# Developing Hydrophobic Silica Aerogels for Cherenkov Detectors



**Makoto Tabata**

**Chiba University**

**Aerogel Factory Co., Ltd.**

Aerogel Factory

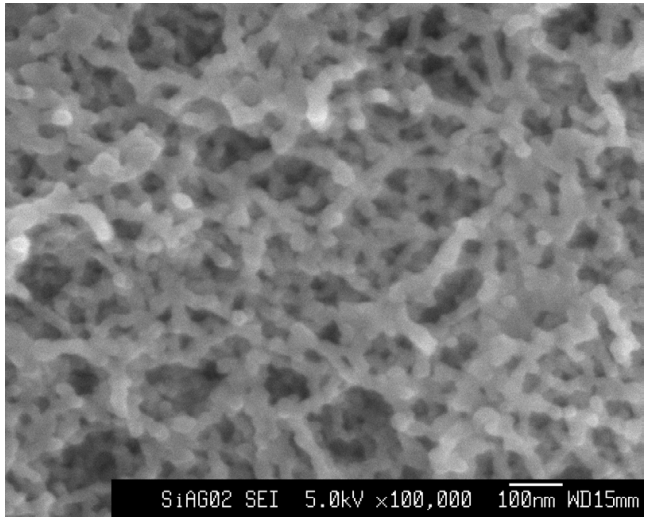


**XII International Workshop on Ring Imaging Cherenkov Detectors (RICH2025)**

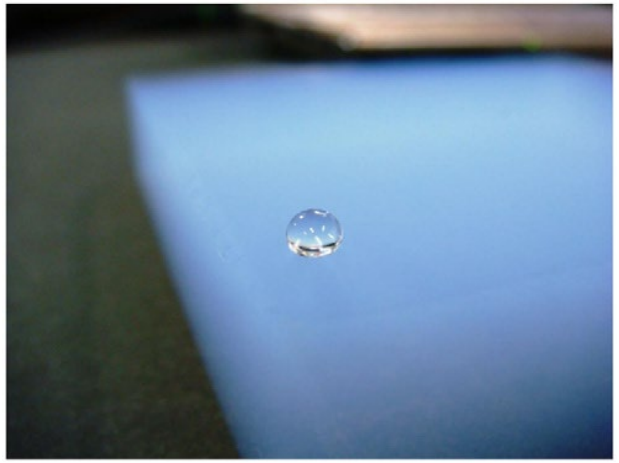
# Outline

- Introduction to Silica Aerogel.
- Review of the achievement of the Belle II aerogels.
- Future direction:
  Lower index & Larger tile production.
  Key technology: Supercritical drying (while eliminating tile cracking).

# Basics of Silica Aerogel



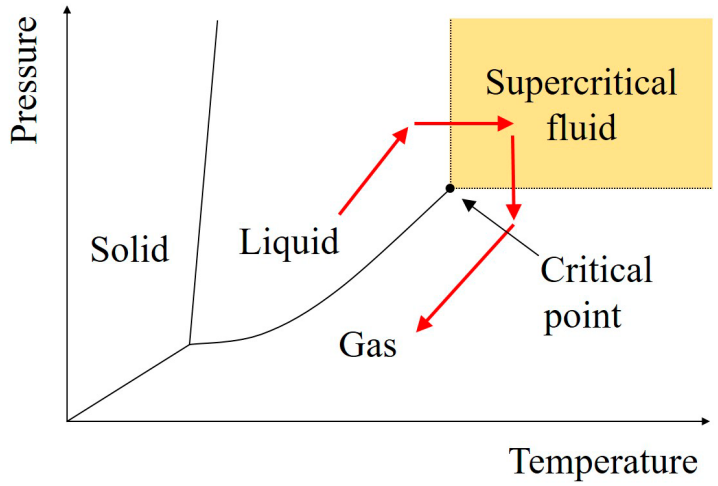

- Nano-structured material made of 3D networks of silica ($SiO_2$) particle clusters.
- Optical properties derived from the distribution of silica clusters and air-filled pores.
- Originally hydrophilic (absorbing environmental moisture).
- One can render aerogels hydrophobic during production by a silane-based reagent.
  Chemical surface modification (not aerogel tile surface but individual silica cluster surface.)
  Including carbon elements derived from methyl groups.
  Can be cut by a water-jet machine.



- Refractive index–density ($\rho$) relationship:
  $n(\lambda)-1=k(\lambda)\rho$
  $k \sim 0.27\ cm^3/g$ for hydrophobic aerogel,
  Slightly depends on the nanostructures.

# Production Procedures

1. Sol–gel polymerization: synthesizing an alcogel block.
2. Aging under an optimized temperature.
3. Hydrophobic modification → Ammonia as a byproduct.
4. Ammonia removal.
   (Still wet alcogel here)
5. Liquid solvent extraction under the supercritical phase.
   (Supercritical “drying” with an autoclave)



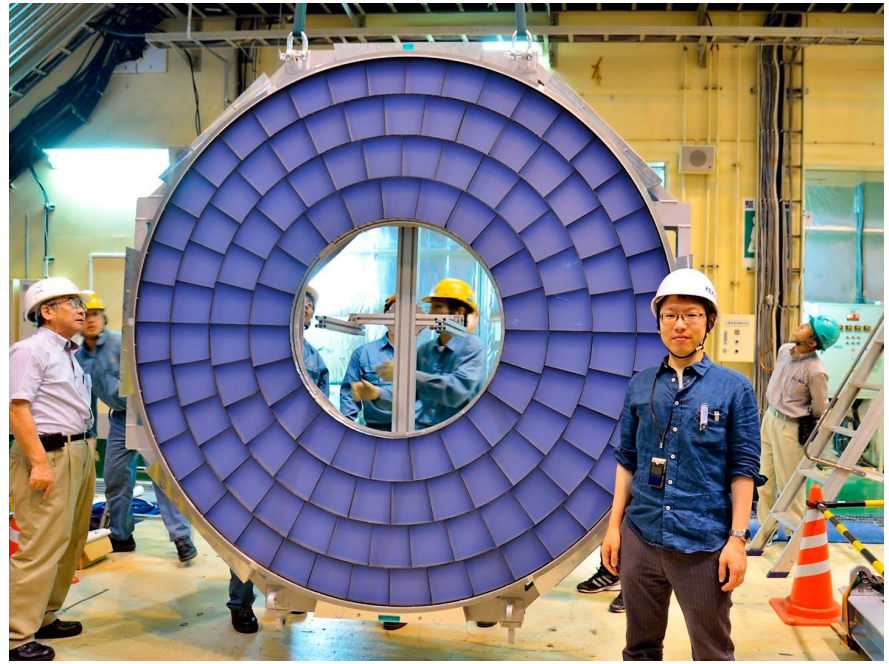

# Review: Belle II ARICH Aerogel

- n=1.045+1.055 (separated two layers).

- 18x18x2 $cm^3$ tiles: ~450 tiles mass-produced, 124+124 tiles installed .
- Water-jet cutting to fan shapes.

