Overview of Gd-loaded and Unloaded Liquid Scintillator for Reactor Anti-ν_e Experiments

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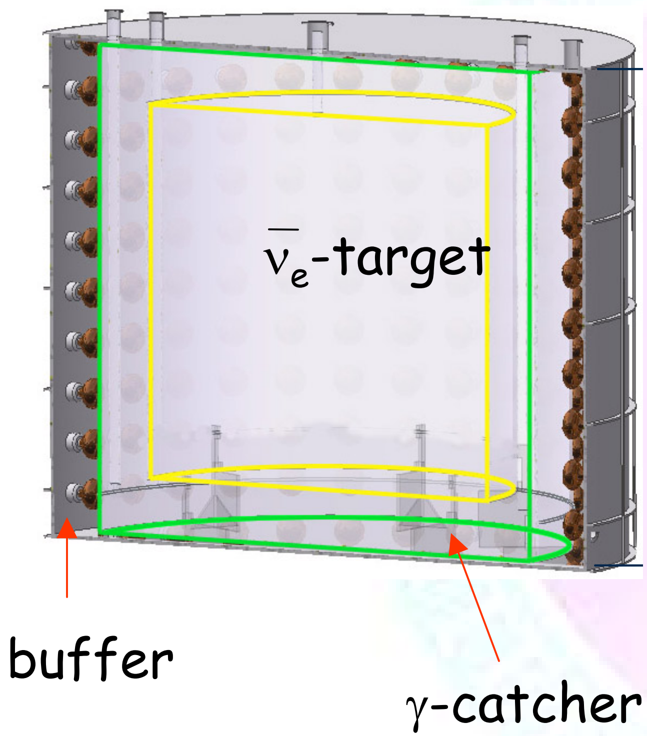
What do we learn from the past?

What are the current developing techniques?

Anti-ν_e Detector



Phase	Type
$\bar{\nu}_e$-target	Gd-loaded LS
γ-catcher	unloaded LS
Buffer	non-scintillation liquid
Veto	RCP, SS, LS or Water

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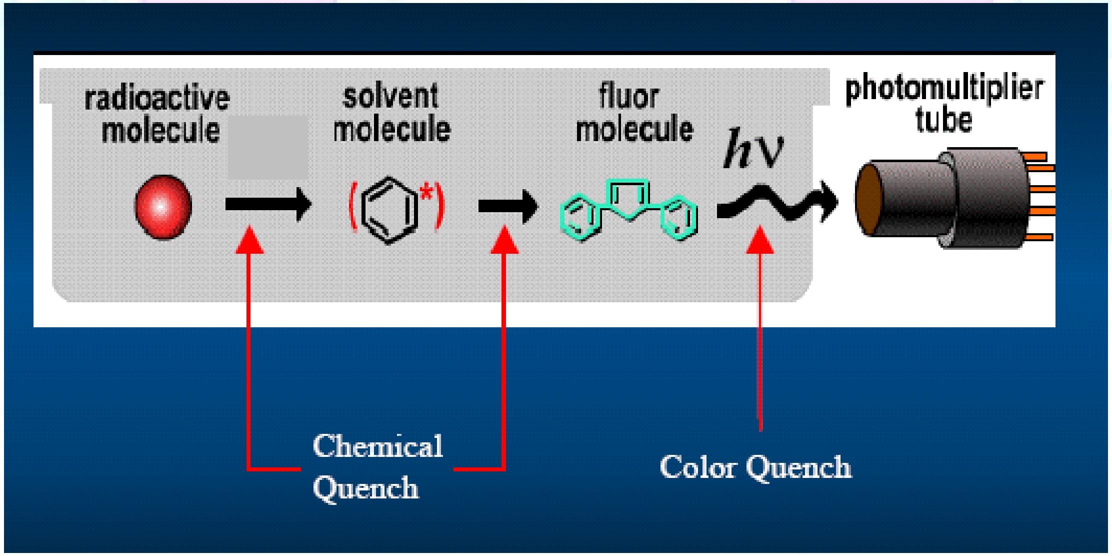
Some Old Selections for Gd-loaded[1] and unloaded[2] LS

