"Liquid scintillator (neutrino)"

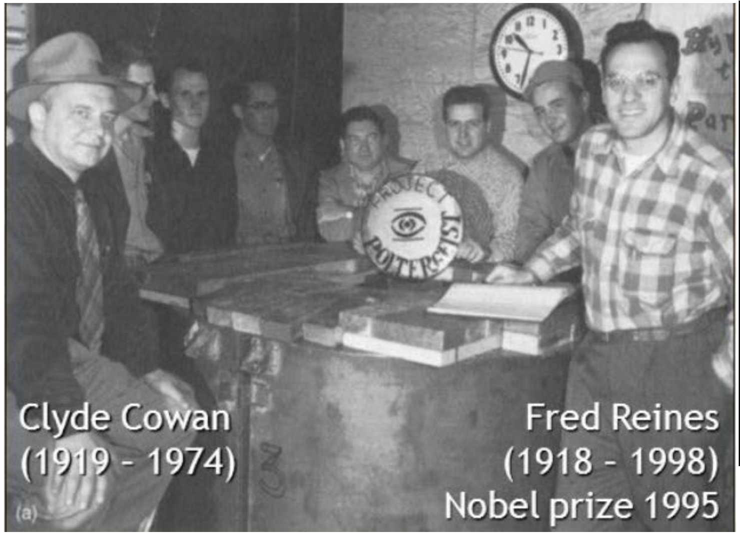
- Mainly focus on
  "Development of metal (Gd)-loaded liquid scintillator"

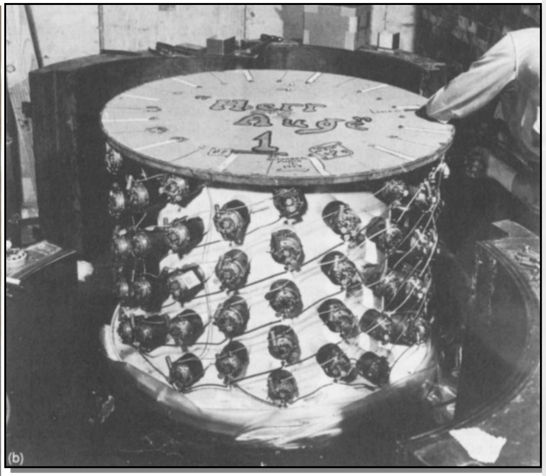
- Others not covered
  - ^{6}Li-loaded liquid scintillator
  - liquid scintillator using water/oil (WbLS, ObLS)

K.K. Joo
Chonnam National University
January 29, 2018

AFAD2018 @ DCC, Deajeon

First observation of neutrinos (1953~1956)





Detector prototype

Frederick Reines (left) and Clyde L. Cowan, Jr. with the control equipment used in their first tentative observations of the neutrino at Hanford, Washington, in 1953. Their definitive detection of the (anti) neutrino was performed at Savannah River, Georgia, three years later. (Courtesy General Electric Co.)

- Reines & Cowan performed experiment @ Hanford nuclear reactor in 1953, but later in 1955 moved to the Savannah River Plant in South Carolina
- Reines won NP (1995), but Cowan had died
- In order to detect neutrinos,
  liquid scintillator (LS) technology was used

Historically, LS plays an important role in neutrinos (especially, in reactor neutrino)

On June 14, 1956, Reines and Cowan sent a telegram to Pauli:

"We are happy to inform you that we have definitely detected neutrinos from fission fragments by observing inverse beta decay of protons…"

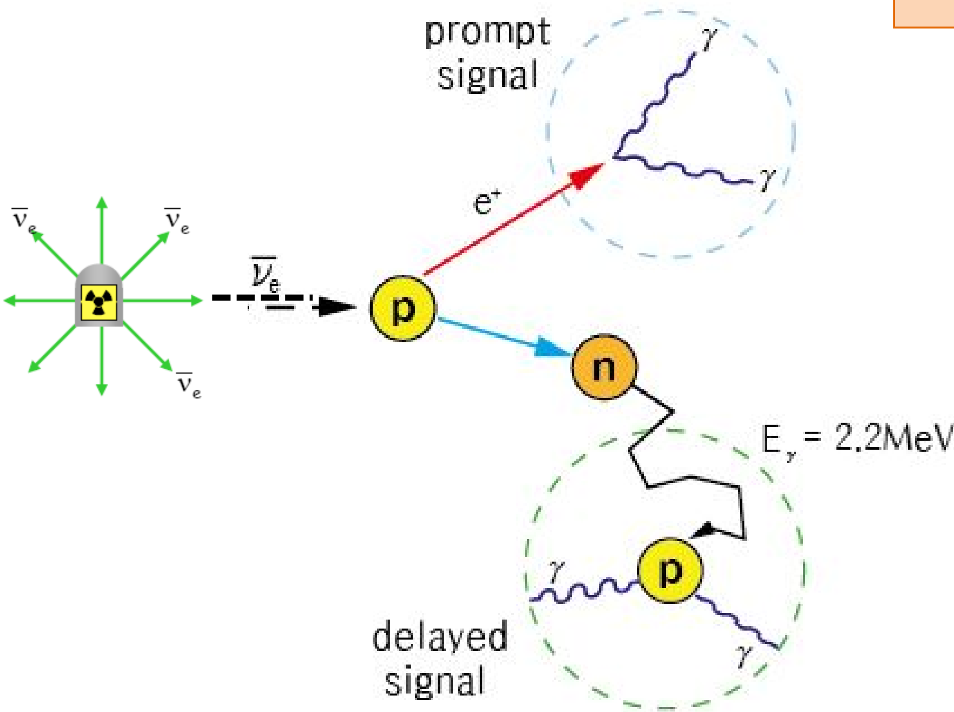
Principle of Reactor Neutrino Detection

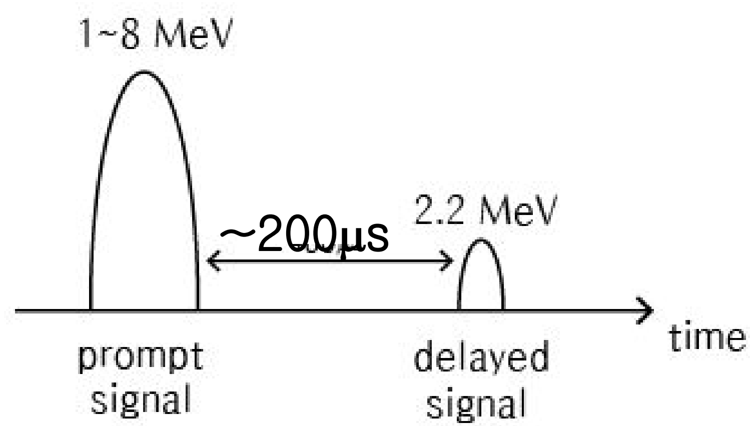
Hydrogen capture

Use Inverse Beta Decay (IBD)

$$\overline{\nu}_e + p \longrightarrow e^+ + n$$

$$n_{thermal} + p \longrightarrow d + \gamma \ (2.2\text{MeV})$$





- Prompt part: subsequent annihilation of the positron to two 0.511MeV gammas
- Delayed part: neutron is captured ~200μs w/o Gd
- Signal from neutron capture ~2.2MeV w/o Gd
- "Delayed coincidence" reduces backgrounds drastically

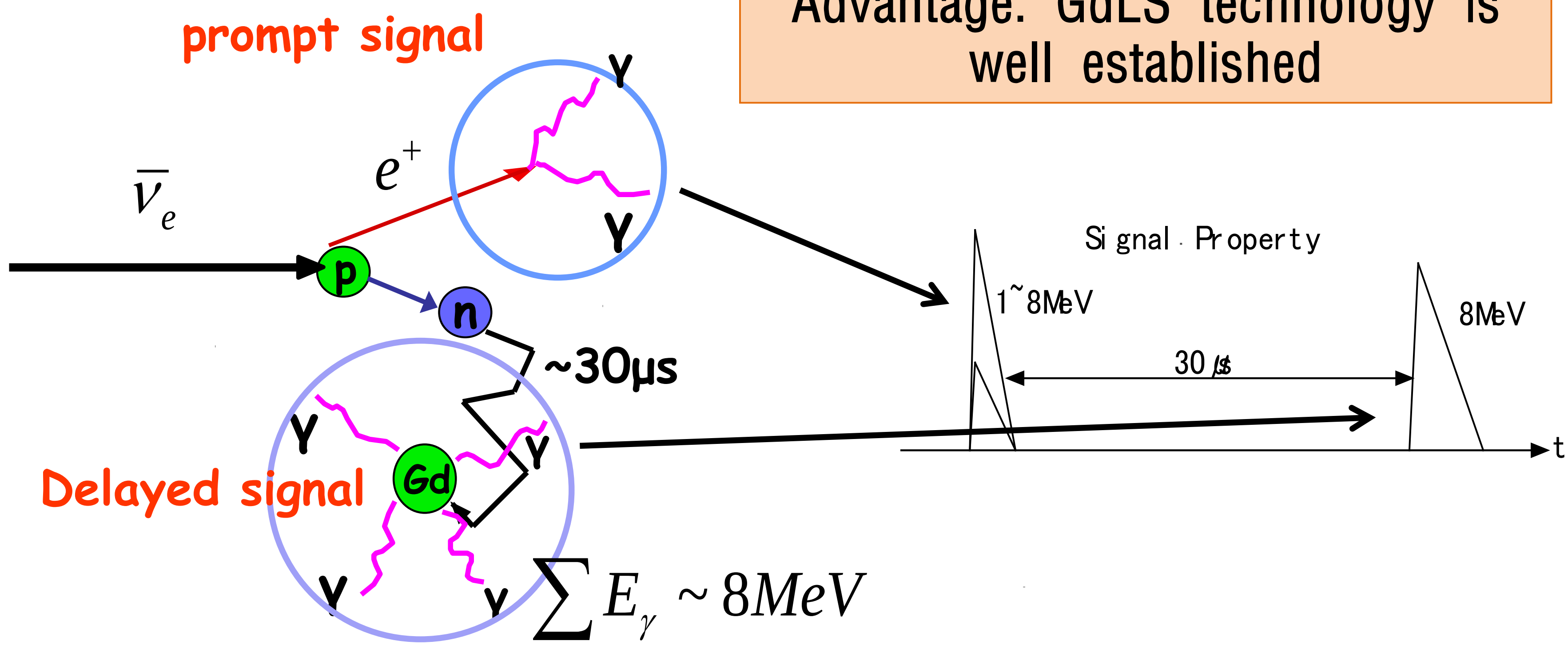
Gd capture

+ 0.1% Gd loading

$$n_{thermal} + {}^{155,157}Gd \rightarrow {}^{156,158}Gd + \gamma s(\sim 8MeV)$$

