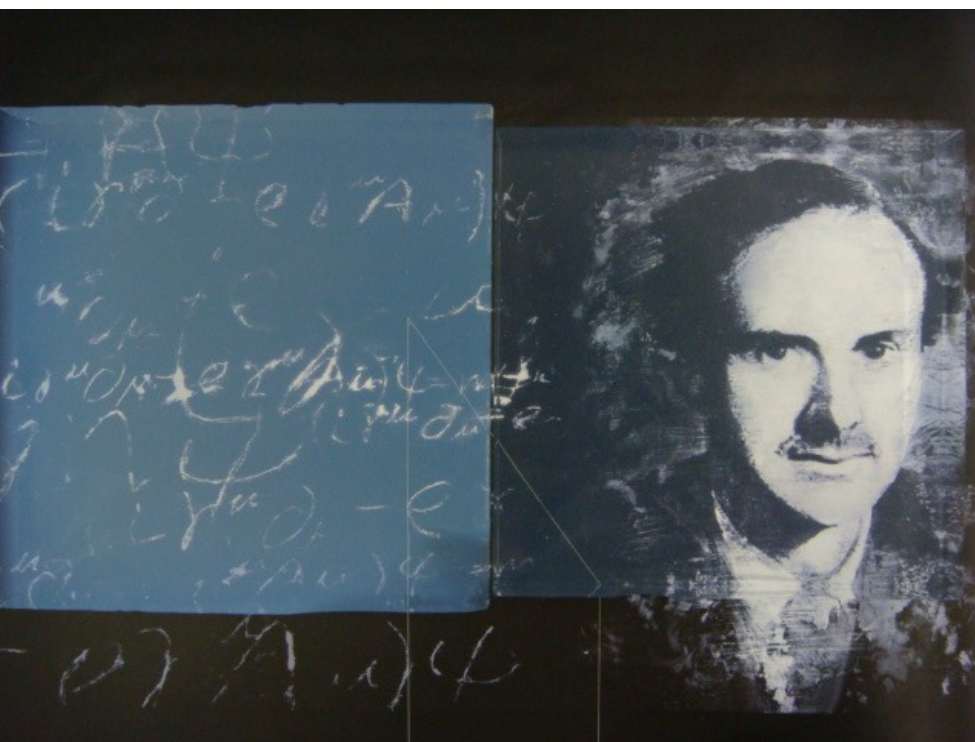


# Status and Perspectives of High Quality Silica Aerogel

Ichiro Adachi

KEK  
2018.08.03




**10th International Workshop on Ring Imaging Cherenkov Detectors**  
on the occasion of the 60th anniversary of P.A. Cherenkov's Nobel Prize  
Moscow, Russia 29 July – 4 August 2018

**RICH2018.org**  
<http://RICH2018.org>

**Topics:**  
• Cherenkov light imaging in particle and nuclear physics experiments • Cherenkov detectors in astroparticle physics • Photon detection for Cherenkov counters  
• Pattern recognition and data analysis • Alternative PID techniques • Technological aspects and applications of Cherenkov detectors • Novel Cherenkov imaging techniques for future experiments • Remembering Pavel Cherenkov

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# Cherenkov detectors with aerogel radiators

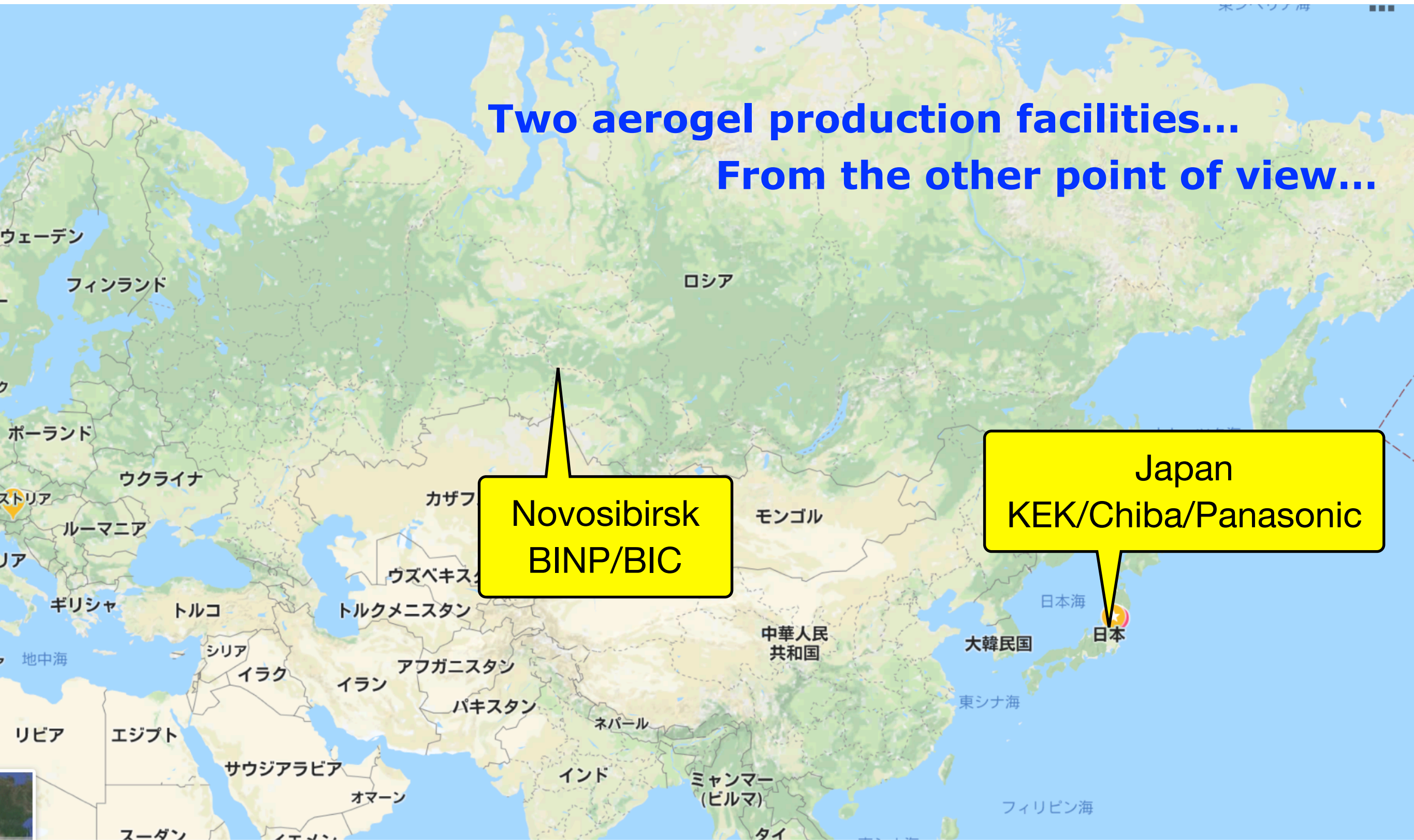
E.A.Kravchenko

*Novosibirsk State University  
Budker Institute of Nuclear Physics*

*We have already had a review talk !*



**Two aerogel production facilities...**  
**From the other point of view...**





# Belle aerogel Cherenkov counters

**1991** KEK and Matsushita started joint effort in producing low refractive index silica aerogels.

- **1992** – seminar of A. Onuchin at KEK on aerogel threshold detector development
- **1994** – approval of KEKB with Aerogel Cherenkov Counters as baseline for PID
- Very intensive R&D both on aerogel production and detector design
- 1124 detectors equipped with 2024 Fine Mesh PMTs (2, 2.5, 3 inch)
- $n=1.01\text{--}1.03$ ,  $V_{\Sigma}=2000$  I, high transparency hydrophobic aerogel
- $N_{pe} = 20\text{--}26(!)$
- In operation 1998-2010
- **Full success of the project!**

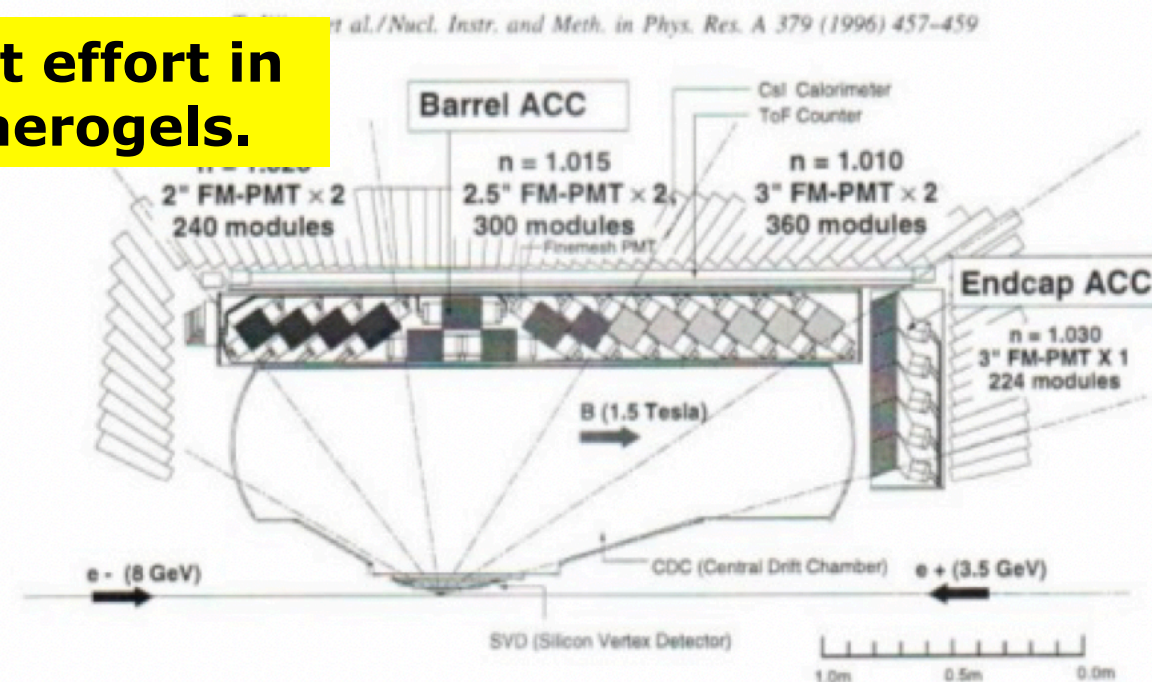


Fig. 1. Design of the BELLE aerogel Cherenkov counter system (as of March 1996).

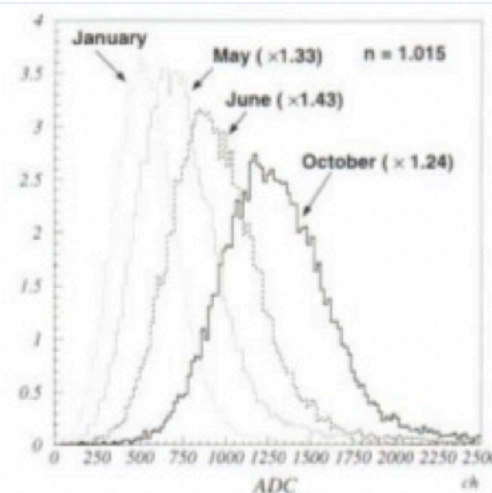
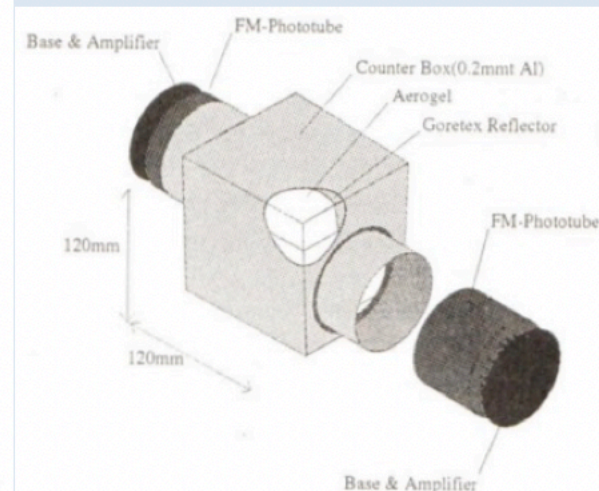


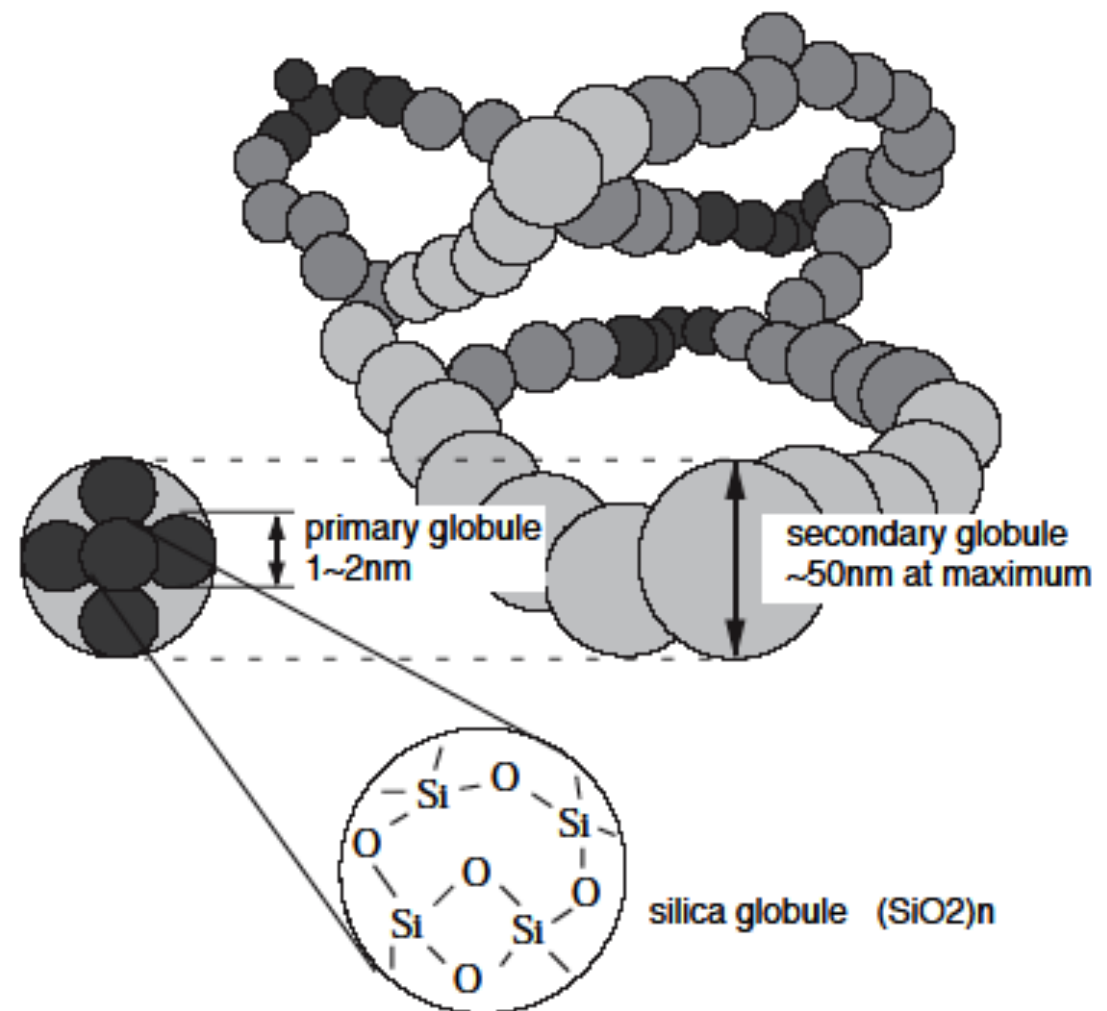
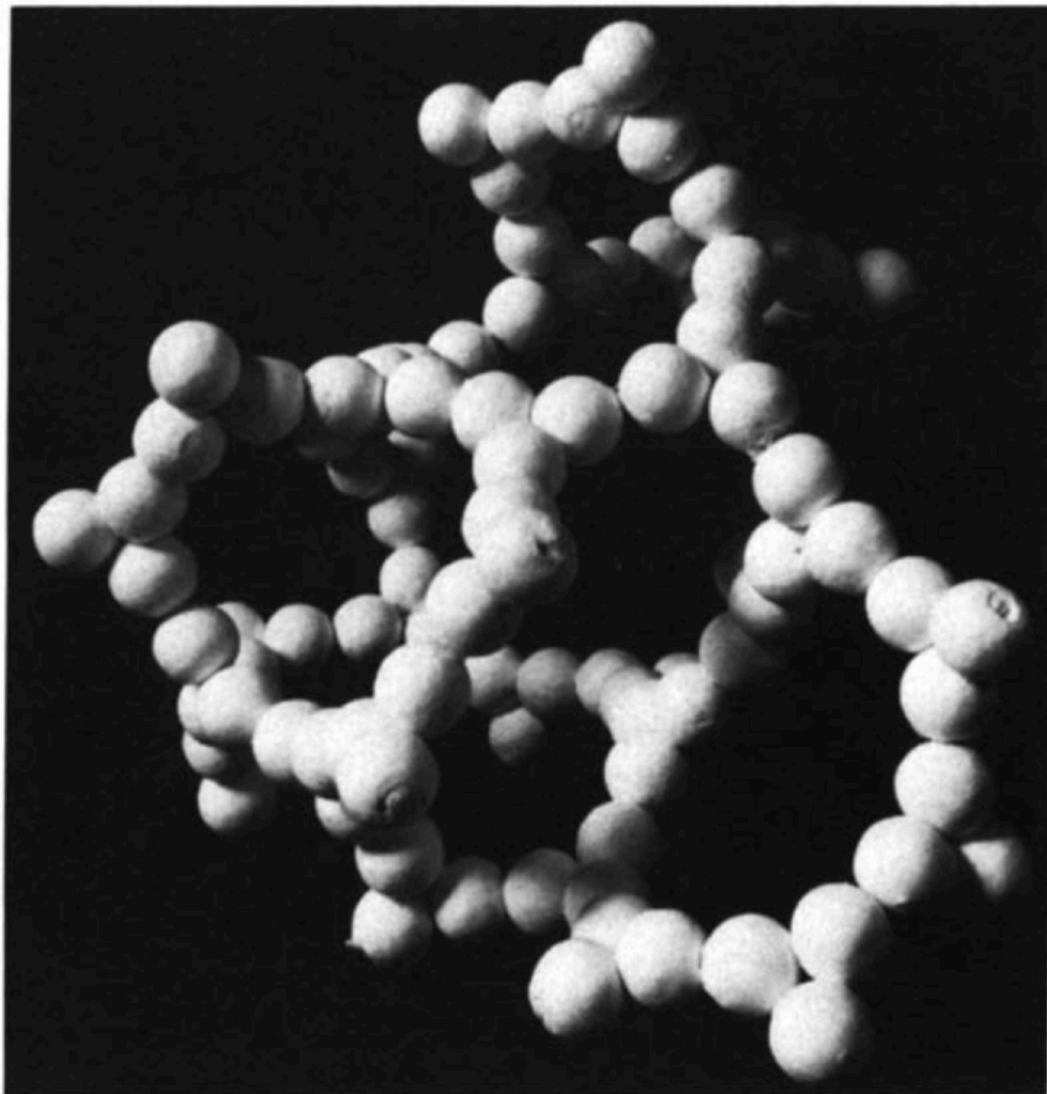
Fig. 2. Pulse height spectra obtained with  $n = 1.015$  aerogels in beam tests in the past one year. The counter configuration and PMT gains are the same for all measurements.





# Silica Aerogel

- Highly porous material of silica clusters
  - 3 dimensional network of  $(\text{SiO}_2)_n$
  - Micro-structure smaller than the wavelength of visible light.
  - More than 90 % air inside volume





# Production Process

- Two subsequent reactions for sol-gel polymerization



**Alco-gel**

- **Hydrophobic treatment**
  - **Introduced in 1990's at KEK**
- Aging
  - Stabilize 3D network of SiO<sub>2</sub> clusters
- Supercritical drying (SCD) to convert from alco-gel to aerogel.
  - Critical point
    - CO<sub>2</sub> : T=31°C & P=7.4MPa
    - Methanol : T=240°C & P=8.1MPa



# Unique Features

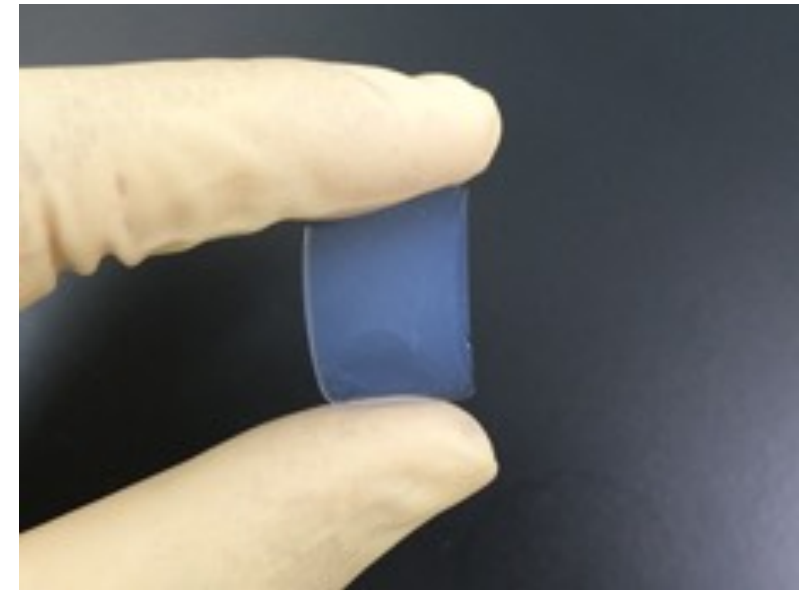
- Excellent thermal insulator



thermal insulation window



- Fragile
  - Especially localized pressure
  - If pressure is widely uniform over aerogel surface, aerogel tiles possess some rigidity.





# Optical Quality

- Refractive Index
  - Density (in other words)
  - **Fraction of silica clusters defines material density.**
- Transparency
  - Transmission length
  - Clarity parameters
  - **Uniformity and dimensions of silica clusters are related to transparency.**
    - For high transparency, synthesis parameters (gelling time etc) should be optimized.



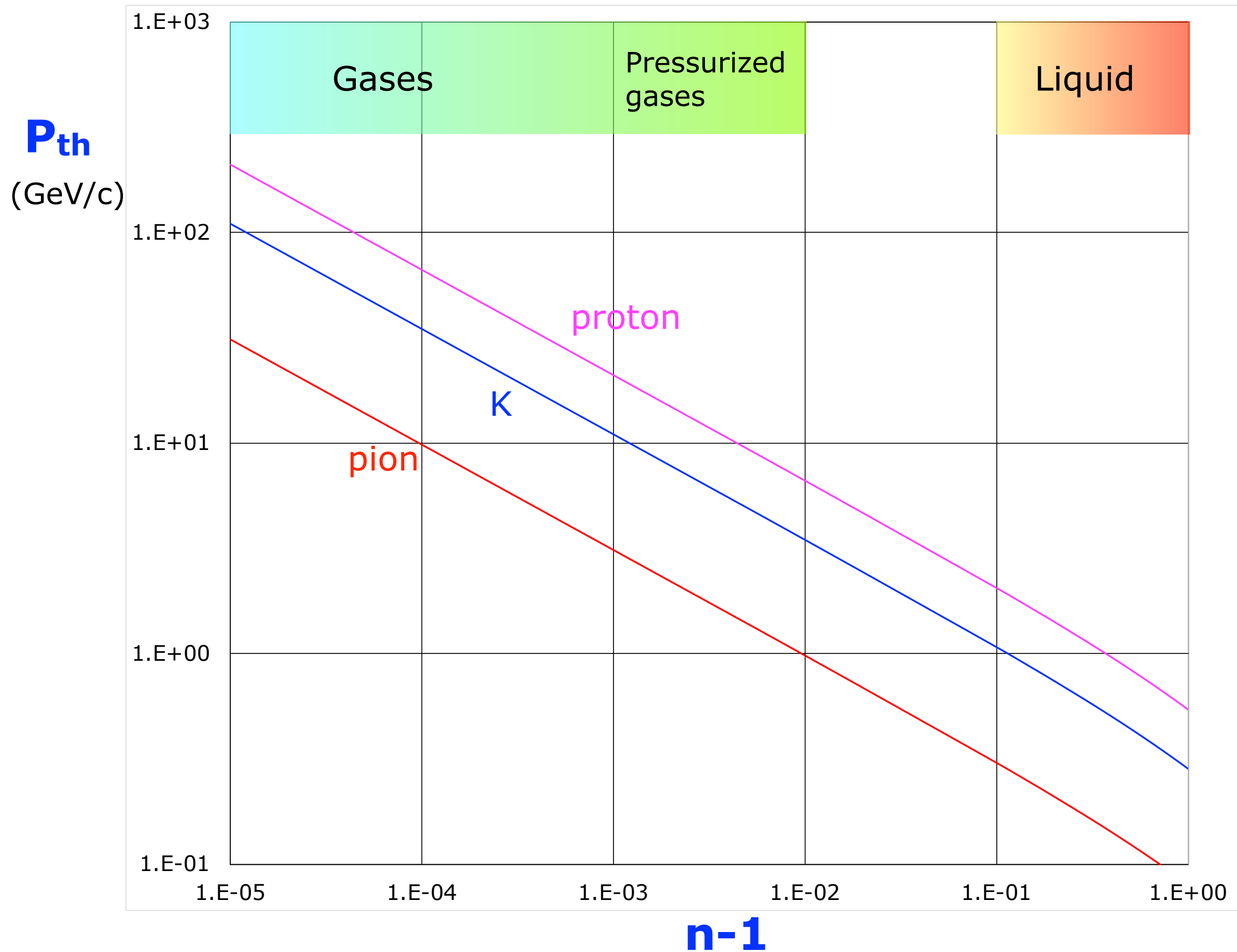
# Refractive Index

- Unique refractive index
  - It is related to quantity of SiO<sub>2</sub> clusters.