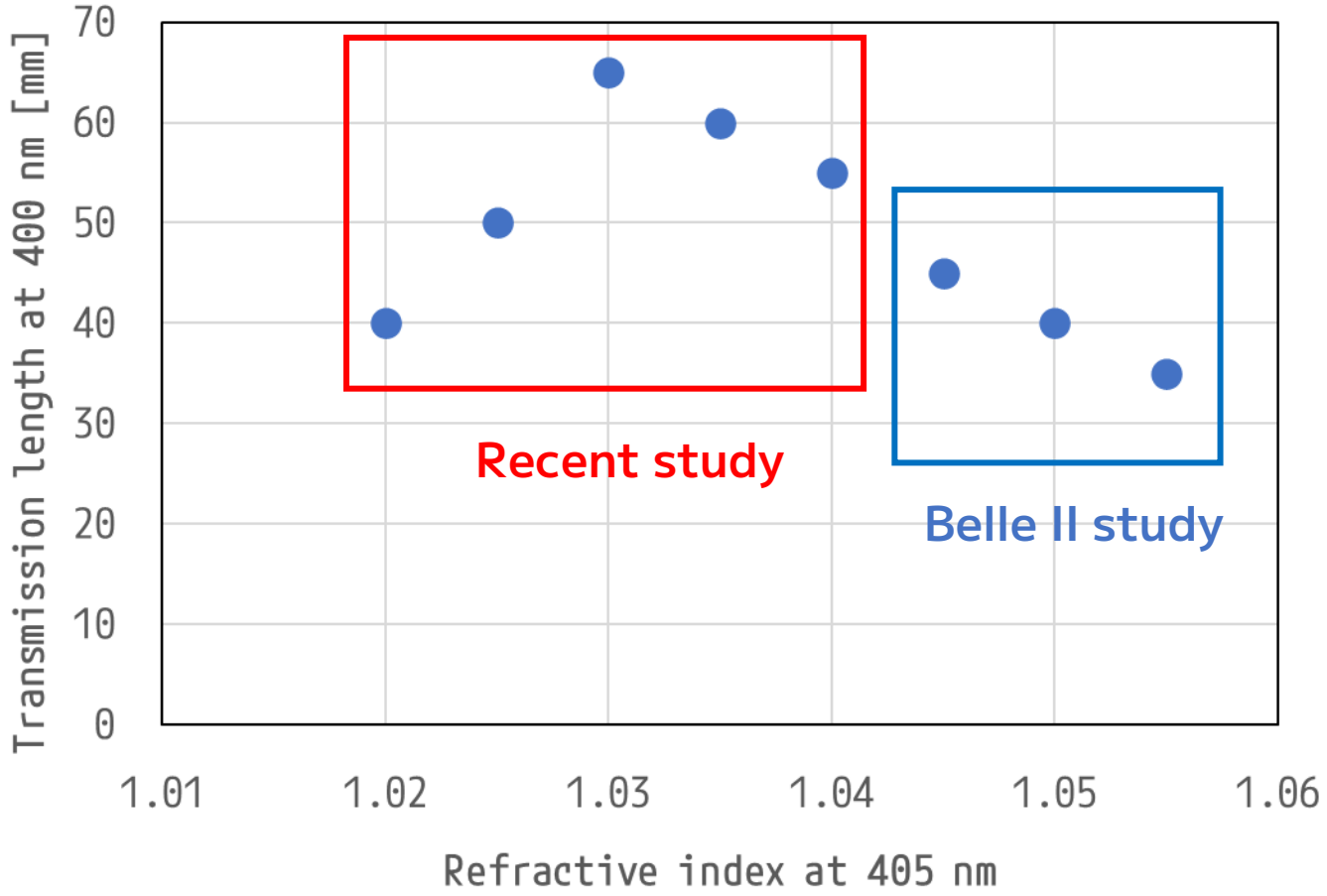
- Transmission length 45 and 35 mm as produced.
  Kept ~40 and ~30 mm after water-jetting.

  Degradation occurs not due to water contact itself, but rather from the machining procedures (handling by machining professionals and transportation).



- Special thanks to Belle II ARICH group!

# Optical Property (Recent Progress)



# Supercritical Drying

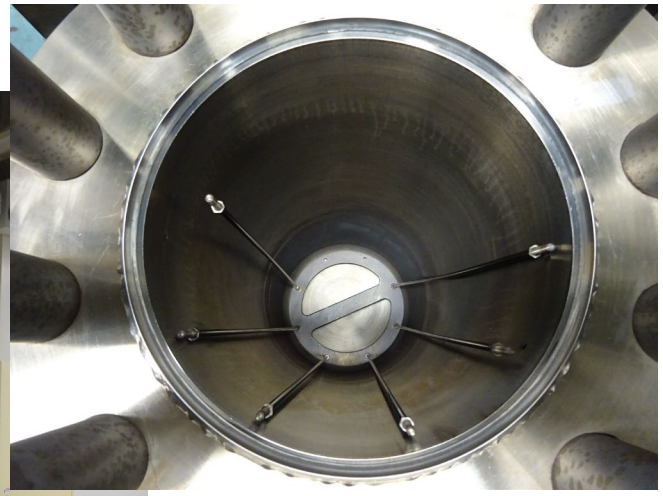
At Chiba Univ.

$CO_2$ autoclave (7.6 L)
Max. 80 °C., 10 MPa during operation.

Alcohol–$CO_2$ exchange in high pressure.
Can be applied in any index.

Alcohol autoclave (31 L)
Max. 260 degrees C., 7.3 MPa

Can be applied in n<1.08.