Signal type γ (1 MeV) – γ (8 MeV) type

Advantage: GdLS technology is well established



Liquid Scintillator at a Glimpse

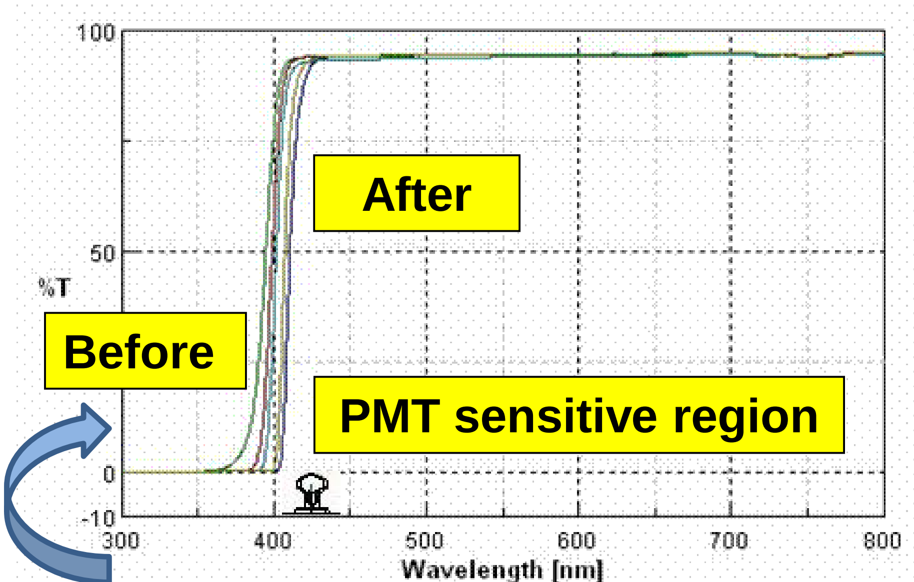
General elements of Liquid Scintillator

Base solvent	For mixing	Fluor	Wave-length shifter
PC, PXE, LAB	MO, Dodecane	PPO, BPO	bis-MSB, POPOP

Required Properties

- Transparency
- High light yield
- Long term stability
- High radiopurity
- Safety matter
- Reasonable price
- Massive quantity available

Base solvent
+ primary scintillating fluor
+ secondary wavelength shifter

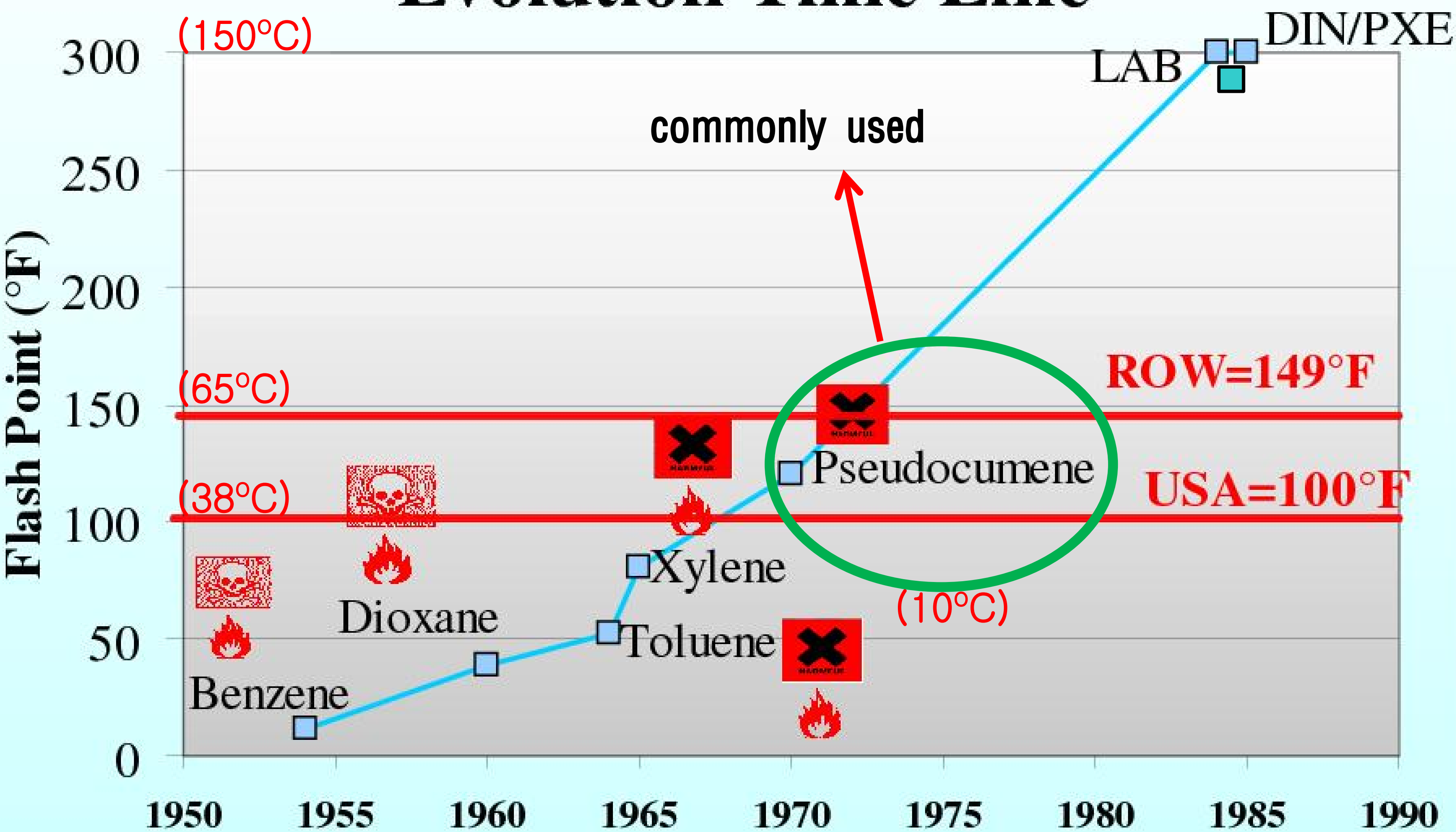


Effect of wavelength shifter

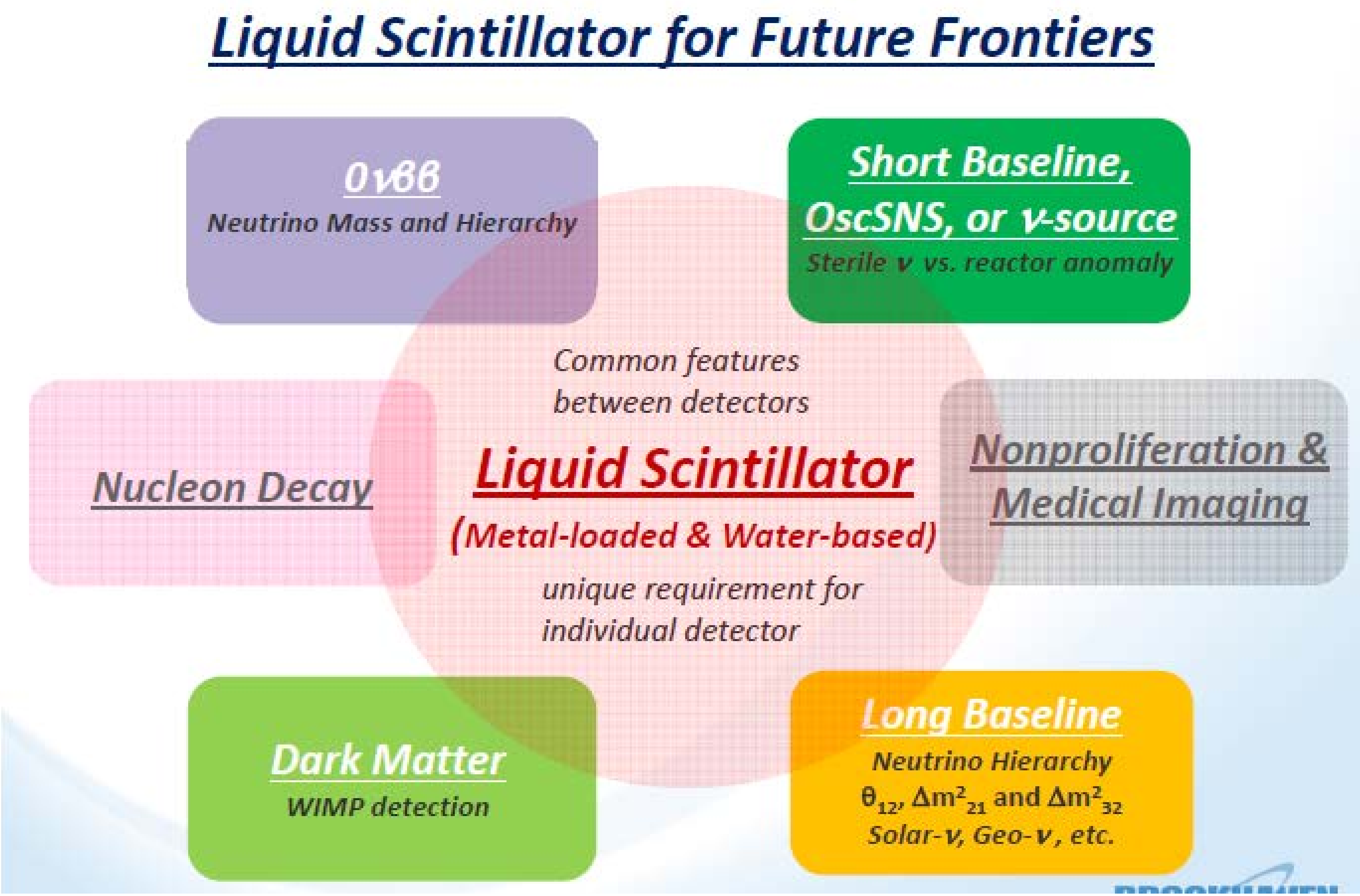
PC is widely used for base solvent for its excellent transparency and fluor dissolution.

LAB (Linear alkyl benzene)
DIN (Di-isopropylnaphthalene)
PXE (Phenylxylethane)
PC (Pseudocumene)

LSC Solvents Evolution Time Line



Minfang Yeh


Liquid Scintillator for Future Frontiers
0νββ
Neutrino Mass and Hierarchy
Short Baseline, OscSNS, or ν-source
Sterile ν vs. reactor anomaly
Common features between detectors
Nucleon Decay
Liquid Scintillator
(Metal-loaded & Water-based)
Nonproliferation & Medical Imaging
unique requirement for individual detector
Dark Matter
WIMP detection
Long Baseline
Neutrino Hierarchy
θ_{12}, Δm^2_{21} and Δm^2_{32}
Solar-ν, Geo-ν, etc.

Various option is possible

Packard	**Ultima Gold** (DIN), **Opti-Fluor** (LAB)
Wallac	**OptiPhase Hi-Safe** (DIN)
ICN	**EcoLume & EcoLite** (LAB+PXE)
ND	**EcoScint** (PXE)
Beckman	**ReadySafe** (PXE)
RPI	**Bio-Safe** (LAB)
Zinsser	**AquaSafe & QuickSafe** (DIN)
Lumac	**LumaSafe** (PXE)

- Commercial LS + Gd/Li loading
- Commercial GdLS
- Synthesis (by yourself)

Past Known LS Cocktails

Institution	LS	Extractant	Fluors
Palo Verde[1]	40% PC + 60% Mineral oil	carboxylate	4 g/L PPO + 100 mg/L bis-MSB
CHOOZ[1]	50% Norpar-15 (paraffinic liquid) + 50% IPB (isopropylbiphenyl)	hexanol	1 g/L p-TP + bis-MSB
Eljen[1]	Anthracene + PC	Unknown	3 g/L PPO + 0.3 g/L POPOP
Bicron[1]	PC or Mix of PC+MO	EHA	unknown
Borexino[2]	PC; PXE	No need	1.5 g/L PPO or p-TP + bis-MSB
KamLAND[2]	20% PC + 80% dodecane	No need	1.52 g/L PPO

[1]Gd-loaded; [2]unloaded

Future of Reactor Experiments

~2020

JUNO

China

?