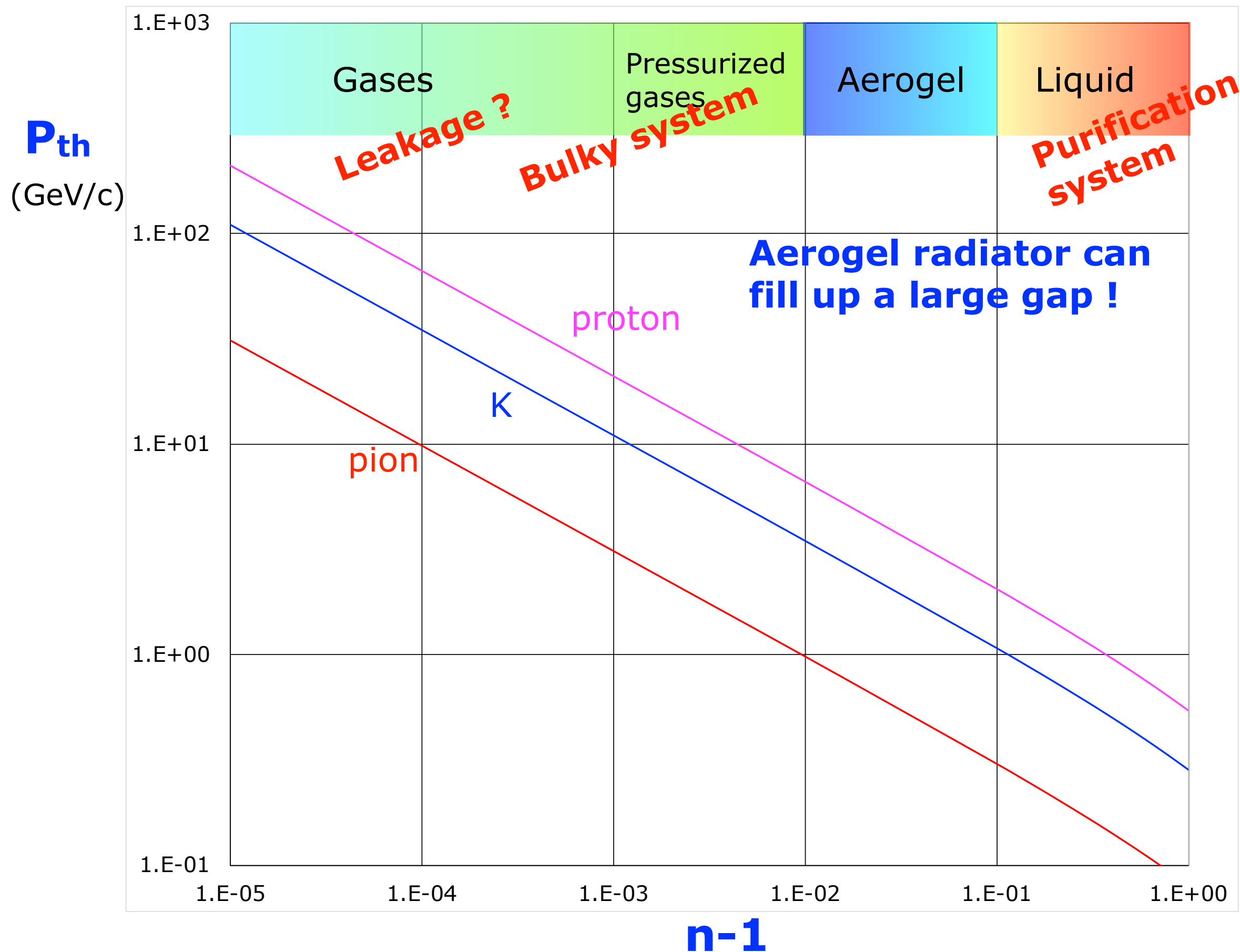
## Cherenkov radiation

Threshold momentum  $P_{th}$  is related to refractive index ( $n$ ) of radiator medium and incident particle mass ( $m$ ) .

$$P_{th} = m / \sqrt{(n^2 - 1)}$$

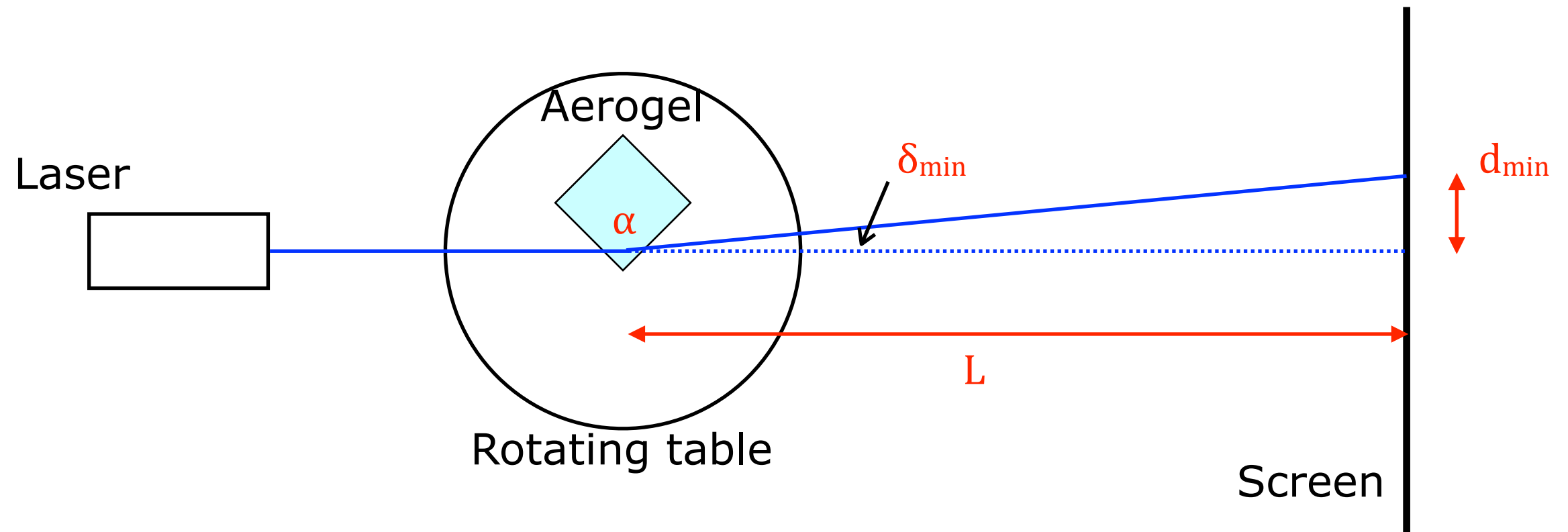






# Refractive Index Measurement

- Refractive index is calculated from a deflection angle in Fraunhofer method.



Prism formula

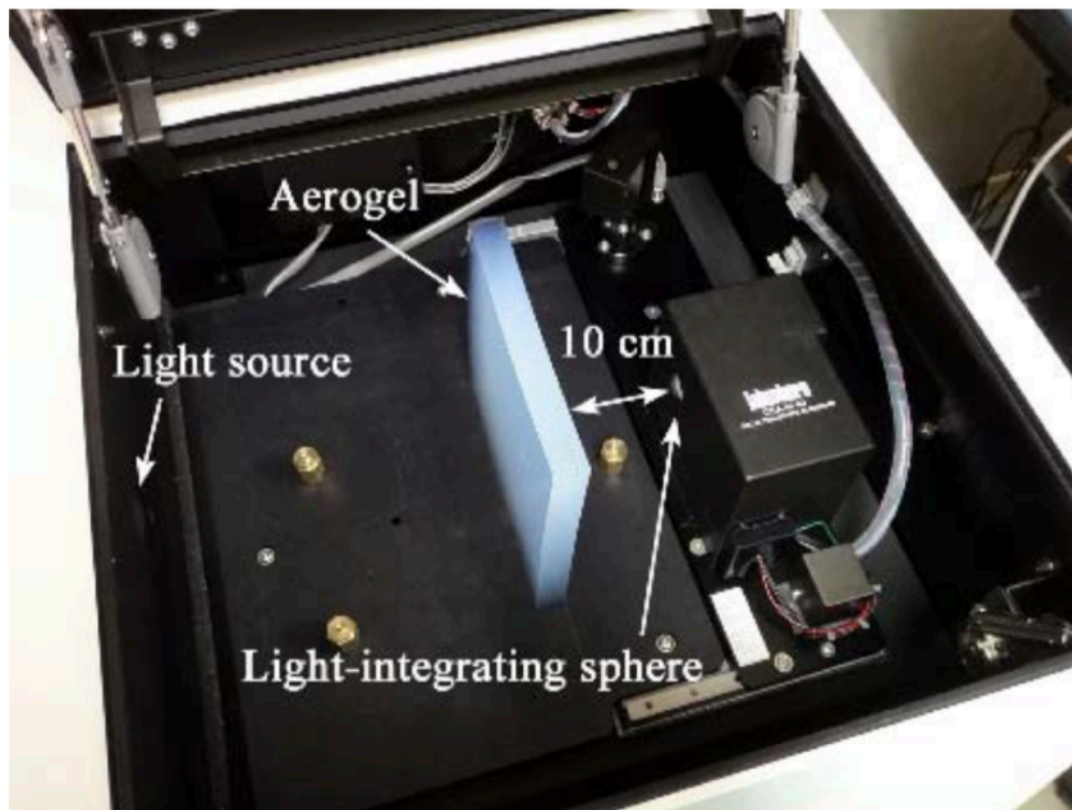
$$n = n_{\text{air}} \cdot \sin((\alpha + \delta_{\min})/2) [1/\sin(\alpha/2)]$$

$$\delta_{\min} = \tan^{-1}(d_{\min}/L)$$



# Transmittance

- Transmittance is measured using a spectrophotometer.
  - At KEK, aerogel tile is **placed by 10 cm** from a light-integrating sphere.



$$T = T_0 \exp(-d/\Lambda)$$

$T_0$ : initial intensity

$d$ : thickness

$\Lambda$ : transmittance length

$$\Lambda(\lambda) = -d/\ln(T/T_0)$$

**$\Lambda(\lambda=400\text{nm})$  often used to characterize aerogel transparency**

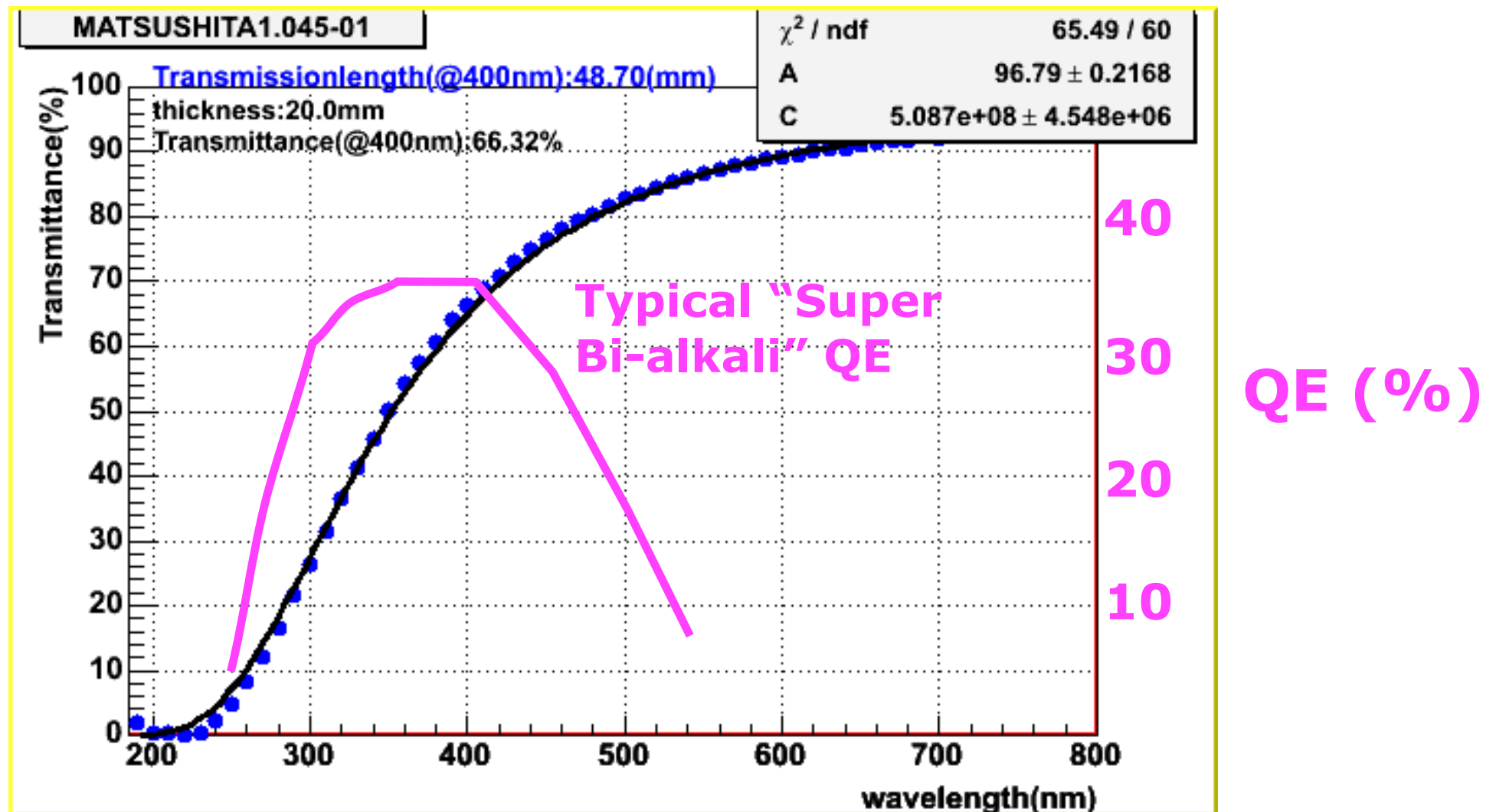
**$\Lambda$  depends on the position, where aerogel tile is placed in the spectrophotometer.**

Clarity parameters also used.

$$T = A \exp(-C \cdot t/\lambda^4)$$

# Transmittance

- Transparent for visible light region
- **This makes photon detection more straightforward.**





# Aerogel Production Centers

- **Japan**

- KEK/Chiba/Panasonic groups
  - Panasonic no longer deal with aerogel production.
  - Technological transfer to Japan Fine Ceramics Center (JFCC).
  - SCD is outsourced to Mohri oil company.
    - Facility with 70-liter dry volume
  - Chiba university has their own SCD facility.
    - Only academic center to handle aerogel production from synthesis steps to SCD.

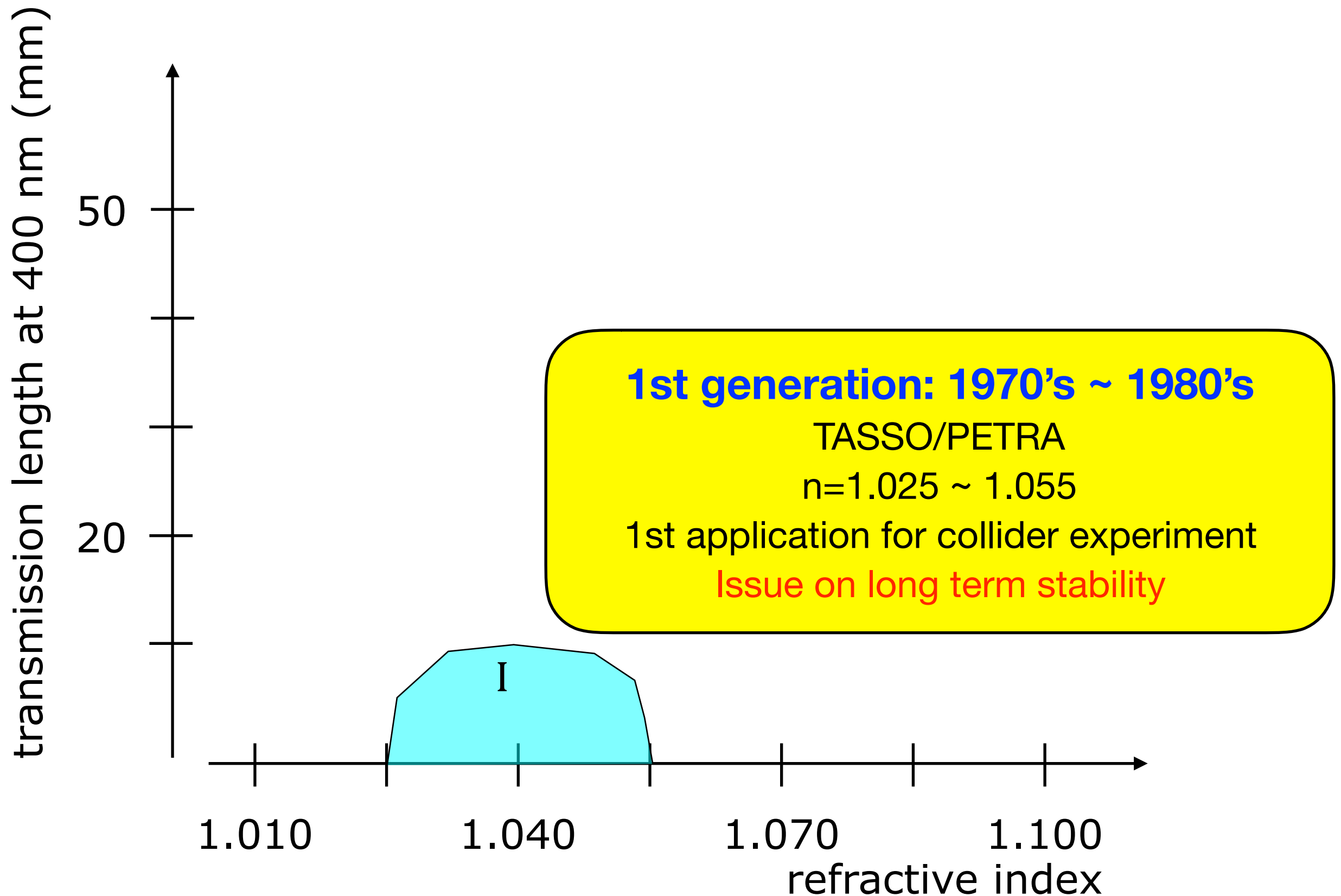
- **Russia**

- Budker INP and Boreskov institute of Catalysis.
- Their own SCD facilities.
  - Please see a nice poster

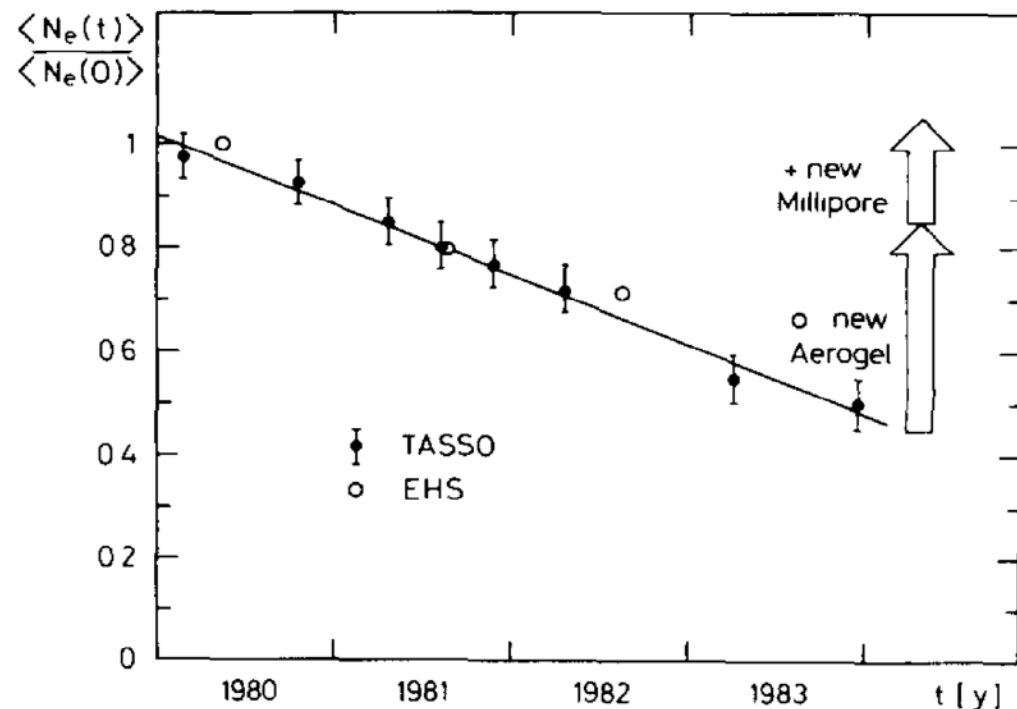
# **History of Aerogel Radiator & Development of Cherenkov Counters**



# History of Aerogel Radiators



# History of Aerogel Radiators



NIMA248(1986)118

$N_{pe}$  decreased by 0.5/yr

Fig. 24. Photoelectron yield of the TASSO aerogel counters for relativistic particles along the time of operation. The yield decreases with a slope of  $0.5 \text{ yr}^{-1}$ . The arrows show the improvement obtained when new aerogel and new millipore was recently installed in one cell. The data from the EHS detector [16] indicate a similar decrease but suggest an exponential decay with a time constant of 7.5 yr.