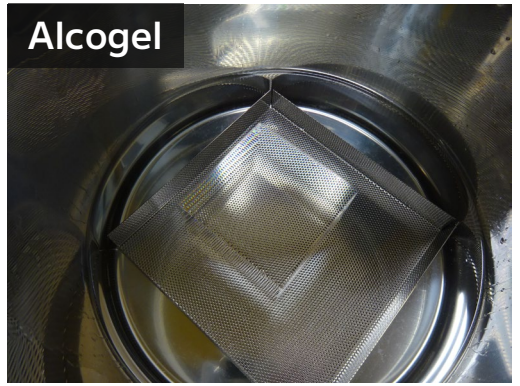


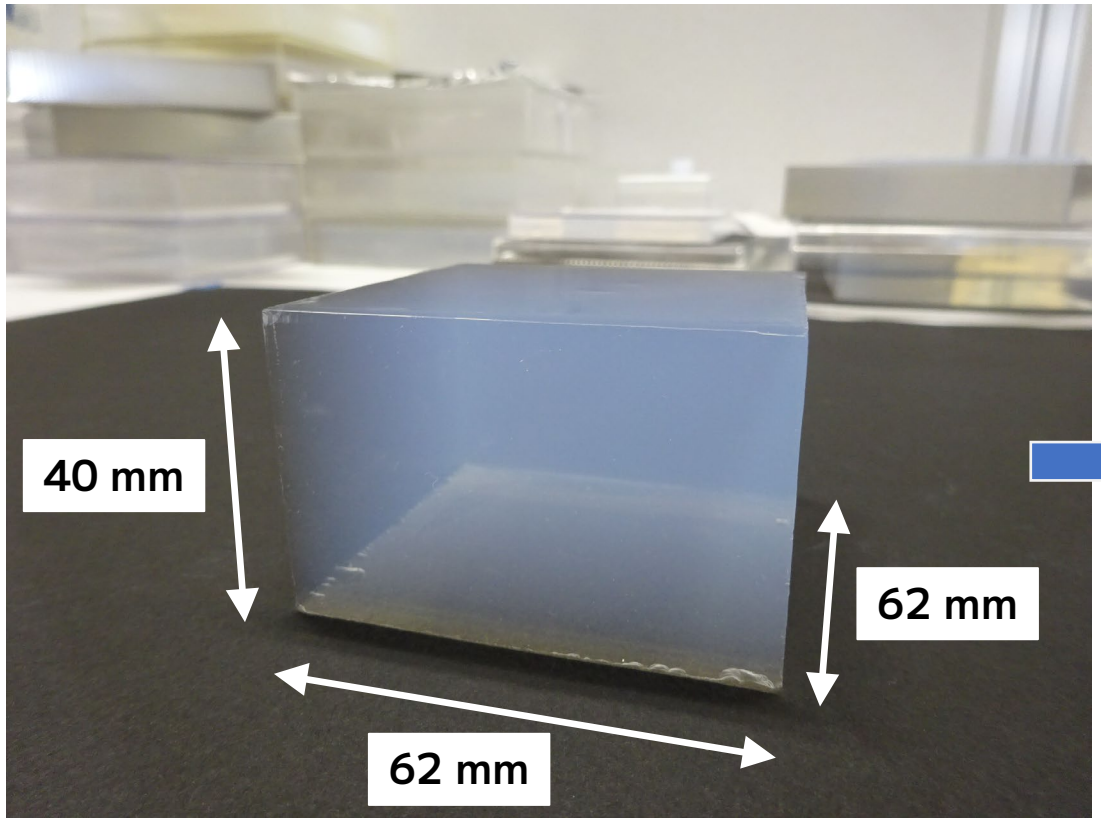
# Increasing Tile Thickness

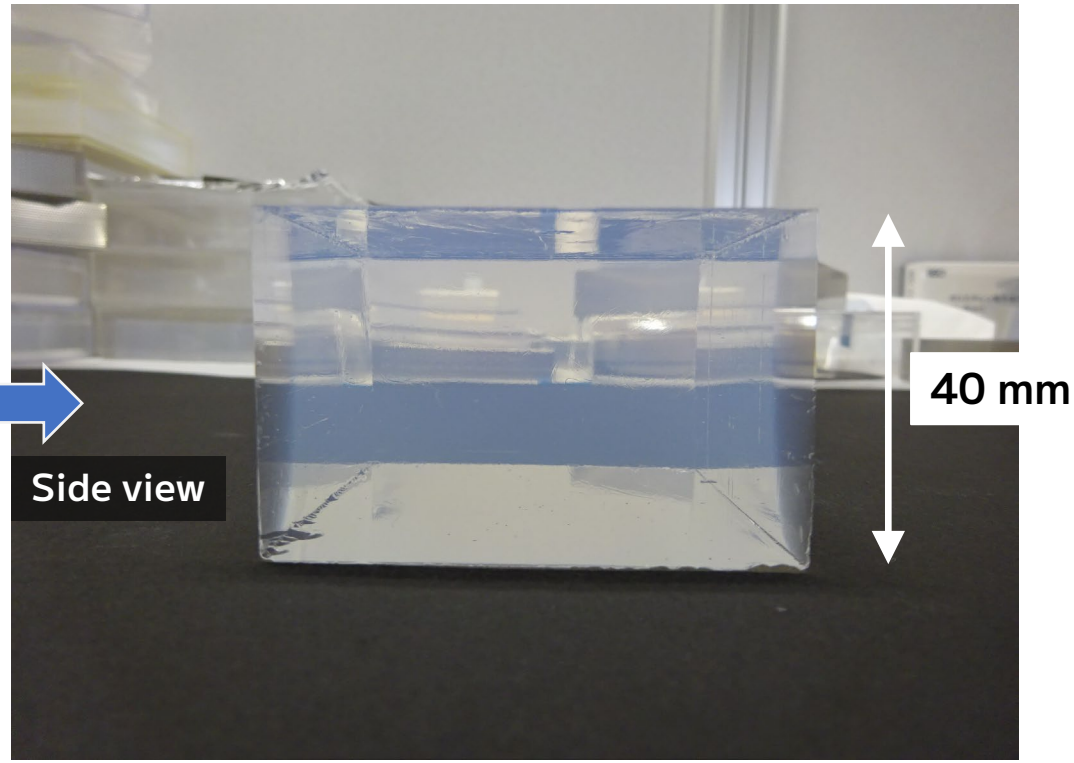
## Beyond 3 cm

# Thick Block with $CO_2$ Drying

- Using the small autoclave at Chiba.
- Applying the low-rate depressurization method.
- n=1.048, TL=43 mm, V=(62x62 mm$^2$, 40 mm thick).
- Crack-free yield is not so high (below 50%) as of Jan. 2024.