2011/2012 - The year of θ_{13}

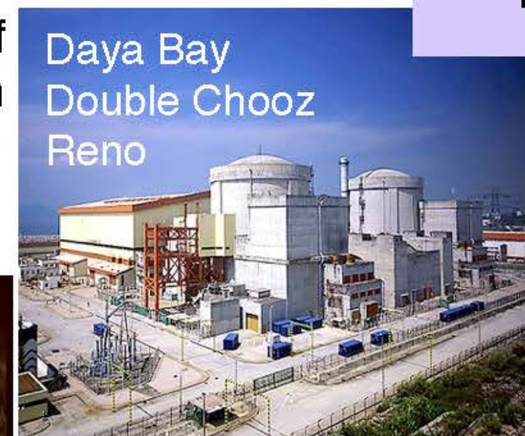
2008 - Precision measurement of Δm_{12}^2. Evidence for oscillation

2003 - First observation of reactor antineutrino disappearance

1995 - Nobel Prize to Fred Reines

1980s & 1990s - Reactor neutrino flux measurements in U.S. and Europe

1956 - First observation of (anti)neutrinos



China
Korea

Japan

Liquid Scintillator

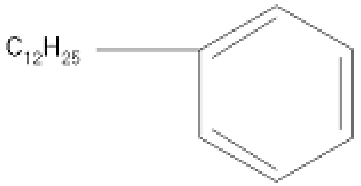
Karsten M. Heeger
University of Wisconsin

LAB-based Gd-loaded Liquid Scintillator

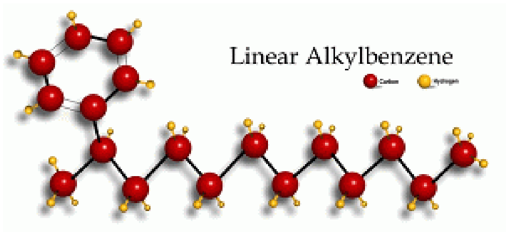
- High Light Yield: comparable to PC
- Good transparency (better than PC)
- High Flashpoint : 147ºC (PC : 48ºC)
- Environmentally friendly & less safety issues (PC : toxic)

LAB (Linear Alkyl Benzene)

C_nH_{2n+1}-C_6H_5 (n=10~14)



Linear Alkyl Benzene (LAB)



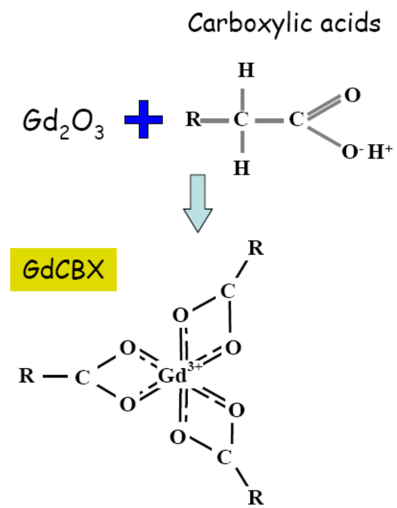
LAB-based LS experiments:

- Daya Bay, RENO, SNO+ , NEOS

Recipe of Gd-loaded for LS @ DB/RENO/NEOS

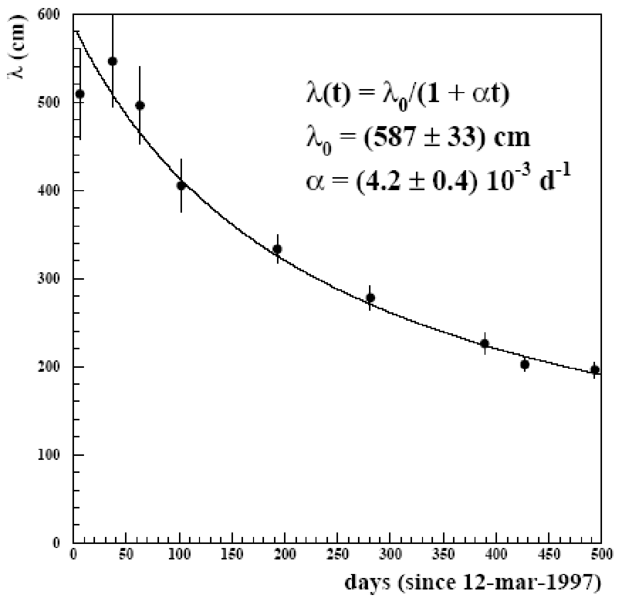
Aromatic Solvent & Flour	WLS	Gd-compound
LAB	PPO + Bis-MSB	0.1~0.5% Gd+TMHA (trimethylhexanoic acid)

❑ 0.1~0.5% Gd compounds with CBX (Carboxylic acids; R-COOH)



Gd-doped Liquid Scintillator Chemistry

Problems reported



From CHOOZ

- 5ton 0.09% Gd-loaded LS
- Color: Not stable (turned yellow after few months of deployment)
- Light output: Degradation of its optical properties.
  very rapid decay of attenuation length was observed
  ~0.4% degradation/day

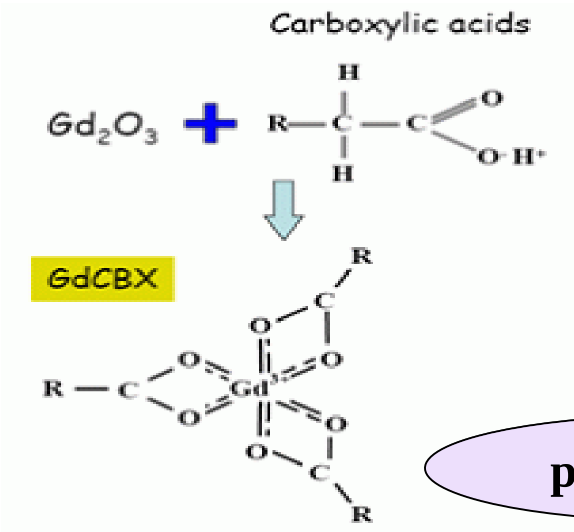
From Palo Verde

- 12ton 0.1% Gd-loaded LS
- Slight deterioration of light output with time
- ~0.03% degradation/day

Lesson: long-term stability is important

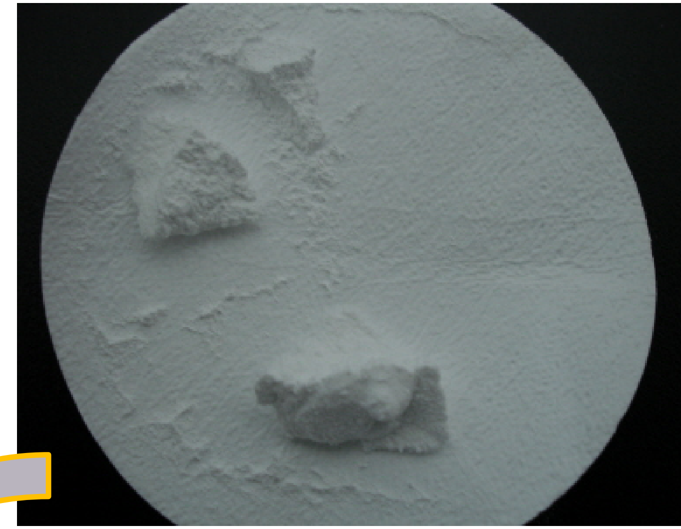
Loading metal into organic solvent is not trivial

Gd needs to transform to Gd-CBX for loading in organic phase



Gd -CBX

precipitation



After filteration

Stored in bottle

Reaction equation

$RCOOH + NH_3 \bullet H_2O \rightarrow RCOONH_4 + H_2O$

$3RCOONH_4(aq) + GdCl_3(aq) \rightarrow Gd(RCOO)_3 + 3NH_4Cl$

Precipitated Gd-CBX powder was filterated and rinse with 18MΩ water several times and dried in desiccator.

Gd loading into LAB

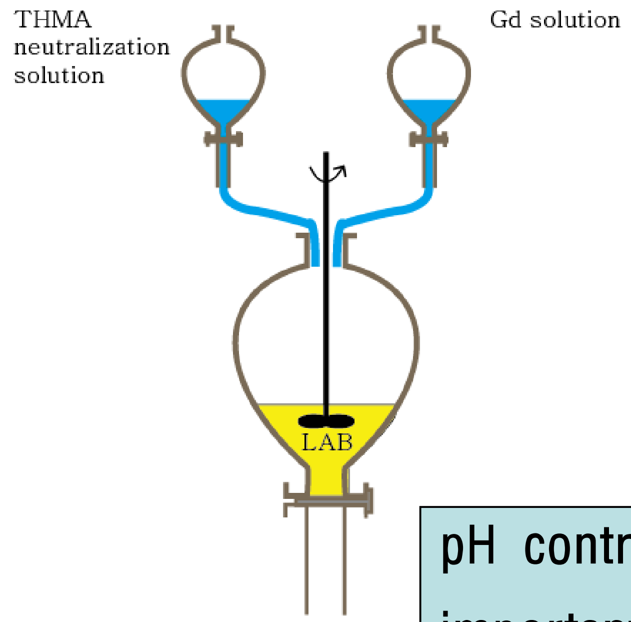
Solvent-solvent extraction

Gd is directly dissolved in LAB

Use neutralization chemical process

$$RCOOH + NH_3 \cdot H_2O \rightarrow RCOONH_4 + H_2O$$

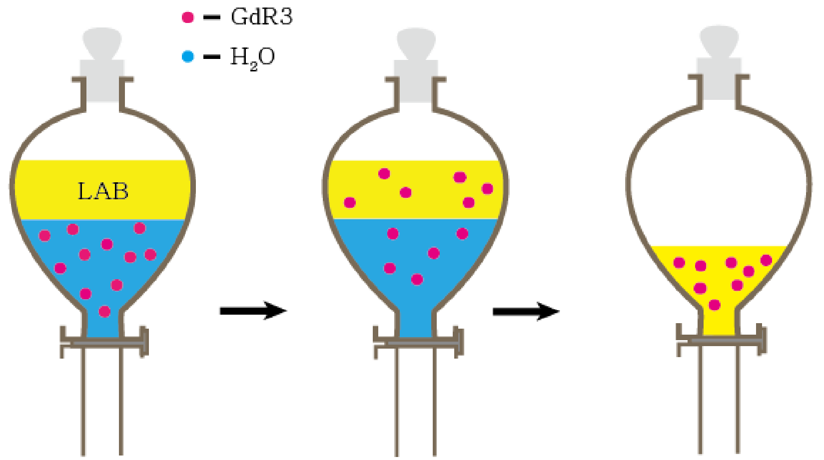
$$3RCOONH_4(aq) + GdCl_3(aq) \rightarrow Gd(RCOO)_3 + 3NH_4Cl$$



pH control is most important factor

Bad:
- Transportation difficult.
- Big synthesis tank.
- Need to remove water.

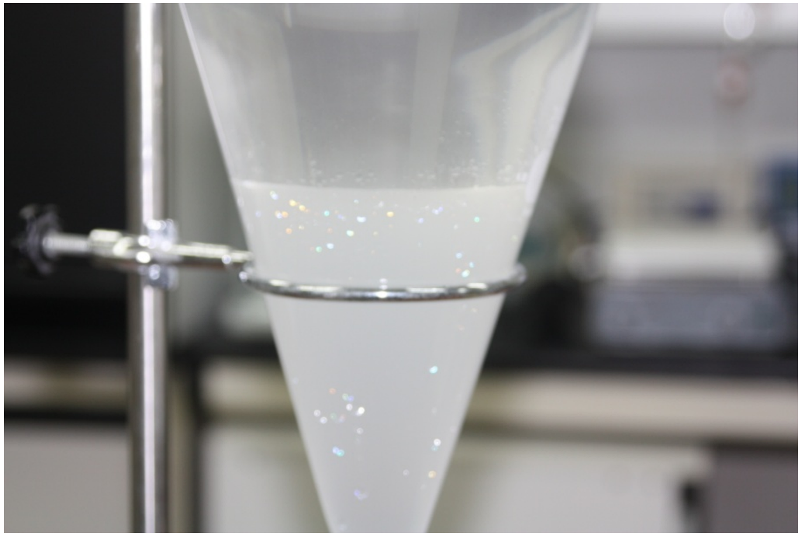
Good:
- Unlike powder, do not need to dissolve Gd into solvent.
- Time saving.