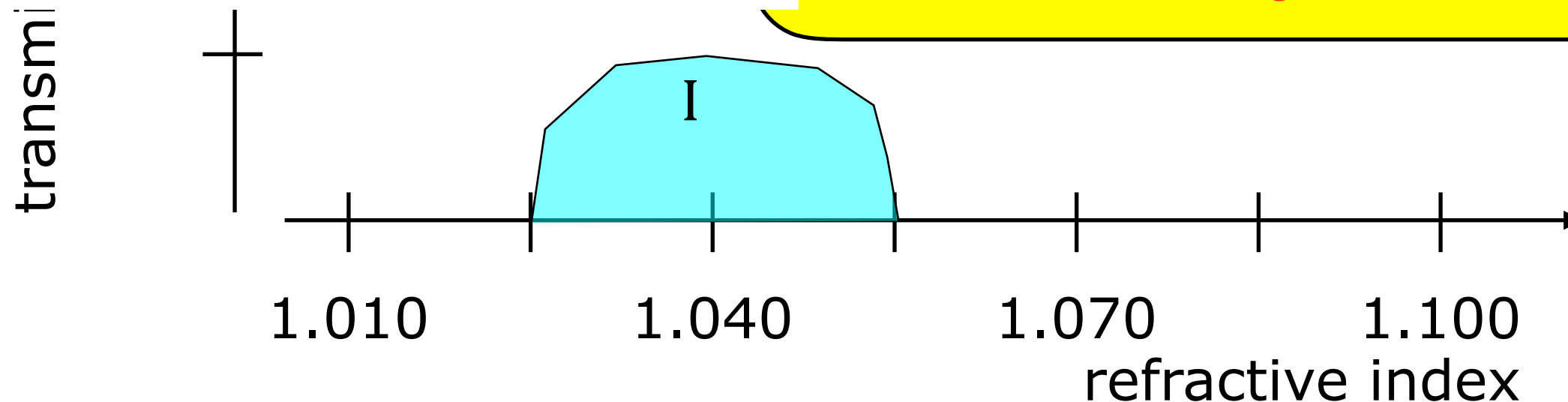
**1st generation: 1970's ~ 1980's**

TASSO/PETRA

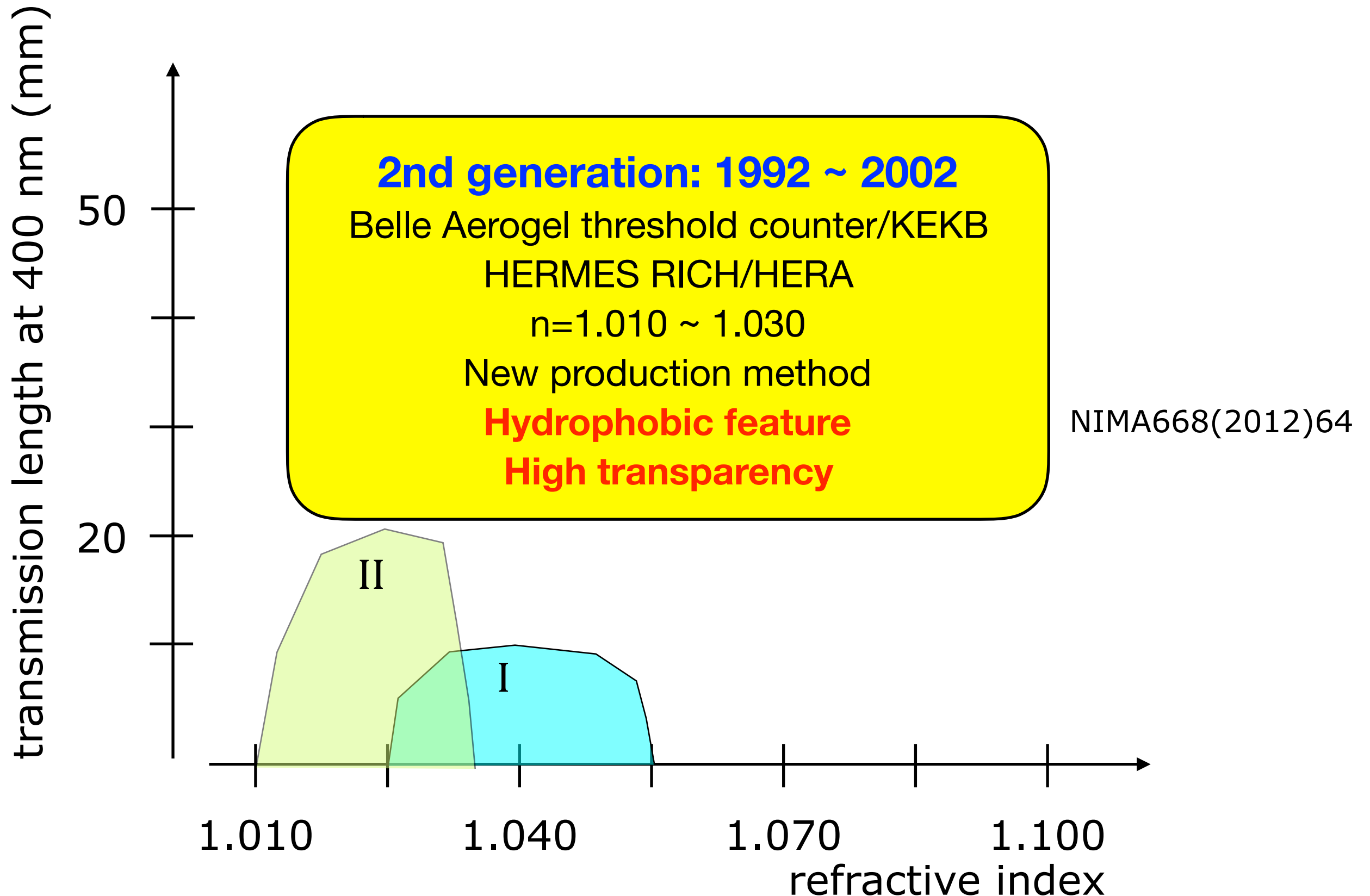
$n=1.025 \sim 1.055$

1st application for collider experiment

Issue on long term stability

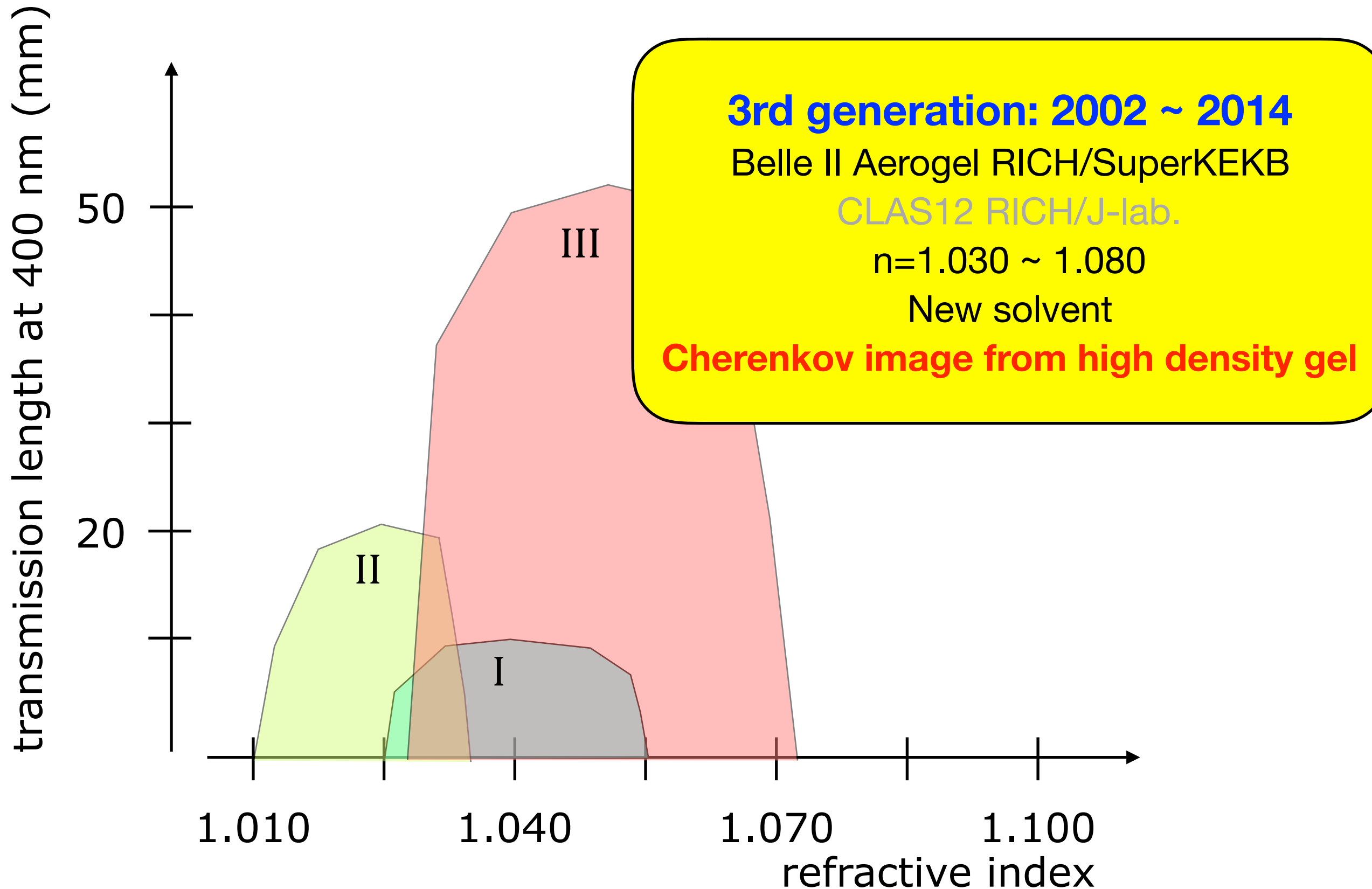


# History of Aerogel Radiators

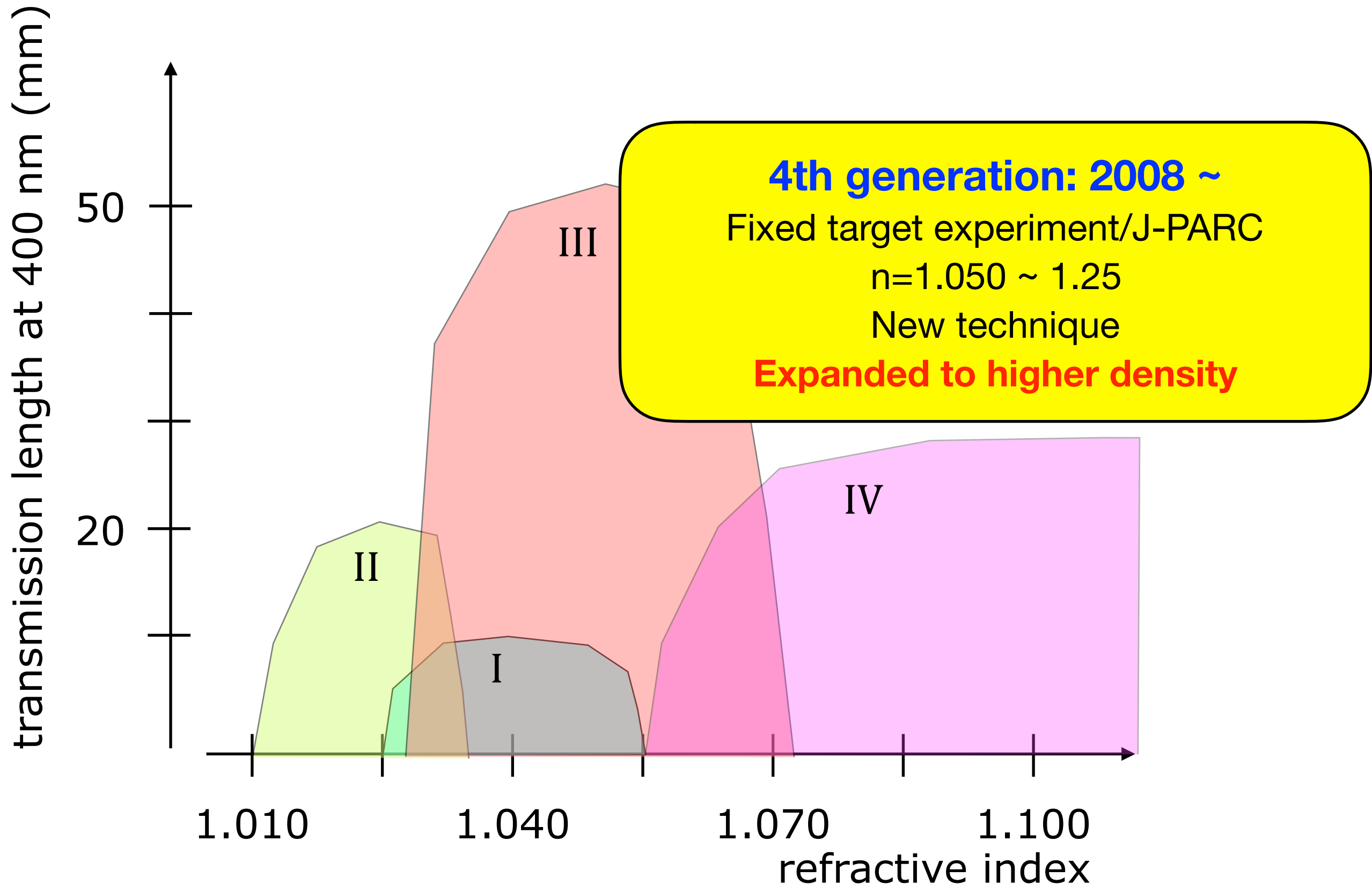




# History of Aerogel Radiators

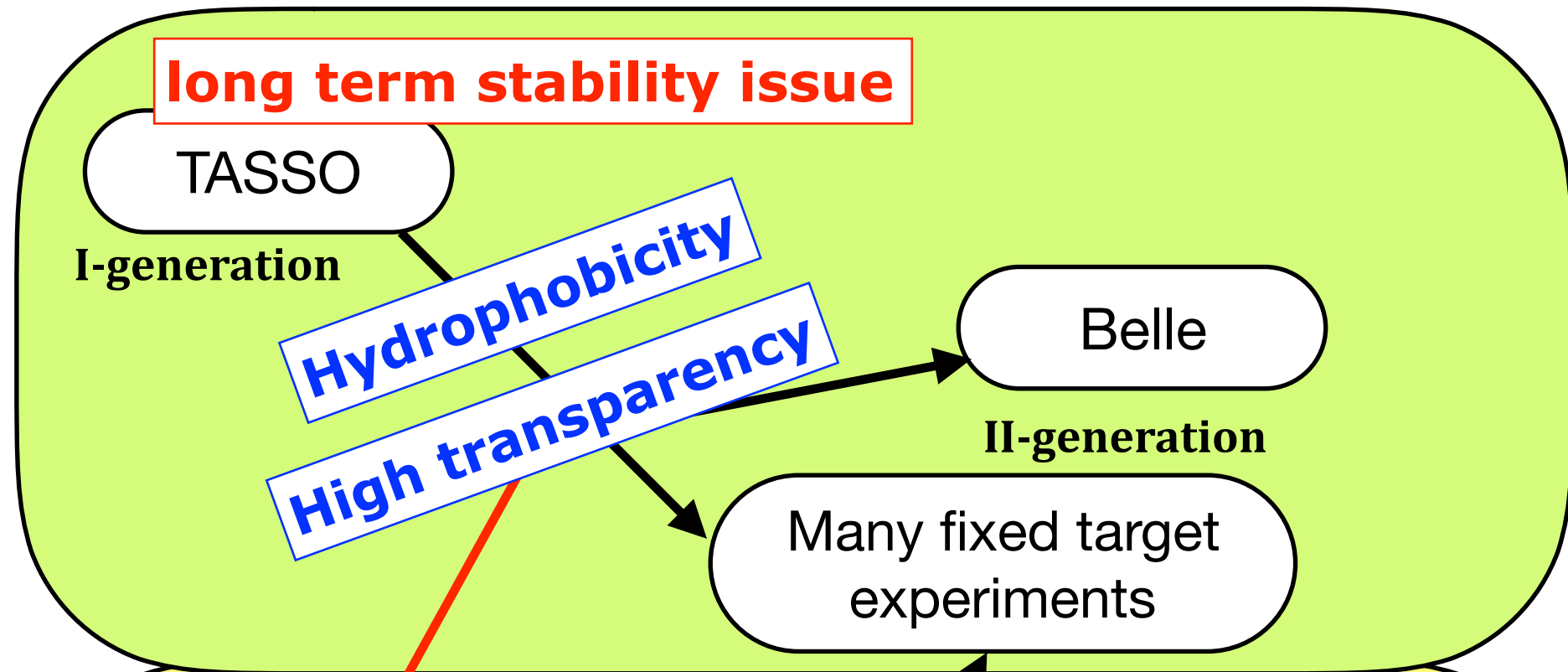


# History of Aerogel Radiators



# Developments of Cherenkov Counters Based on Aerogel

## Threshold Counter



## Imaging Counter

# Belle ACC

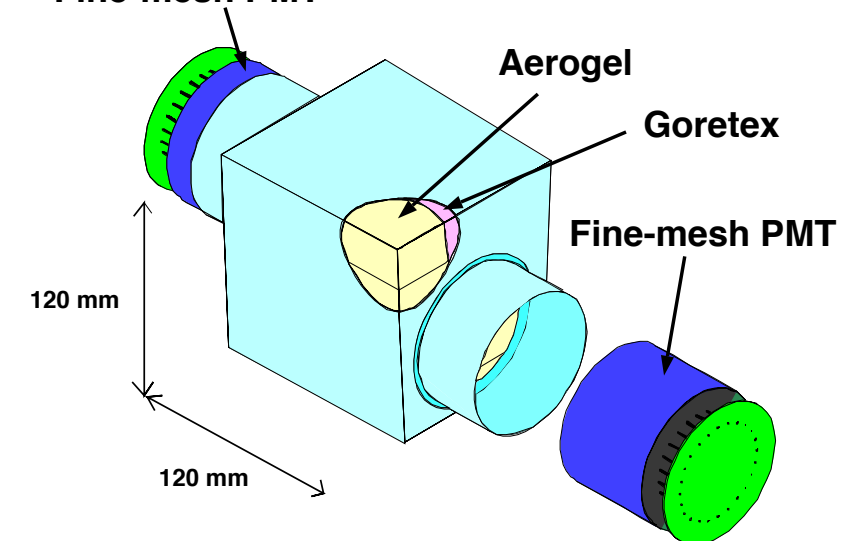
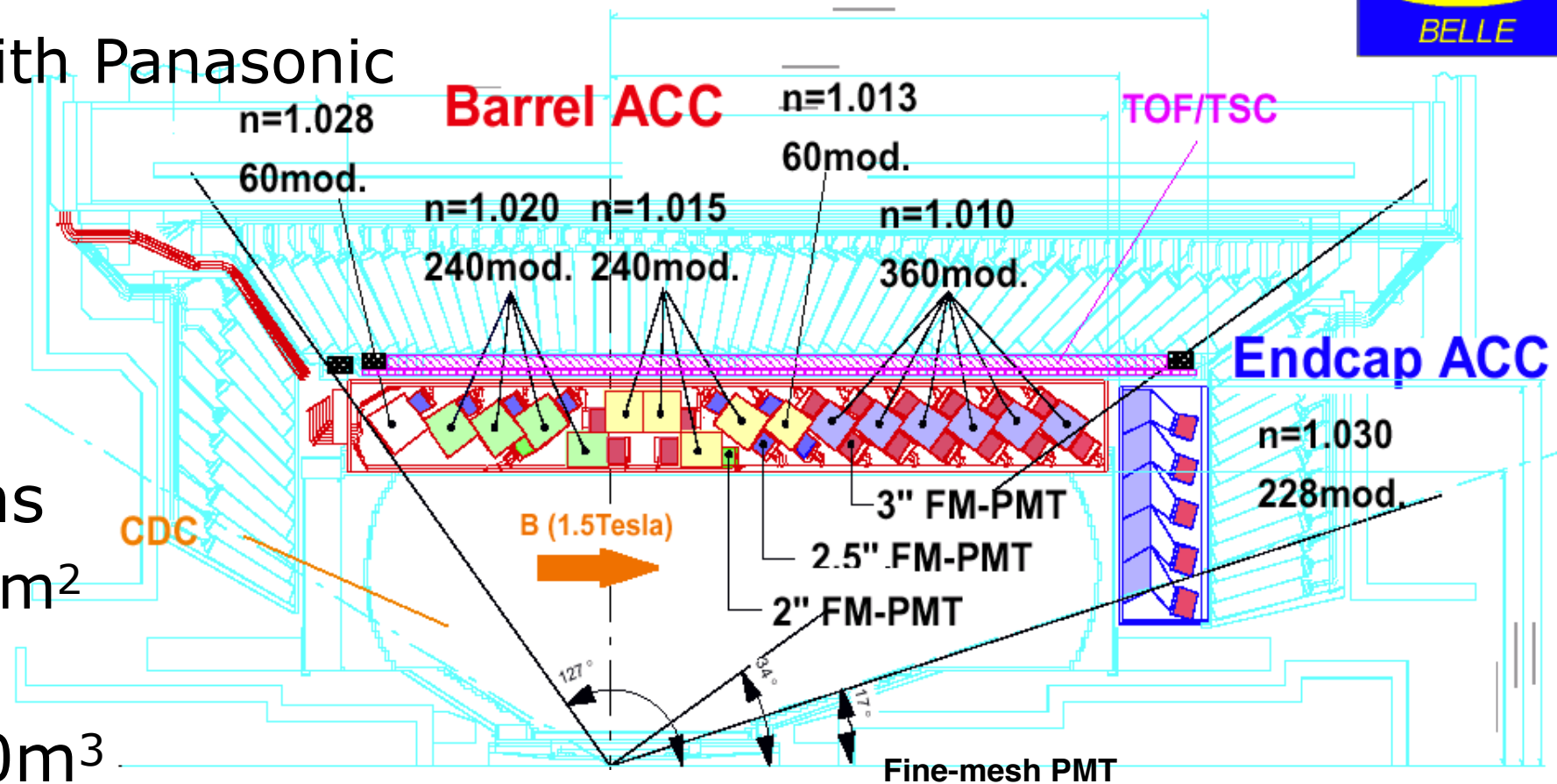
- 1st large detector using the 2nd generation hydrophobic aerogels.

- 1st corporation with Panasonic

- Low index
  - 1.010~1.030

- Tile size dimensions
  - $\sim 100 \times 100 \times 20 \text{ mm}^2$

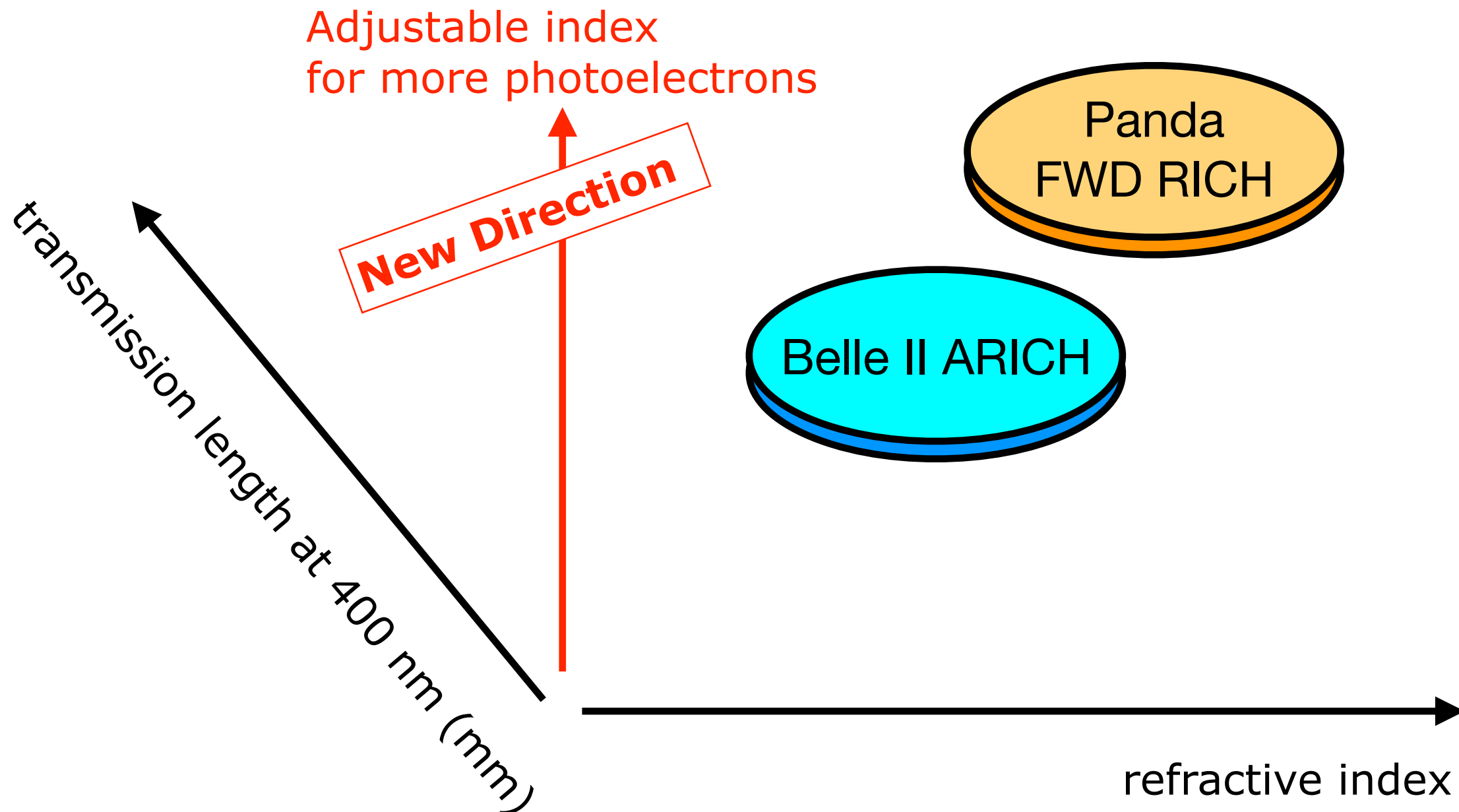
- Total volume  $\sim 1.0 \text{ m}^3$ 
  - Home-made





Highly transparent aerogels in the 3rd generation allows us to have another direction.

Refractive index in aerogel can be tunable.