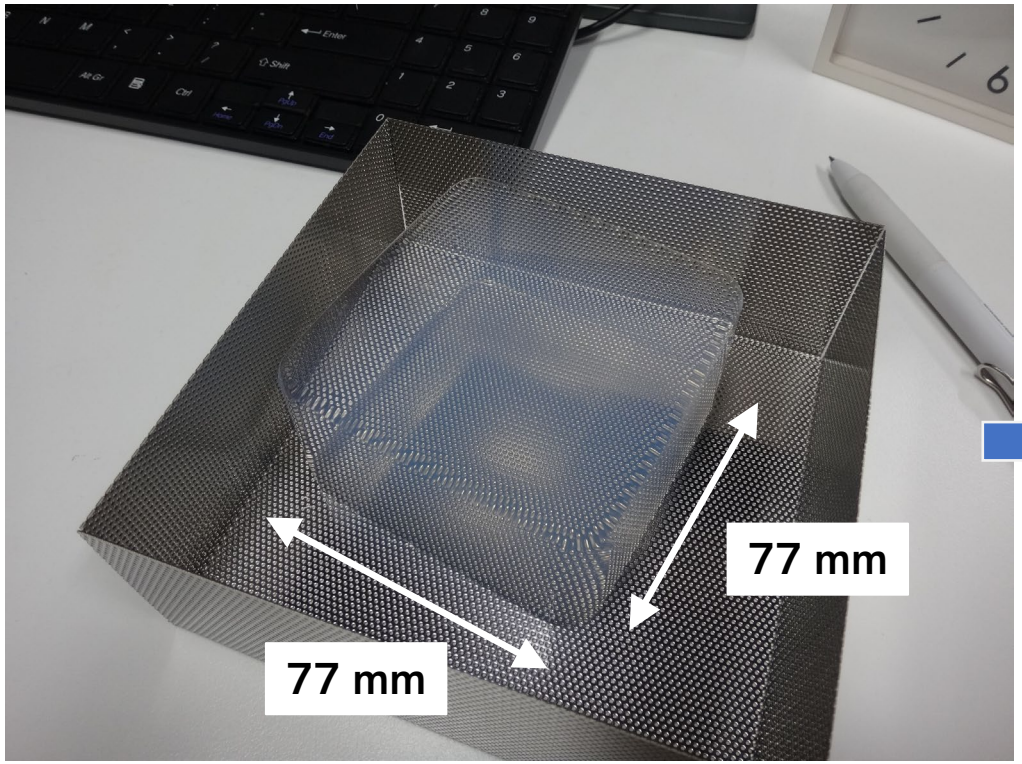


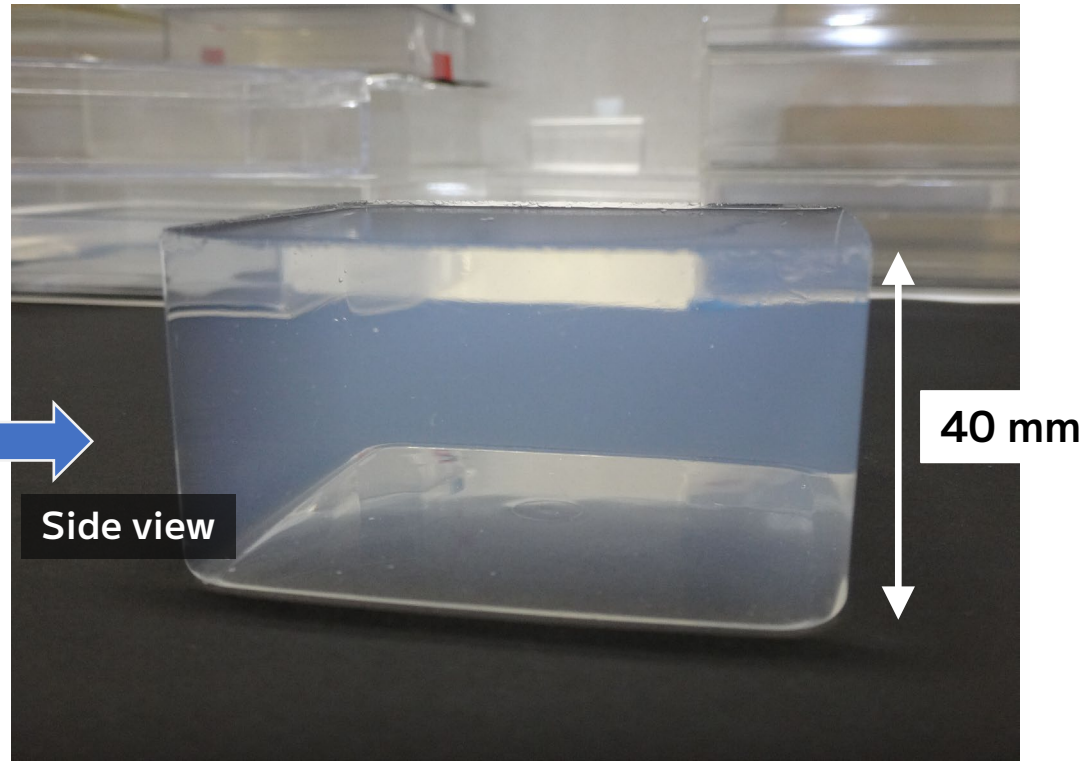



# Thick Block with Alcohol Drying

- Using the medium-sized autoclave at Chiba.
- Applying the low-rate depressurization method.
- n=1.039, TL=49 mm, V=(77x77 $mm^2$, 40 mm thick).
- The first crack-free block in Aug. 2025. → Next steps: 50 mm thick and yield check.



# Increasing Tile Area

## Beyond 18x18 $cm^2$

# Choice of Drying Method

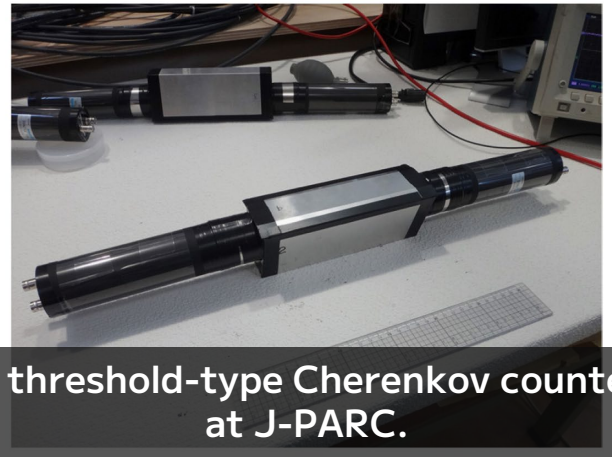
1. $CO_2$ drying has the disadvantage of tiles being likely cracked during solvent exchange (alcohol* to $CO_2$) under high pressure.
   (* Alcohol used as ammonia removal)

2. The $CO_2$ autoclave used in Belle II aerogel production was commercially operated by a company in Japan.
   → Continuous availability is not ensured.

3. If we build a new autoclave in the future, an alcohol autoclave has advantages in building costs and compliance with laws and regulations in Japan.

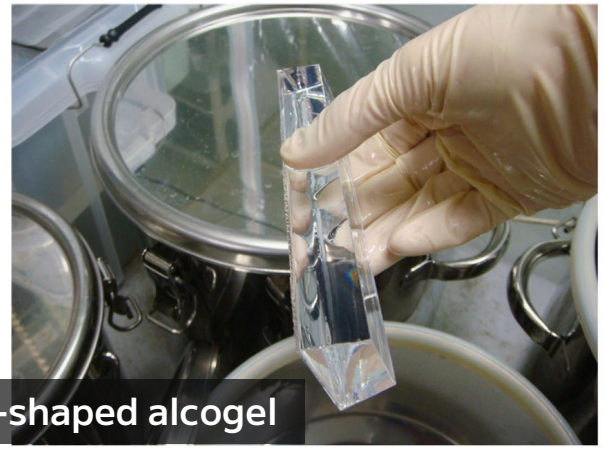
We chose alcohol drying for later study, aiming at larger tiles.

# A Proved Method: Vertical Storing in $CO_2$ Autoclave NIMA 795 (2015) 206.



A threshold-type Cherenkov counter at J-PARC.

**Fig. 2.** Assembled AC counter module. Two PMTs are oriented to the kaon beam axis. The length of the counter housing (aerogel box) is 18 cm.