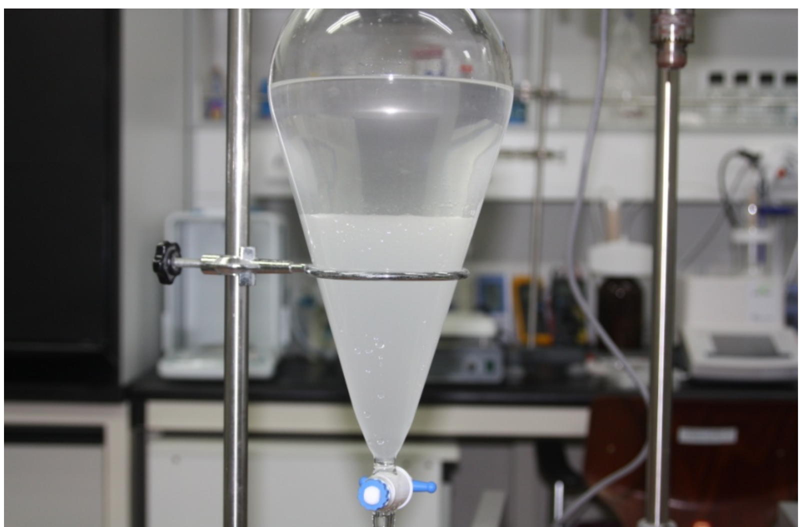


Small scale synthesis

Initially

N_2 flush





Time elapsed

Two layers are separated & bottom layer removed

N_2 flush again

↓

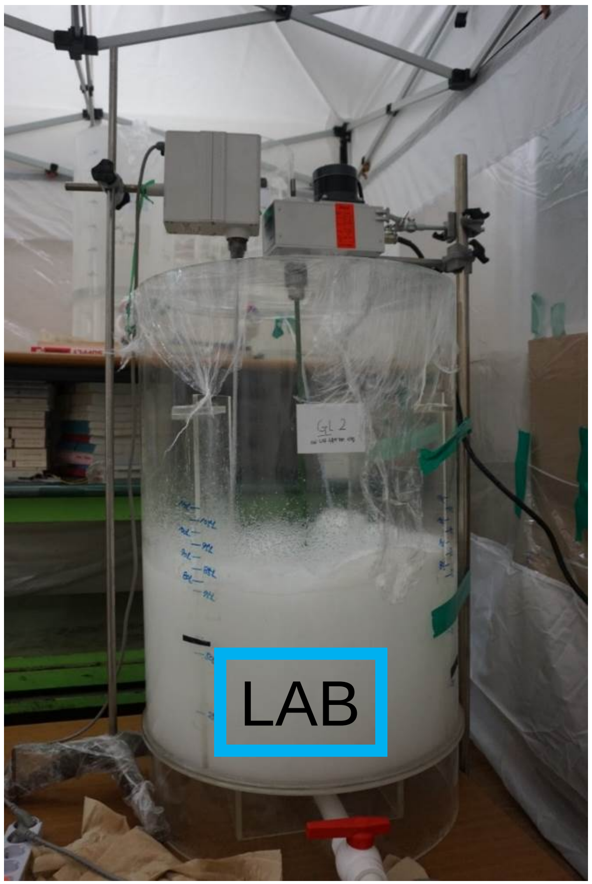
Transparent by eye & measurements

Final GdLS

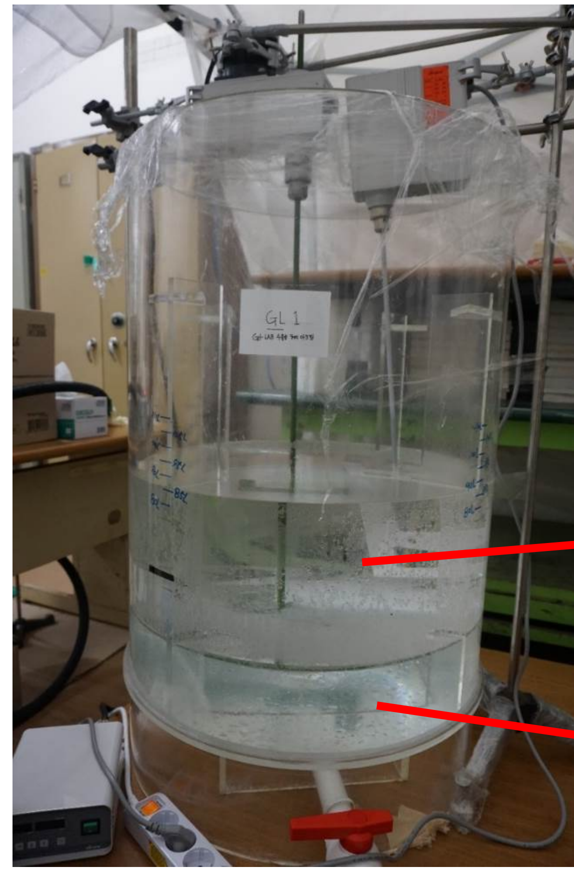
Half-ton scale mass production

Gd-sol

TMHA-sol



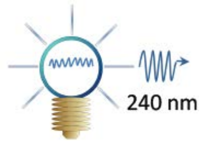
~12 hrs
layer
separation

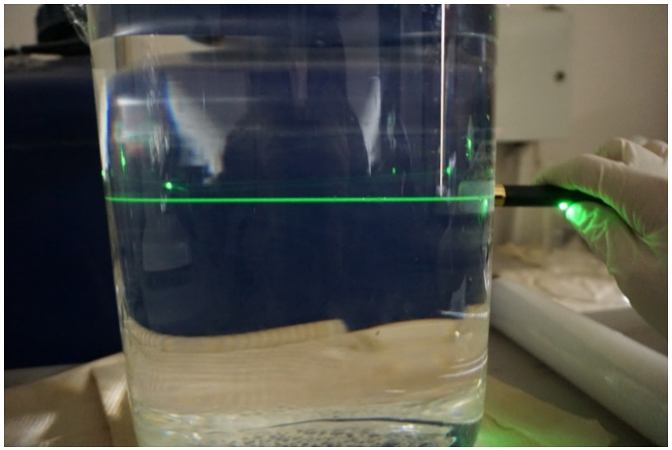


Gd-LAB

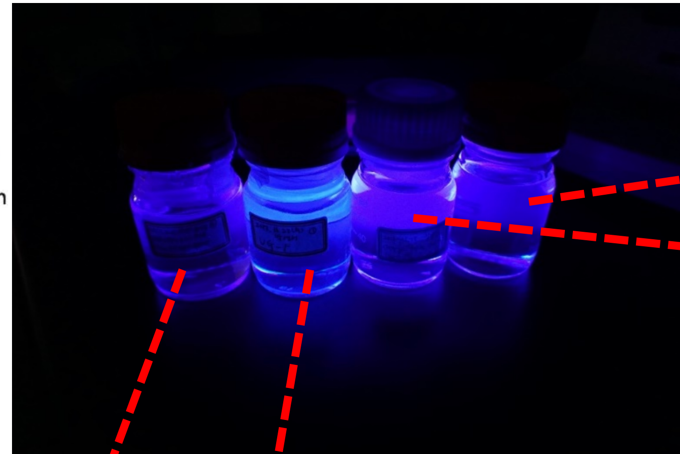
Water

Eye inspection
&
Laser penetration





UV irradiation



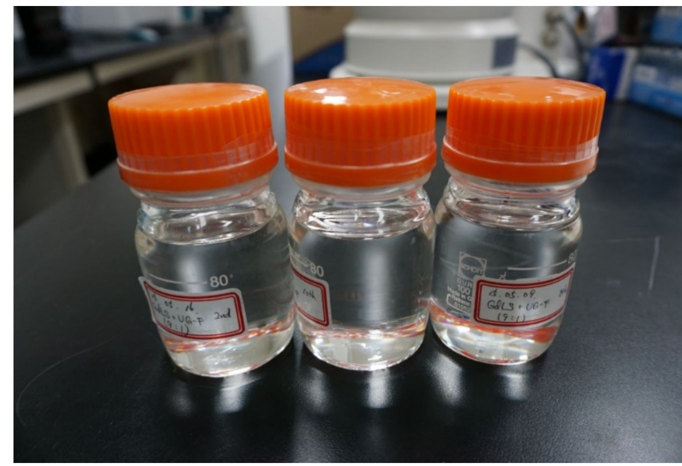
NEOS

GdLS

LS

UG-F itself

NEOS samples
(2015.05 synthesized)



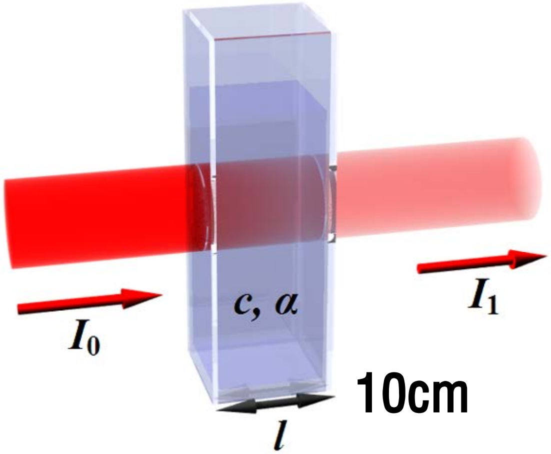
All transparent. No sign of aging

[Note] LS = LAB+3g/L PPO+30mg/L bis-MSB

[**Caution & important**]
GdLS should be kept in hermetic condition.
In particular, DIN(UG-F) is very vulnerable to any air contact

➡ We have experiences up to 3% high concentration GdLS
More than 3%, it becomes like jell

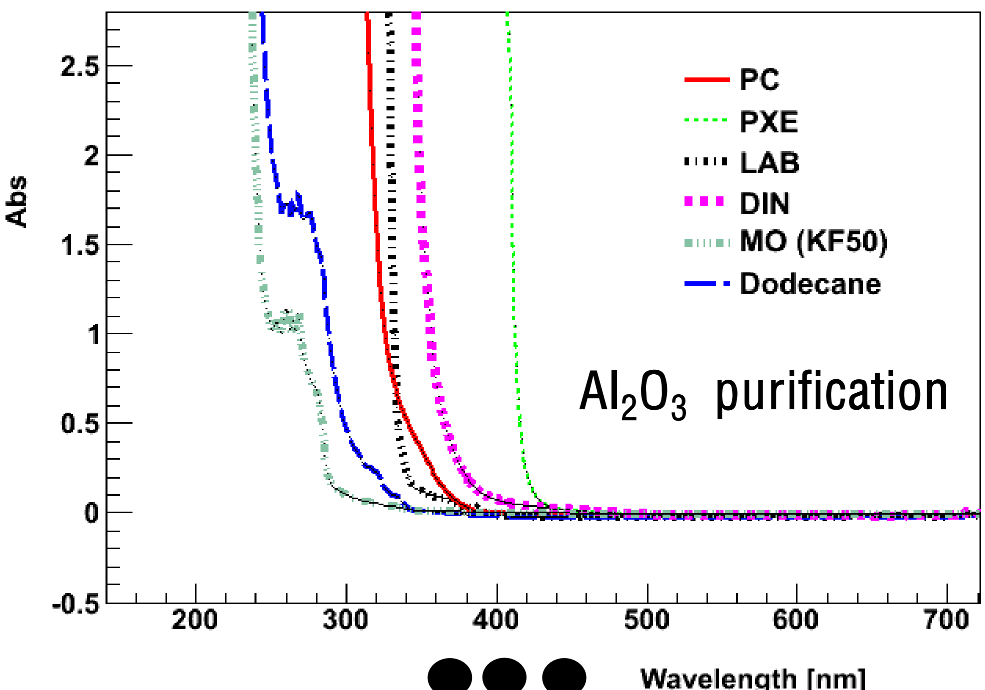
Absorption [Abs]



