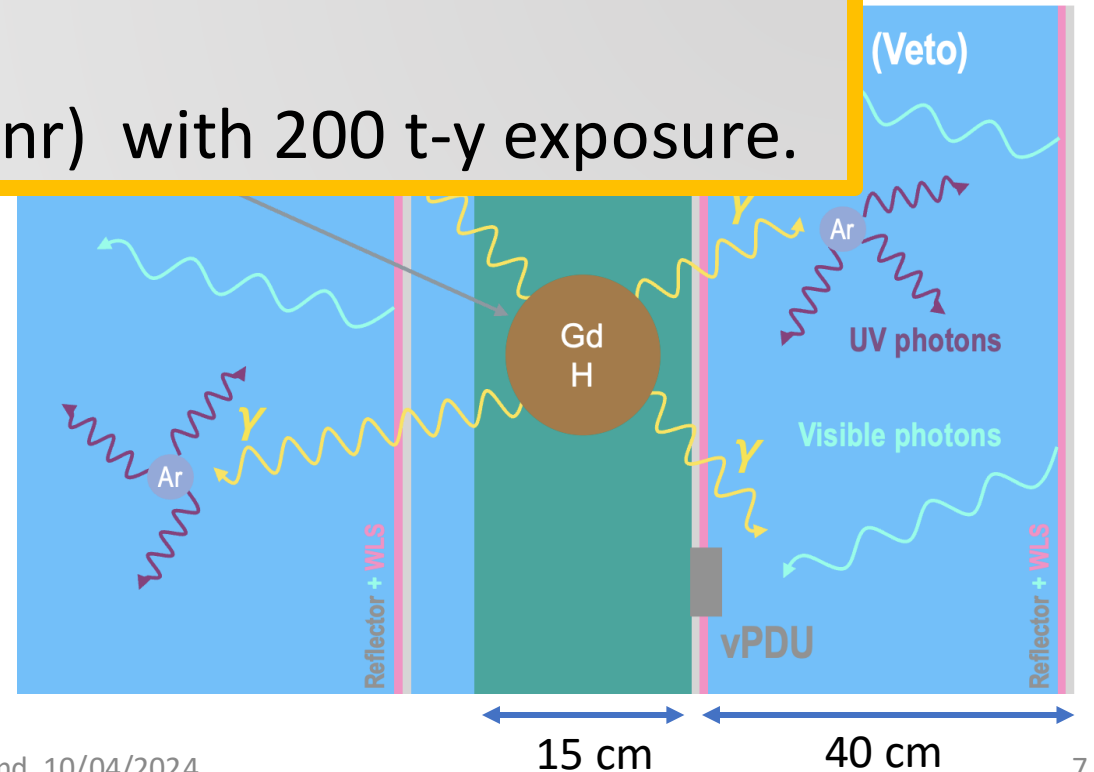
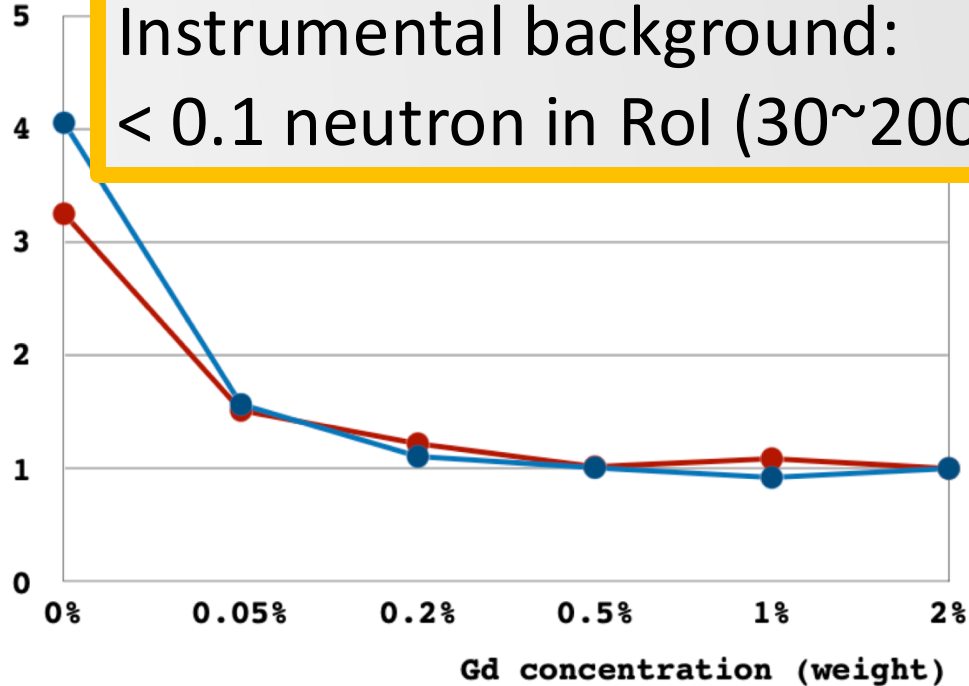
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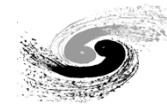
- Product
- (40 cm

For the case of DarkSide-20k, with Gd-PMMA 4-pi covered. Instrumental background: < 0.1 neutron in RoI (30~200 keVnr) with 200 t-y exposure.



The Development of Gd-PMMA

- Recipe development;
- Radiopurity;
- Industrialization;
- Residual stress & Annealing.



Development of Recipe

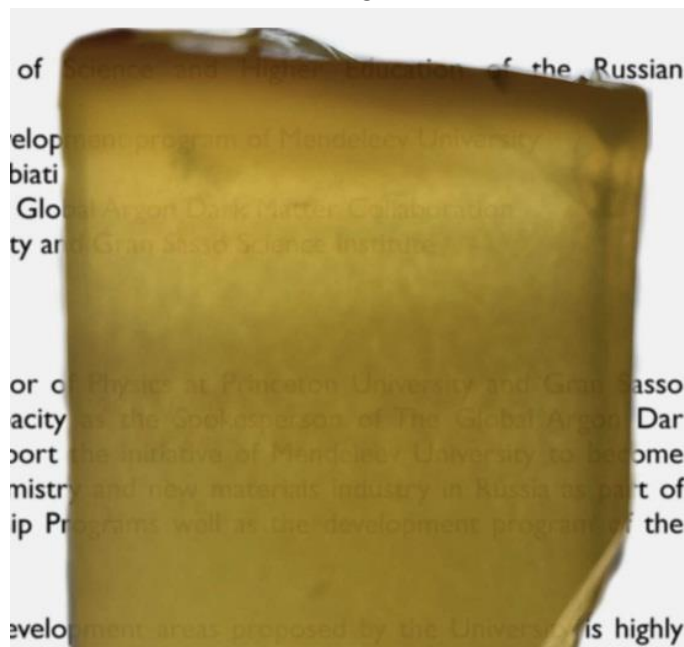
- Three approaches have been developed for Gd-PMMA:
 - Gd_2O_3 recipe: mechanically mix Gd_2O_3 nano grain with MMA for polymerization;
 - $Gd(acac)_3$ recipe: “directly” dissolve $Gd(acac)_3$ into MMA for polymerization;
 - $Gd(MAA)_3$ recipe: “indirectly” dissolve $Gd(MAA)_3$ into MMA for polymerization.