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

Components of unloaded LS

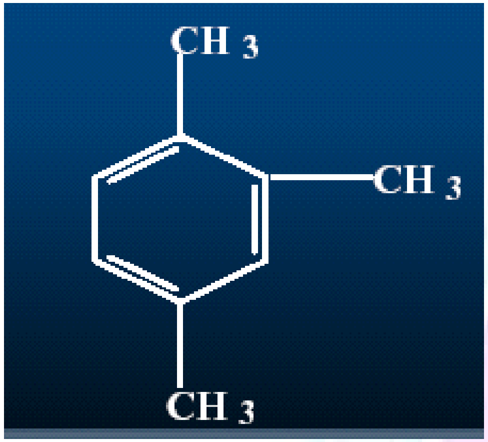
- **Aromatic solvent** that contains a high density of π-electrons for energy transfer
- **Fluor** that transfers the energy (<400 nm) to light (>400 nm) within the optimal detection range of PMT



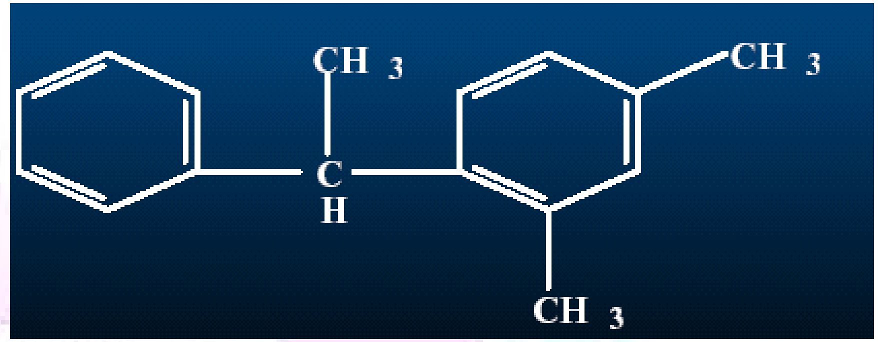
NOνA 22k-tons unloaded-LS

Component		% (w./w.)
Mineral Oil	diluent	94.4
PC	scintillator	5.5
PPO	primary shifter	1.2×10^{-1}
bis-MSB	secondary shifter	1.7×10^{-3}
BHT	Anti-oxidant	~$2x10^{-6}$
Performance	Attenuation Length	2m – 10m
	Light Yield	28% anthracence

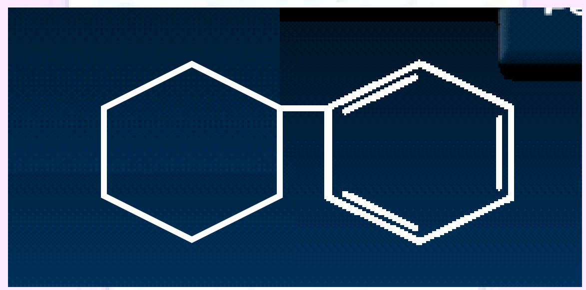
Selected Aromatic Solvents



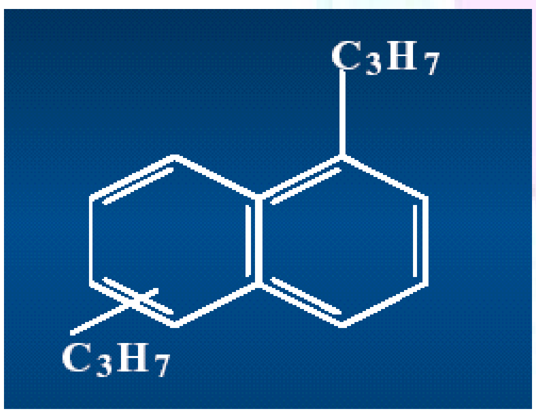
Pseudocumene (PC)
(1,2,4-trimethylbenzene)