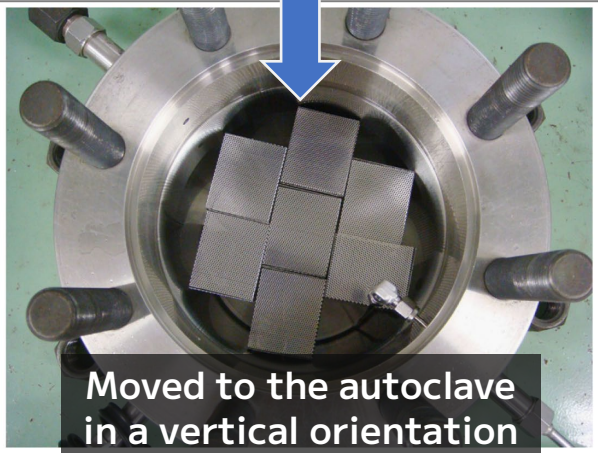


Bar-shaped alcogel

**Fig. 5.** A wet gel bar detached from the mold. When the aerogel density was more than $\sim 0.1$ g/cm$^3$ ($n=1.08$ corresponding to $\rho=0.27$ g/cm$^3$), direct handling of the wet gel was possible. The wet gel needed to be returned to the ethanol bath within several tens of seconds to avoid cracking because of drying.

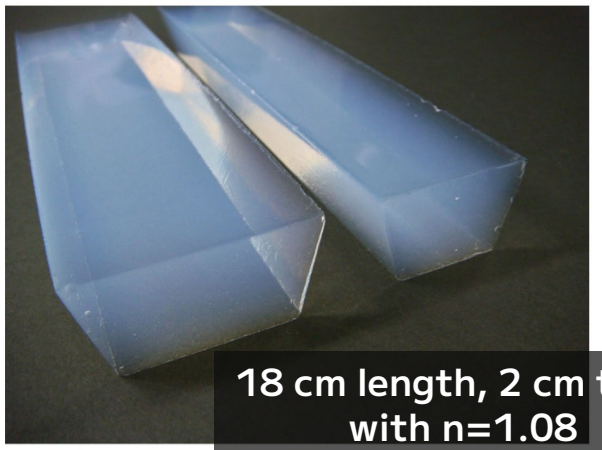
Stored in punched SUS containers

**Fig. 6.** Wet gel in the hydrophobic treatment process. The wet gel bars were placed in the punched trays and soaked in a solution for the hydrophobic treatment.



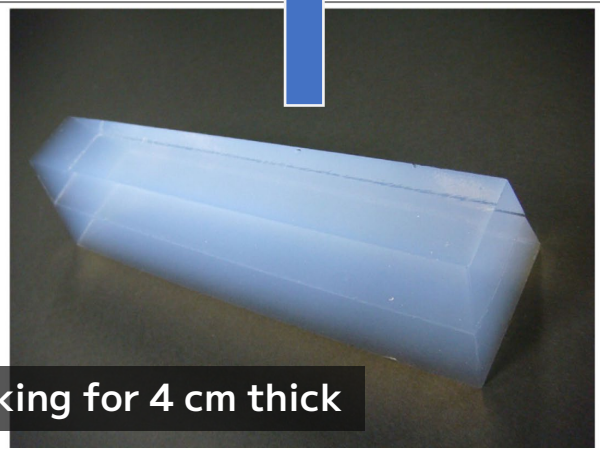
Moved to the autoclave in a vertical orientation

**Fig. 7.** Punched trays placed in the autoclave of supercritical carbon dioxide drying apparatus. Each tray contained two wet gel bars. The autoclave was filled with ethanol at the beginning of the drying operation.



18 cm length, 2 cm thick with n=1.08

**Fig. 8.** Crack-free aerogel bars for (left) downstream and (right) upstream layers obtained for the final production. Both the aerogel bars had a longitudinal length of approximately 18 cm and a thickness of 2 cm.

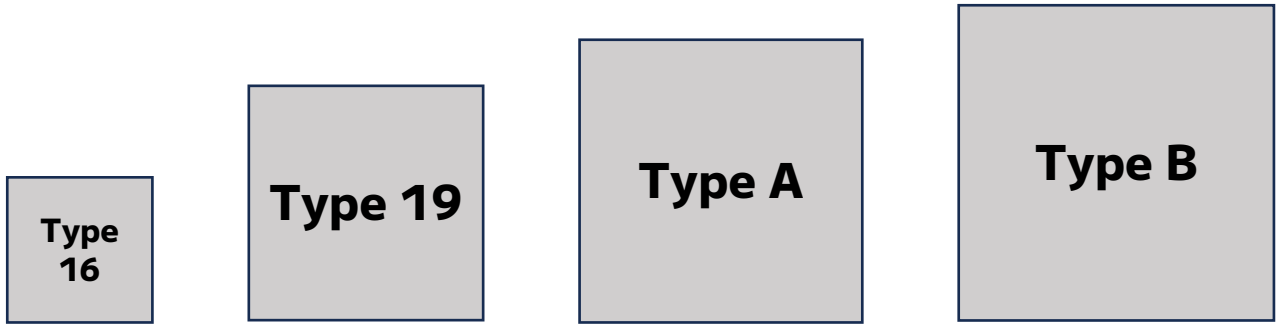


Stacking for 4 cm thick

**Fig. 13.** Upstream aerogel bar stacked on top of a downstream one. The longitudinal length and total thickness are approximately 18 cm and 4 cm, respectively.

# Polystyrene Molds for Alcogel Production

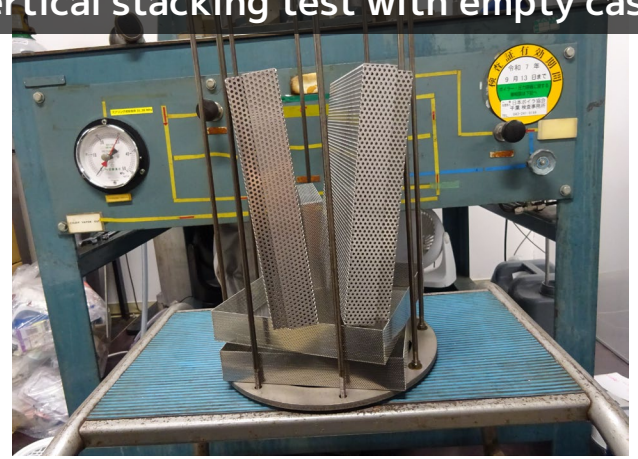
| Type | Inner size | Utilization | Final aerogel size (Approximate) |
|---|---|---|---|
| Type 16 | 96 mm | Basic R&D size | 9 cm |
| Type 19 | 155 mm | Fit alcohol autoclave | 15 cm |
| Type A | 187 mm | Belle II actual use | 18 cm |
| Type B | 207 mm | Belle II extra R&D | 19 cm |