Gd_2O_3 recipe



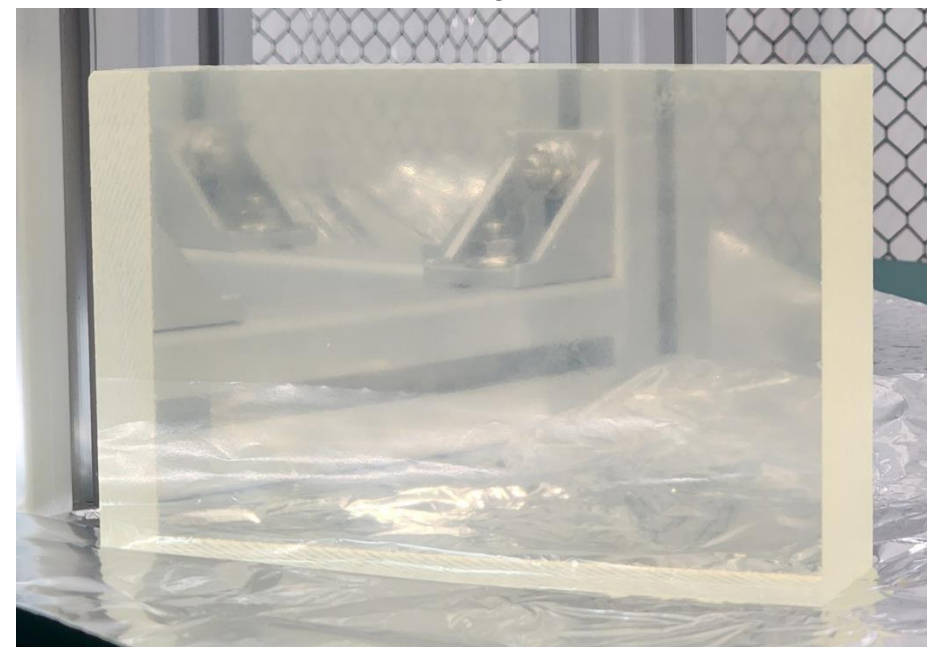
2024 JINST 19 P09021

$Gd(acac)_3$ recipe

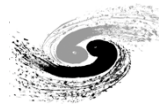


Materials 2021, 14, 3757

$Gd(MAA)_3$ recipe



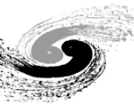
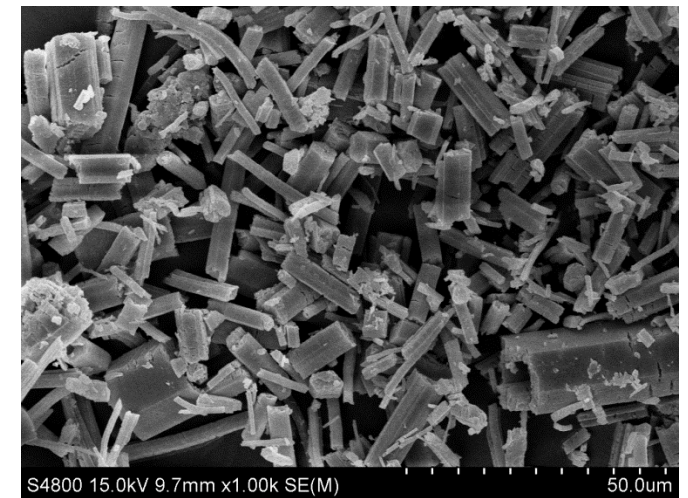
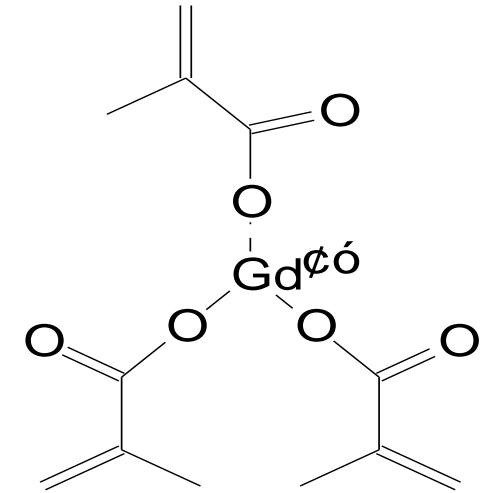
Rare metals 2010 34(4):568-573



Gd(MAA)₃

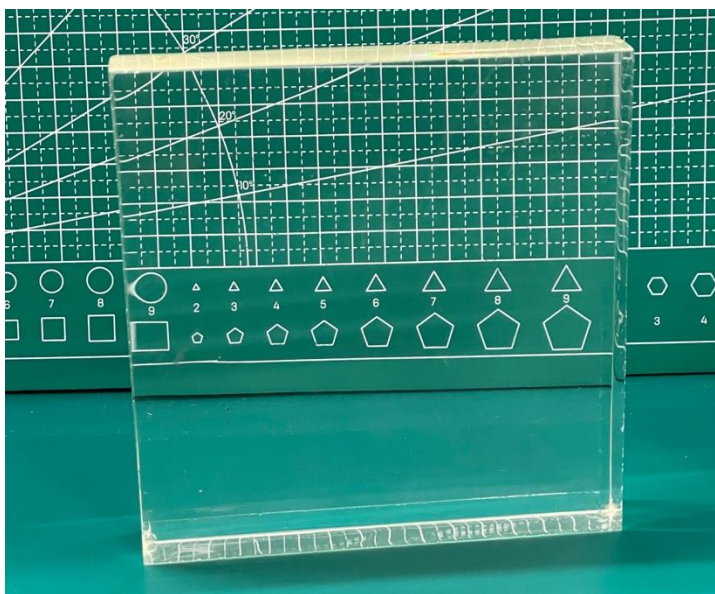
- Gadolinium methacrylate;
- It can be dissolved in liquid MMA monomer using a dedicated dissolving recipe;
- Good chemical stability due to the molecular connection between Gd(MAA)₃ and MMA;
- **Max.** ~30% Gd(MAA)₃ can be dissolved in liquid MAA (~10% Gd by mass fraction).

The dissolving recipe was developed by Yangzhou University & IHEP:

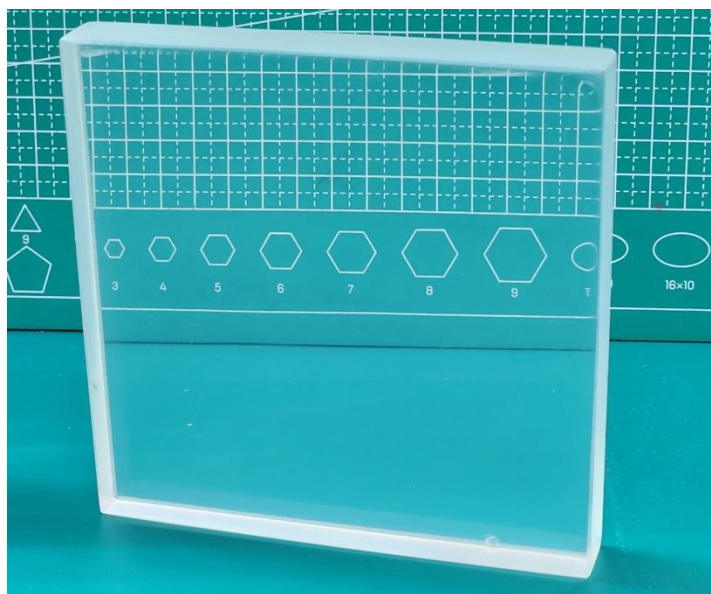


Gd-PMMA

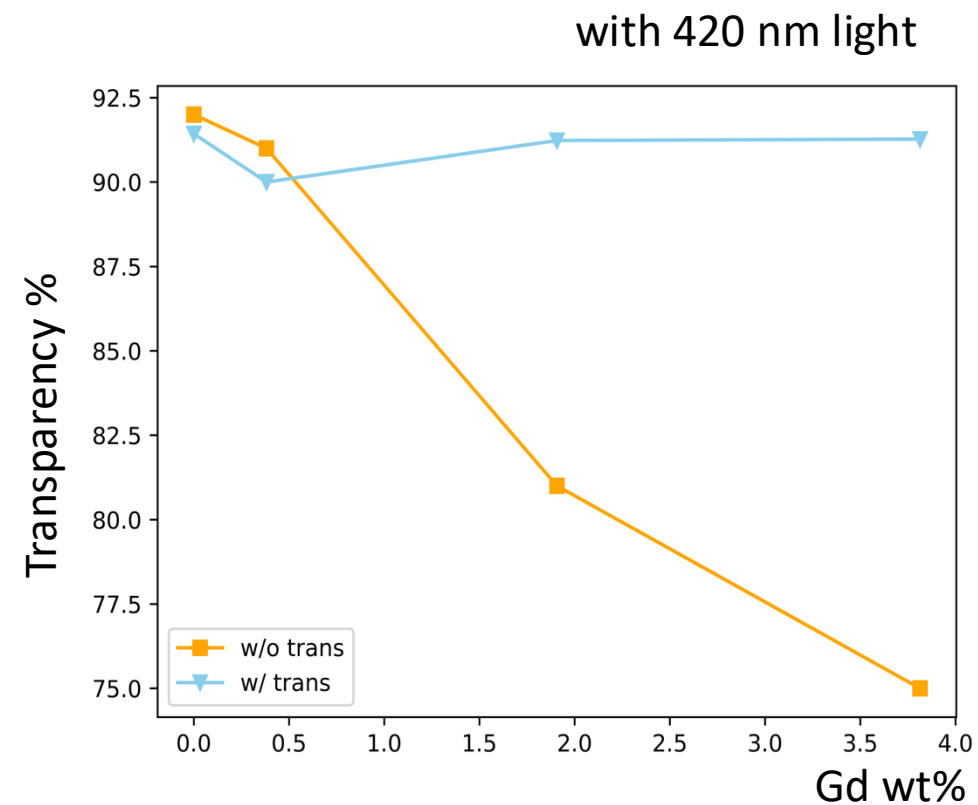
- Polymerization of $\text{Gd}(\text{MAA})_3$ doped liquid MMA;
- A dedicated initiator recipe was developed to prevent self-inhibition and implosion during polymerization;
- High optical transparency is achievable.