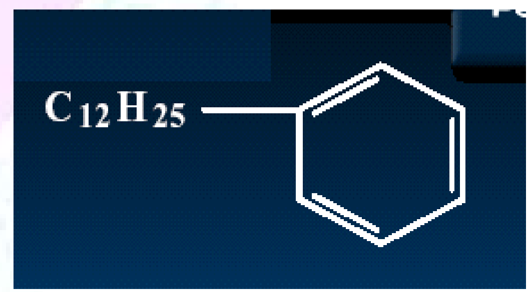
1-phenyl-1-xylyl-ethane (PXE)



Cyclohexylbenzene (PCH)



Di-isopropylnaphthalene (DIN)



Linear Alkyl Benzene (LAB)

Properties of Selected Scintillator

	Gd Loading	d (g/cm^3)	UV Abs^{430} before / after	Abs_{260}	n^{20}	Light Yield	H atoms‡ per c.c	Flash Point
PC	Yes	0.889	0.008 / 0.002	2	1.504	1	5.35×10^{22}	48 C
PCH	Yes	0.95	0.072 / 0.001	1.7	1.526	0.46	5.71×10^{22}	99 C
DIN	Yes	0.96	0.040 / 0.023	>10		0.87	5.45×10^{22}	>140 C
PXE	Yes, but not stable	0.985	0.044 / 0.022	2.1		0.87	5.08×10^{22}	167 C
LAB	Yes	0.86	0.001 / 0.000	1	1.482	0.98	6.31×10^{22}	130 C
Mineral Oil C_{24}~C_{28}	No	0.85	0.002 / 0.001	1	~1.46	~	6.73 – 8.00 $\times10^{22}$	215 C
Dodecane	No (<20%)	0.75	0.001 / 0.000	1	1.422	~	6.89×10^{22}	71 C

Comparison of Liquid Scintillators

Criteria	Needs	LS
Physical Performance	optical transparency	LAB > PC > PCH > DIN ~PXE
	light yield	PC > LAB > DIN~PXE > PCH
Safety	flash point and toxicity	PXE > DIN > LAB > PCH > PC
Cost	Cheap and availability	LAB
Compatibility	vs. acrylic, PVC, …	LAB
Neutrino Interaction	H atoms per c.c	LAB > PCH > DIN > PC > PXE
Density	Buoyant force	Dependent on ρ_{buffer}

Preferable Unloaded LS

	PC	LAB
Flash Point	48°C (extra concern raised)	130°C
n^{20}	1.50	1.48
Density	0.89 g/mL	0.86 g/mL
Attenuation Length	> 20 m	> 25 m
Light Yield	100% PC or 55% anthracene	~98% of PC
Compatibility (acrylic)	Negative, need diluent (i.e., dodecane)	Stable so far; need further study
Cost and availability	availability not sure	Cheap and available
Fluor Dissolution	Very Good	Might need help from PC

Components of Gd-LS

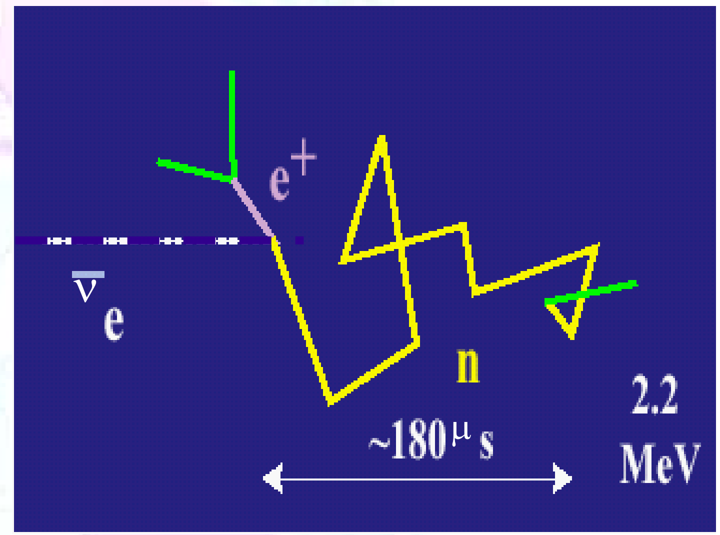
- H atoms to interact with $\overline{\nu}_e$ and to capture the n's (0.3 barn):