Absorption of a beam of light as it travels through a cuvette of size ℓ

- Beer-(Lamvert-Bouguer) law
- $\lambda = 0.4343\ (L/A_{420})$

  where L is pathlength in the cell
  A is the absorption at 420~430nm



Solvents	Attenuation length (m)±0.3
PXE	1.3
Dodecane	16.2
PC	8.2
MO	N/A
LAB	15.2

(one thing)

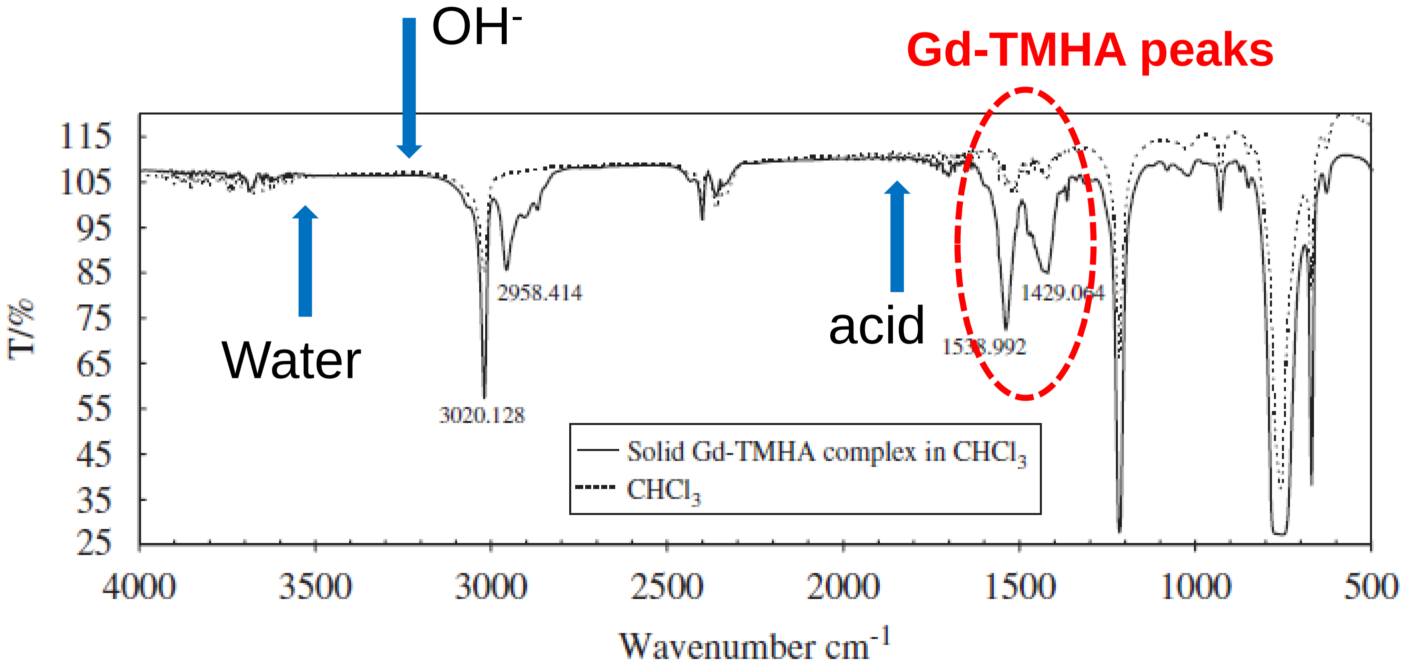
→ Path-length is too short

Spectrum Gd-TMHA by FT-IR

Fourier Transform Infrared Spectroscopy

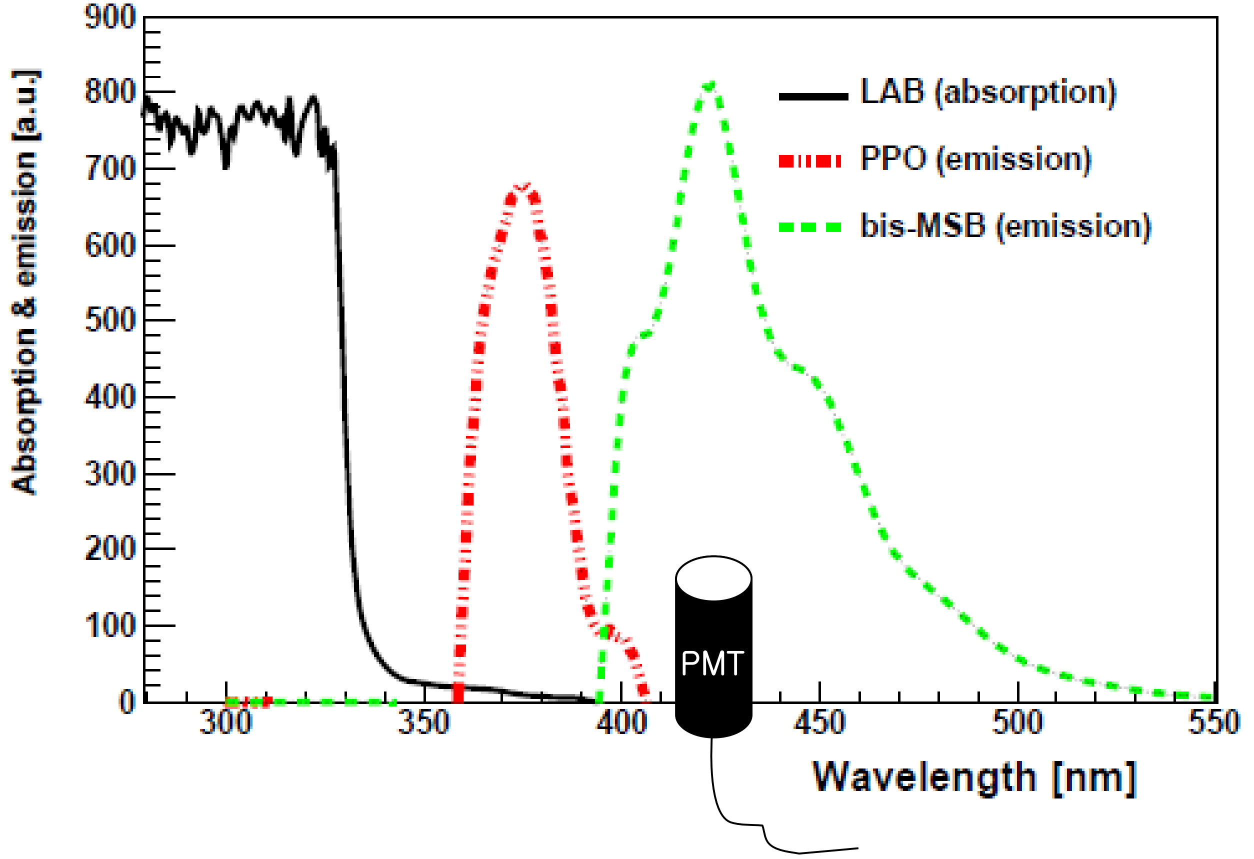
Requirements for Gd-TMHA

- No OH^- group peak (3200~3500 cm^{-1})
- No free acid peak (~1700 cm^{-1})
- No water-content peak (~3500 cm^{-1})
- Gd-TMHA carboxylate peaks (~1420 and 1580 cm^{-1})



Emission Spectrum

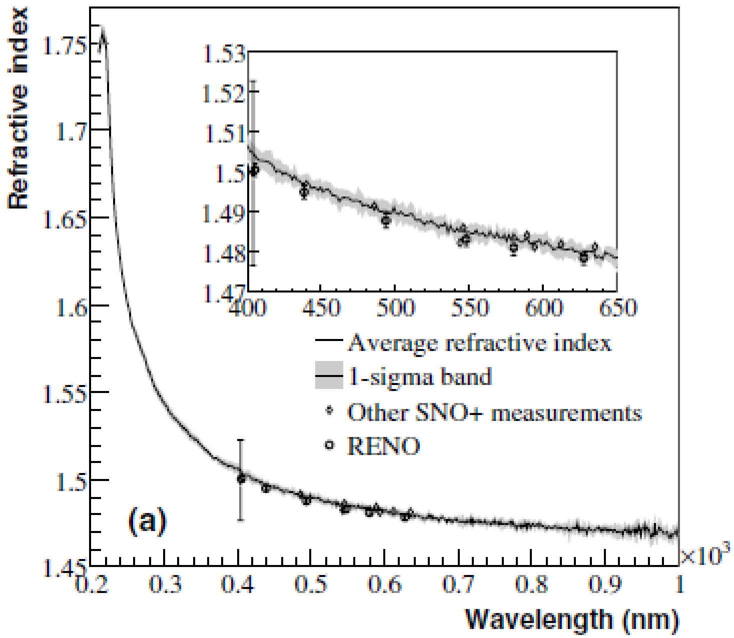
Apparatus: Varian Cary Eclipse Fluorescence Spectrophotometer



- Need to know energy transfer between LAB & LAB-flour mixture
- Also need to optimize to the sensitive wavelength region of PMT used to each experiment

Refractive Index

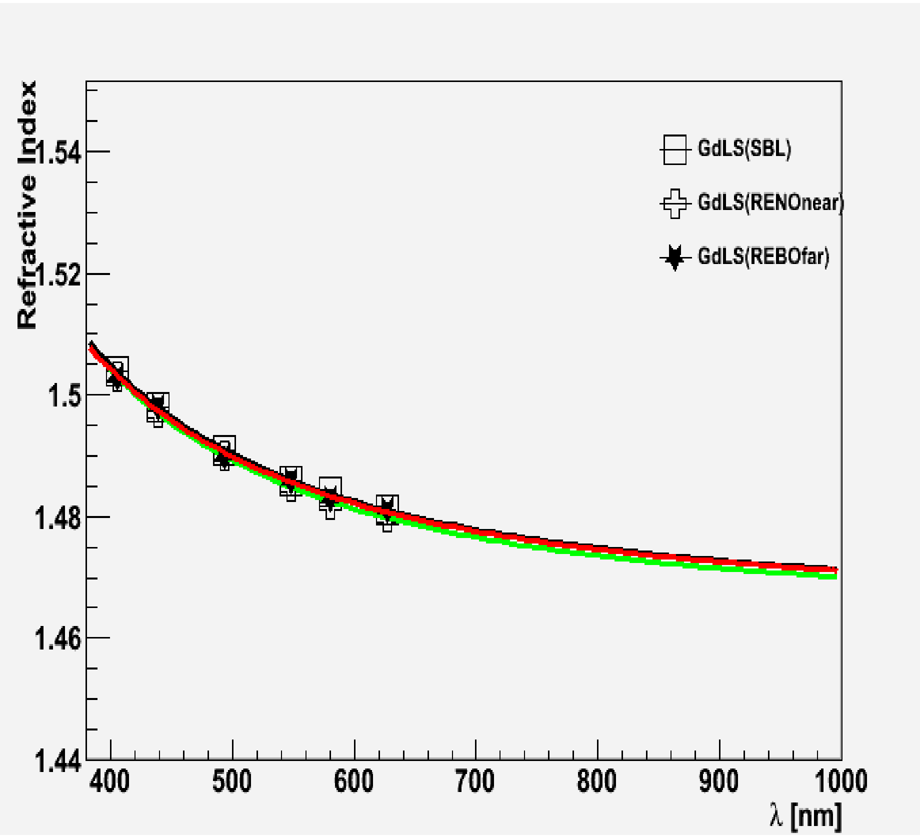
Ellipsometric measurements of the refractive indices of linear alkylbenzene and EJ-301 scintillators from 210 to 1000nm