High optical transparency



Normal optical transparency



Radiopurity Control

- PMMA is essentially pure, thanks to the radiopure PMMA production line developed by Donchamp for the JUNO experiment;
- 5N Gd₂O₃ from ShinEtsu is selected for Gd-PMMA production for low background.

JUNO PMMA

Isotopes	mBq/kg
Th232_Ra228	< 0.14
Th232_Th228	< 0.078
U238_Ra226	0.05 ± 0.02
U238_Th234	< 2.1
U238_Pa234m	< 1.8
U235	< 0.07
K40	< 0.41
Cs137	< 0.025

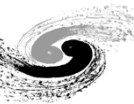
ShinEtsu 5N Gd₂O₃

Isotopes	mBq/kg
Th232_Ra228	< 0.5
Th232_Th228	0.4 ± 0.1
U238_Ra226	0.5 ± 0.1
U238_Th234	< 33
U238_Pa234m	< 7.4
U235	< 0.31
K40	4 ± 1
Cs137	< 0.079

Gd-PMMA (under validation)

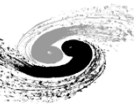
Isotopes	mBq/kg
Th232_Ra228	< 0.33
Th232_Th228	< 1
U238_Ra226	< 1.3
U238_Th234	< 8.3
U238_Pa234m	< 49
U235	< 0.54
K40	< 11
Cs137	< 0.12

Radiopurity assay is still ongoing at LNGS.



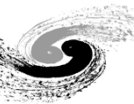
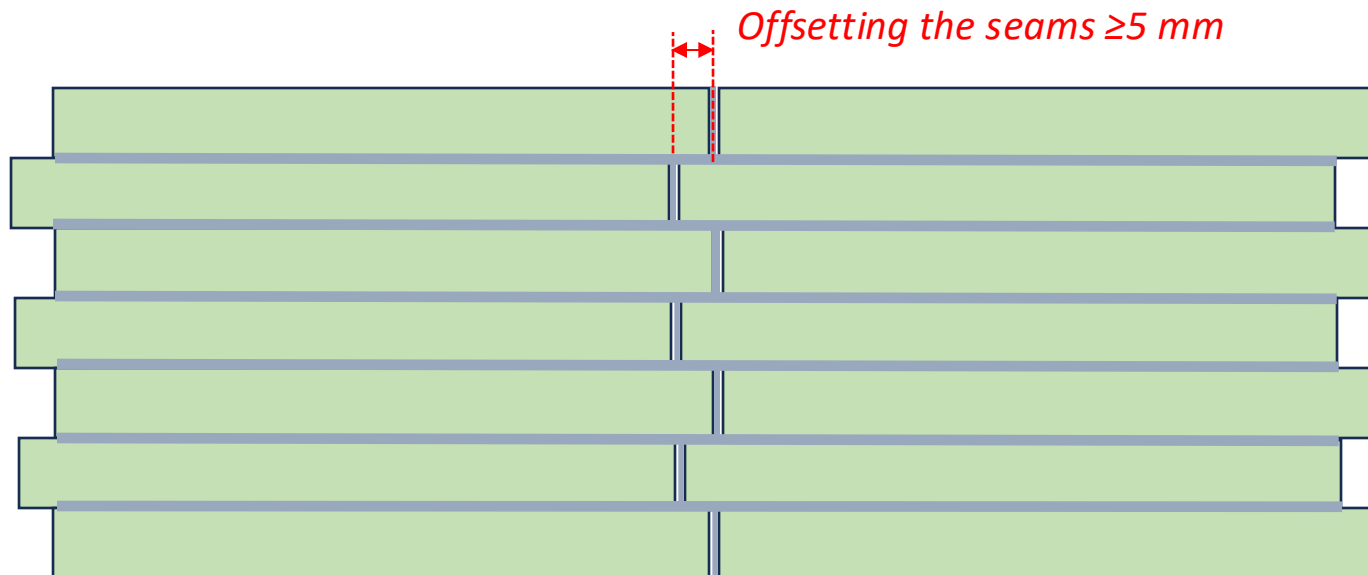
Production of Large Dimension Panels

- Developed recipe has been implemented to an industrial scale production line.
- Dimension options for single-cast sheet (largest so far):
 - For high optical transparency: 100 cm x 70 cm x 4 cm;
 - For normal optical transparency: 200 cm x 200 cm x 2 cm.



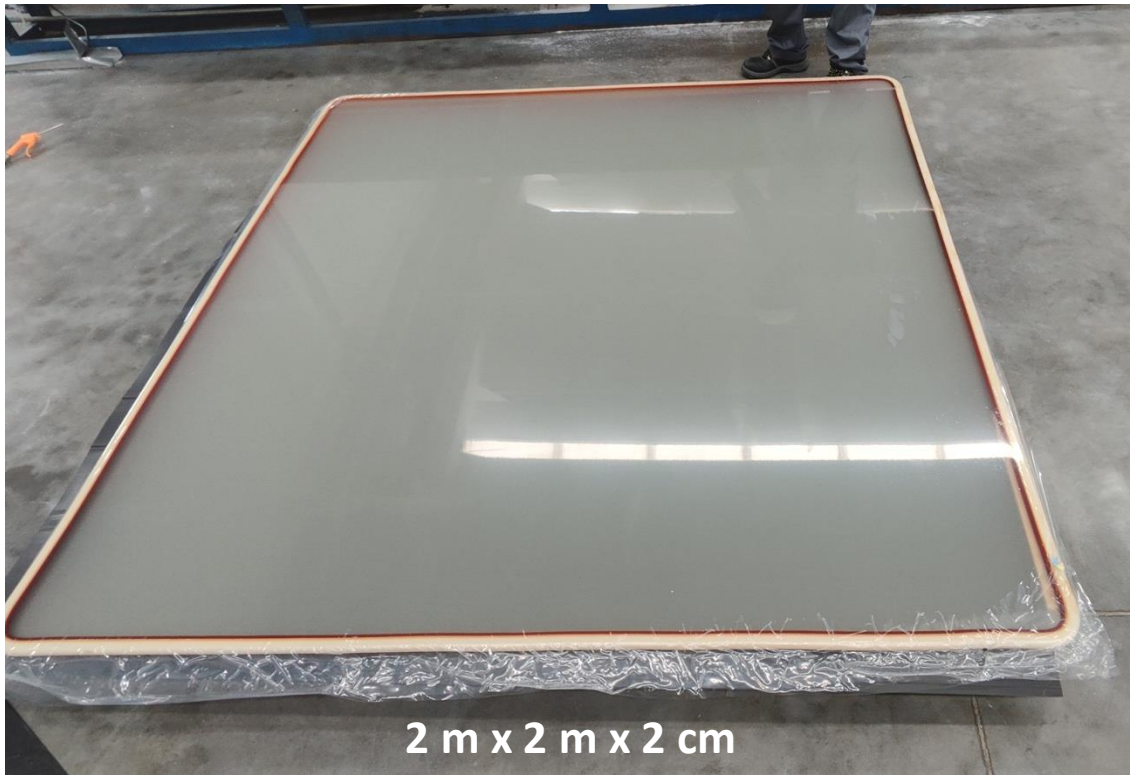
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 - For high optical transparency: 100 cm x 70 cm x 4 cm;
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- A bonding and lamination procedure has been developed. The adhesive uses the same ingredients as Gd-PMMA, specifically $\text{Gd}(\text{MAA})_3$ dissolved in MMA.