  $n + p \rightarrow D + \gamma$ (2.2 MeV)

- 0.1~0.2% of Gd in organometallic form to enhance the n-capture cross-section (49000 barn):

  $Gd \rightarrow Gd^* \rightarrow Gd + \gamma$'s (8 MeV)

- Fluors to transfer the light via the delocalized π-electrons of the aromatic solvent into visible region.



Early Inspiration on M-loaded LS

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LENS

Low **E**nergy **N**eutrino **S**pectrometer

M-loaded LS (M=Yb,In) in carboxylate or β-dikitonate

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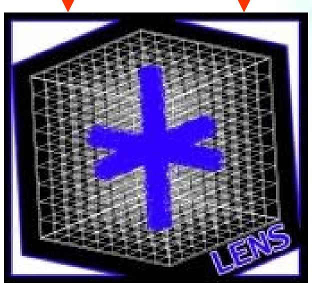
INFN LNGS/JINR



DOUBLE CHOOZ

Gd-LS Preparation

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- Solvent-Solvent extraction vs. organometallic solid dissolution:

	Solvent-Solvent Extraction	Organometallic Solid Dissolution
Synthesis	One step (~few hrs); simpler and gentle chemical system; the procedures are very consistent and reproducible; easier to be adopted at different institutions.	Multiple steps (~days); increase the chances of contamination; normally require the aids of P=O compounds and/or under harsh chemical condition (high pH or Temp.)
Transportation	Specific tanks and handling required	Easy to be transported

- High concentration (1~2%) of Gd-LS to be diluted at percent of interest on experimental site.

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Gd-LS in Reactor Experiments

- Past Experiments
  - CHOOZ (direct dissolution)
  - Palo Verde (Bicron)
- Newly Proposed Experiments
  - Double-CHOOZ (LNGS, MPIK)
  - Braidwood (BNL)
  - Daya Bay (BNL, IHEP, JINR)

Palo Verde and CHOOZ Gd-LS

	CHOOZ	Palo Verde
Gd in LS	$Gd(NO_3)_3$	Gd_2O_3
Liquid Scintillator (LS)	5 tons of 50% Norpar-15 (paraffinic liquid) + 50% IPB (isopropylbiphenyl)	11.3 tons of 40% PC + 60% mineral oil
Loading Methods	dissolved in hexanol + LS	converted to 2-ethyl hexanoate dissolved in LS
[Gd]	0.9 g/L	1 g/L
Density	0.846 g/L	~0.87 g/L
L_{λ}^{att}	4 m	11 m
Light loss	0.4% per day	0.03% per day

CHOOZ. Phys. Lett. B 420 (1998) 397- 404.
Palo Verde, NIM in Phys Res. A 432 (1999) 392 – 398.