Phys. Scr. **84** (2011) 035701 (4pp)

H Wan Chan Tseung and N Tolich

RENO/Hanaro SBL Sample



Fitting function: $a + bx^{-2} + cx^{-4} + dx^{-6}$

Water Contents

Karl-Fischer (titration) Coulometer

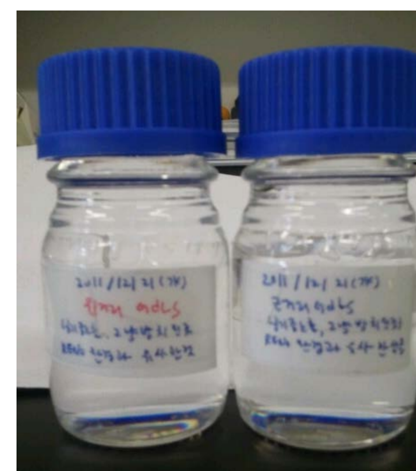
	Hanaro SBL GdLS	RENO GdLS (ND)	RENO GdLS (FD)
Water contents (ppm)	223.6 (± 5.0)	48.5 (± 5.0)	50.3 (± 5.0)

[Apparatus : METTLER TOLEDO]

Ex) 1g: 100ppm = ~16ton : x

x=~1.6kg

➡ ~1.5 bottles of 1L coke



Density Measurement

- # of protons in Target
- Minimize buoyancy force between layers

For cross-check

- Portable density meter
- Ohaus balance
- Hare's Apparatus

T=24.5±0.2℃

$\pm 0.001 \frac{g}{cm^3}$

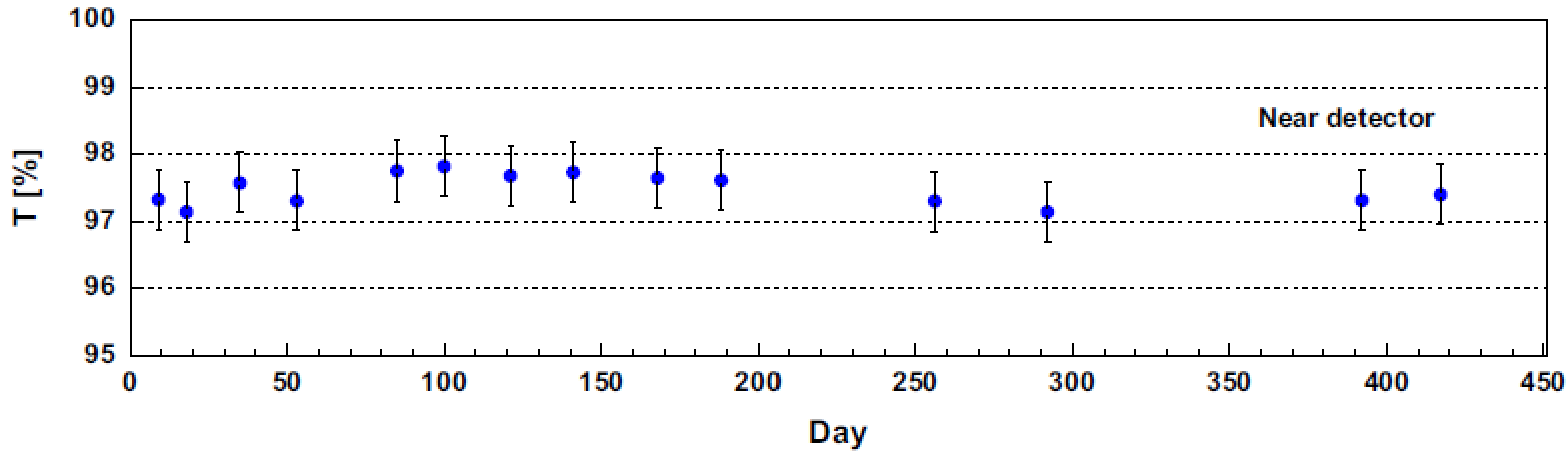
	Near detector	Far detector
GdLS	0.856	0.856
LS	0.855	0.855
MO	0.819	0.820

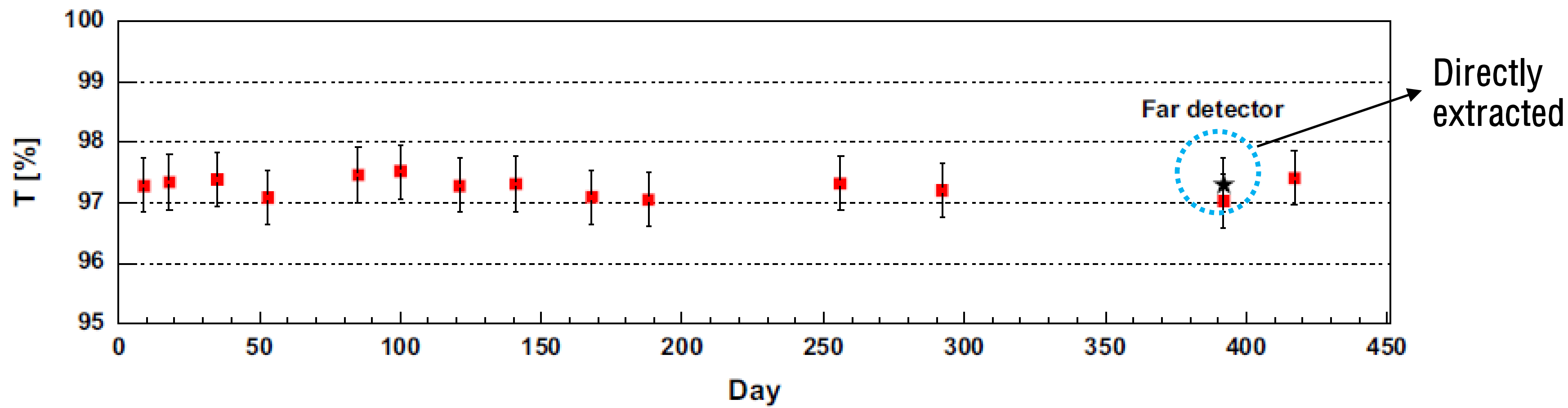
Reference [specific gravity]
-LAB: Isu chemical Co. 0.856~0.866 @15.4^{o}C
-MO: Seojin chemical Co. KF50 0.800~0.840 @15.4℃

→ ND & FD same within reasonable measurements & uncertainties

Stability of Transmittance [T]

* Shimadzu UV-1800 spectrophotometer





* Stable transparency at 430 nm

* Stable : currently ~4 years (2011.09 ~ present)

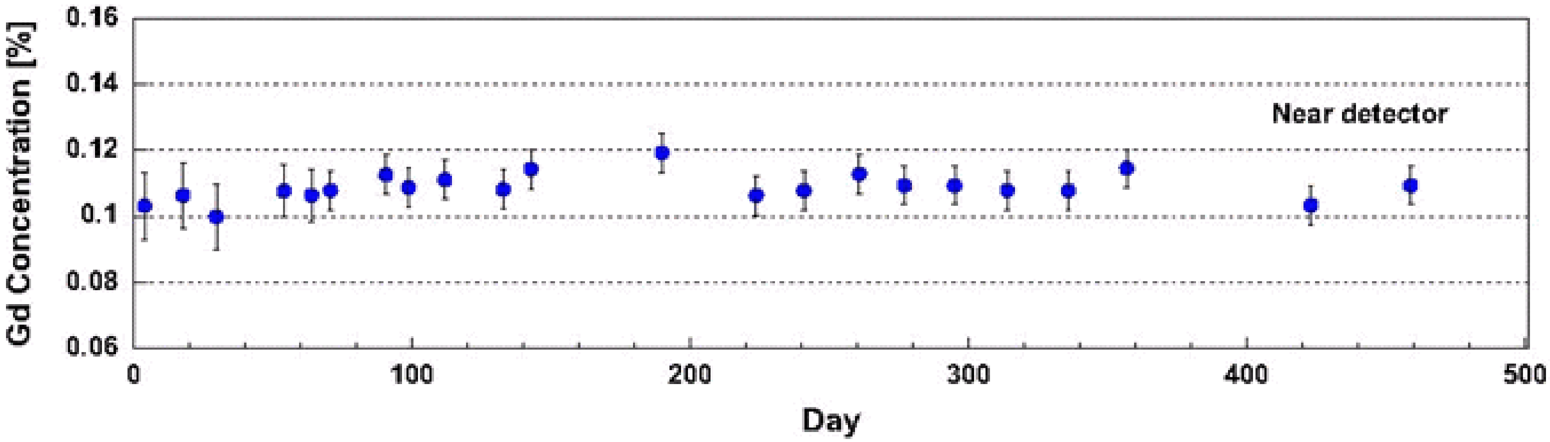
Stability of Gd Concentration

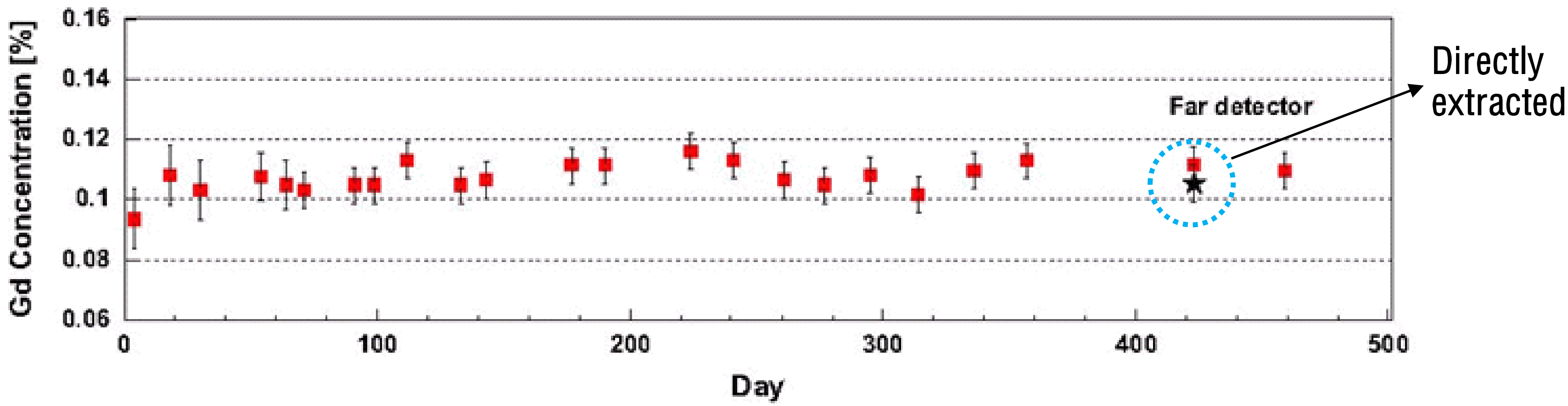
EDTA titration method

* Stable Gd concentration (0.11%)