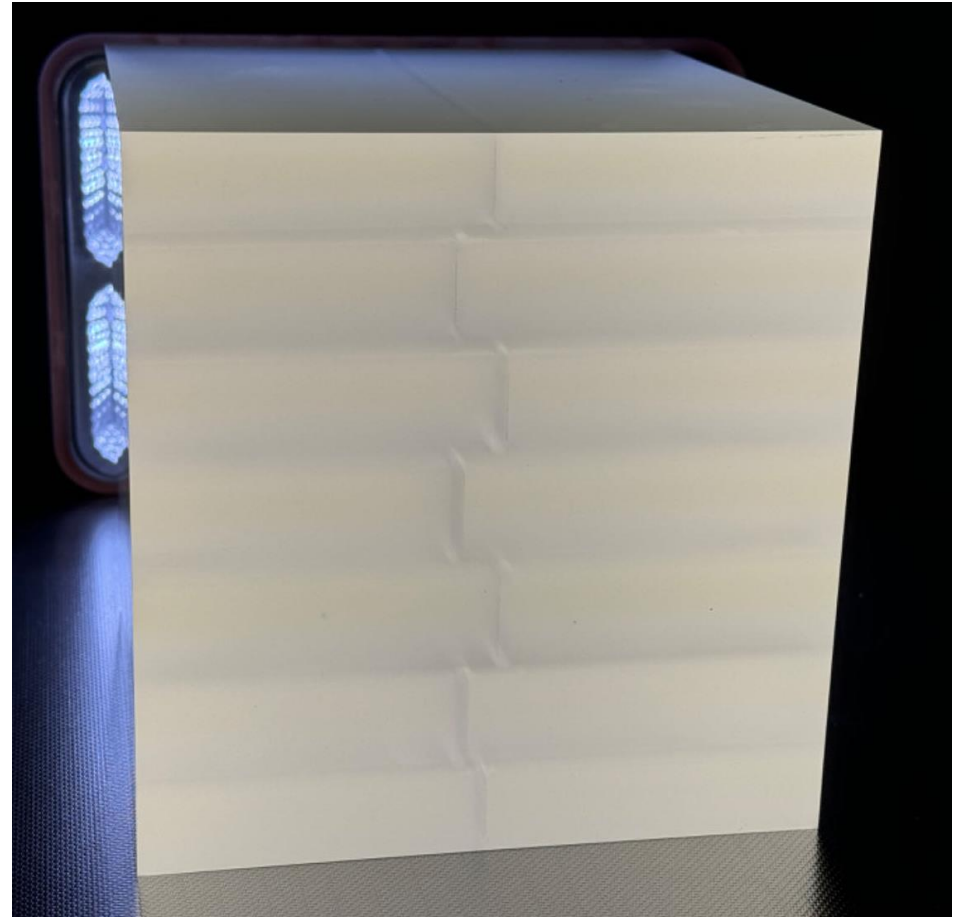


Production of Large Dimension Panels

- The first full-dimension Gd-PMMA single-cast panel, was manufactured at Donchamp in China in spring 2024.



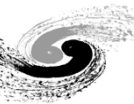
- Several 15 cm³ bonded & laminated cubes are at IHEP for annealing and cooling tests.



- A relatively large bonded & laminated sample, measuring 80 cm x 80 cm x 16.4 cm, has been produced. This sample will be used to study the large piece anneal at the University of Alberta.

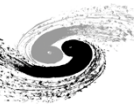
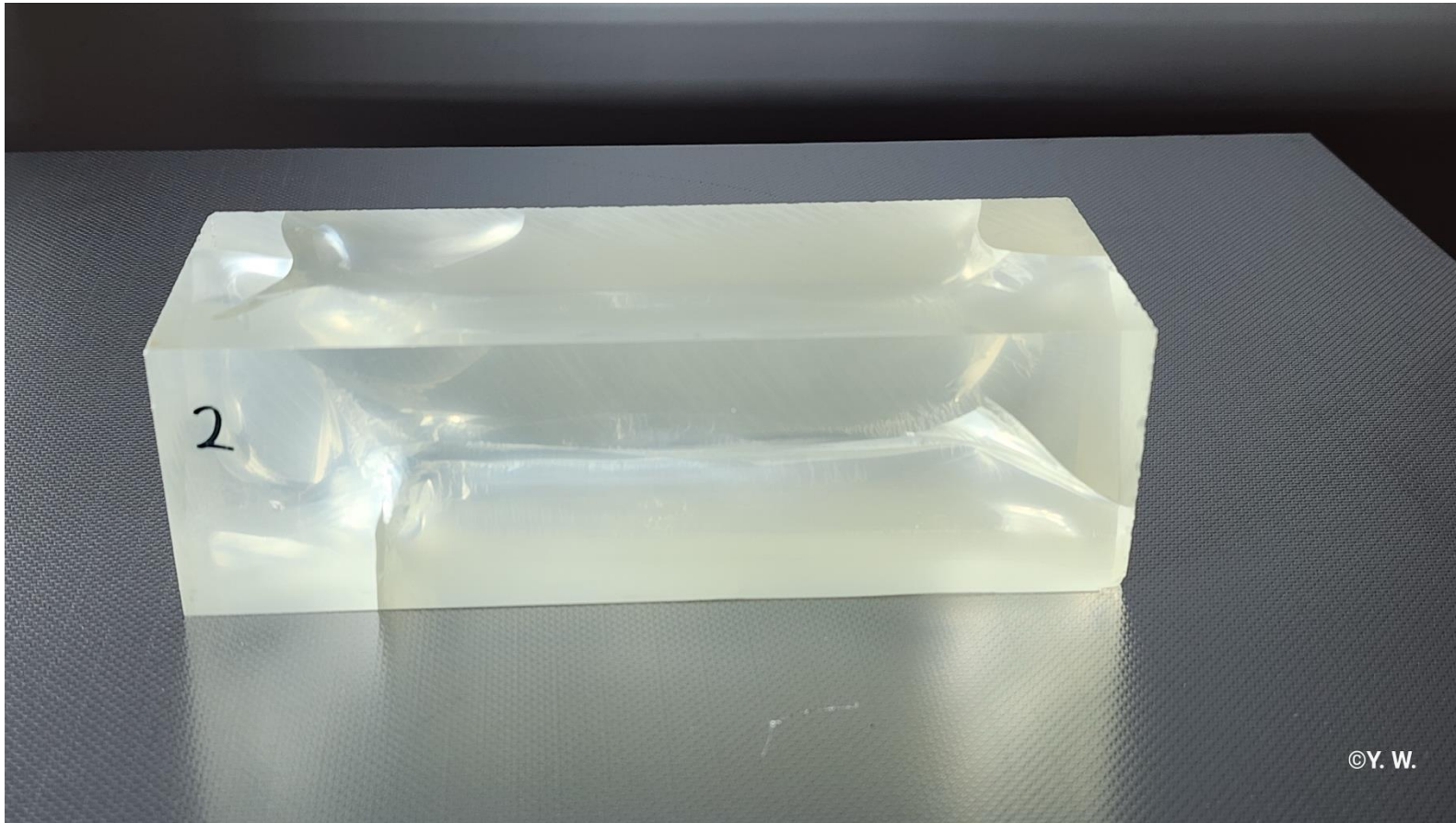


- The next step is finalizing the annealing procedure, which is the most important step for mitigating residual stress in applications at low temperatures.