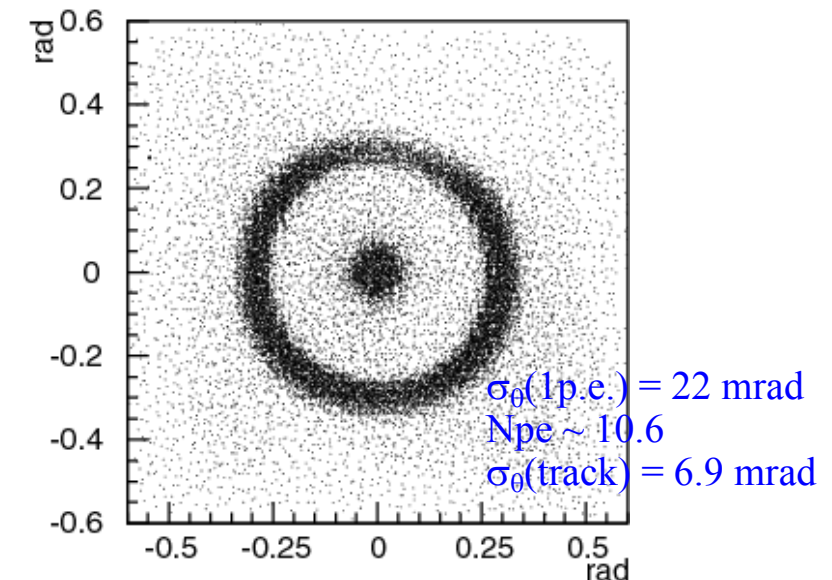
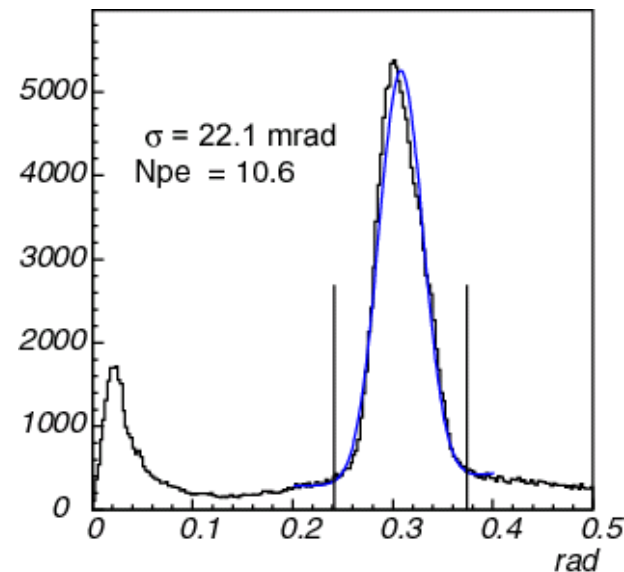
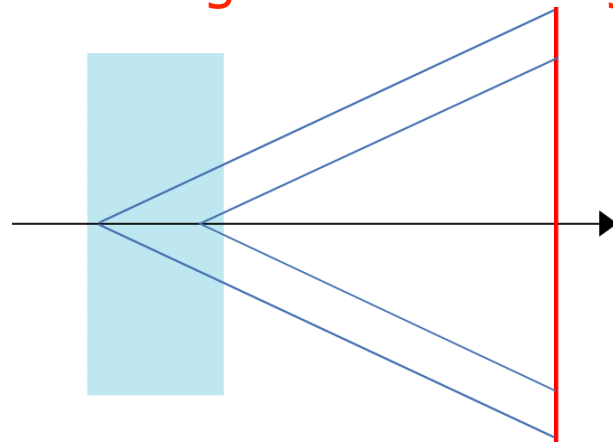


# Belle II RICH

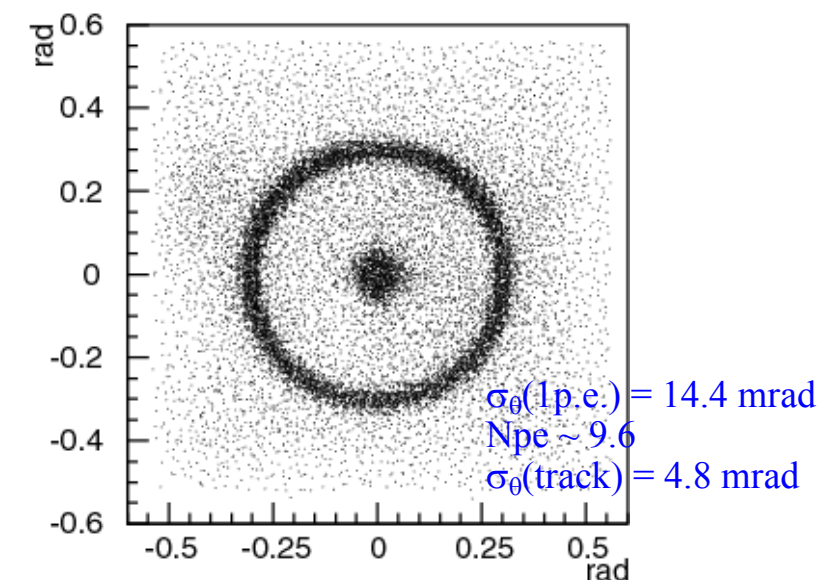
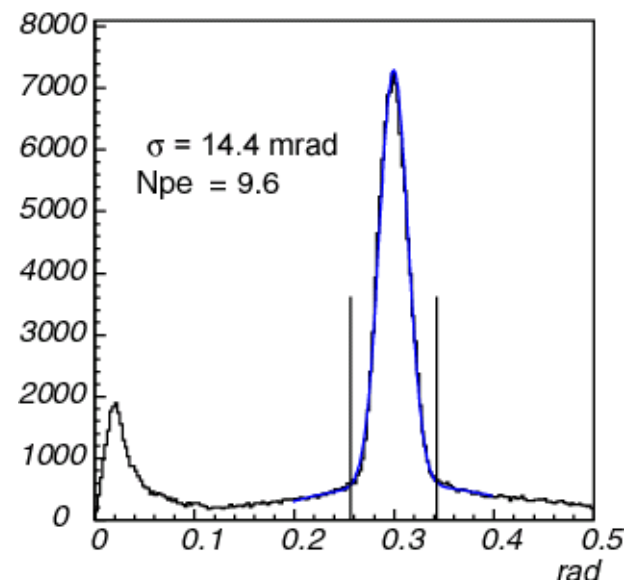
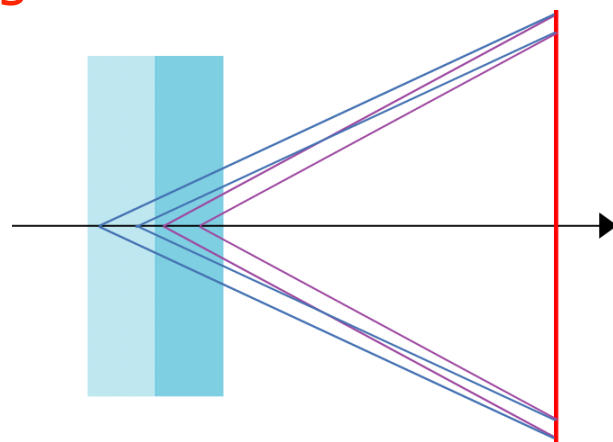
NIMA548(2005)383

- Aerogel in the 3rd generation used.
  - Highly transparent tiles with refractive index  $> 1.03$
  - Dual Cherenkov radiators consisting of 1.045 (upstream) and 1.055 (downstream)

4cm thick single index aerogel



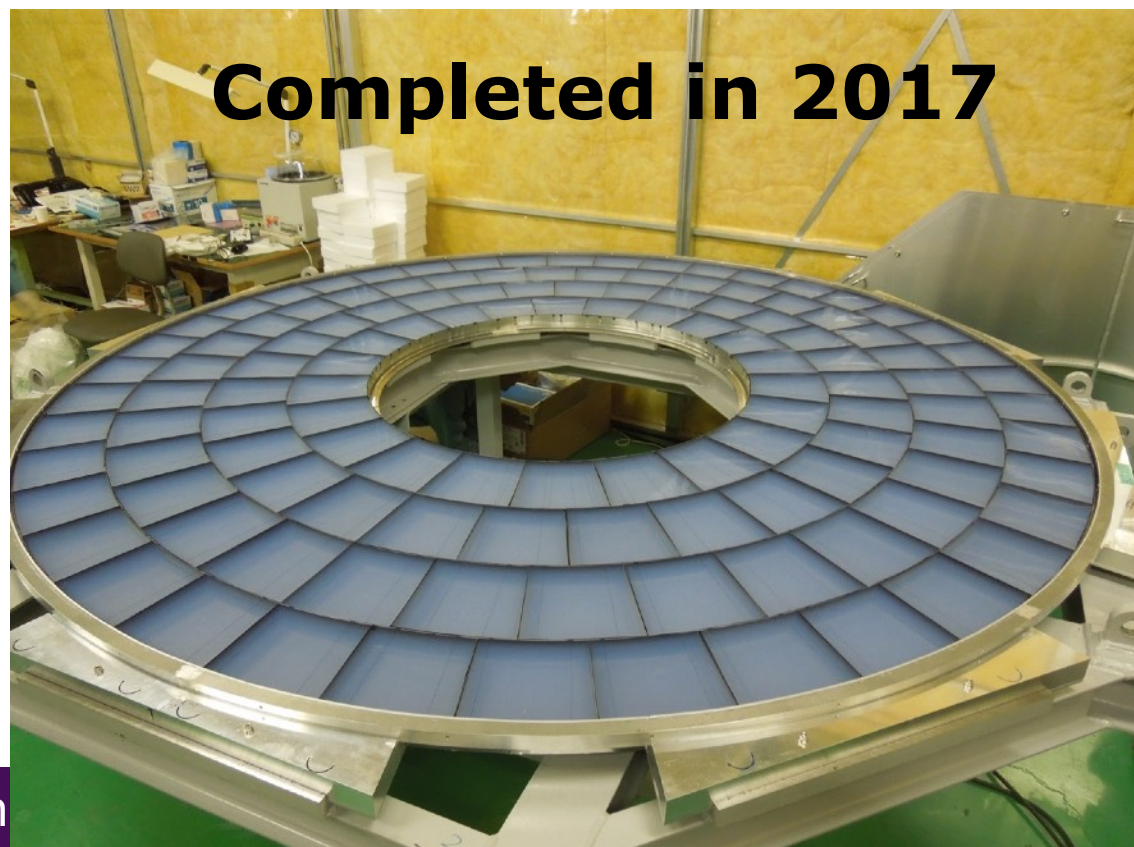
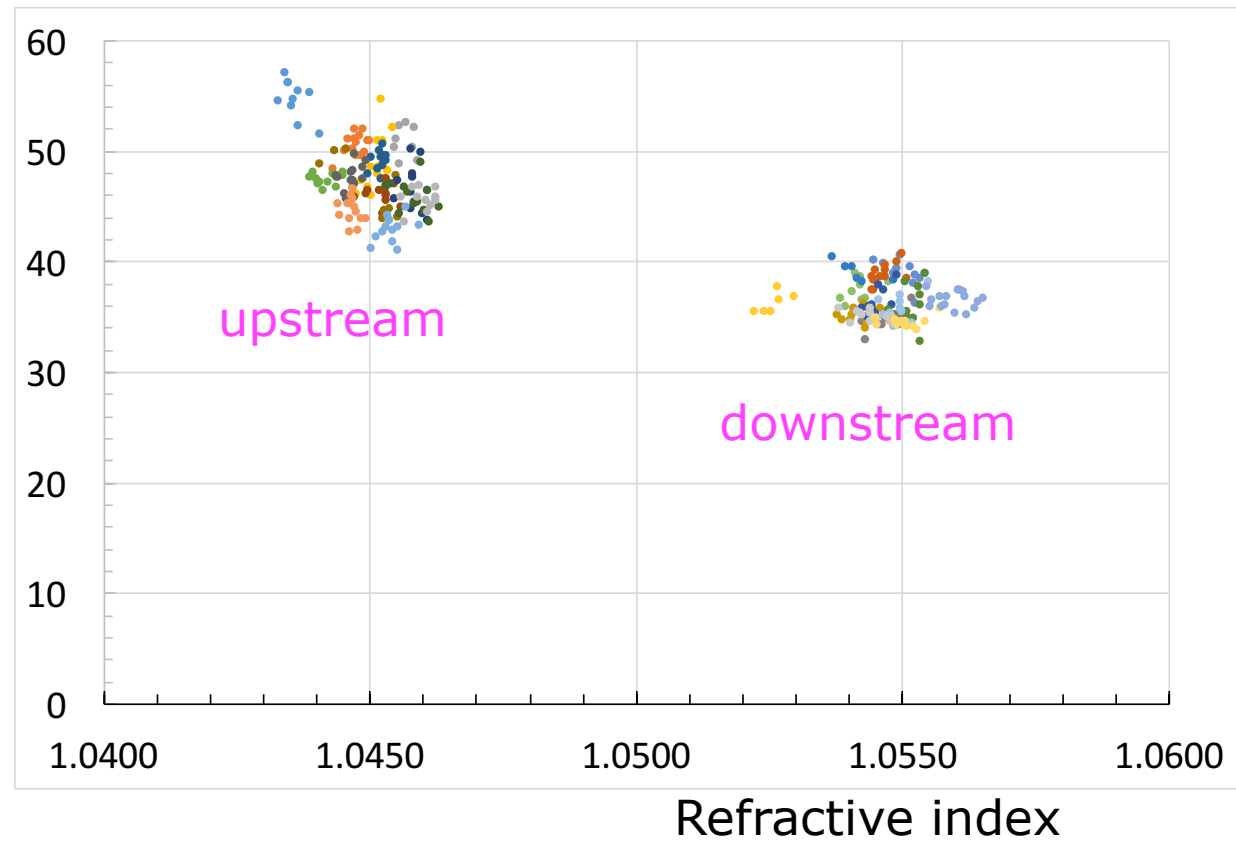
Focusing 2cm+2cm dual aerogel



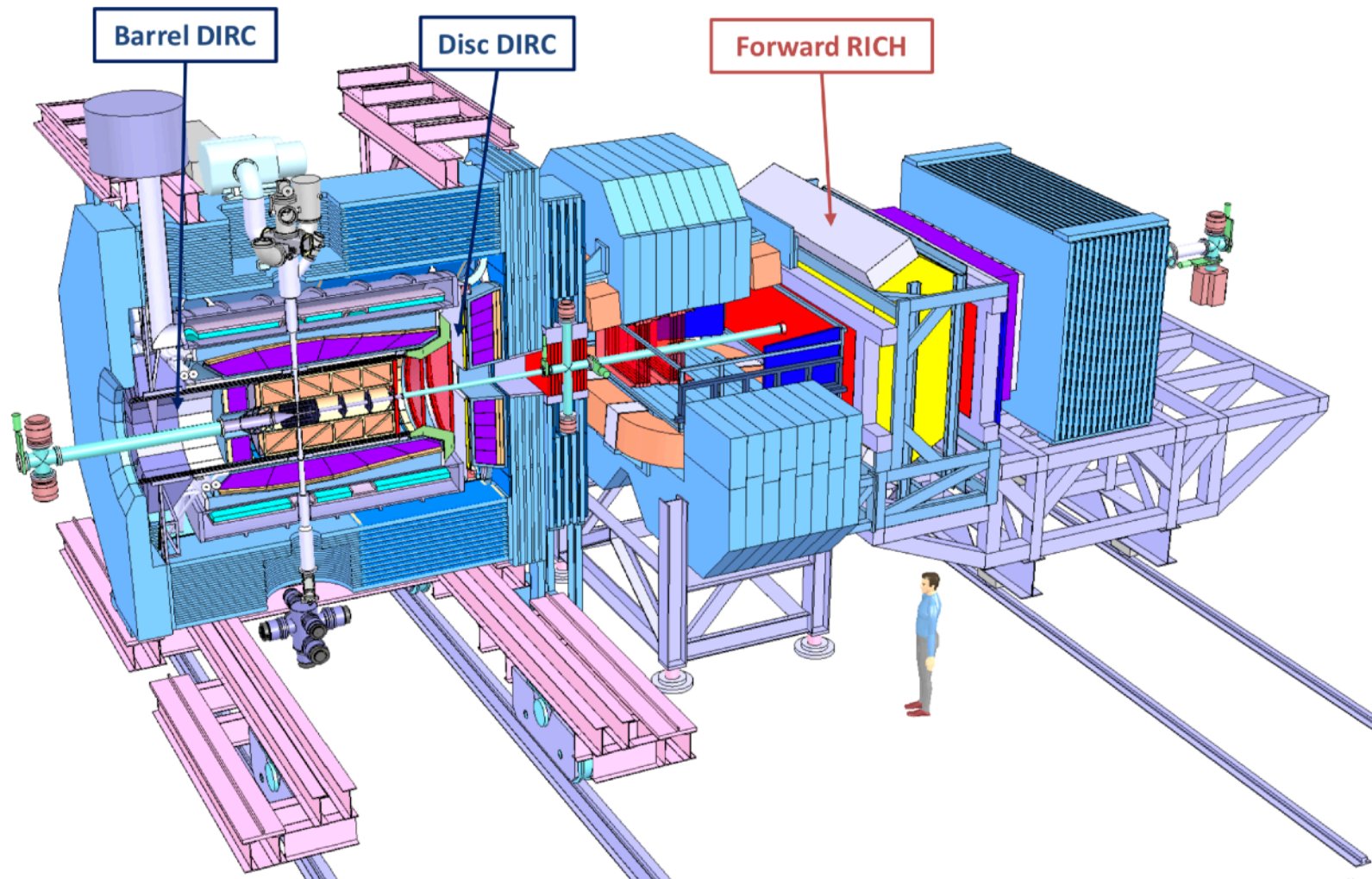


# Belle II RICH

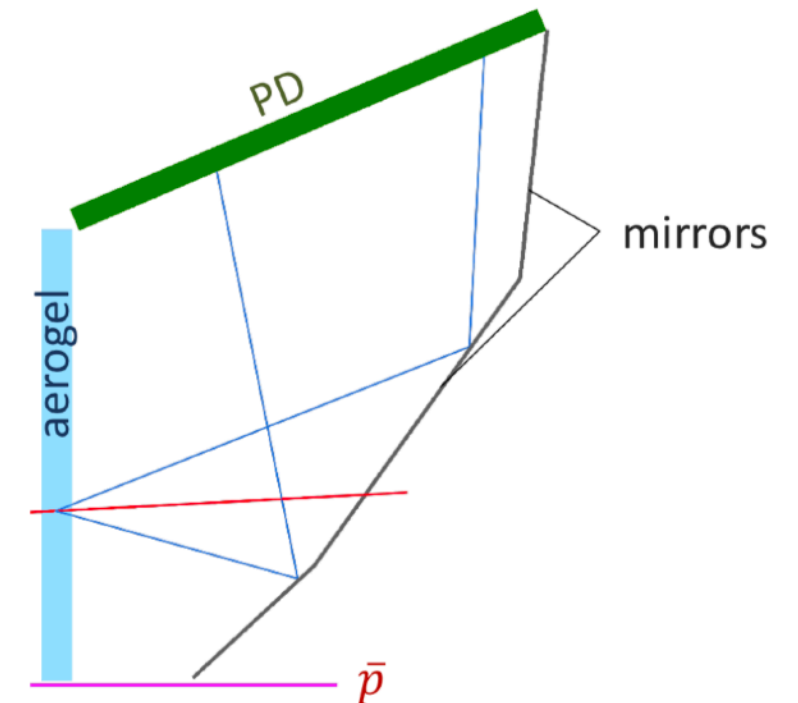
Transmission length at  $\lambda=400\text{nm}$  (mm)



# Panda FWD RICH



Panda spectrometer

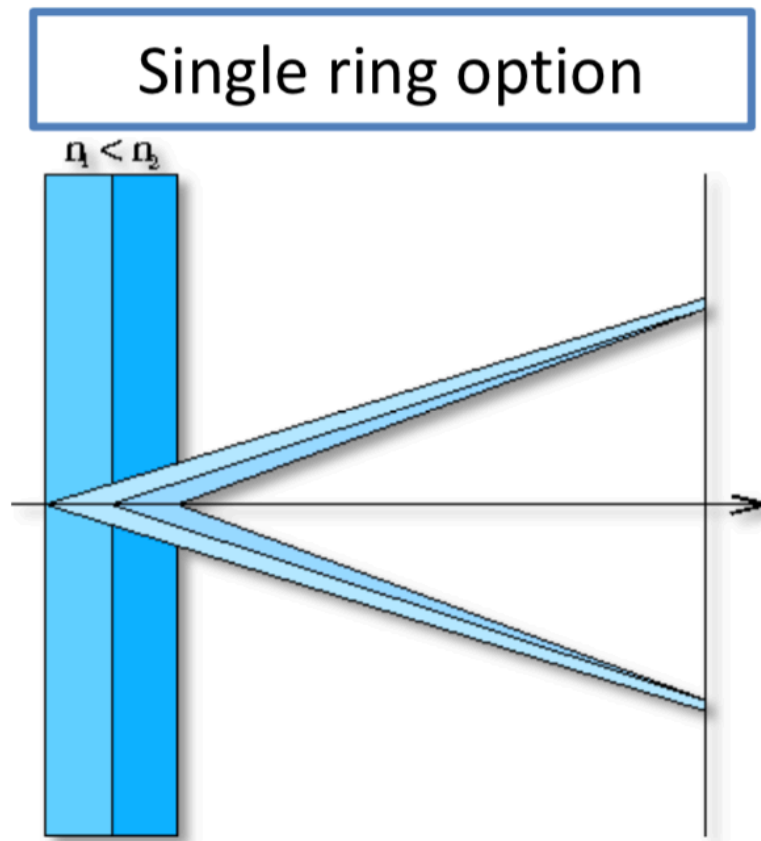


## Radiator

- Focusing 2- or 3-layer aerogel
- 40 mm thick
- No gaseous radiator



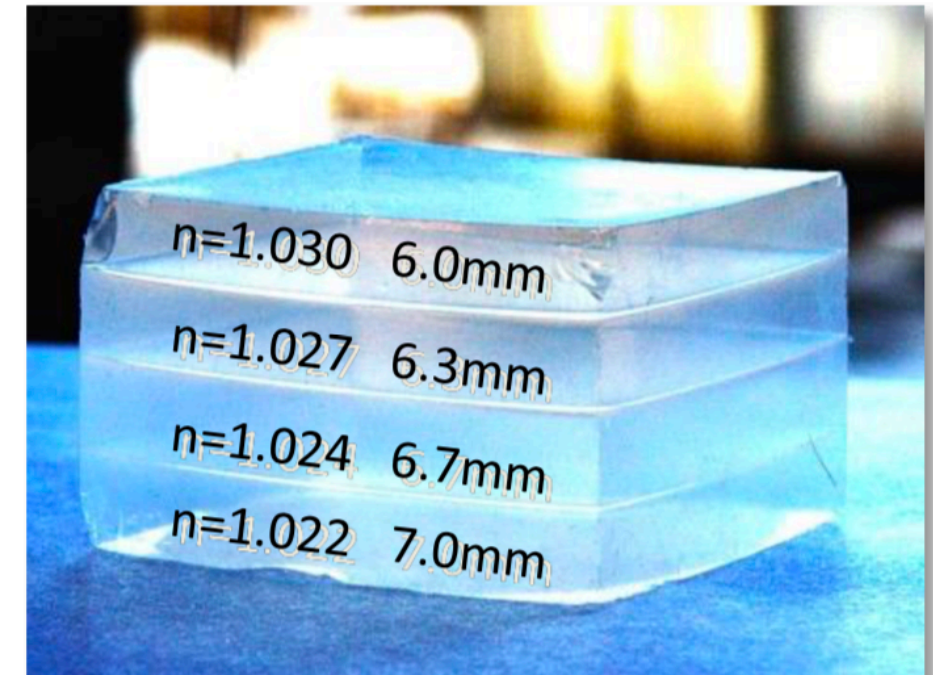
# Panda FWD RICH



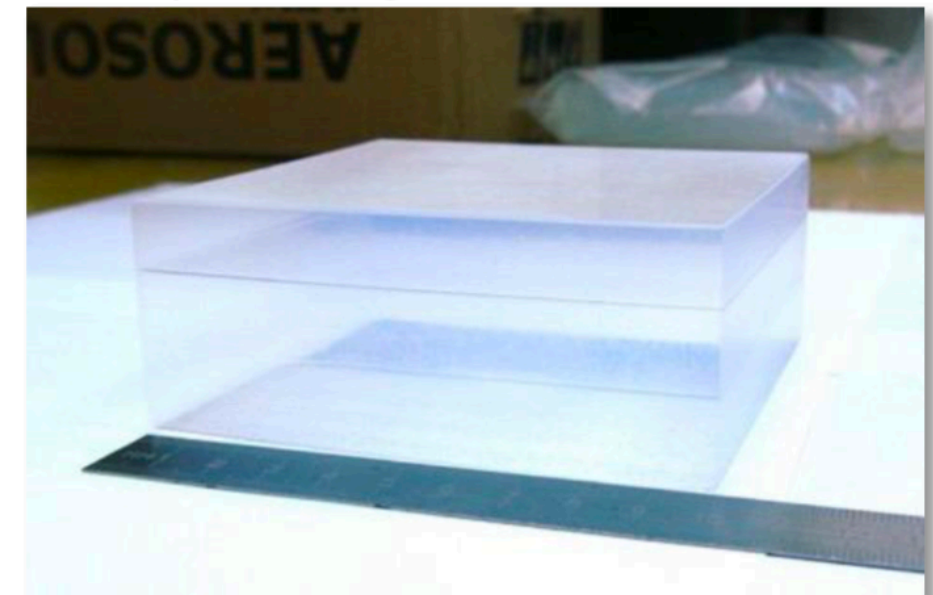
Focusing aerogel improves proximity focusing design by reducing the contribution of radiator thickness into the Cherenkov angle resolution

Multi-layer monolith aerogels have been being produced by the Boreskov Institute of Catalysis in cooperation with the Budker INP since 2004.