Double CHOOZ 1st Gd-LS

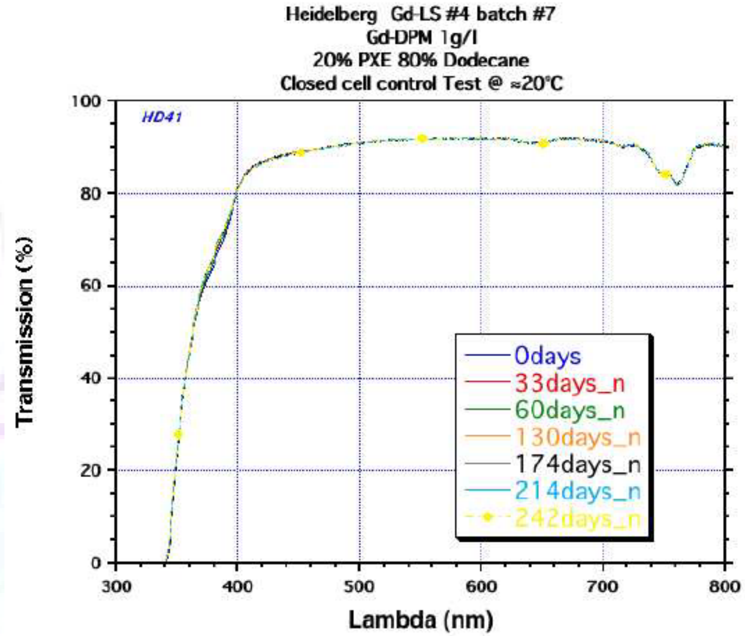
Gd in LS	Gd-CBX ($Gd(OH)R_2$)	Gd-CBX ($GdR_3.nHR$)		
LS	35%PC + 65%dodecane	35%PC + 65%dodecane	20%PXE + 80%dodecane	LVD (C_nH_{2n+2}, <n> = 9.6)
Loading Methods	Gd salt dissolution			
[Gd]	1 g/L			
Density	0.81 g/L		0.79 g/L	~0.8 g/L
L^{att}_{λ}	-	~10m	-	~11 m
problem	Not stable; polymerization	Stable, but not compatible with acrylic	Not stable; thin-gel formation	Stable; but light yield only ~10% of PXE

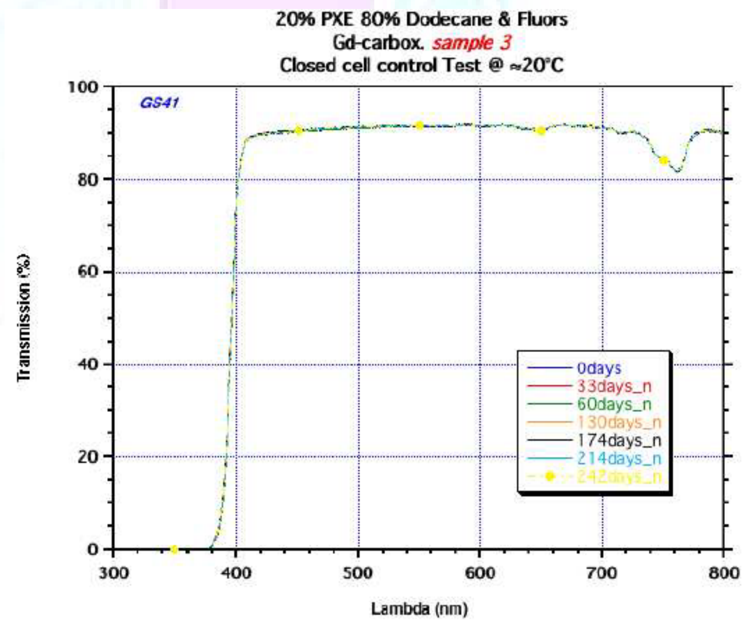
Need a new recipe with the aid of P=O compounds: **$GdR_3 \cdot nNPOC$**

C. Cattadori et. al., Ivth Future LENeuEs, Angra dos Reis, 2005

Double CHOOZ 2nd Gd-LS

Gd in LS	Gd-CBX. $(NPOC)_n$	Gd-BDK
LS	20%PXE + 80%dodecane	
Loading Methods	Gd salt dissolution	
[Gd]	1 g/L	
Density	0.79 g/L	
L_{λ}^{att}	90%T ~ 1 m?	
Stability	>250 days	