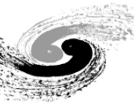
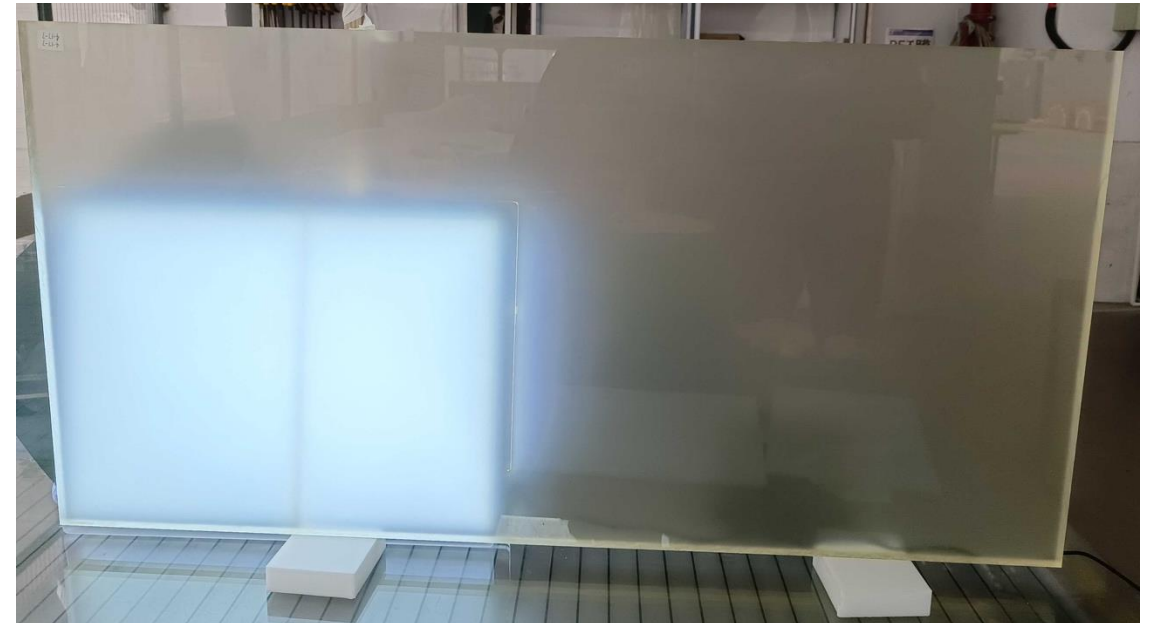
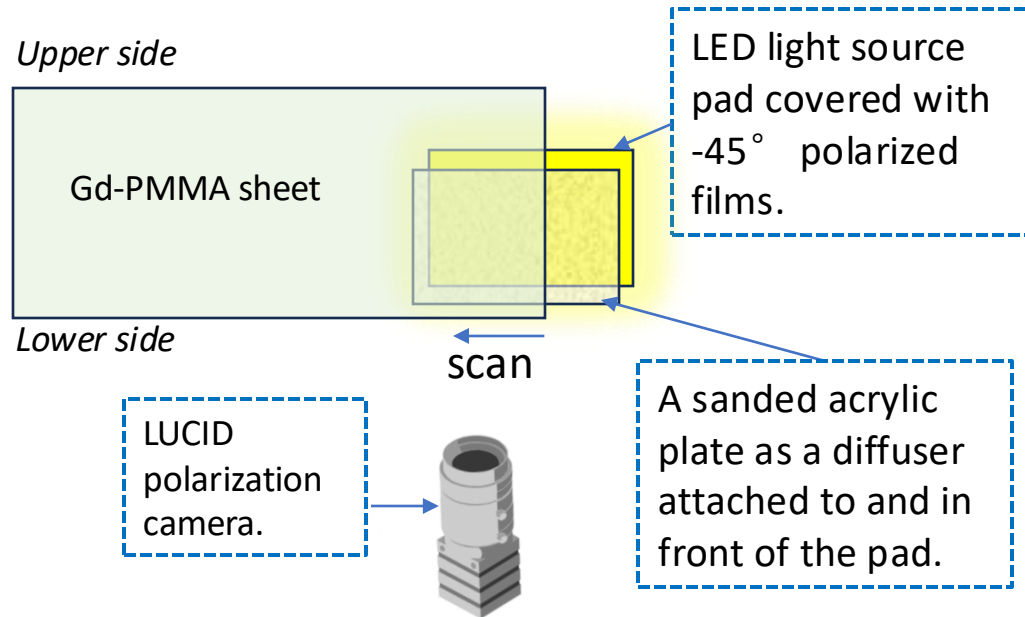
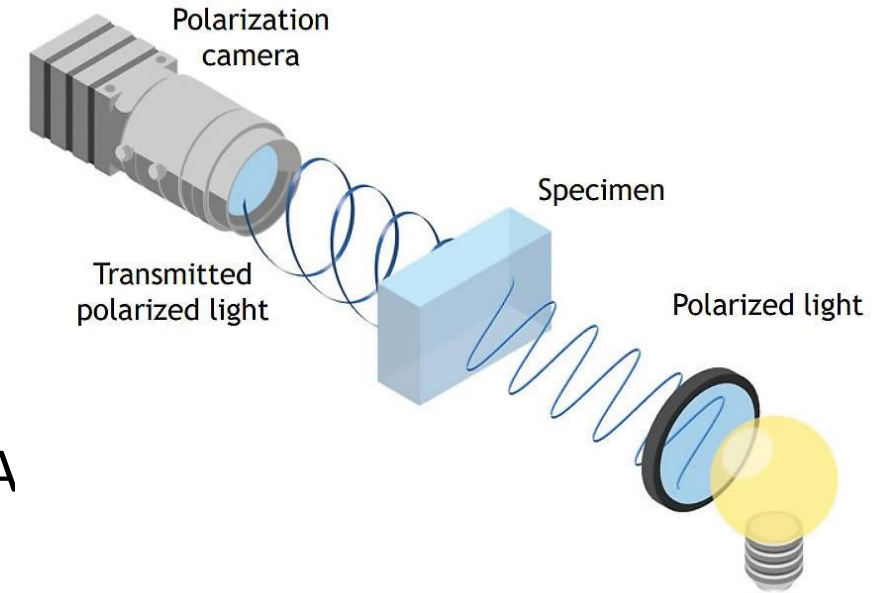
What if there are stresses ?

- Acrylic will crack if we do not carefully manage the stresses.



Stress Measurement

- Use a polarimeter to measure the stress.
- A polarization camera is not capable of scanning large areas.
- A segmented setup is used for scanning large Gd-PMMA sheets.

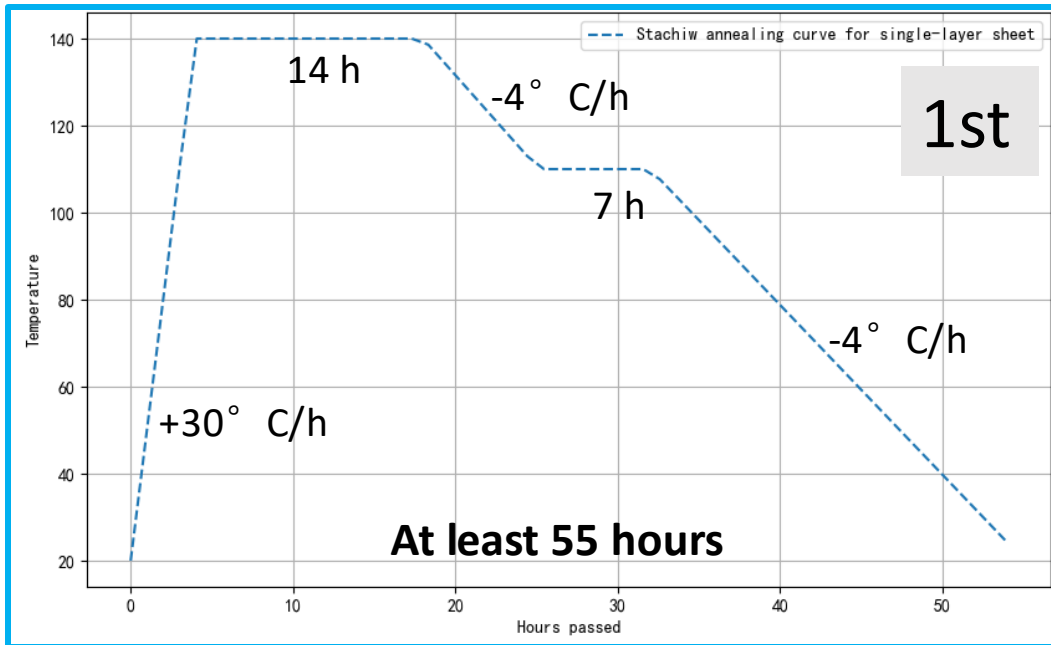


Annealing

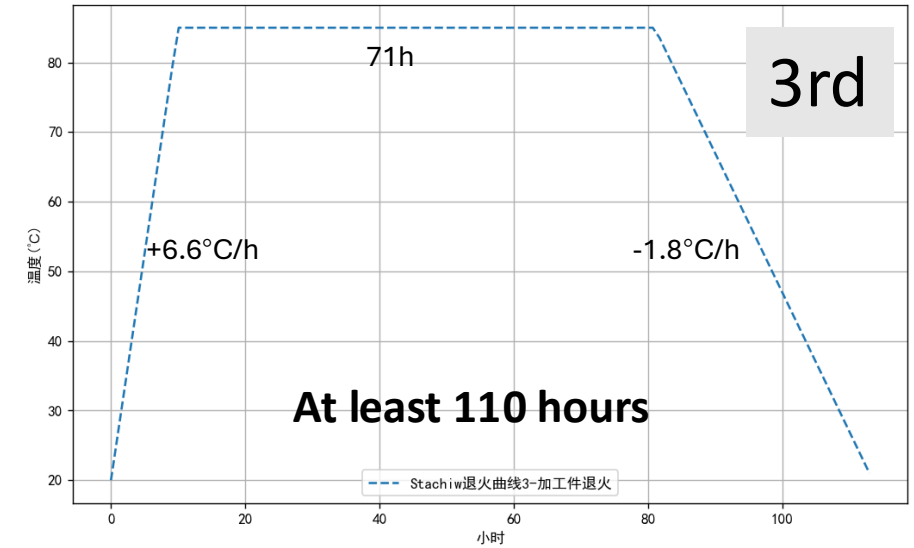
- 1st annealing for single-cast sheet;
- 2nd annealing for bonded & laminated panel;
- 3rd annealing after machining.