First sample of 4-layer aerogel



3-layer aerogel 115x115x41 mm<sup>3</sup>

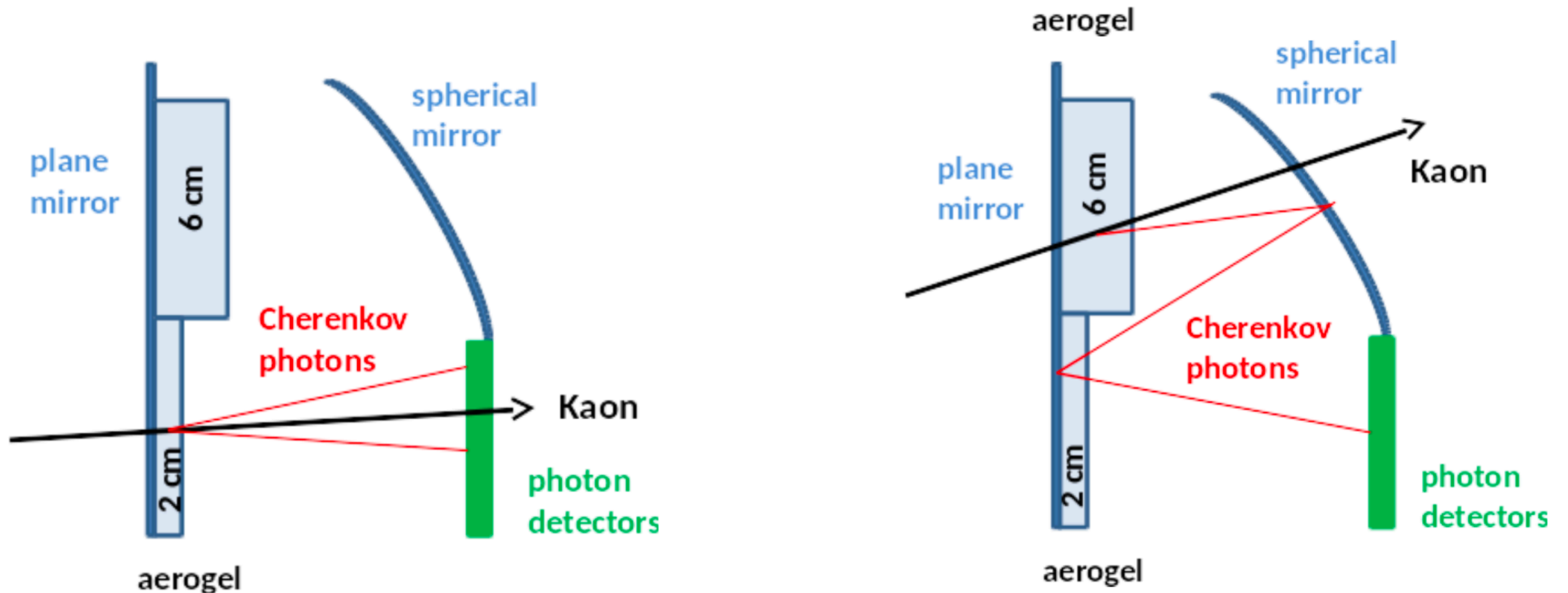


T.Iijima et al., NIM A548 (2005) 383

A.Yu.Barnyakov et al., NIM A553 (2005) 70

# CLAS12 RICH

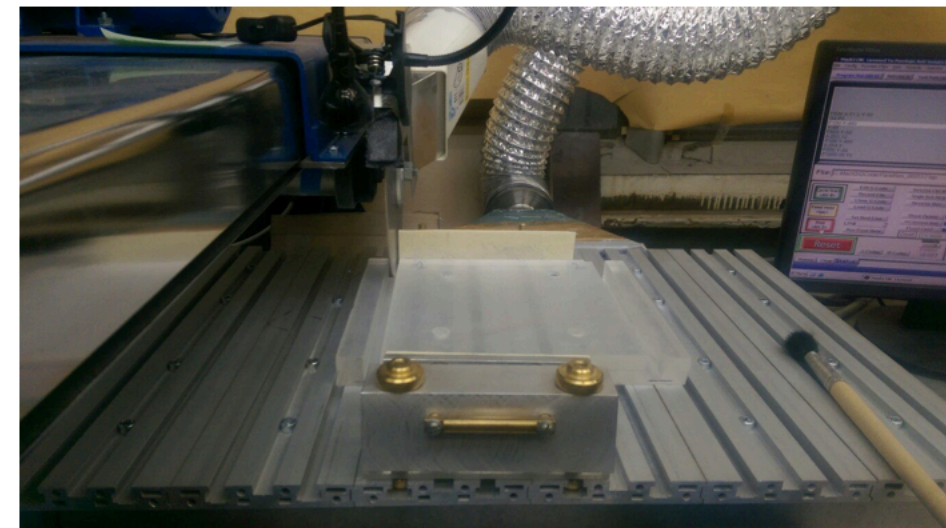
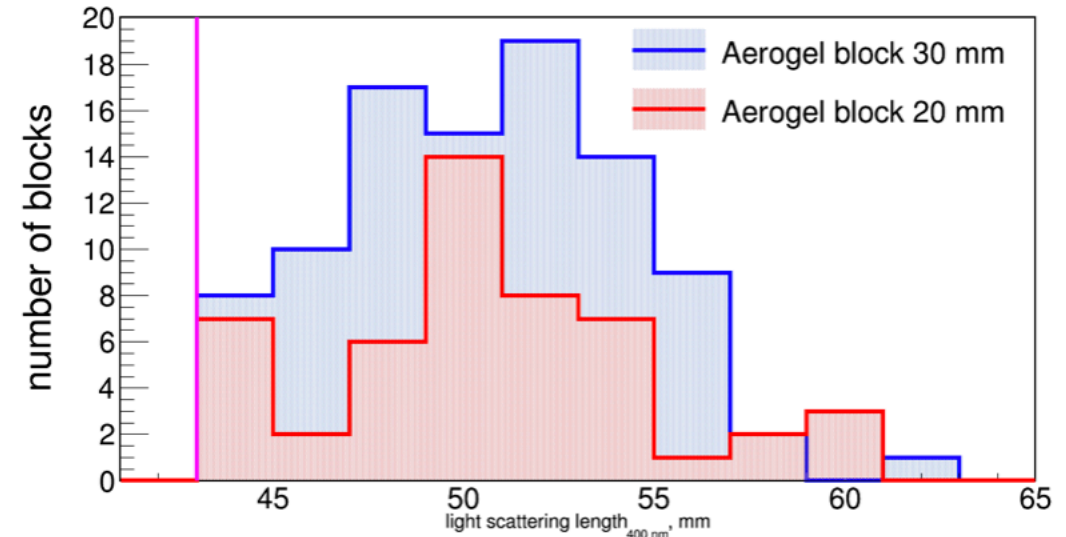
- Challenging design due to the constraints from the present CLAS spectrometer.
- 2 layers of 30 mm thick slice aerogels ( $n=1.05$ ). Two reflections (one for spherical mirror and the other planer through aerogel medium) before photon detector



# CLAS12 RICH

## Production of large aerogel tiles

- 200x200 mm<sup>2</sup>, 20 and 30 mm thickness
- $L_{sc}(400\text{nm}) > 43\text{ mm}$   
(mean  $\sim 50\text{ mm}$ )
- New CNC cutting machine (diamond wheel). The cutting accuracy is 0.25 mm.

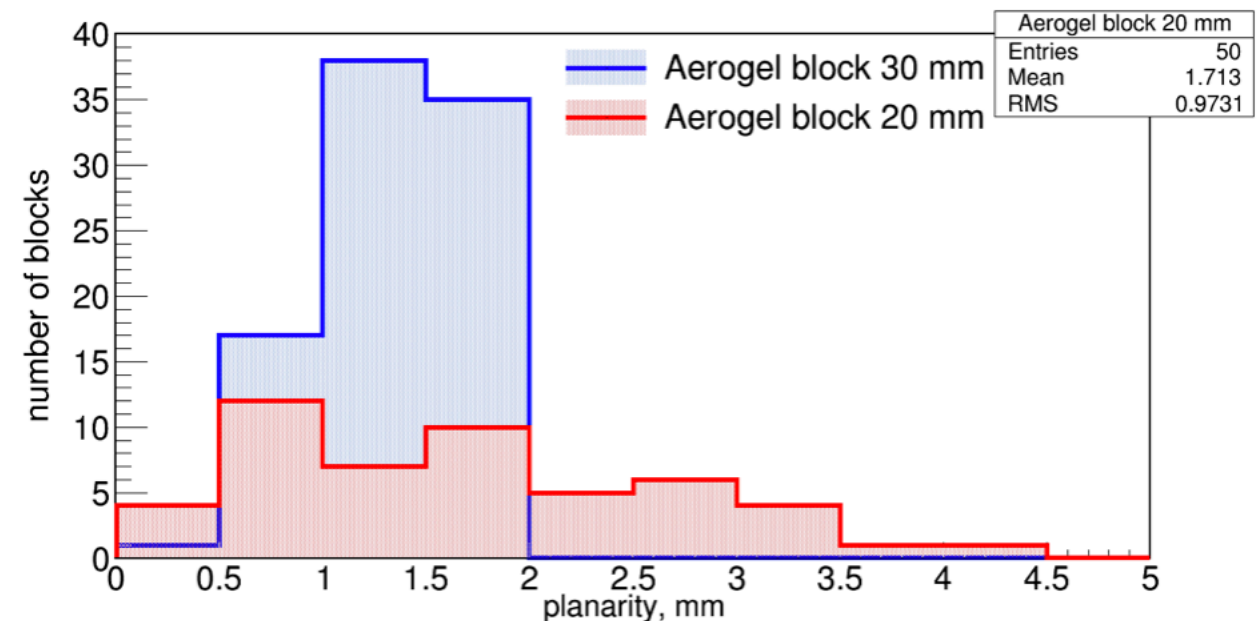




# CLAS12 RICH

## Planarity of large aerogel tiles

- The aerogel tile planarity is measured mechanically in 49 points (7x7 grid with step 30 mm). The planarity criteria is defined as the difference between the maximum and minimum values of the coordinate field after alignment procedure. Selection criteria:
  - the aerogel tile 30 mm, planarity up to 2 mm;
  - the aerogel tile 20 mm, planarity up to 4.5 mm, but the average planarity of all blocks does not exceed 3 mm.

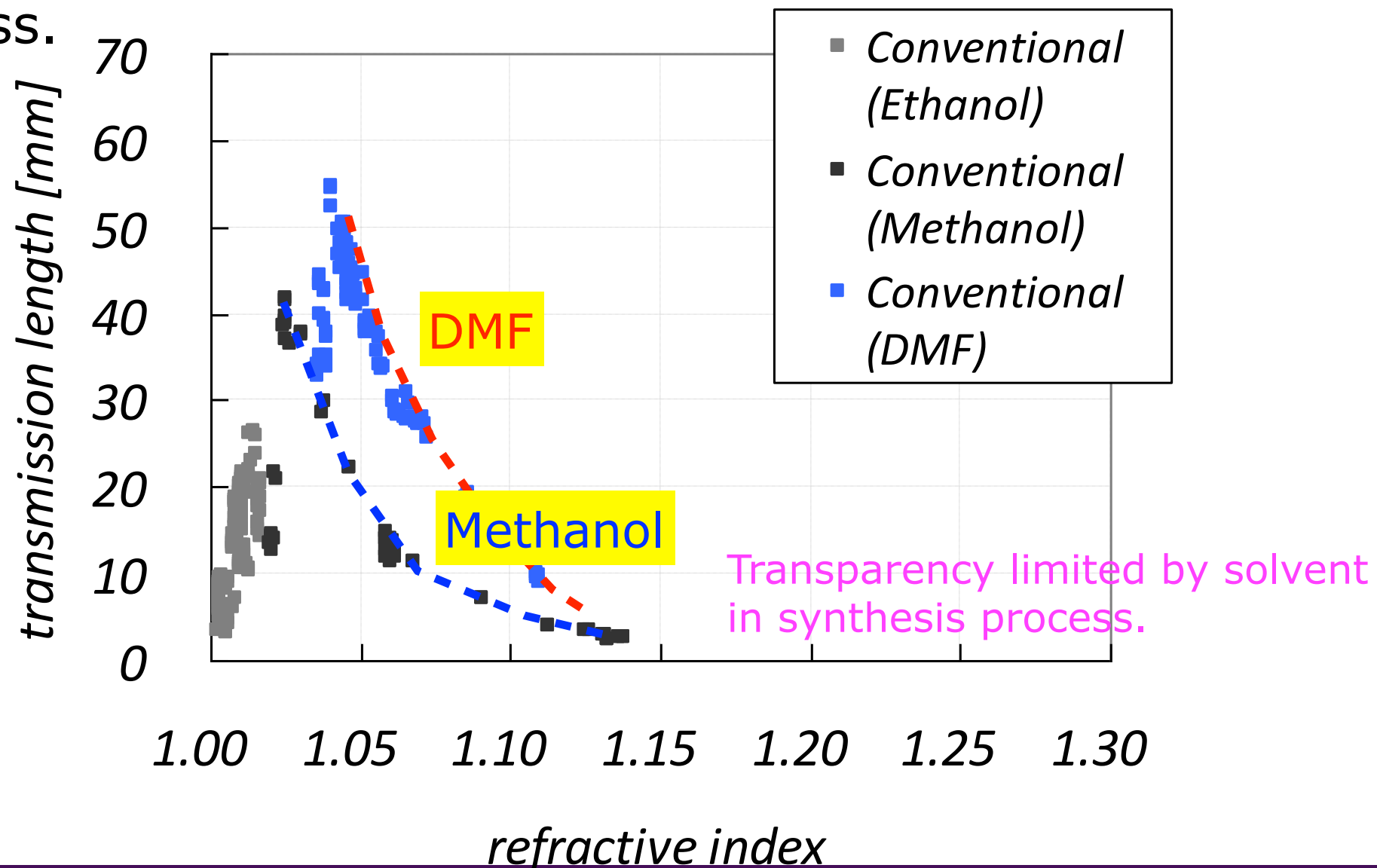


Long-term stability  
Monitoring is essential.



# 4th Generation

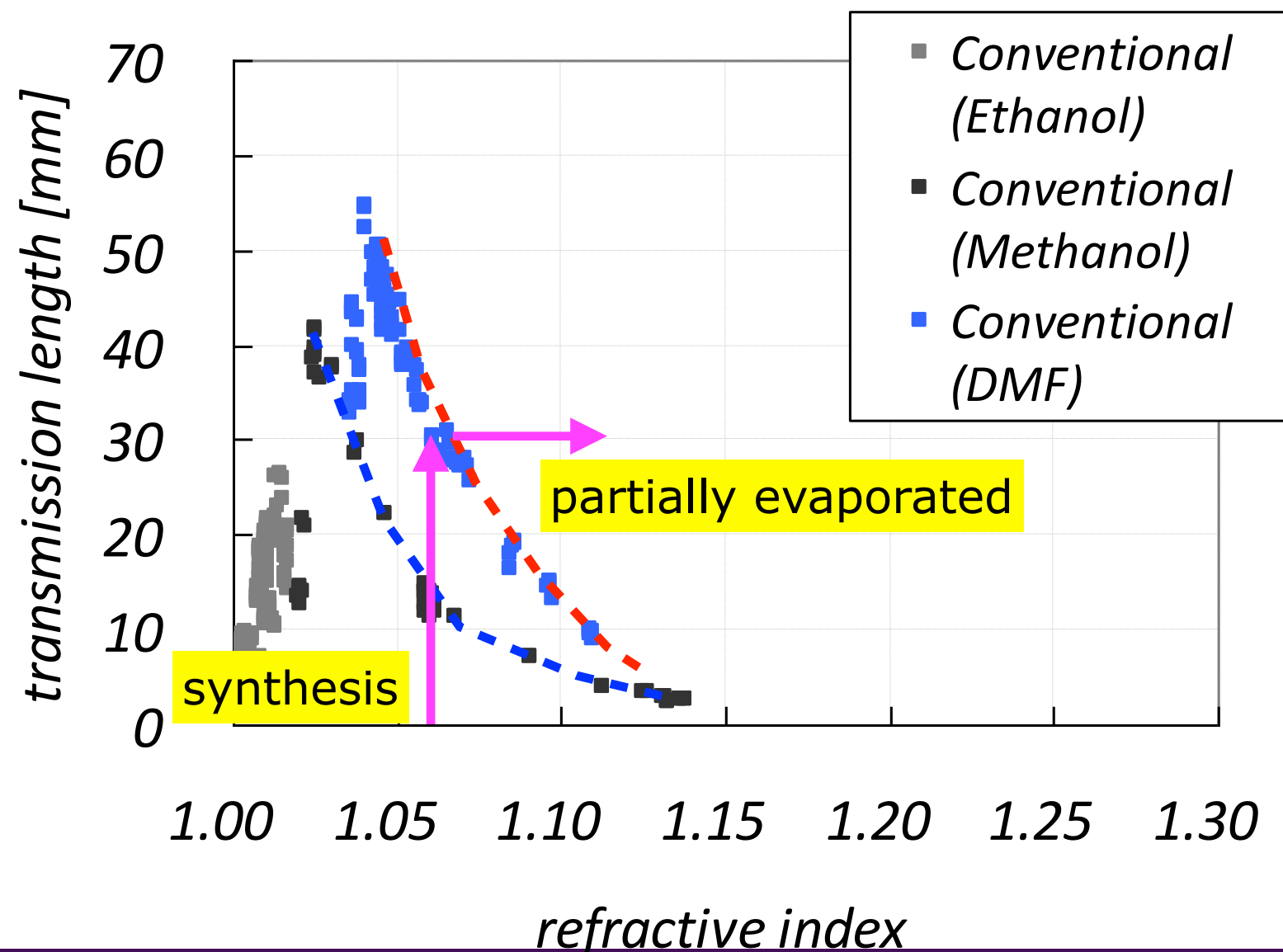
- High refractive index ( $> \sim 1.14$ )
  - Transparency limited by solvent in synthesis step.
  - Difficult to produce alco-gels since too much silica particles can not undergo smooth gelation.
  - This results in non-transparent aerogel after supercritical drying process.



# 4th Generation

Synthesis process targeting  $n \sim 1.06$ , then alco-gel obtained is partially evaporated in controlled environment to get target index(=density).

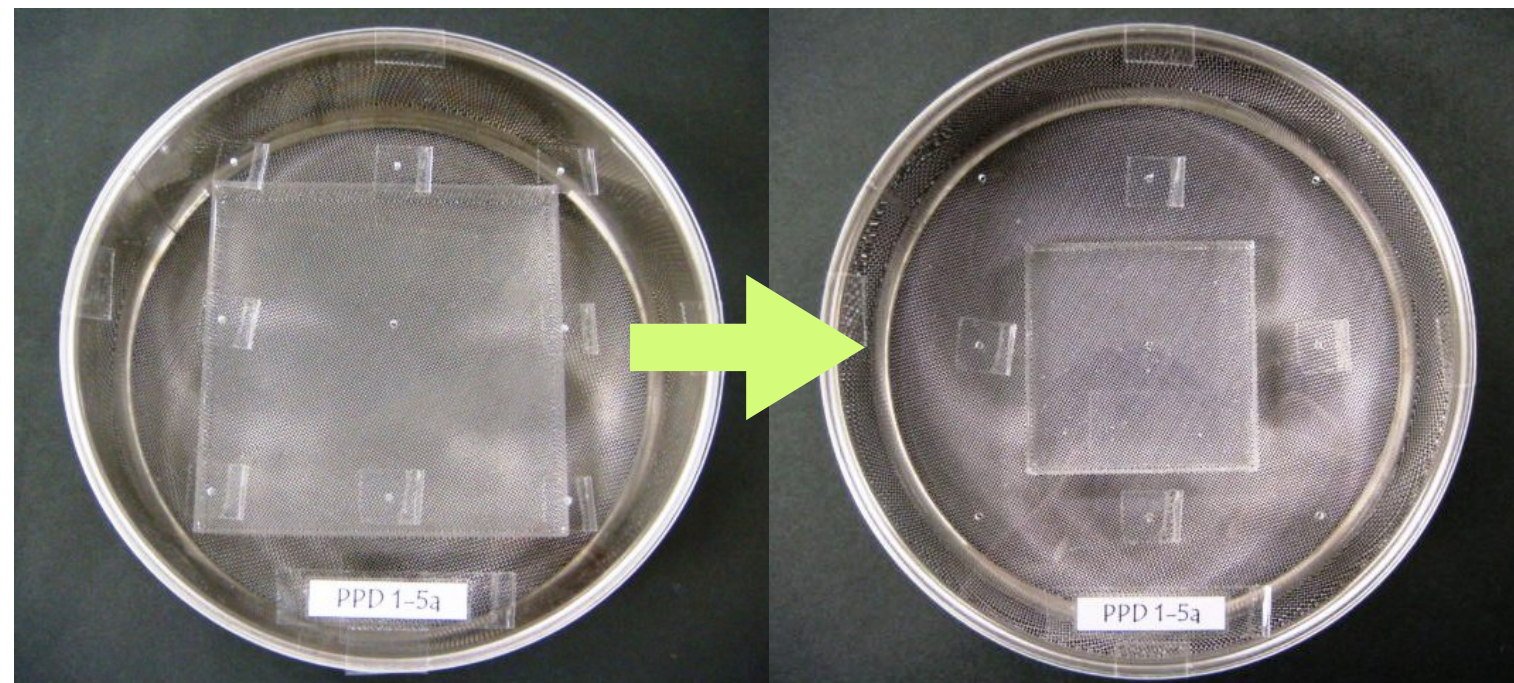
## Pin-drying(PD) method by Chiba university group



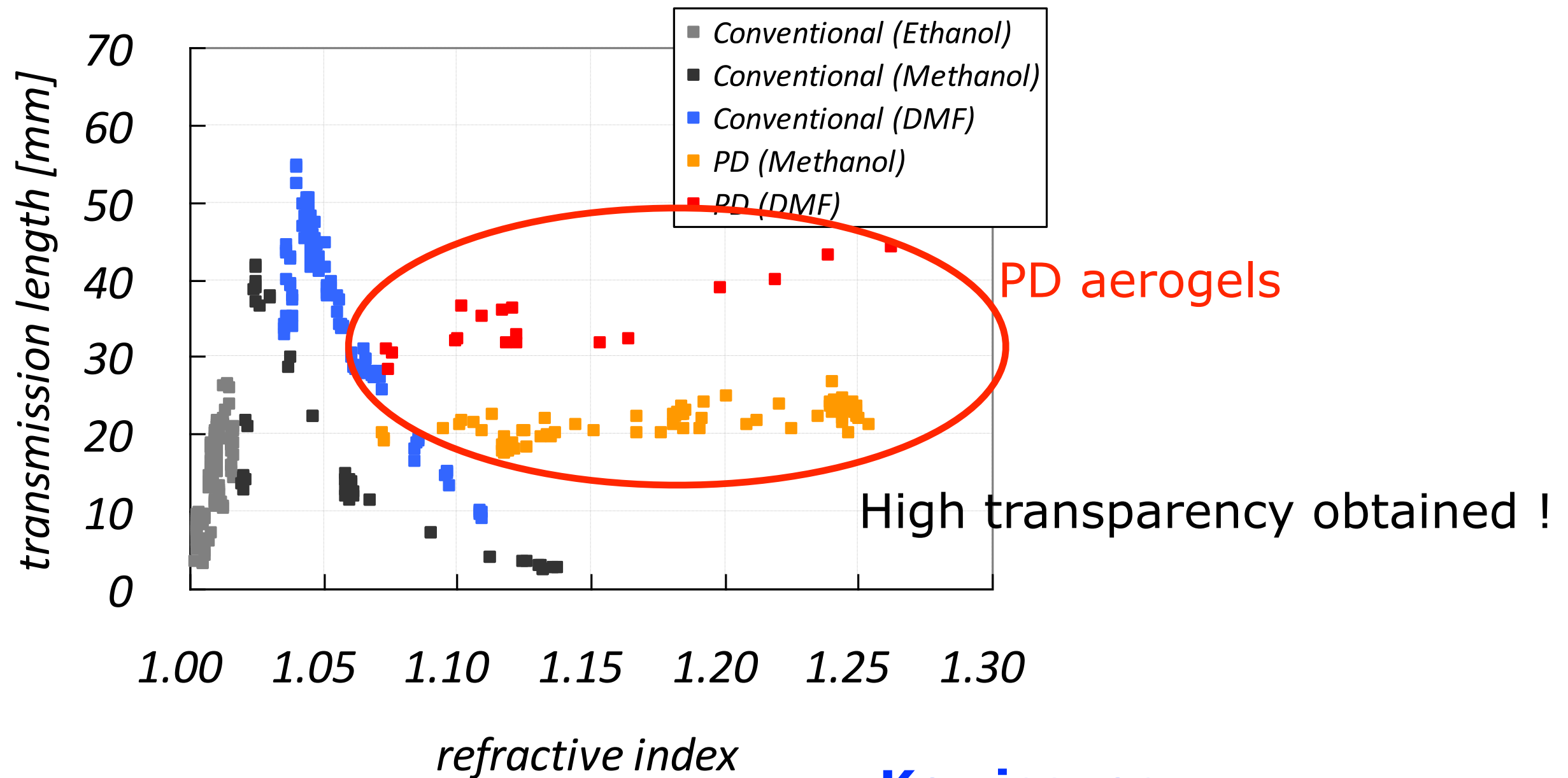
# 4th Generation

Synthesis process targeting  $n \sim 1.06$ , then alco-gel obtained is partially evaporated in controlled environment to get target index(=density).