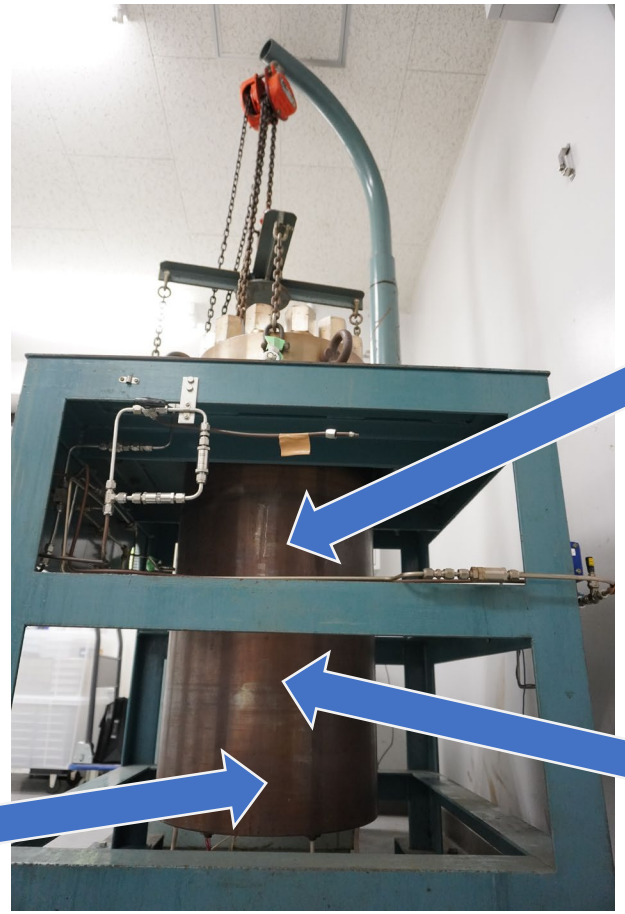



- Try to use types A and B with alcohol drying

# Alcohol Autoclave Setup

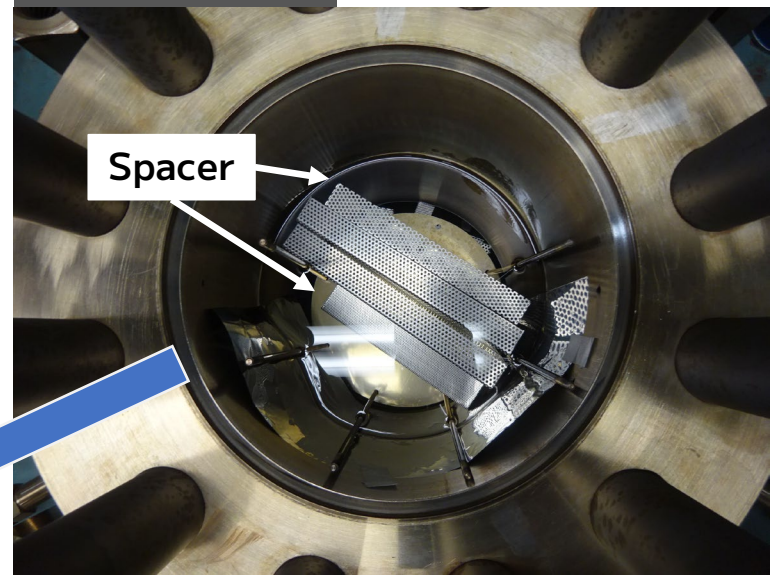
Vertical stacking test with empty cases



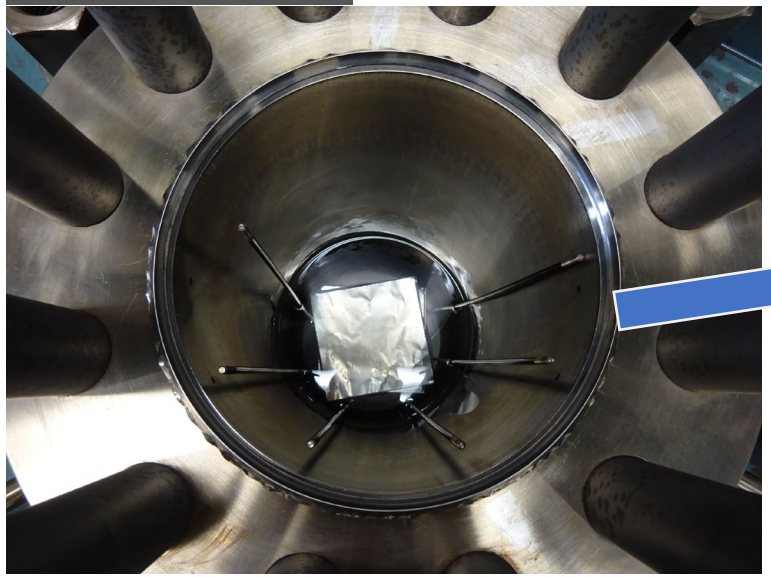


While filled with alcohol

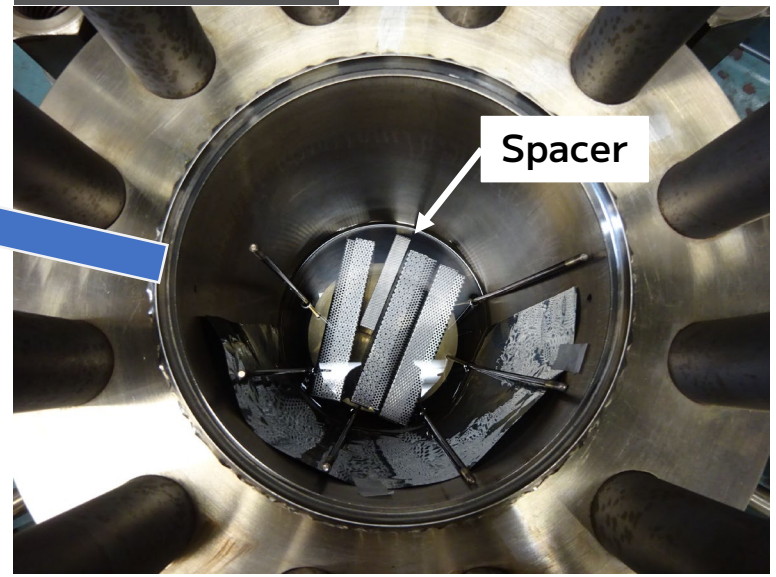
Type B (2 tiles)




Type 19 (2 tiles)