Stachiw's handbook ISBN 1-930536-15-1

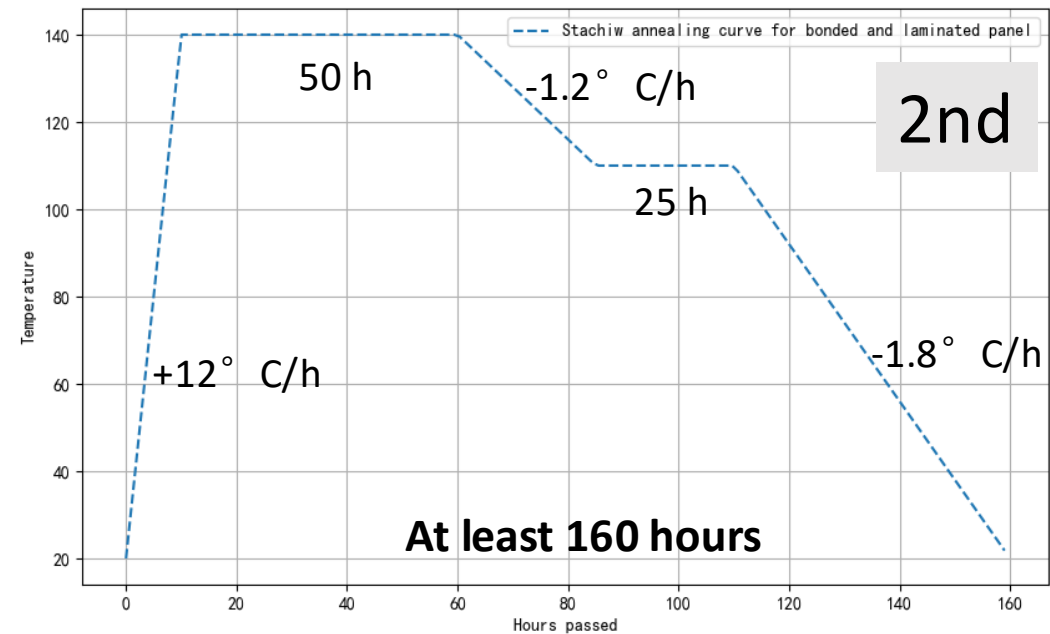
Stachiw annealing curve for single-layer sheet



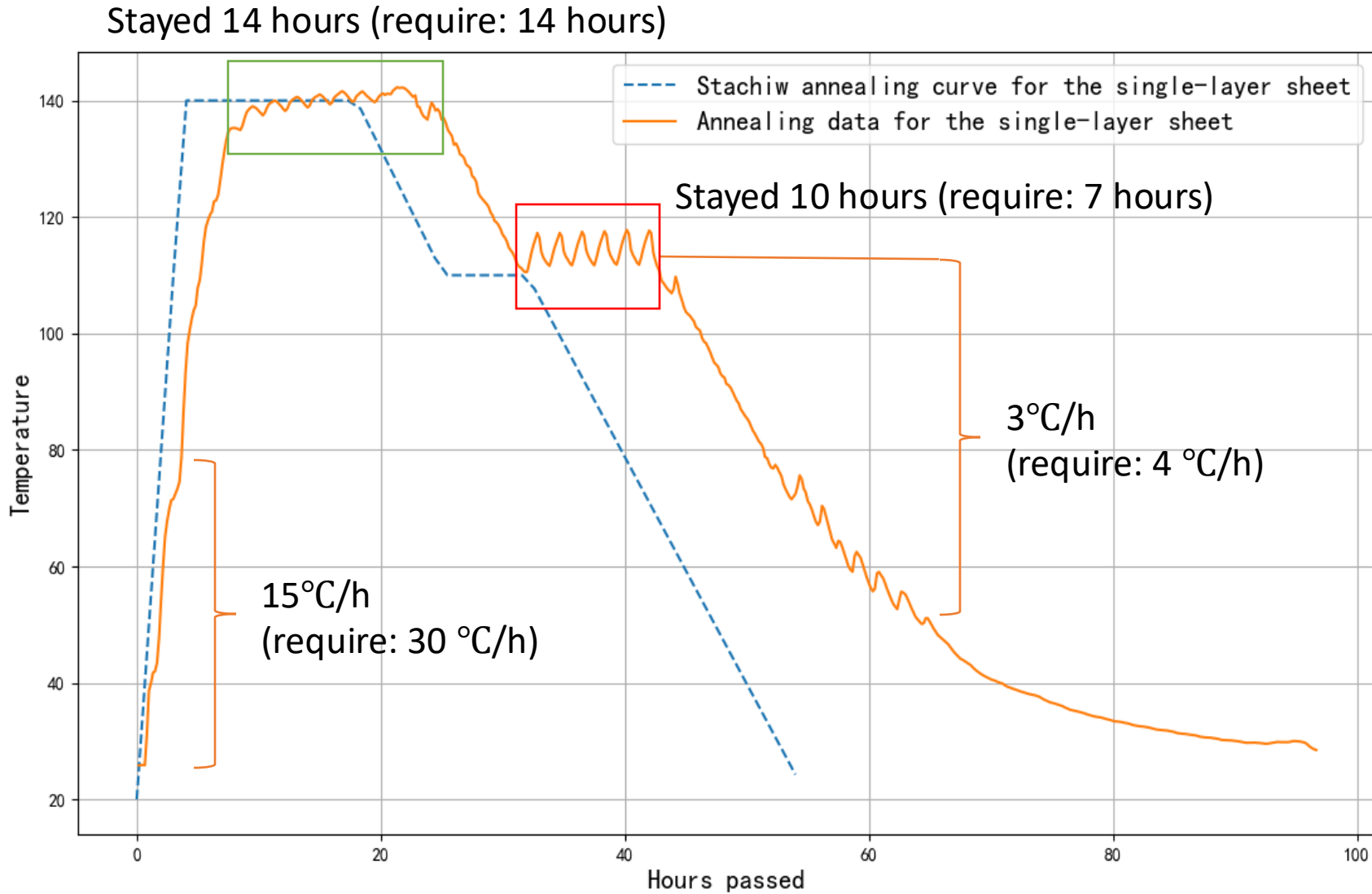
Stachiw annealing curve machined panel



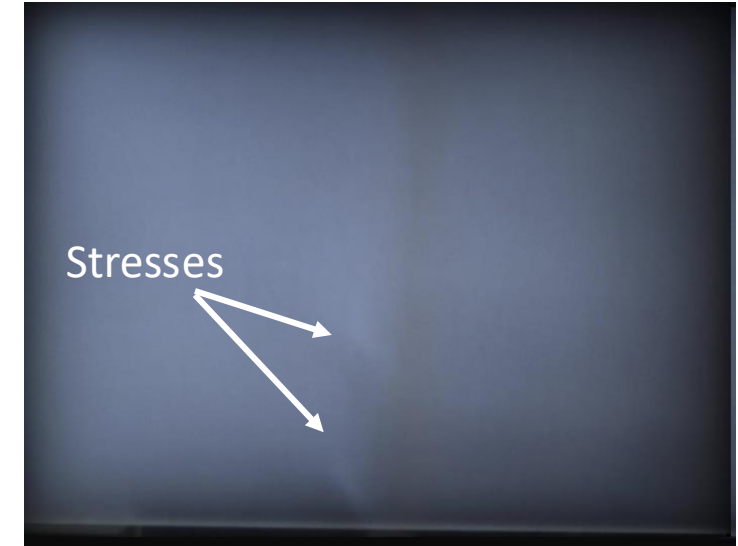
Stachiw annealing curve for bonded & laminated panel