**Pin-drying(PD) method by Chiba university group**



# 4th Generation



## Key issues

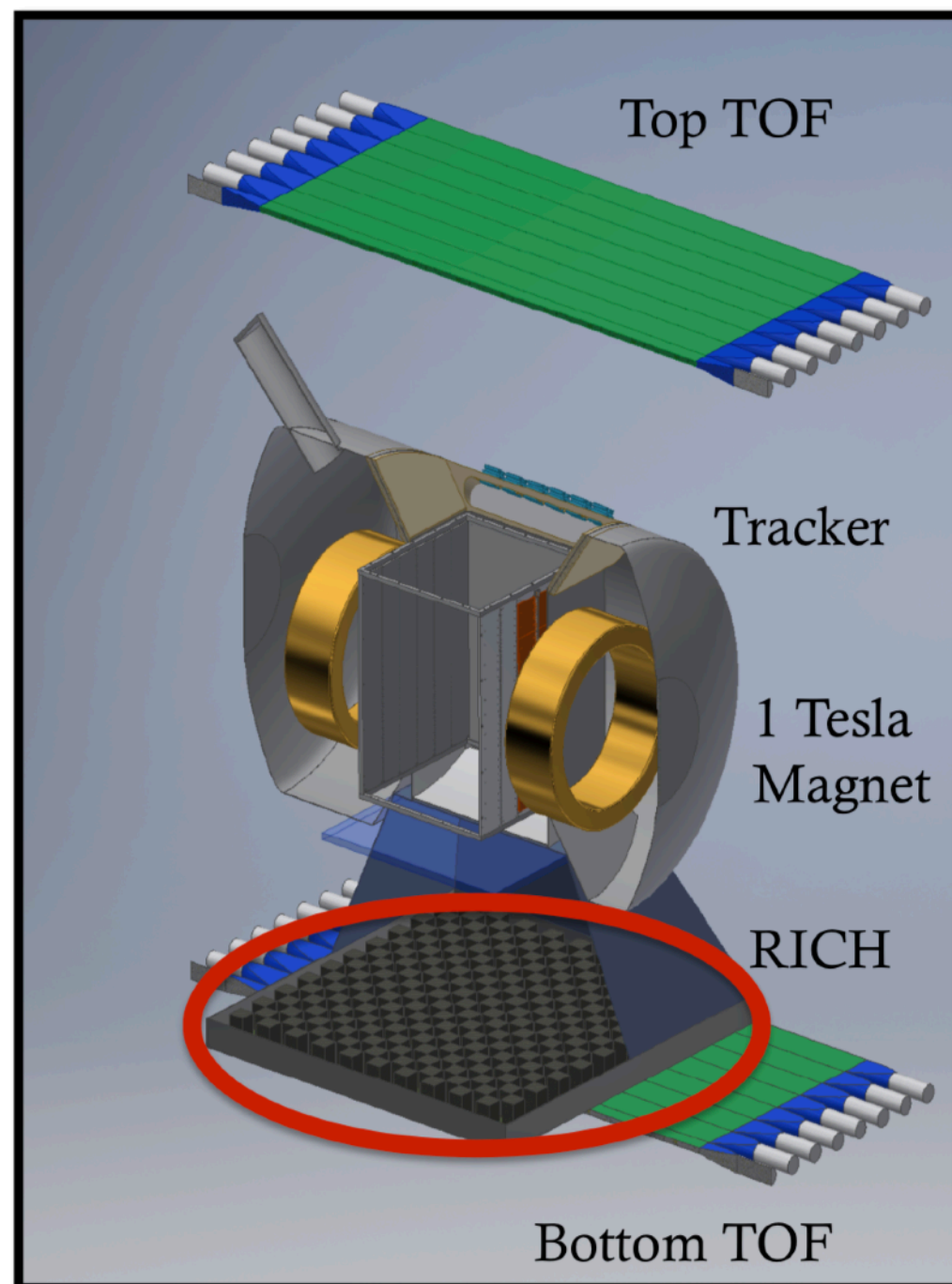
Tile size and uniformity  
Duration of production



# HELIX RICH

- HELIX is a magnet spectrometer for a balloon experiment around the south pole. Launch ~ 2019-2020.

Univ. Chicago ...



## The Ring-imaging Cherenkov Detector

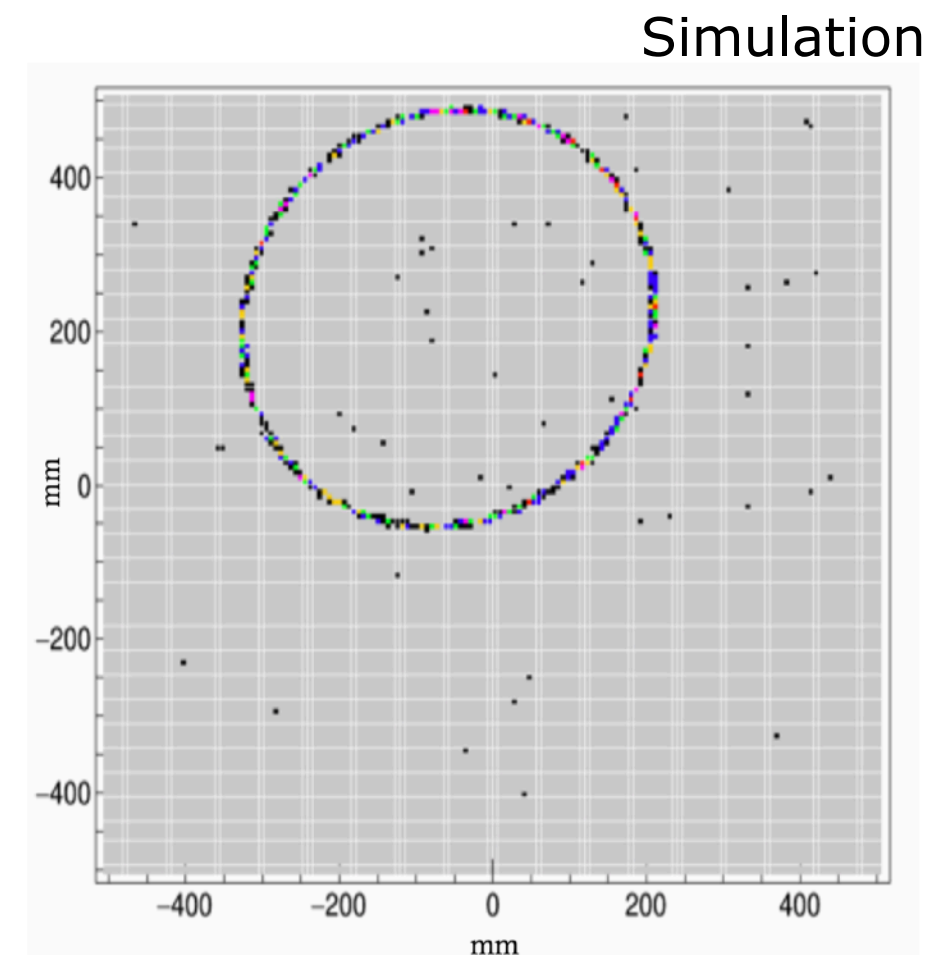
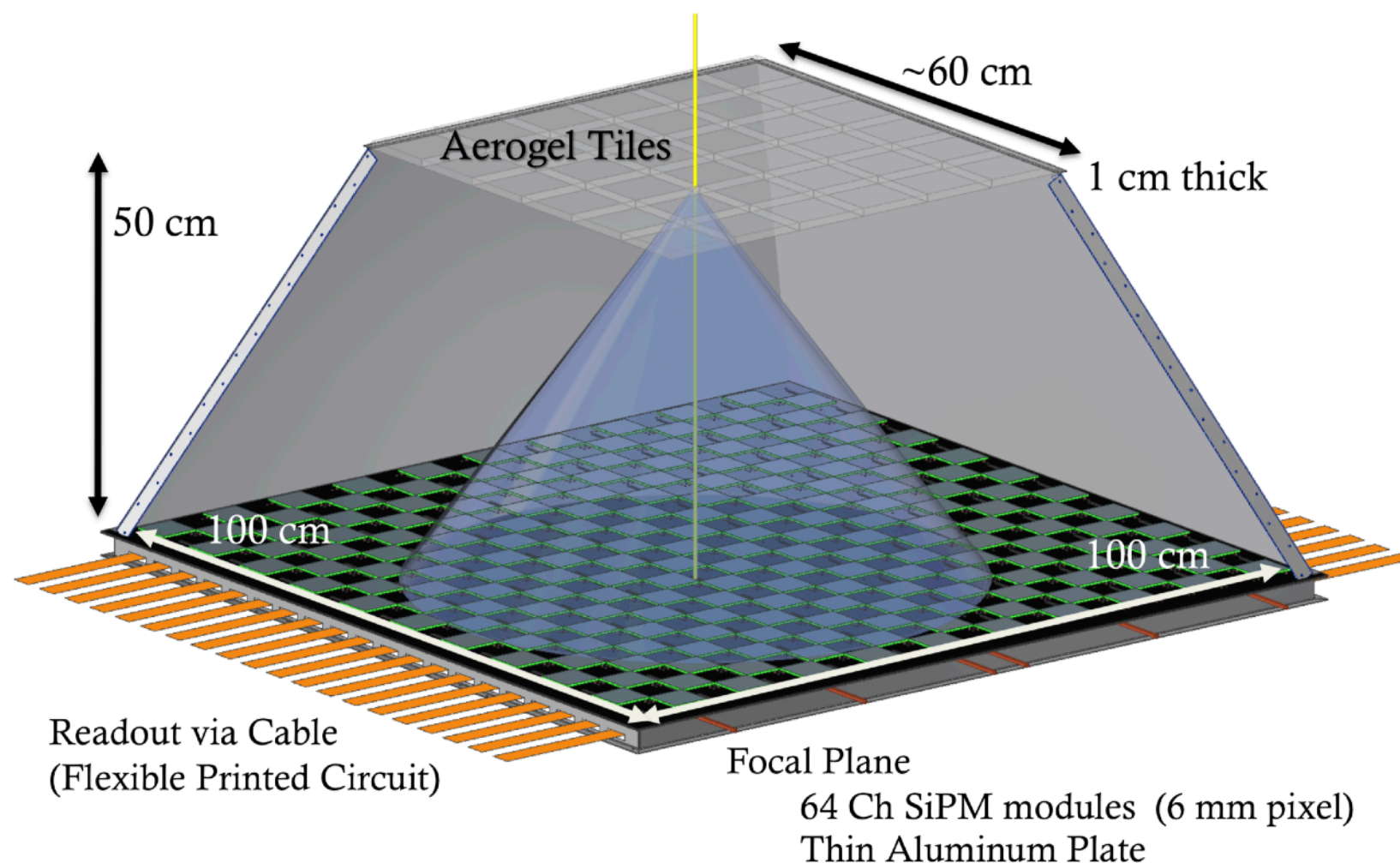
- Proximity-focused RICH w/ **SiPM** readout
- Design goal :  $\Delta\beta/\beta \sim 4 \times 10^{-4}$  for  $Z > 3$
- Requires detector developments to reach goals

Presented at ICRC 2017

# HELIX RICH

## Cherenkov radiator system using 4th generation aerogel

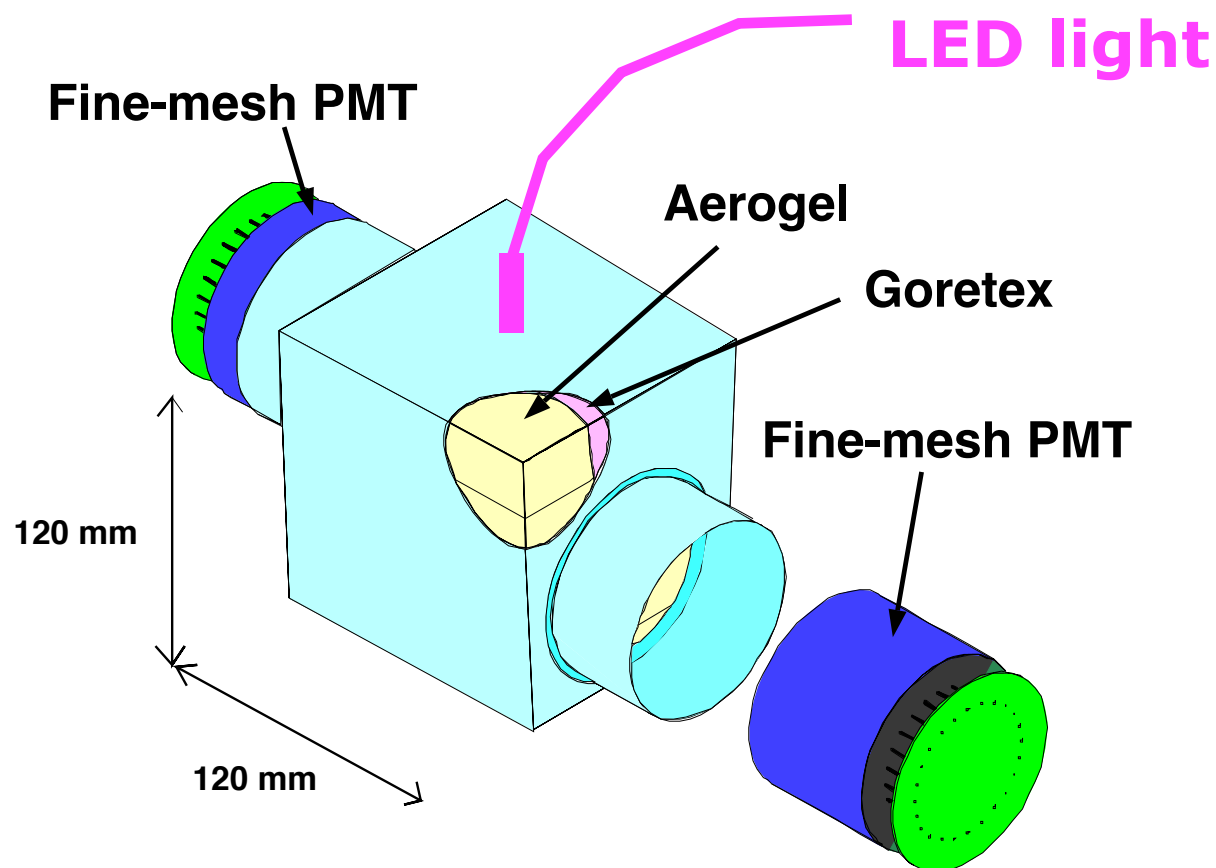
- $n=1.15$  Aerogel from Chiba university group
- 36 tiles with  $100 \times 100 \times 10 \text{ mm}^3$
- Target transmission length at 400 nm  $> 30 \text{ m}$



# **Application Related Issues**

# Long-Term Stability

- In Belle aerogel Cherenkov counter we measured light output from PMTs for each counter by eliminating LED light.



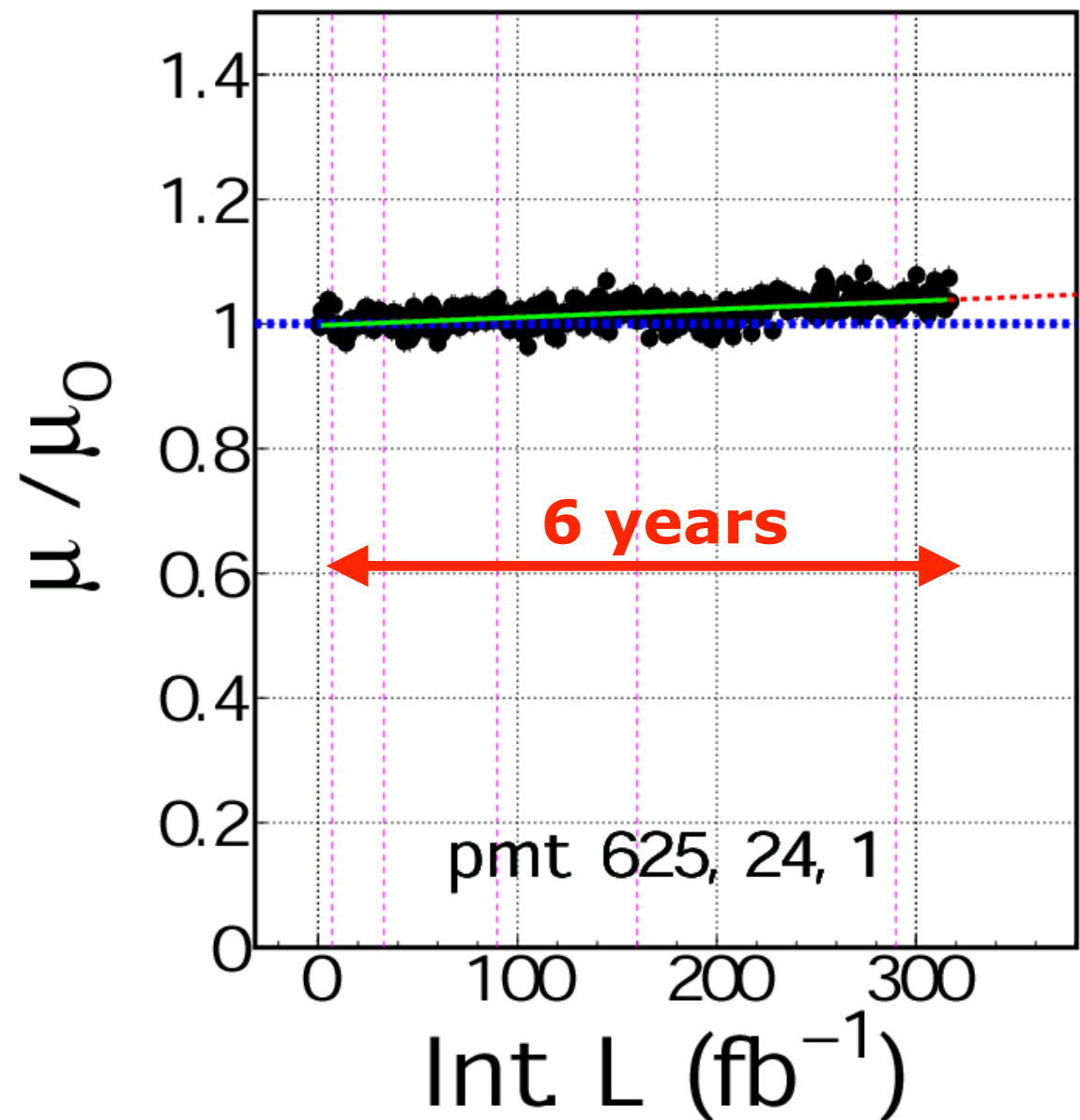
Aerogel tiles of  $n=1.01-1.03$   
They are hydrophobic

- Check long-term stability.

# Long-Term Stability

- Very stable output has been obtained.
- This is a mixture of stabilities of:
  - Aerogel transparencies
  - PMT gain

**Transparency for hydrophobic aerogel tiles at Belle is stable.**

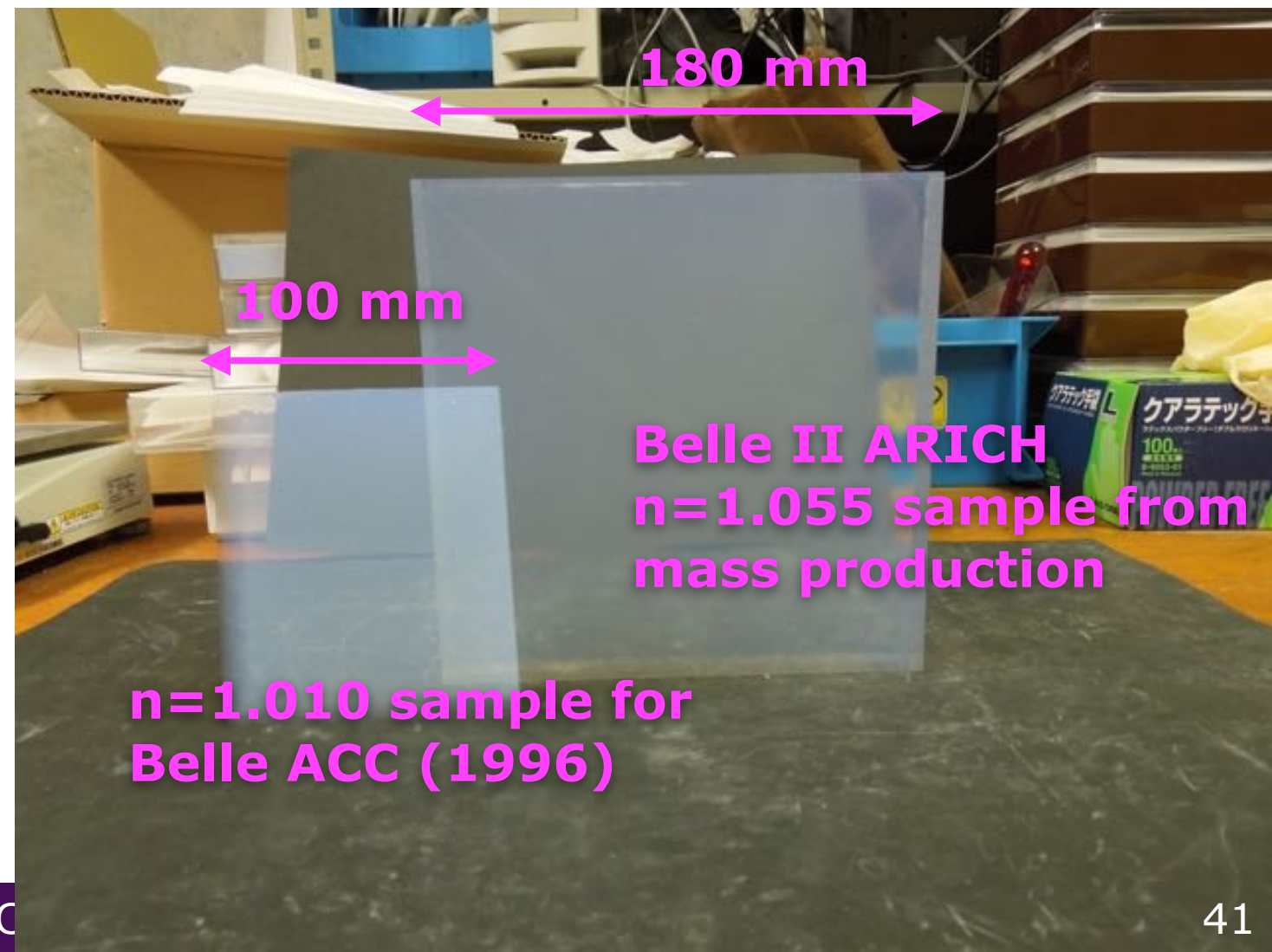




# Tile Dimensions

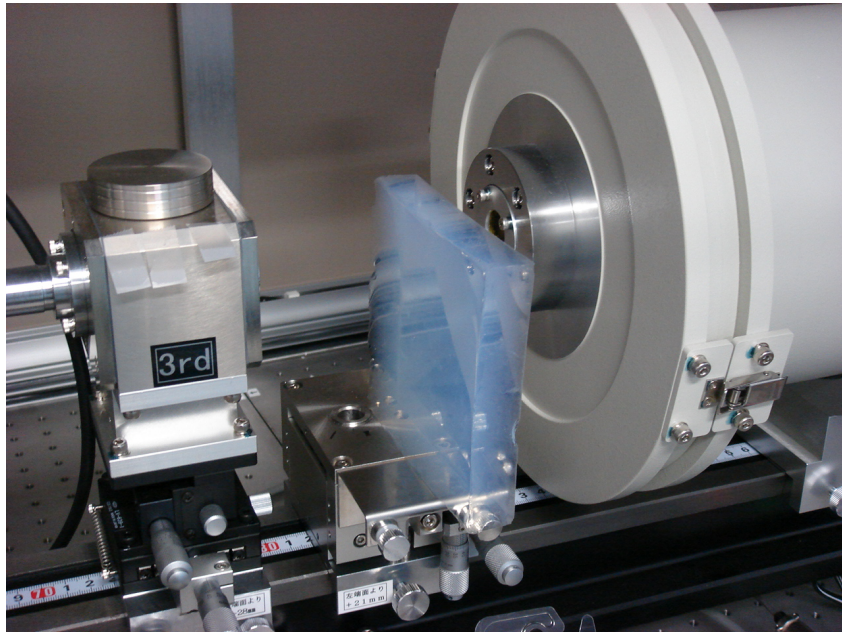
- If radiator medium needs to cover large area, basically big size tile reduces boundary region, where less Cherenkov photon yield is expected.
- Handling point of view, large tile needs more attention not only in production process but also in construction stage.
  - Crack-free yield/Tip-lost-at-corner yield/Related to thickness
- Depends on tile index.
- Optimizations in SCD step.
  - Pressure control

*M.Tabata et al., The Journal of Supercritical Fluids, Vol.110, April 2016, Pages 183-192*



# Density Uniformity

- Uniformity scan was done using X-ray tomography device.