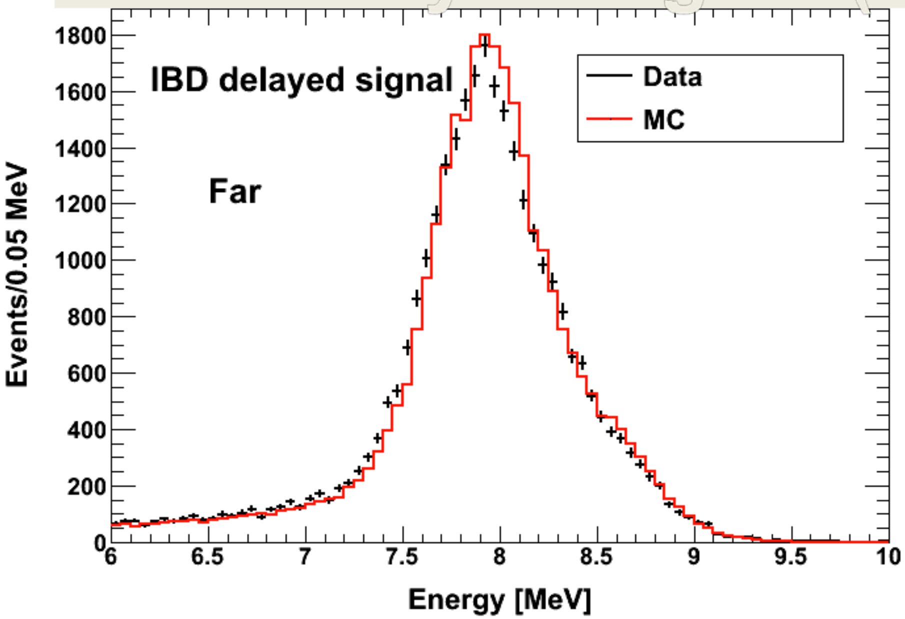
- Request outside company for consistency check (ICP-AES). Same results

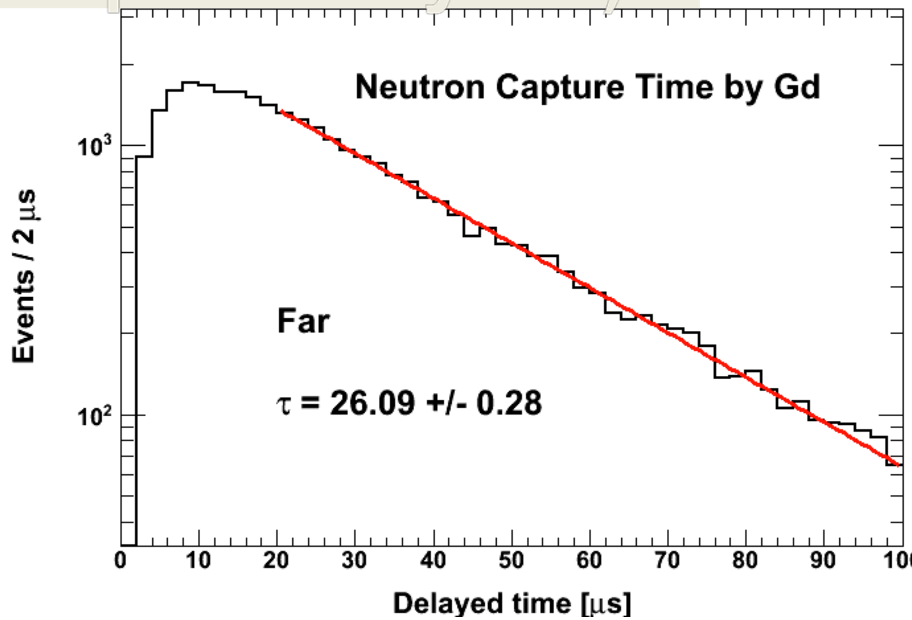
Nuclear Instruments and Methods in Physics Research A, 707, 45-53 (2013. 4. 11)

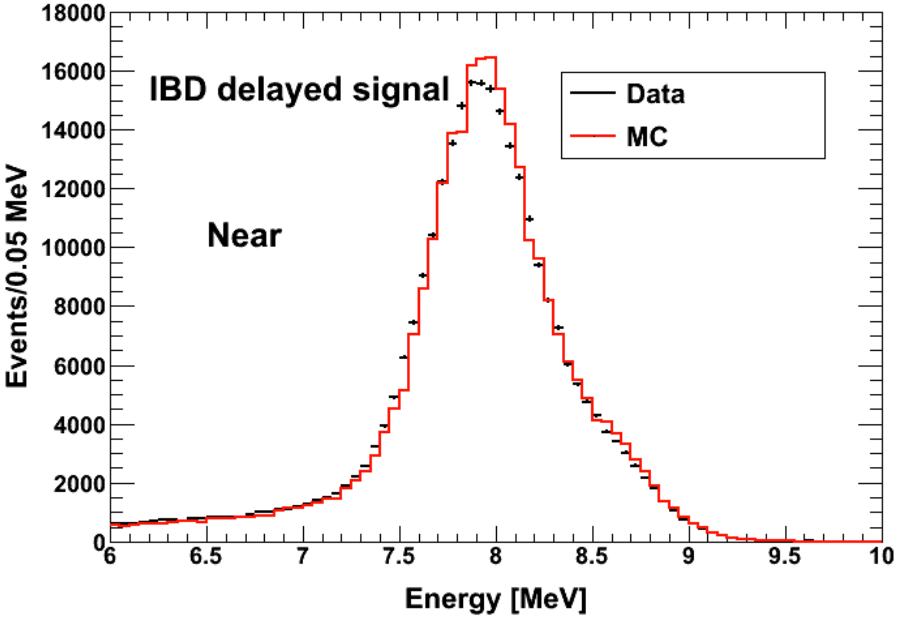


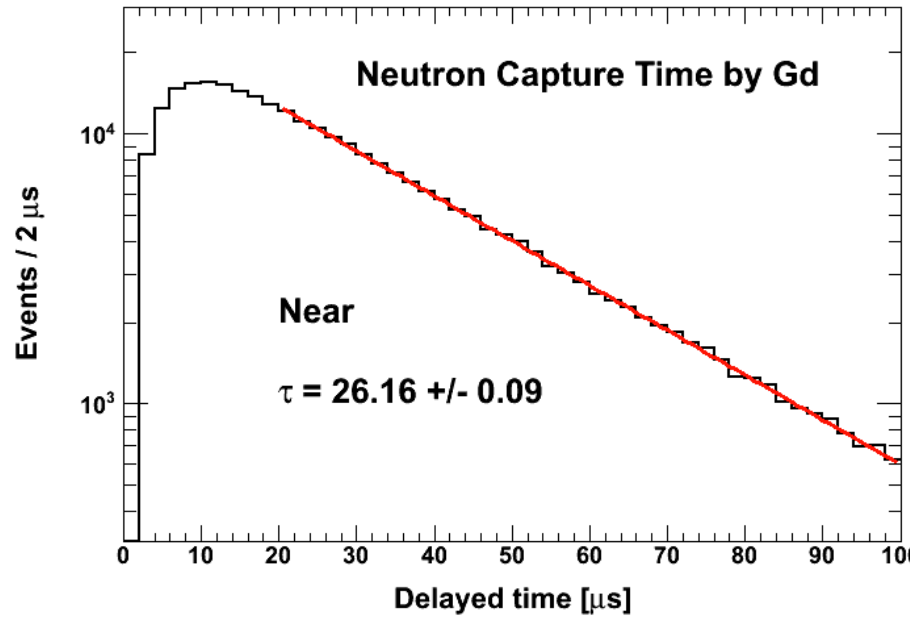


Observed Spectra for Delayed Signal (n captured by Gd)