Annealing for Single Sheet



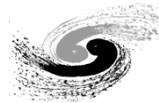
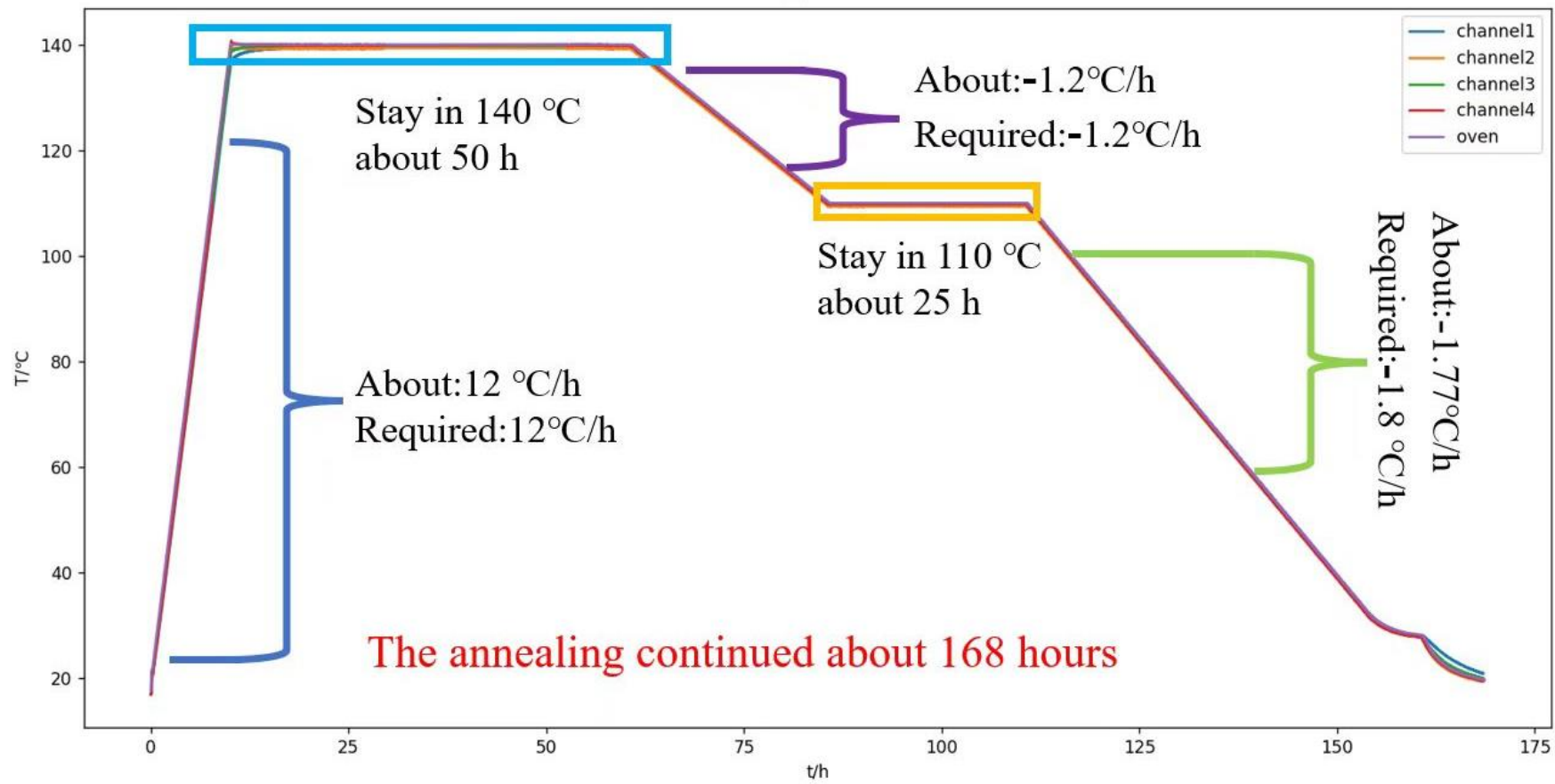
Before annealing



After annealing



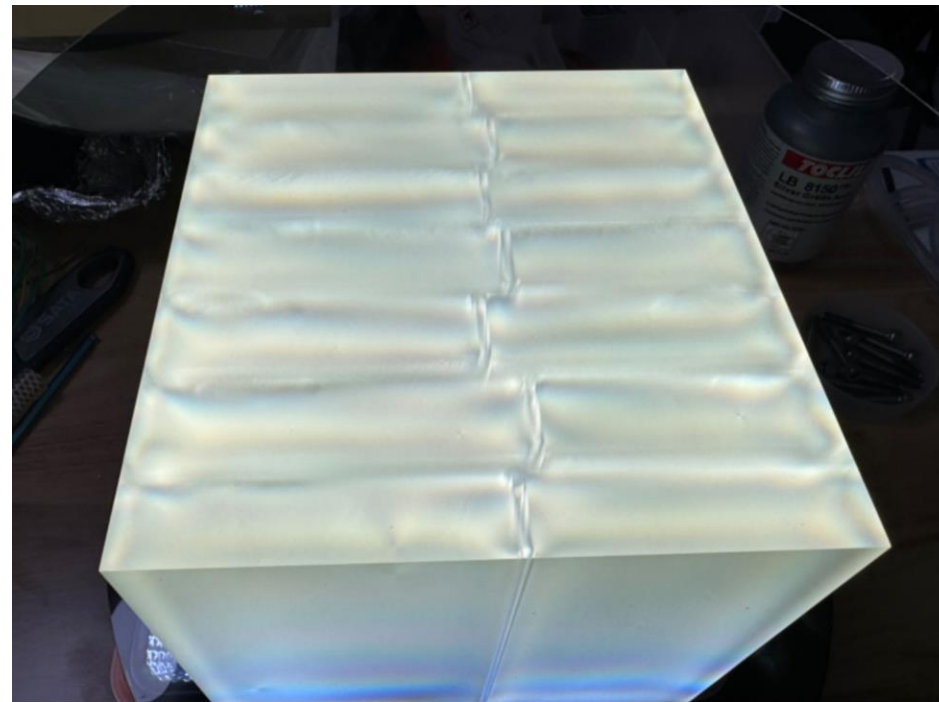
Annealing for 15 cm Thick Piece



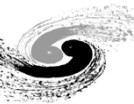
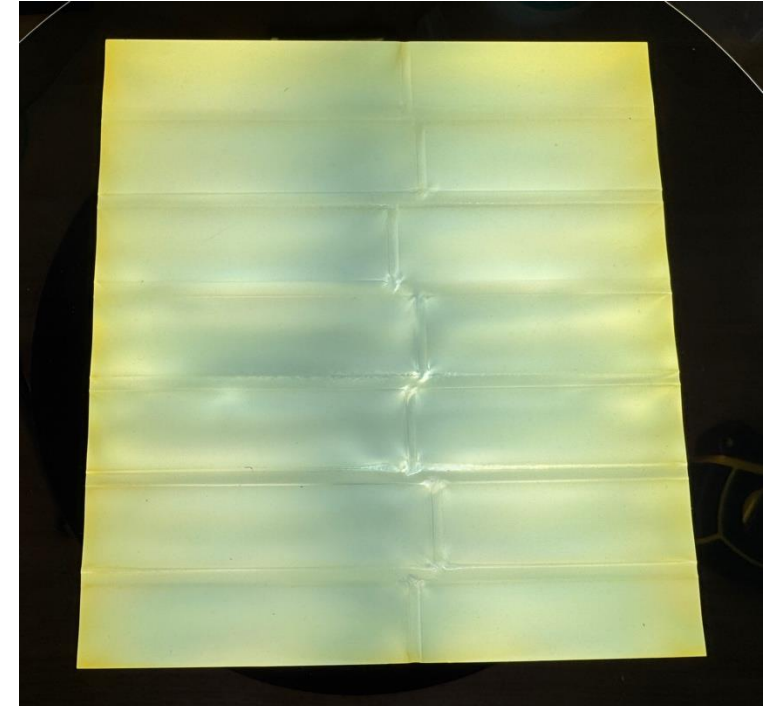
Annealing for 15 cm Thick Piece

- The residual stresses are significantly mitigated after the annealing process.