X-ray  $\lambda=0.156\text{nm}$

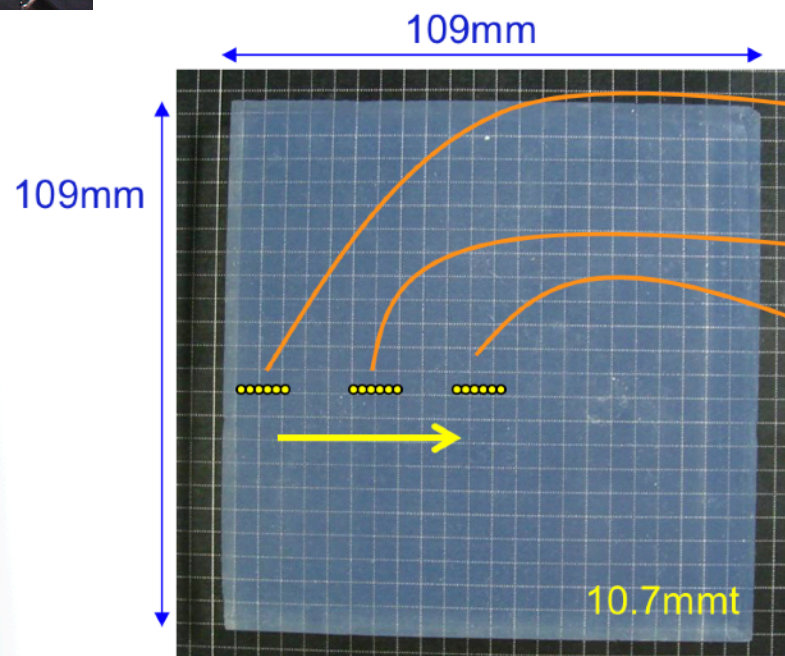
$\phi$  beam spot  $< 1\text{mm}$

density relative uniformity

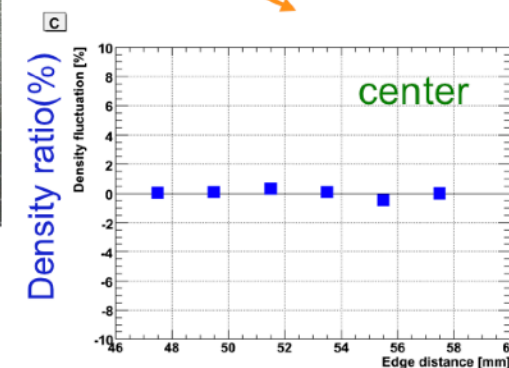
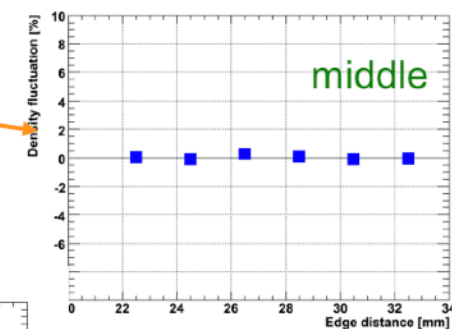
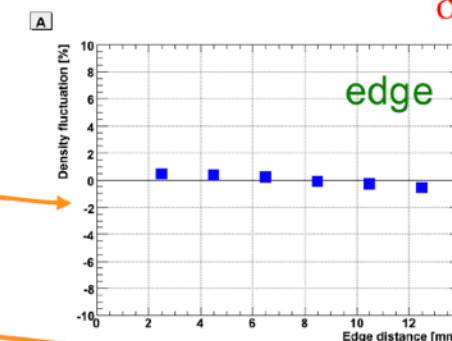
preliminary value:

$$\delta(n-1)/(n-1) \sim \pm 0.02$$

need further studies



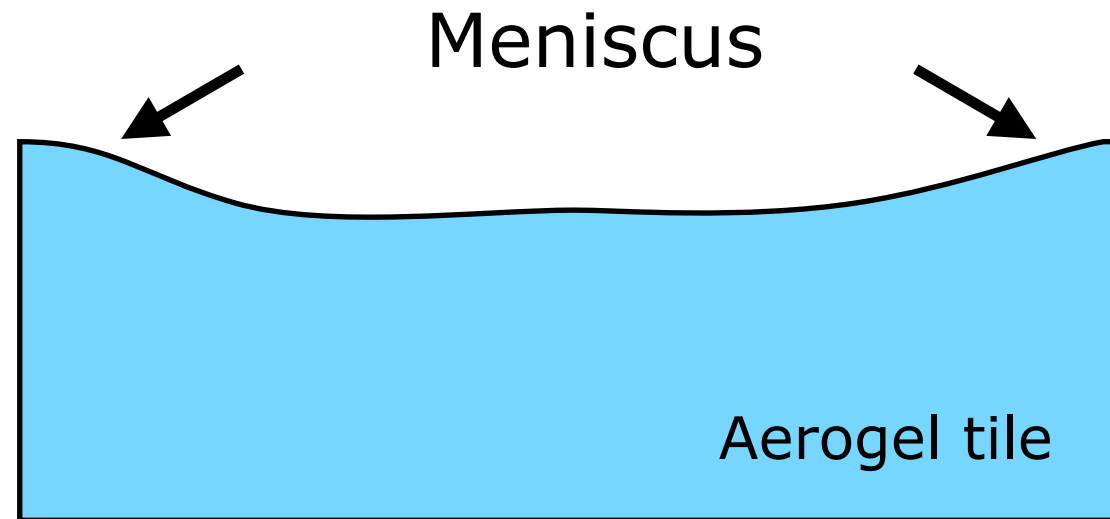
Index (Fraunhofer method at 405nm)  
= 1.0577  $\pm$  0.0006



Distance from edge(mm)

# Meniscus (1)

Aerogel tile has meniscus structure.  
Meniscus structure partially related to production process.

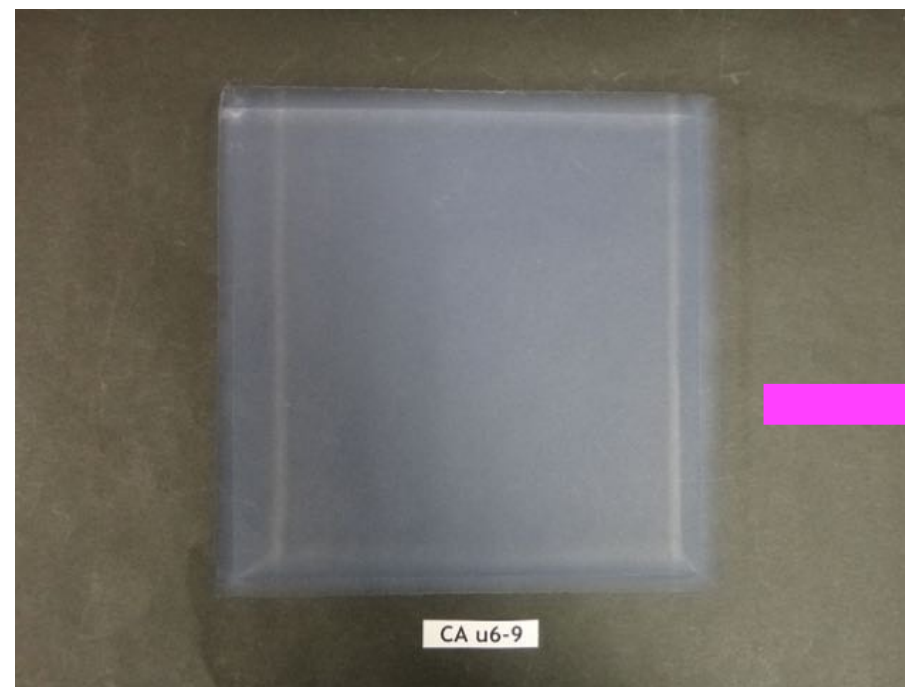


This could be a (*potential*) problem when Cherenkov radiator is organized by stacking # of aerogel tiles.

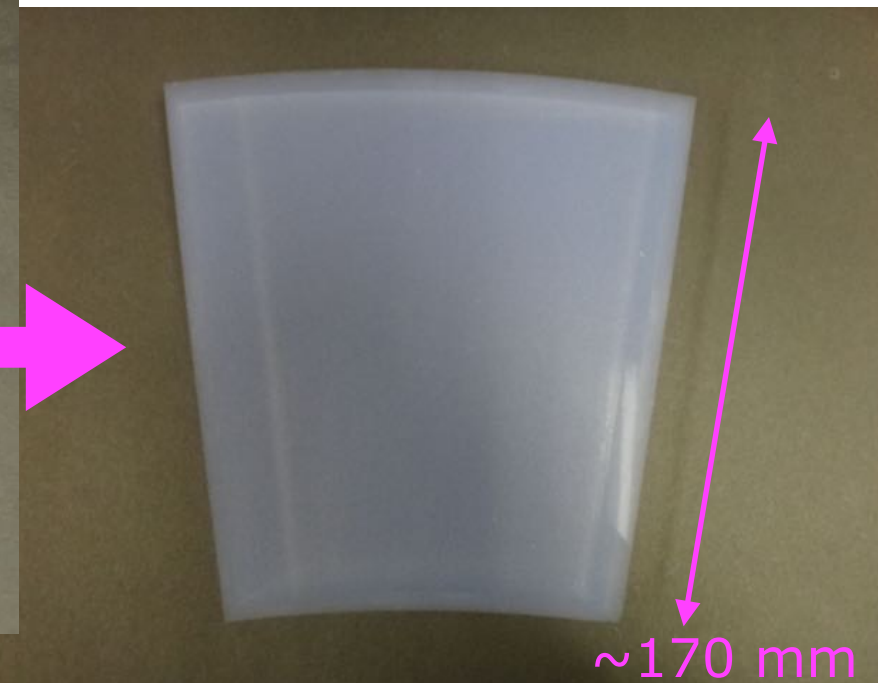


# Meniscus (2)

- The best way is to remove tile edge regions, where there is meniscus.
- Hydrophobic feature allows us to use a water-jet machine without deteriorating optical transparency.
- In case of hydrophilic tiles, diamond cutter can be used for machining (like CLAS12 aerogels from BINP).



Belle II case



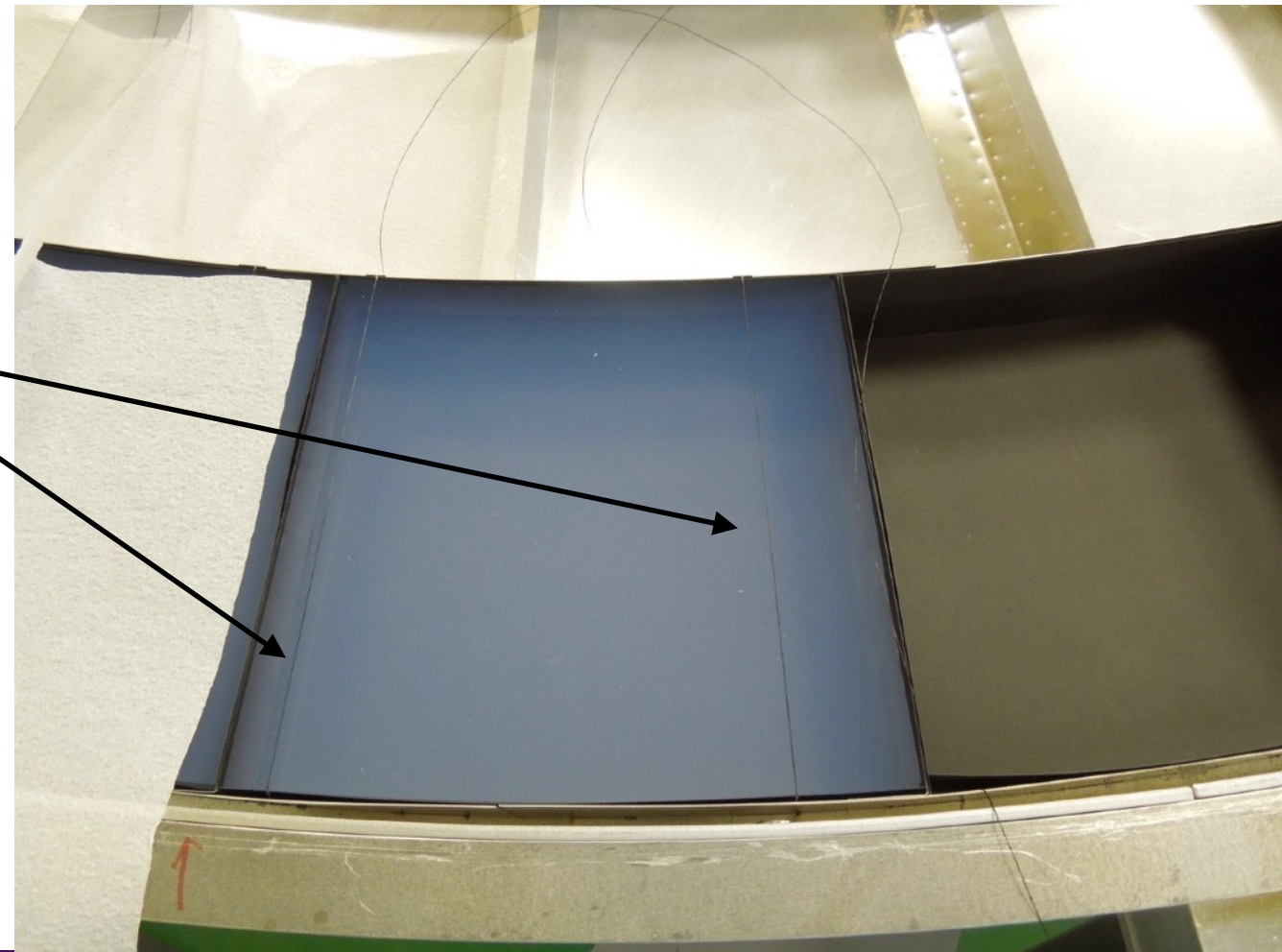


# Housing

- To construct aerogel radiator system, each tile has to be fixed into the detector container.
- Aerogel is fragile and the way to fix tiles is rather limited.
  - Metal screws can not be used.
  - Wrapping transparent film and glue
  - Directly glue
    - One can not replace tiles.
  - Strings

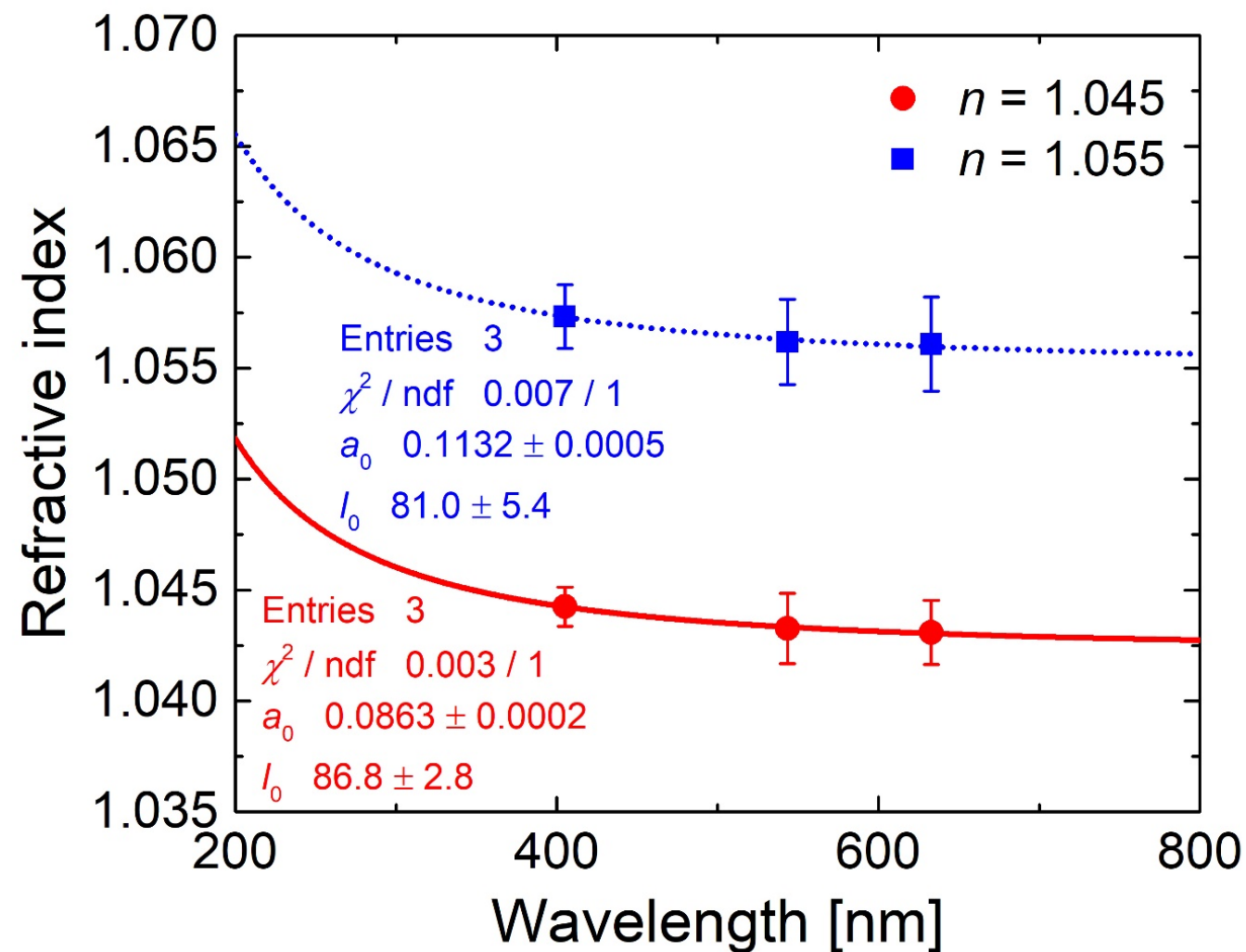
Belle II case  
Glass fiber

**It is not a problem, but  
(potential) worry...**



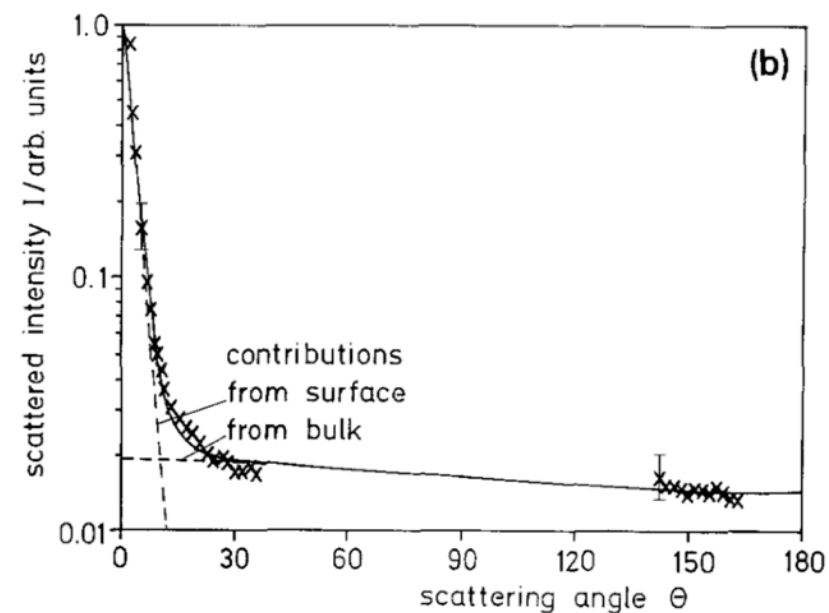
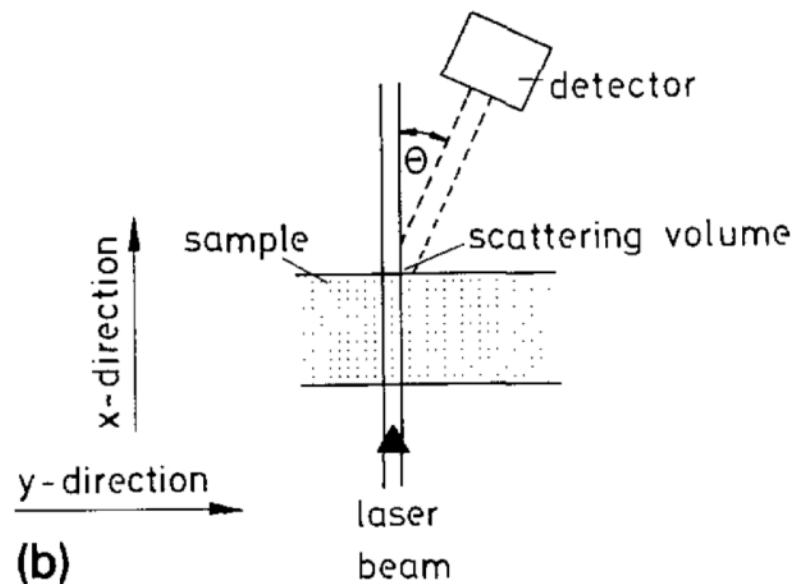
# Chromaticity Dispersion

- Refractive index measured with lasers having different wavelength.
- Fitted with Sellmeier equation :  $n^2 - 1 = a_0 \lambda^2 / (\lambda^2 - \lambda_0^2)$



# Forward Scattering

- Forward scattering mainly on aerogel surface reported by P.Wang et al. in J. Non-Crys. Solids 145(1992)141.
  - Sharp peak in forward, unlike Rayleigh scattering.