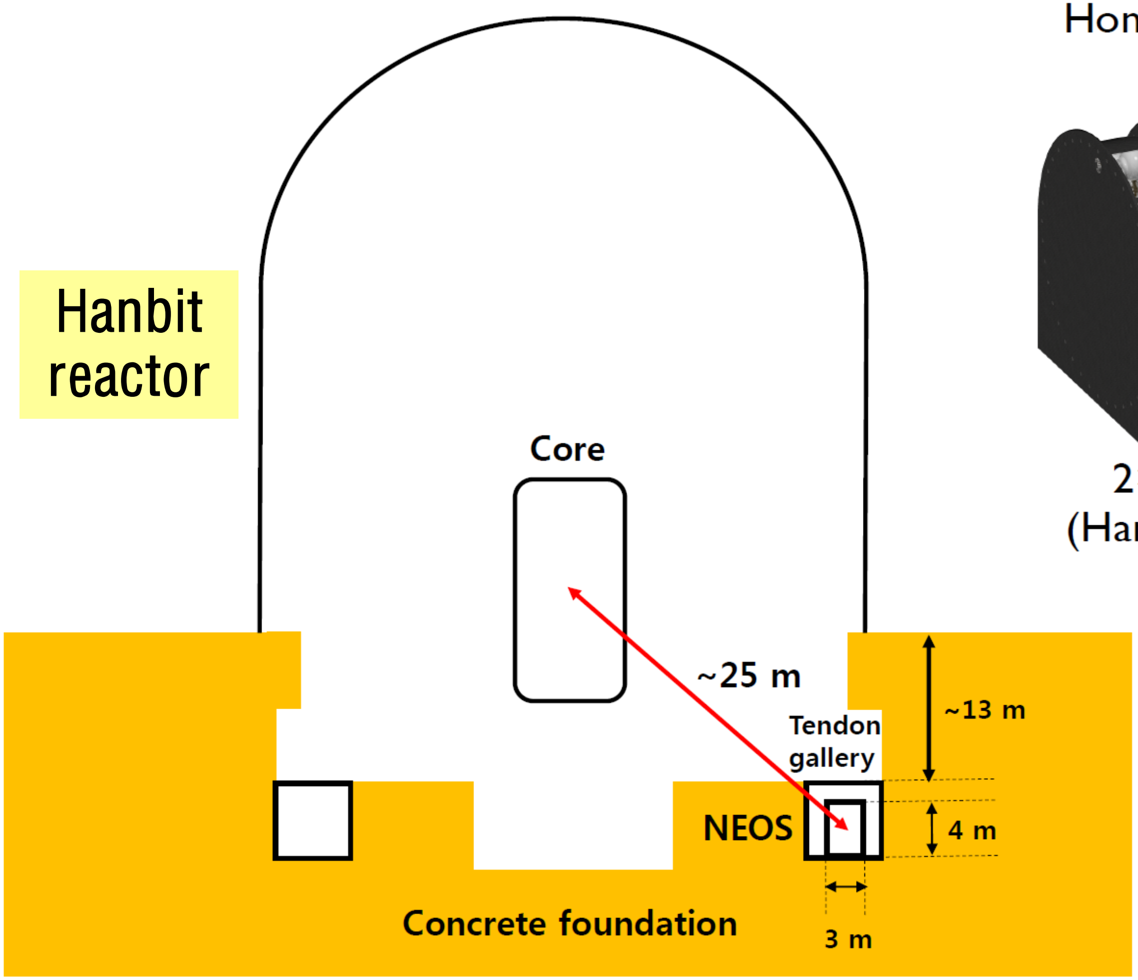


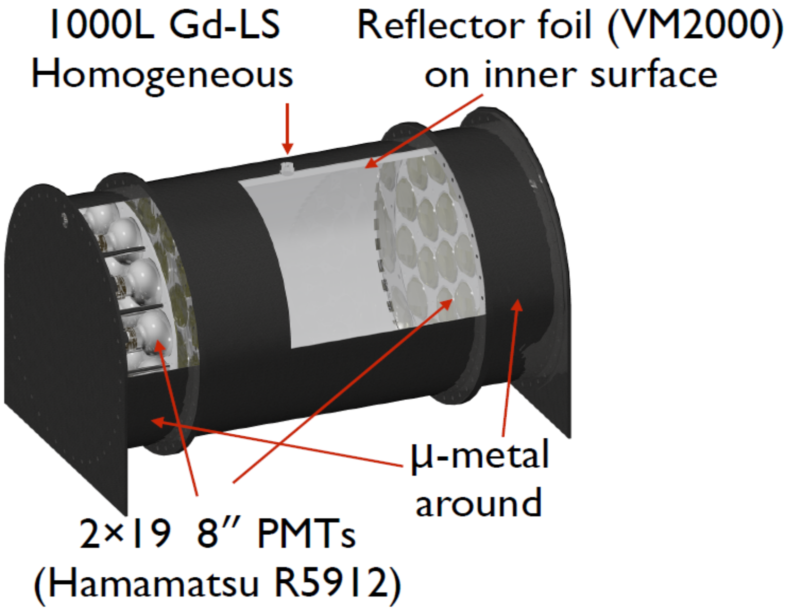


NEOS (NEutrino Oscillation at Short-baseline) Experiment

To investigate a reactor antineutrino anomaly

Hanbit reactor



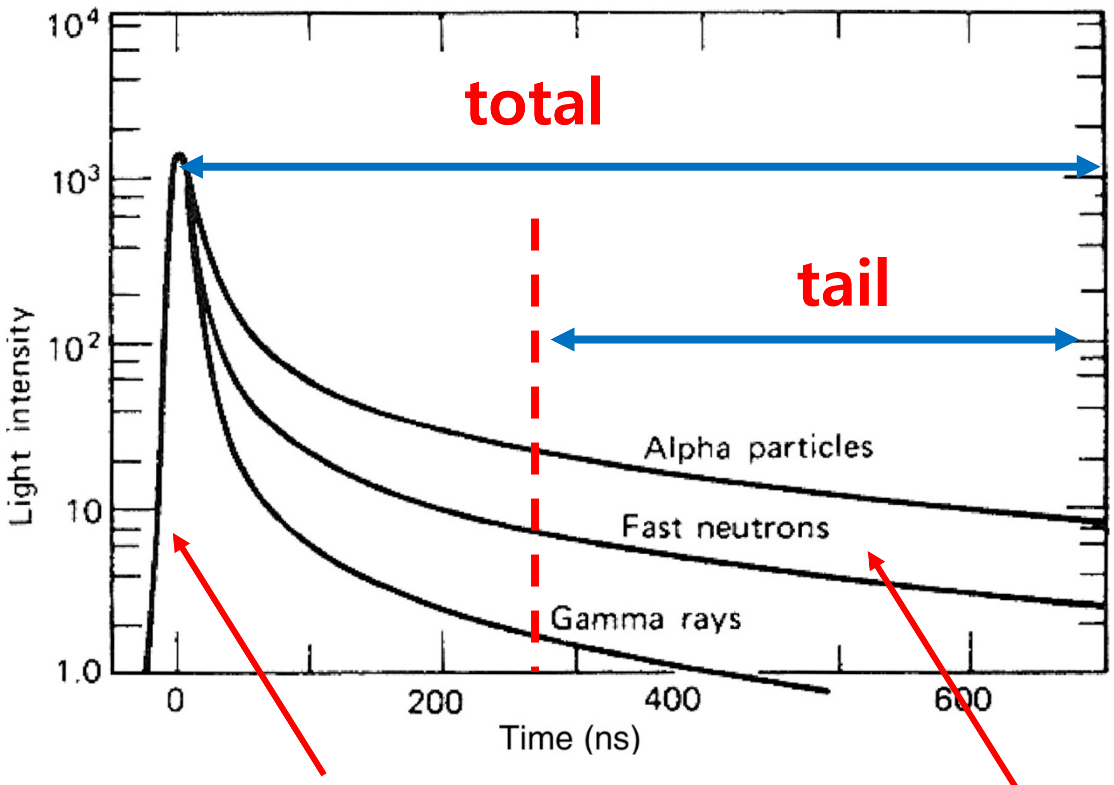


- 1-ton LAB-based LS
- ~0.5% Gd-loaded
- **~10% DIN added**

Pulse Shape Discrimination (PSD)

The waveform observed varies depending on the particle in the LS. Using this, it is possible to specify the particle is detected by the waveform of the event.

Tail shape is different, depending on particles



fast component

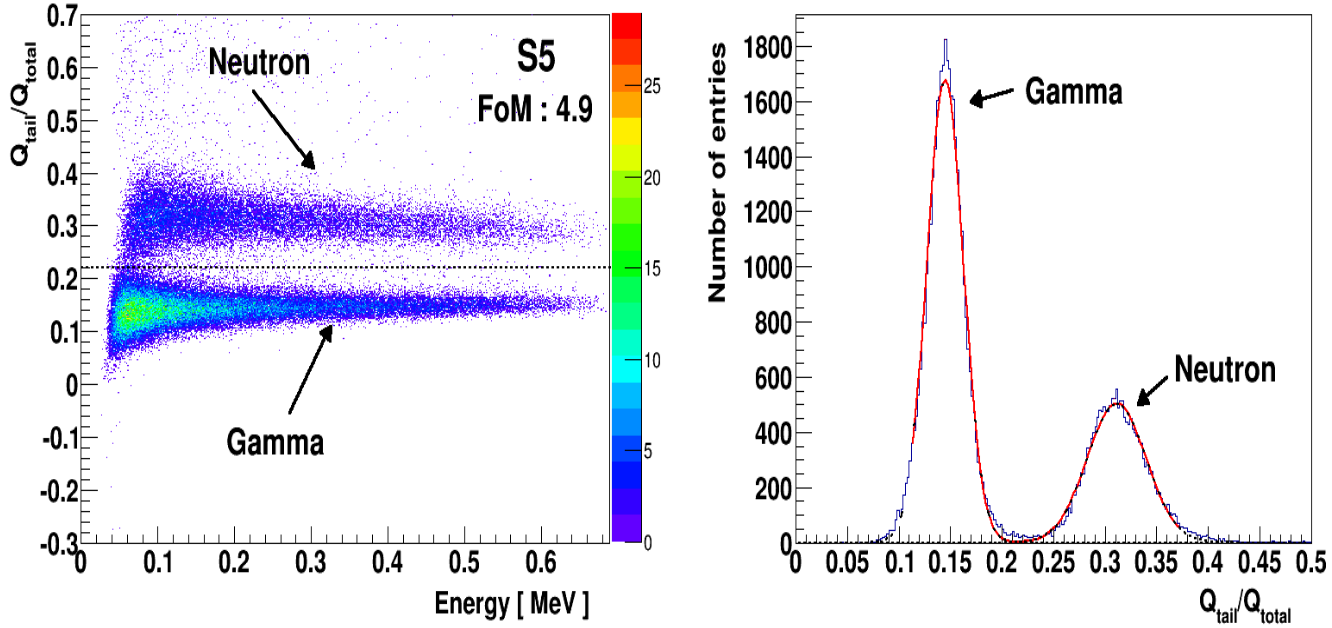
slow component

PSD parameter

$= Q_{tail}/Q_{total}$

Figure of Merit (FoM)

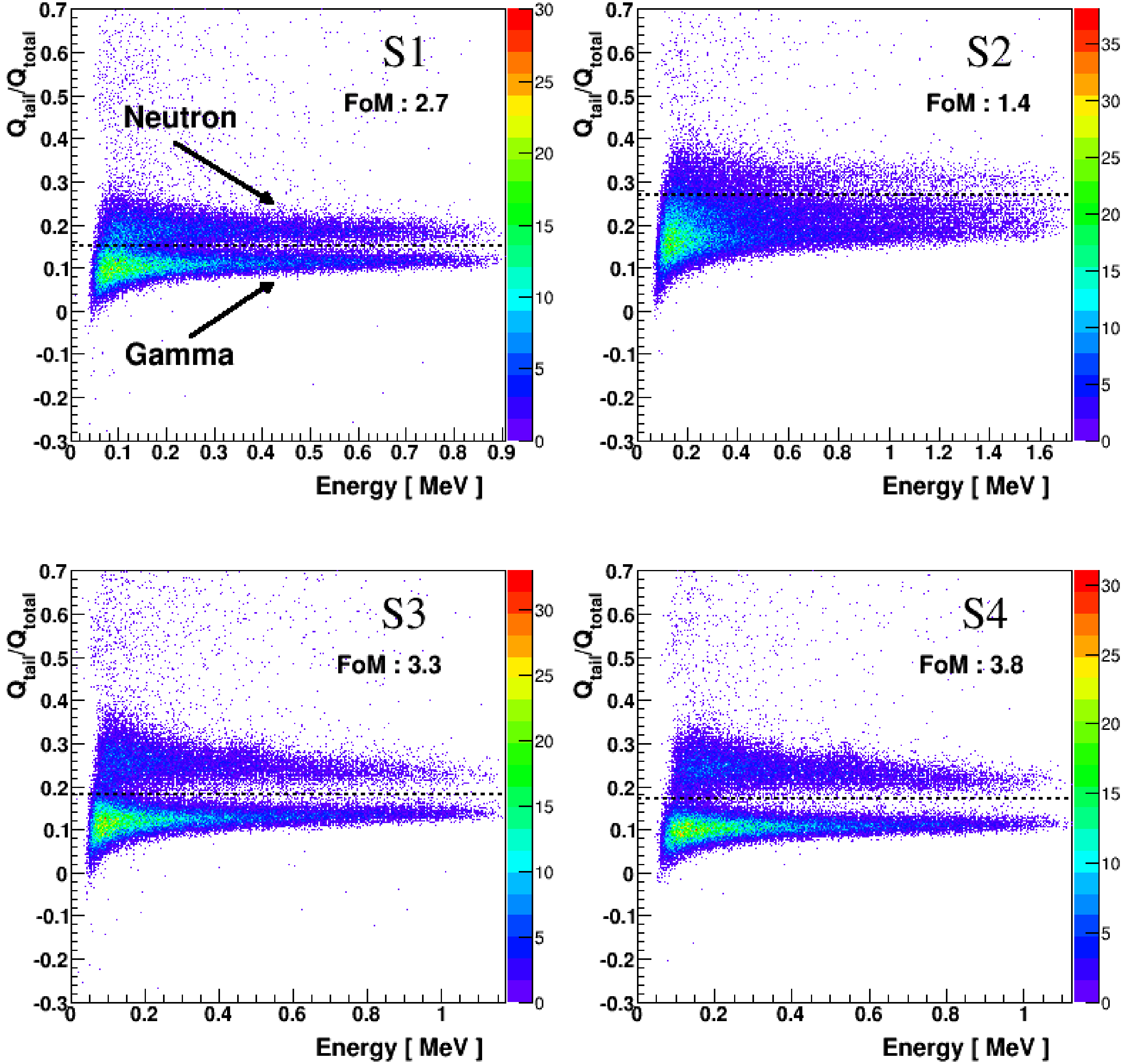
$$\mathrm{FoM} = \frac{\Delta S}{\sqrt{\sigma_x^2 + \sigma_y^2}}$$



- S5 sample [UG-F (DIN) + LAB-based 0.3% GdLS] = 10: 90%

Adding even a small portion of DIN-based liquid scintillation coctail (UG-F) into a LAB-based GdLS sample, LY improves, and the FoM power is drastically improved

FoM



S1
FoM : 2.7
Neutron
Gamma
S2
FoM : 1.4
S3
FoM : 3.3
S4
FoM : 3.8
Q_{tail}/Q_{total}
Energy [MeV]

Summary

- Historically, LS plays an important role in neutrino community (especially, in reactor neutrino)
- LS has been widely used in various neutrino experiments

❑ Loading metal into LS is not trivial, but it has been well established and became well-known technology

- Especially, transparency & long-term stability of GdLS is very important

- For next generation neutrino experiment, purification, purification, purification will be important matter