Before annealing

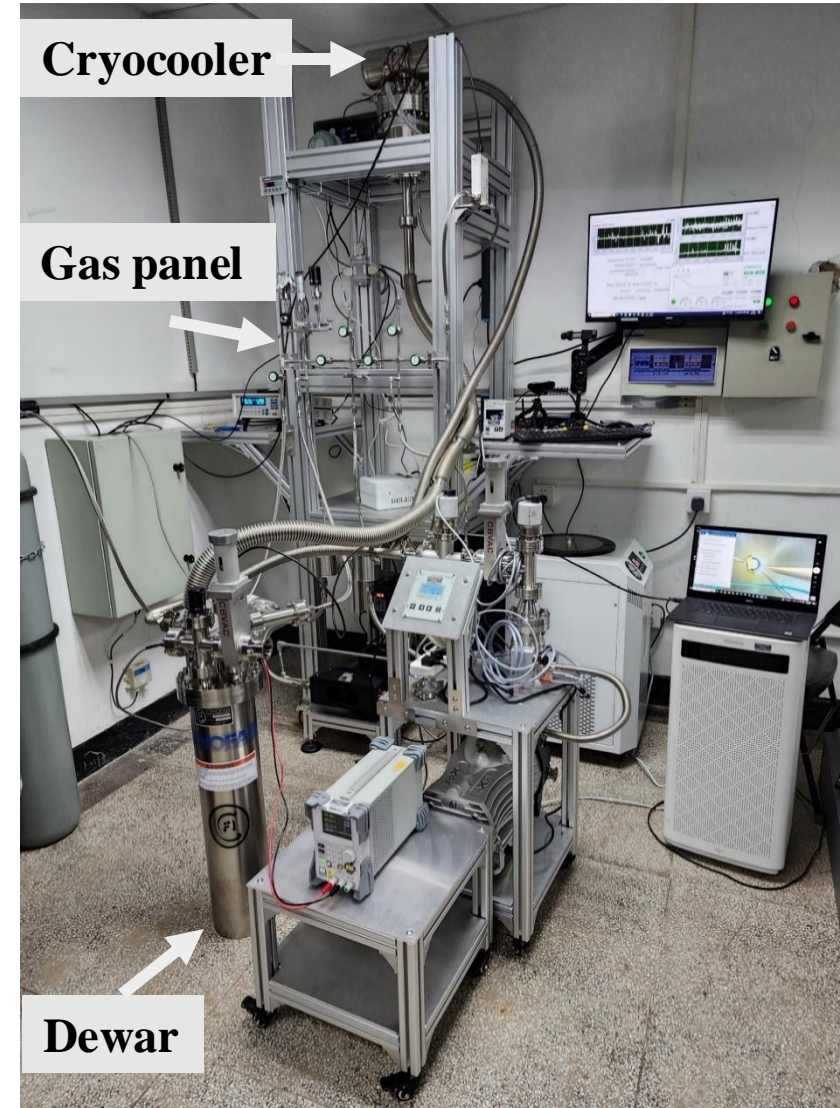
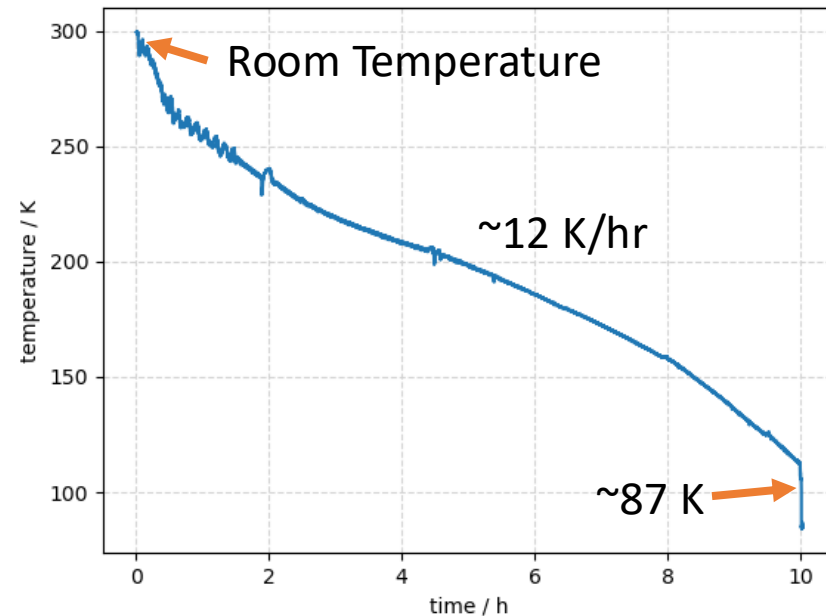


After annealing



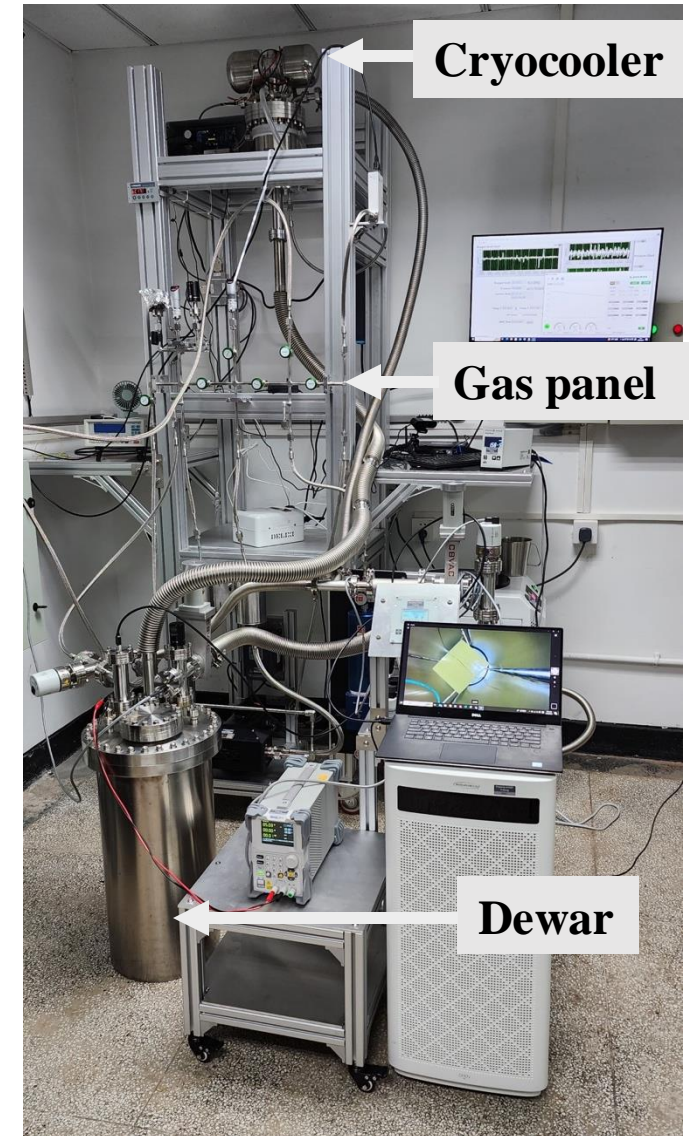
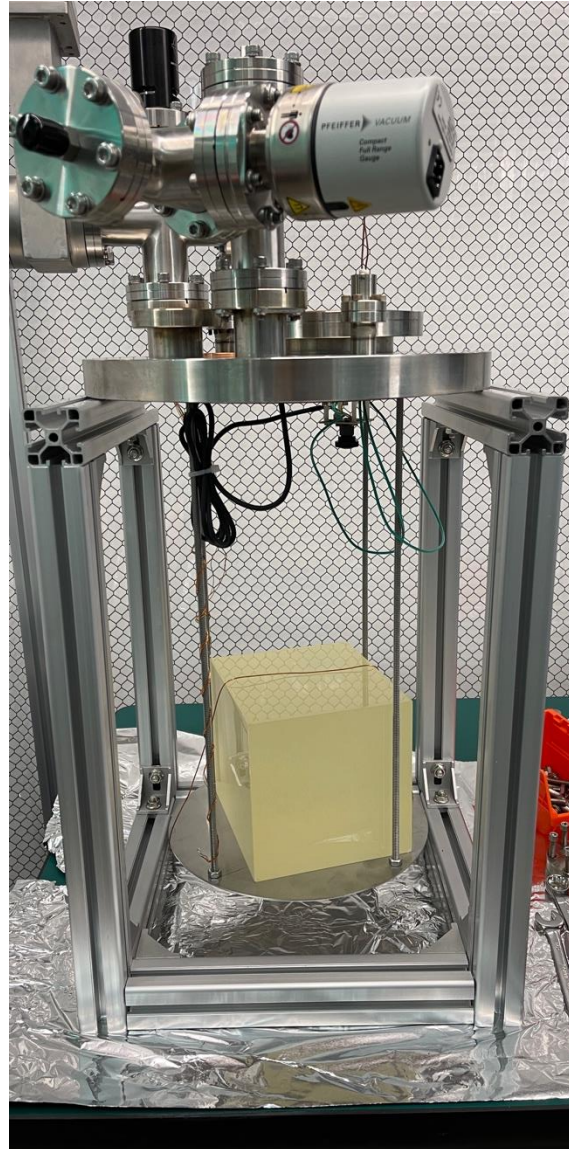
Cooling Test

- Study the cooling speed that can safely put Gd-PMMA in LAr;
- A test cooling procedure has been performed with a two-layer laminated Gd-PMMA sample (not annealed).
- The average cooling rate was ~ 12 K/hr, while the slowest cooling rate of the cryogenic system is ~ 5 K/hr.
- No defects have been observed after the test.



Cooling Test for the 15 cm Thick Sample

- A full thickness sample:
a 15 cm³ cube
- Annealed;
- This test is scheduled to begin soon.



Summary & Discussion

- Dual-phase Ar TPC is a promising technique for background-free DM searches.
- A novel active neutron veto detector with Gd-PMMA was proposed.
- Gd-PMMA, based on the $\text{Gd}(\text{MAA})_3$ recipe, has been successfully developed.
- Industrial-scale production of Gd-PMMA sheets has been validated.
- Extensive studies on stress management and the relevant annealing procedures are being conducted to ensure material reliability.

Beyond Gd-PMMA:

Lead-doped acrylic: successfully developed!

Boron-doped acrylic: development in progress...

Boron-doped plastic scintillator: development in progress...

For more information, please contact wangyi90@ihep.ac.cn.