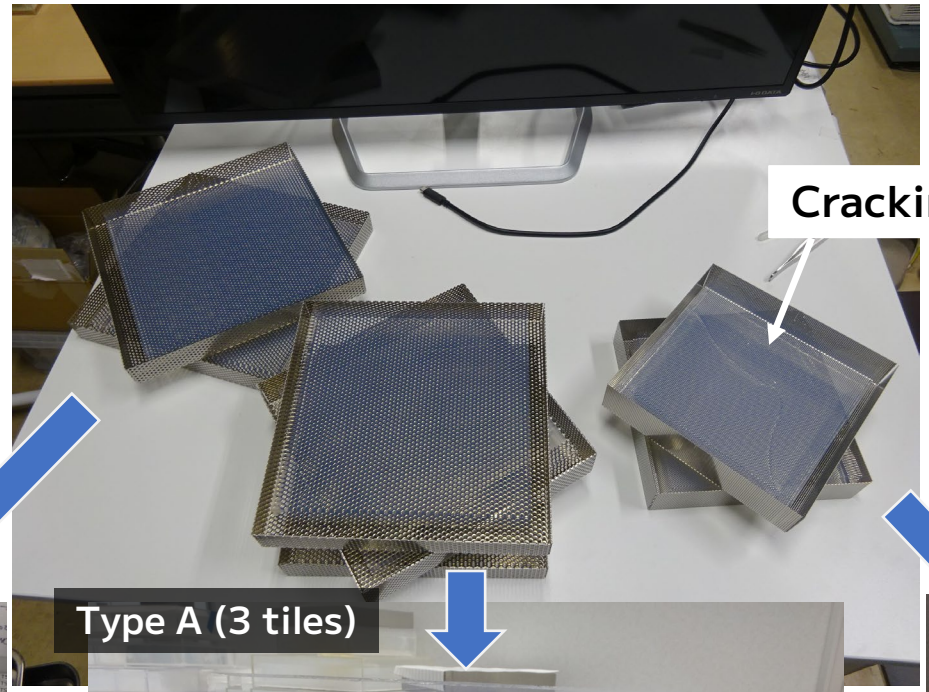


Type A (3 tiles)

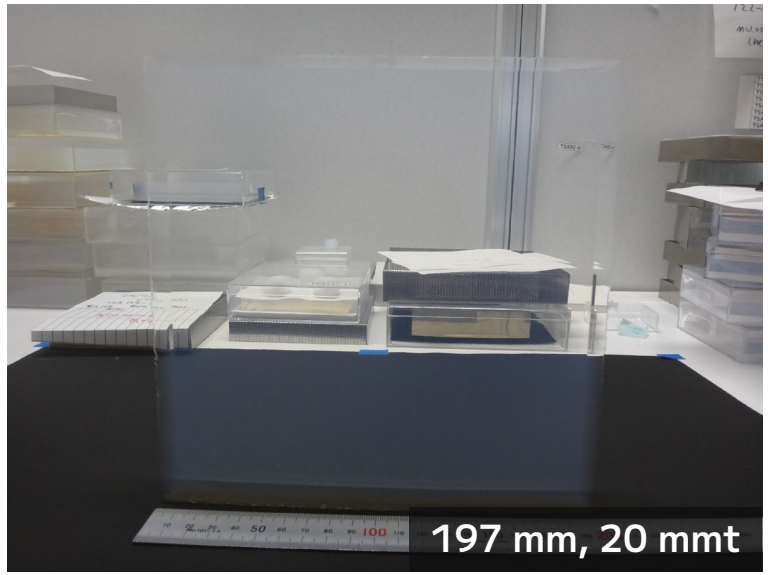


# Results from a Single Batch (n=1.026)

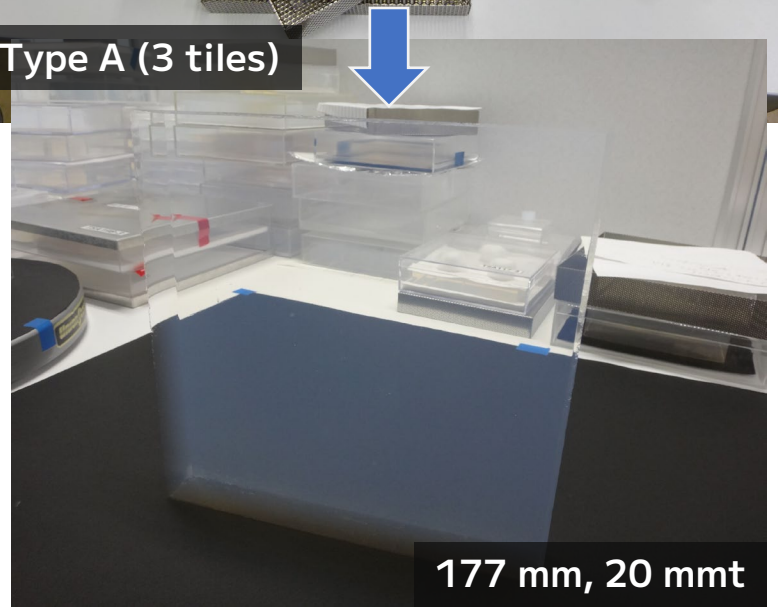
As of Dec. 2024



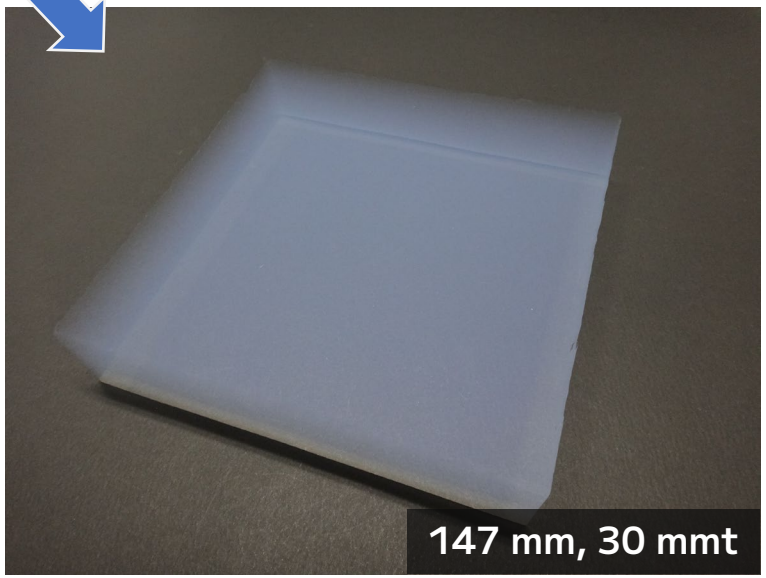

Type B (2 tiles)