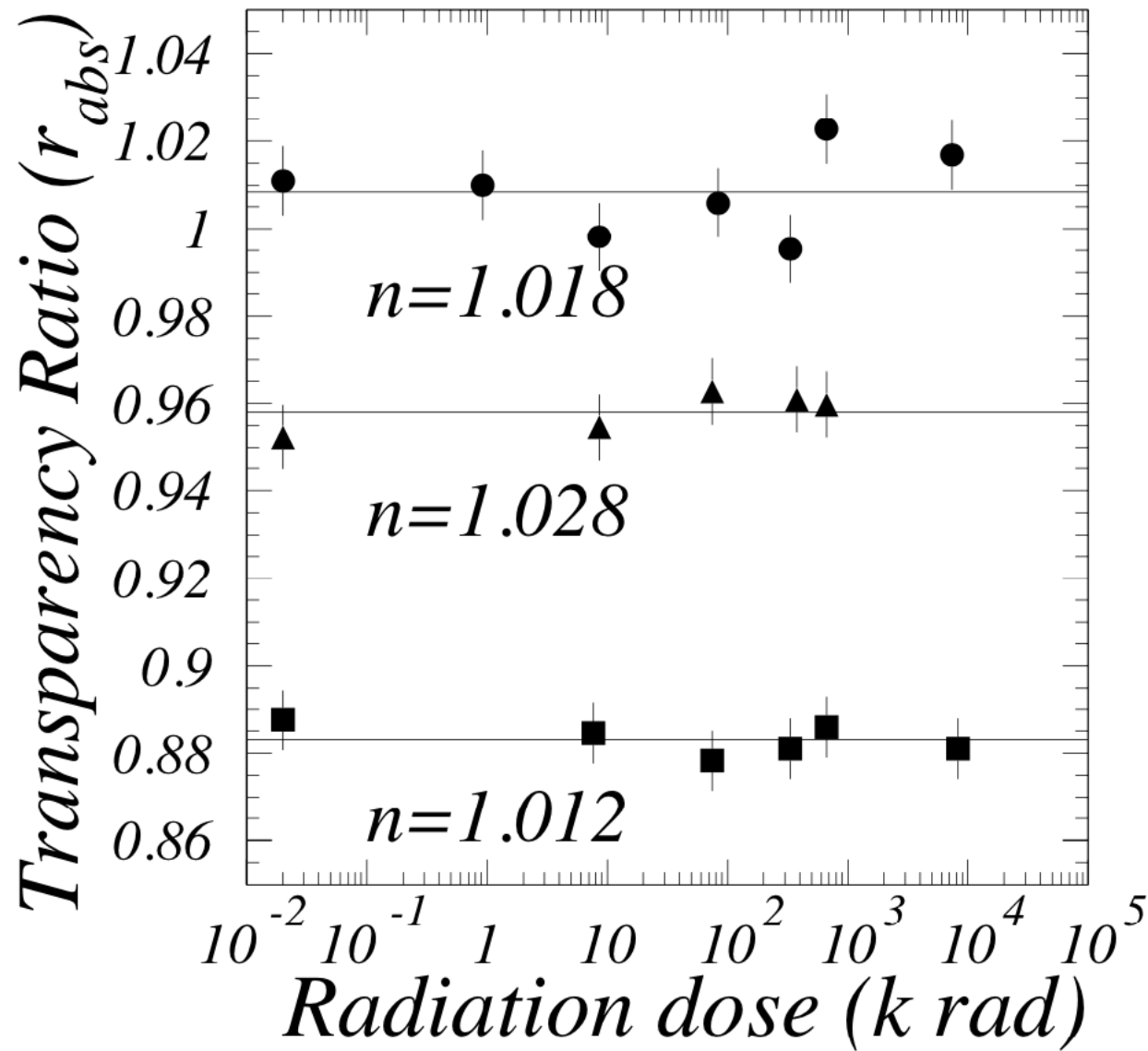


- Further systematic studies done by R. De Leo et al. NIMA457(2001)52. Matsushita aerogel used.
- This effect contributes as one of components in background.

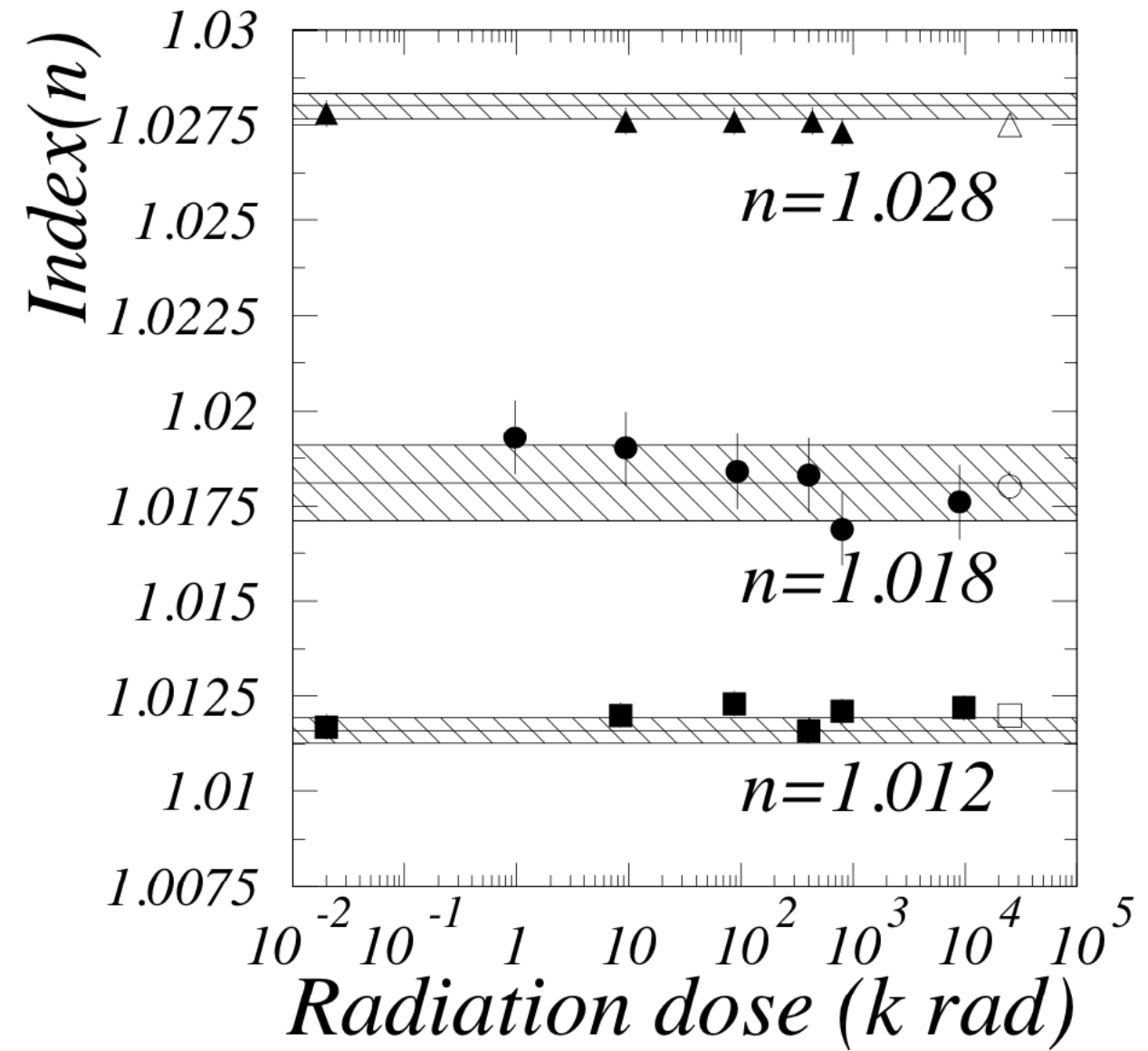
Need to measure this effect for recent samples

# Radiation Damage

Transmittance



Refractive index



**No deterioration up to 9.8 Mrad**



# Summary

- Aerogel is a unique material and has been developed as Cherenkov radiators since 1970's.
- Aerogels in the new generations from Russia and Japan are widely used as Cherenkov detectors in various experiments.
- Transparent tile allows us to take the new direction.
  - Focusing scheme of multiple aerogel layers
- For the 4th generation aerogels, experience at HELIX RICH gives us further information on applications.



**Silica aerogel is attractive  
material as Cherenkov radiator !**

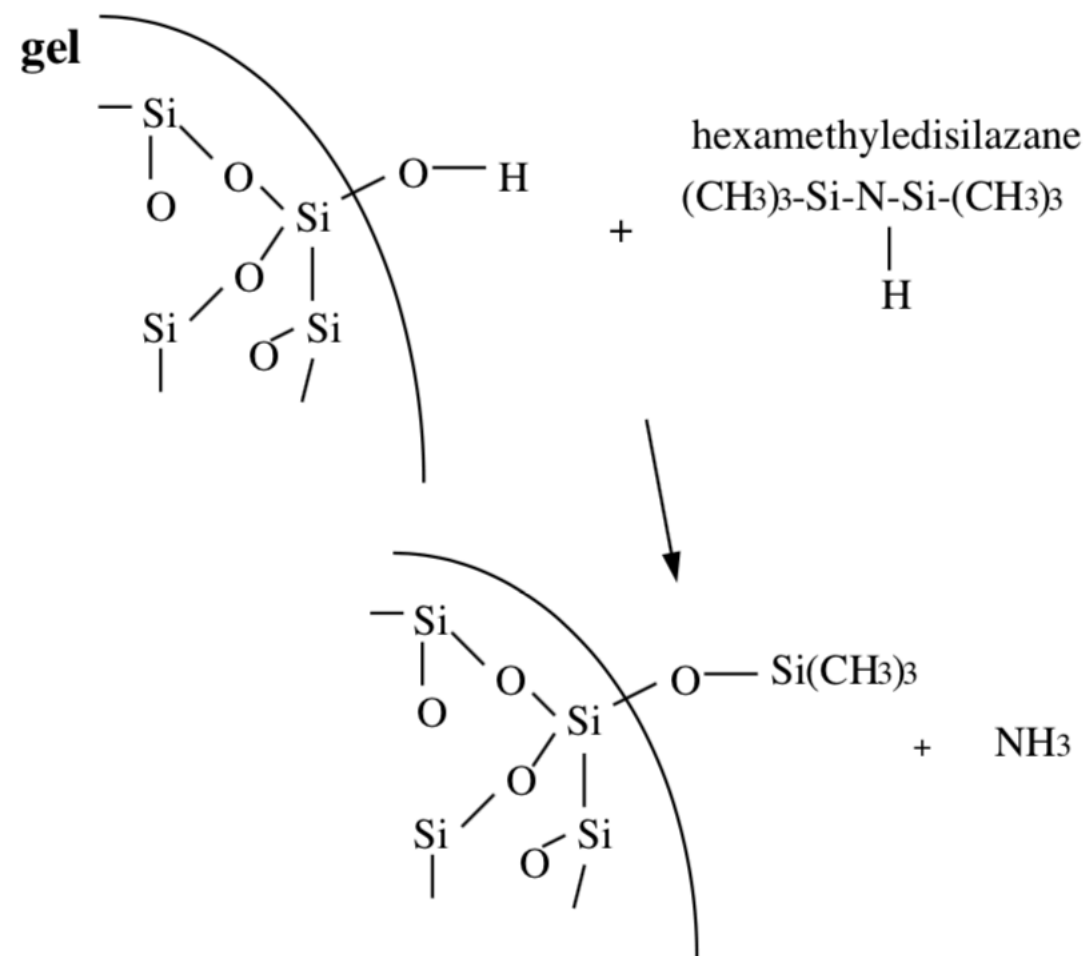
**Thank you very much !**





# Hydrophobic Treatment

- -OH group in alco-gel is replaced into -O-Si(CH<sub>3</sub>)<sub>3</sub>
- -OH group is likely to be charged and reacts with water.

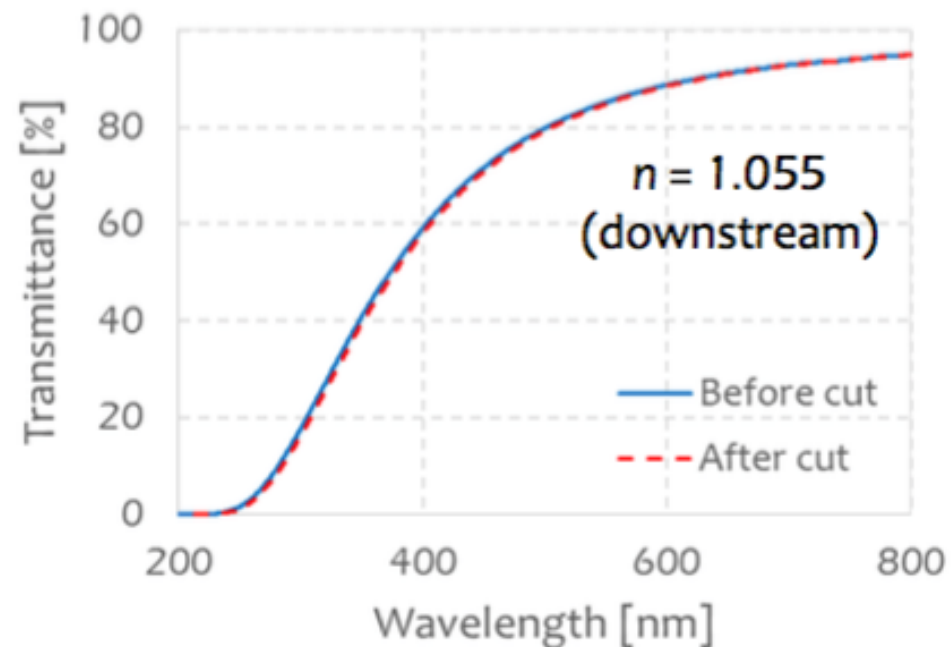
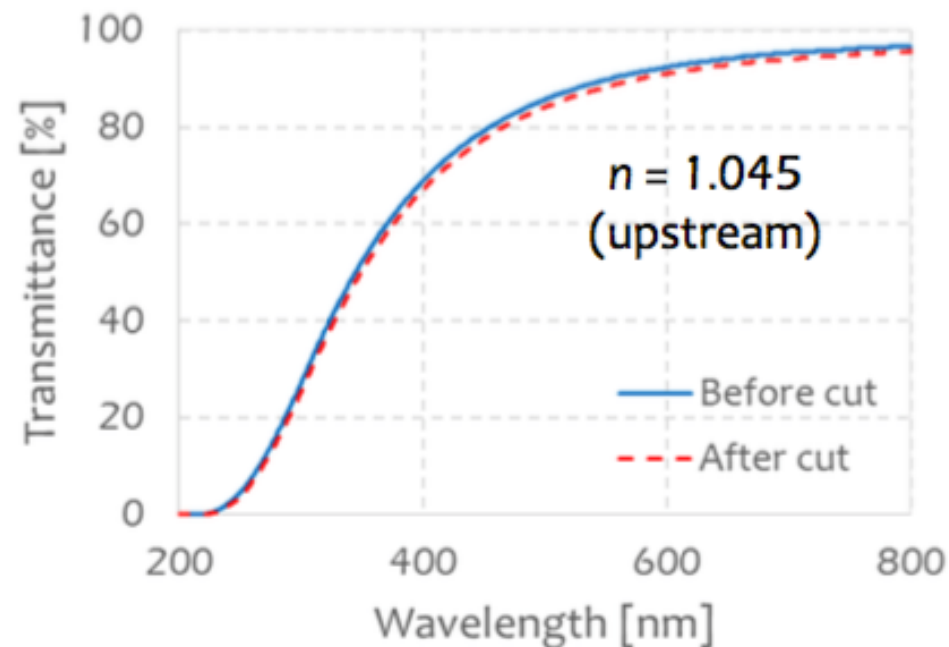




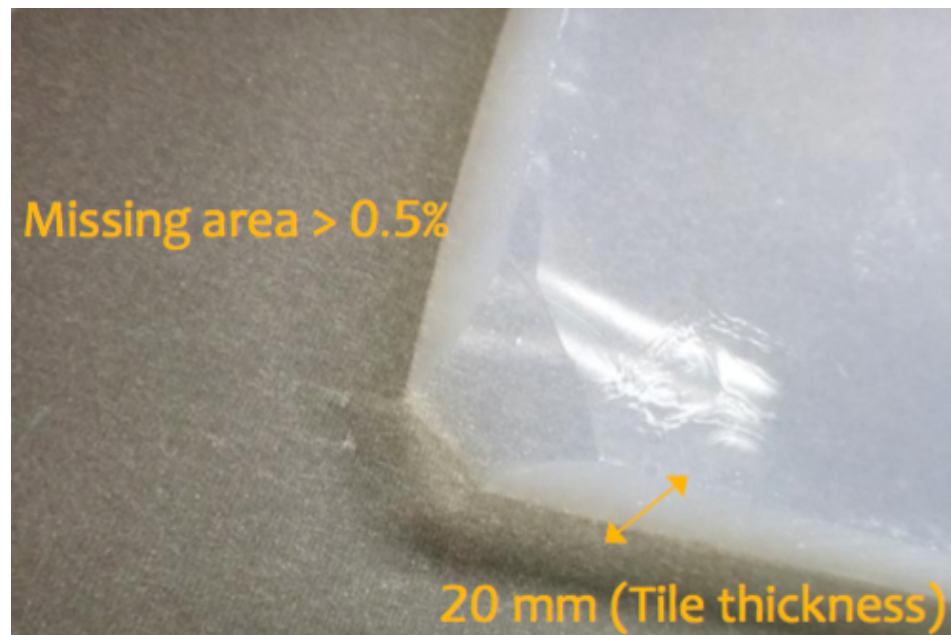
# Tile Machining

**No degradations in transparency due to this machining**

UV-Vis spectra for typical aerogel tiles



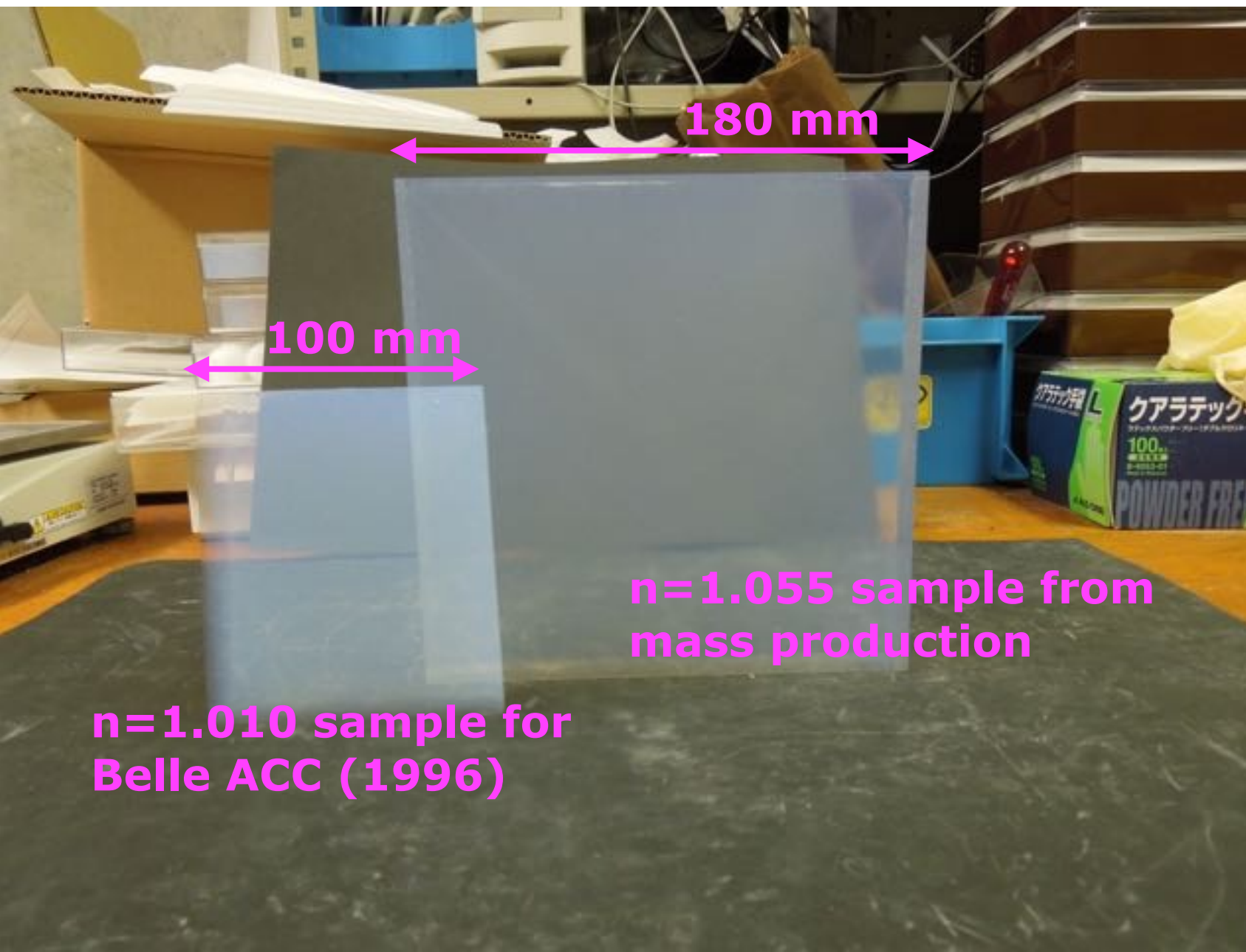
**Small chips found in some tiles**



If missing area > 0.4 % w.r.t. whole surface ( $\sim 1.0 \text{ cm}^2$ ), the sample is removed from candidates.

# New Aerogel Tile

- Large tile of  $180 \times 180 \times 20 \text{ mm}^3$  with no cracks
  - Optimization of pressure control in supercritical drying process.
  - 3 times longer duration from operating point to atmospheric pressure introduced by Chiba university group.



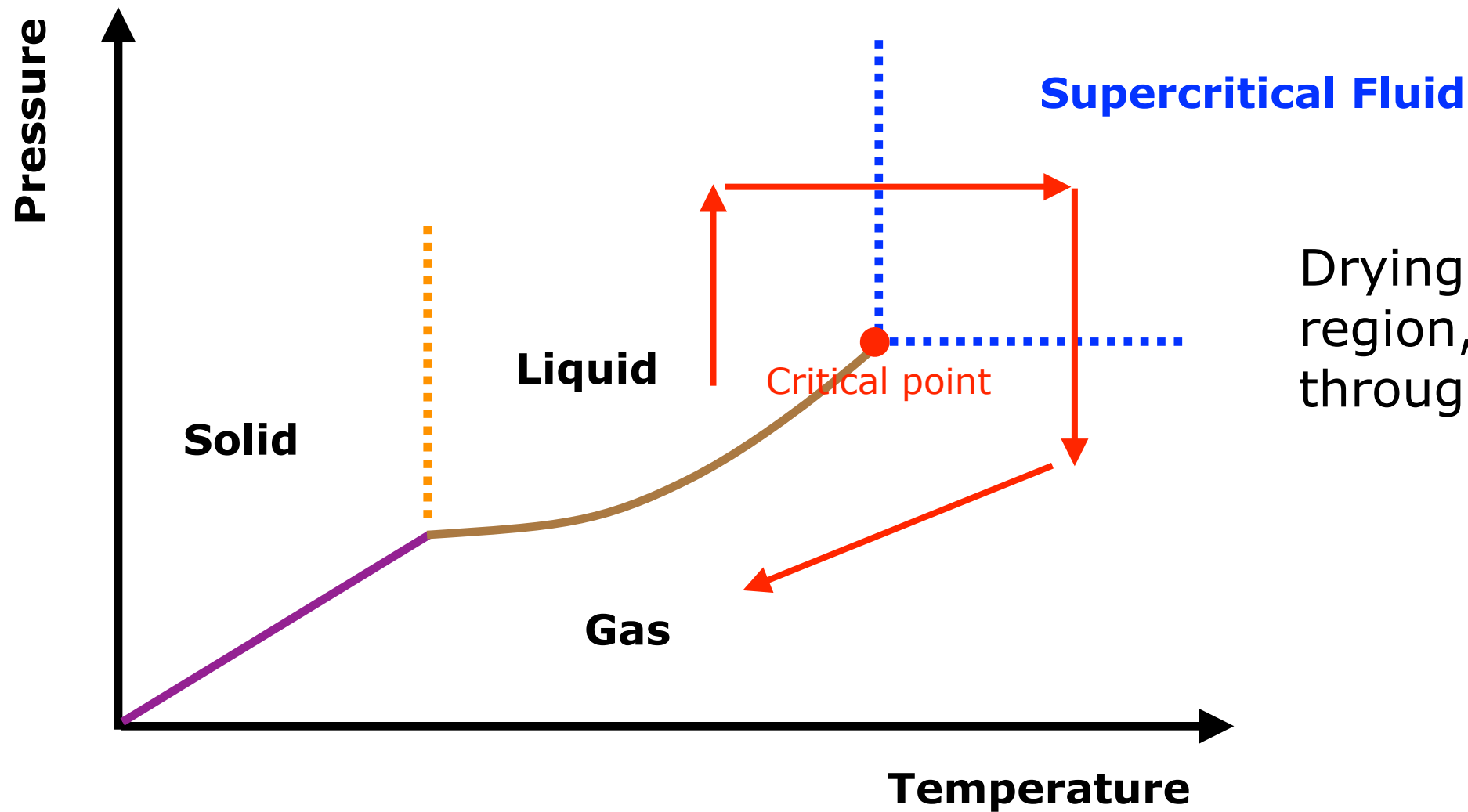
Crack-free yield  $\sim 87\%$

Drawback is to need more time for tile production.

*M. Tabata et al., The Journal of Supercritical Fluids, Vol. 110, April 2016, Pages 183-192*

# Supercritical Drying

Phase diagram



Drying through supercritical region, instead of passing through liquid-gas boundary.