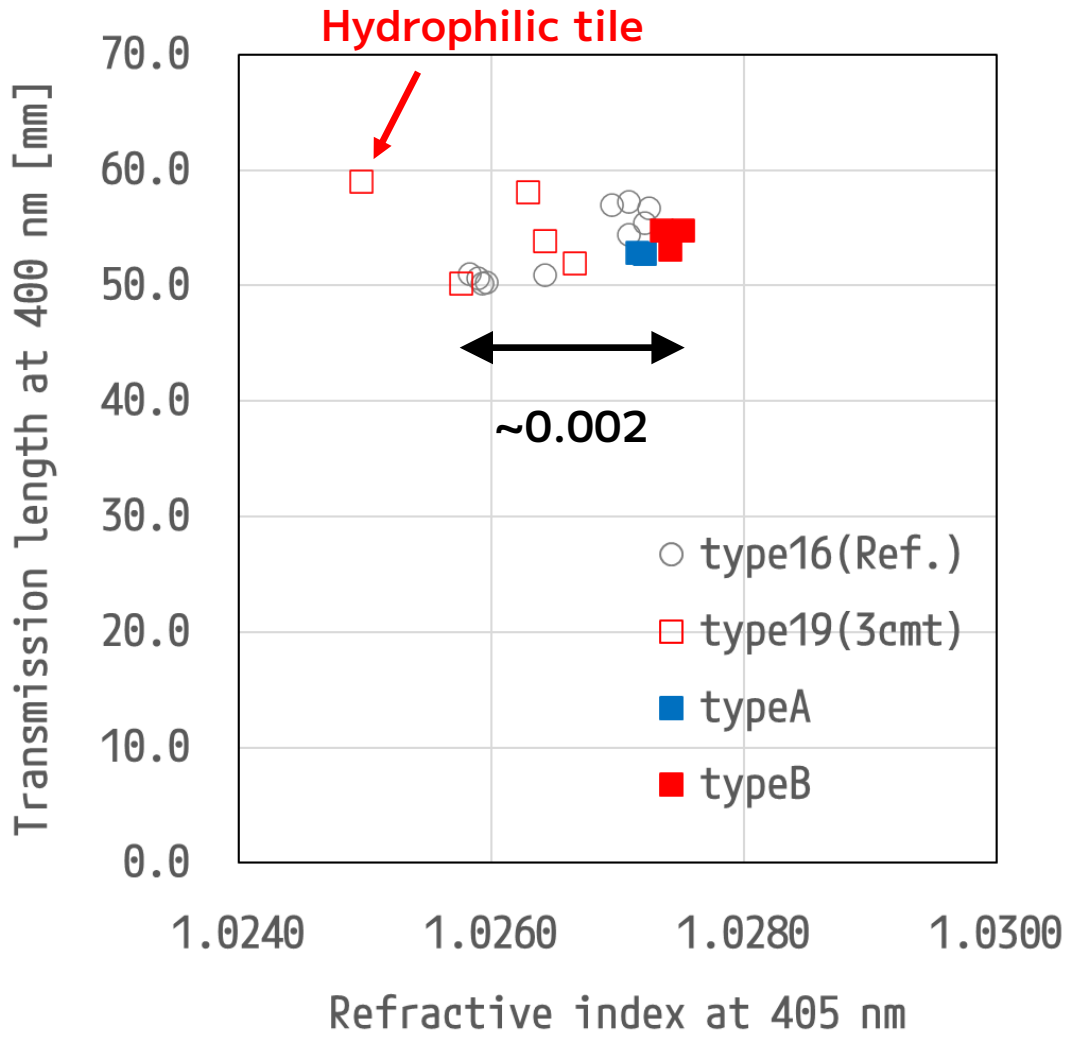

Type A (3 tiles)




Type 19 (2 tiles)



# Optical Properties of Lager Tiles




- No significant differences compared with small tiles.
- Refractive index distribution within the production tolerance (reproducibility ±0.001).
- Comparison with hydrophilic tile.
  Low refractive index and high transmission length.

# Summary

- Results from large-volume aerogel tile R&D were reported.
- As of today, ~20x20x2 $cm^3$ hydrophobic tiles were obtained with no cracking using the alcohol drying method.

Next step.

- An alcohol-type large autoclave is under design.
- Inner diameter = 47 cm, depth = 66 cm (~115 L)
- Maximum size of ~30x30 $cm^2$ can be stored.
- The first engineering operation is expected in May 2026.

- Hopefully, results from the large autoclave will be shared at the next RICH conference!

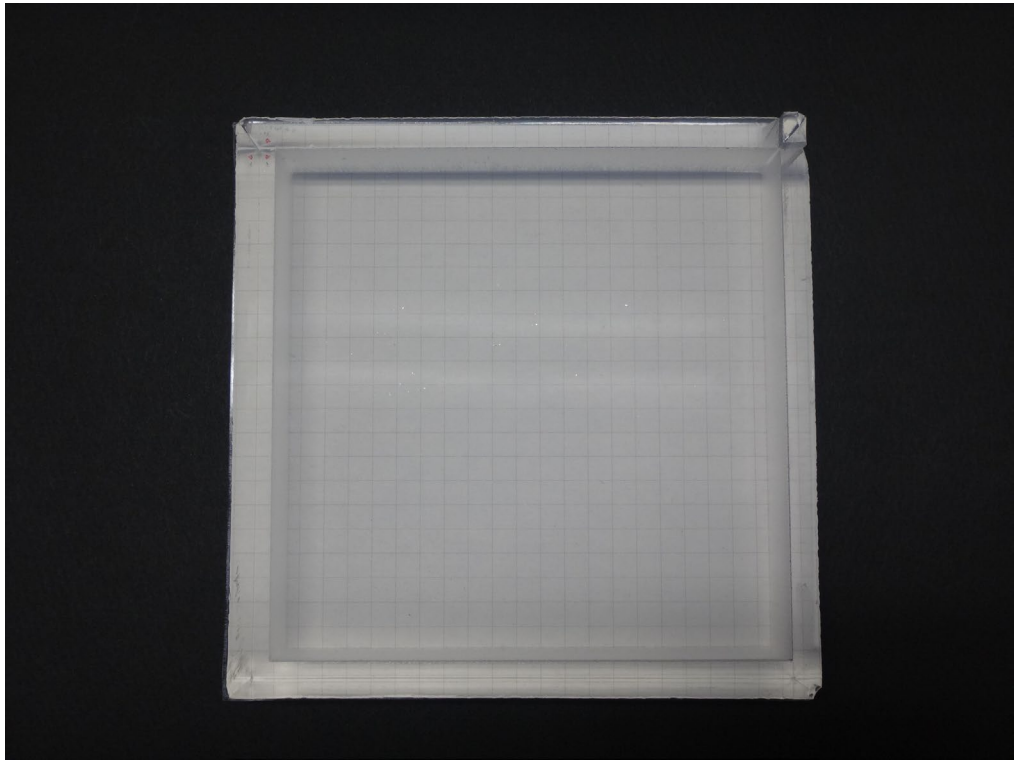
# Backup Info

- Excellent review by I. Adachi (KEK)!
  Selected slides are attached to the following pages.

- In the workshop, fruitful focused characterizations in the individual experimental collaborations!
  ePIC dRICH & pfRICH, ALICE3, HELIX, etc.
  Each feedback is vital for improving aerogel production!

# Water-jetting

- Continuous line cutting (Recommended).
- Need extra volume outside the designed shape (~7mm or more).
- Ex. preparing a 114mm tile for getting a 100mm square.
- The success of cutting proves the hydrophobic property.



- Basically, no degradation of optical properties by cutting itself.
- However, potential surface damage from the handling of tiles by cutting experts in a company. Also, during transportation.