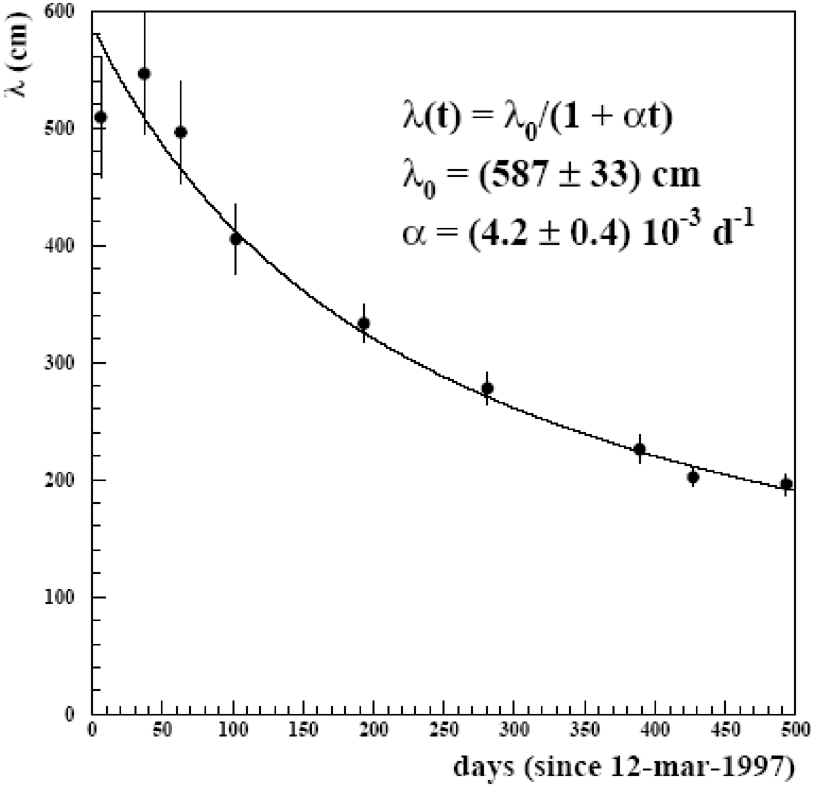




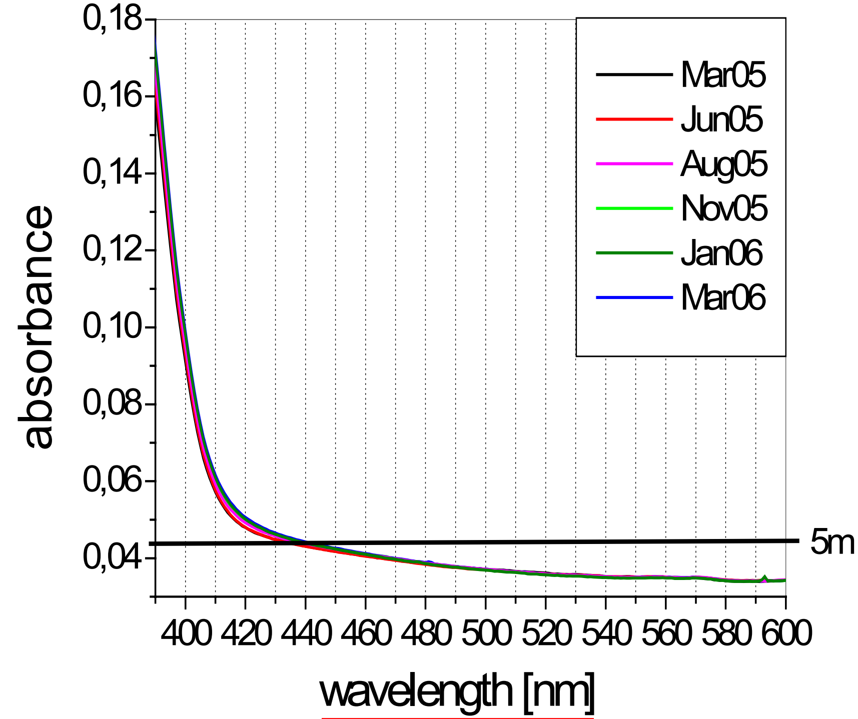
C. Buck., MPI, TAUP 2005
T. Lasserre, CEAS&APC, NO-VE 2006

Long Term Stability: Solved

Eur.Phys.J. C27 (2003) 331-374



Chooz



Double Chooz

Daya Bay-IHEP Gd-LS

- $Gd(TMHA)_3$ can be directly dissolved in LAB
- $Gd(EHA)_3$ can only be dissolved in the mixtures of mesitylene + LAB or dodecane

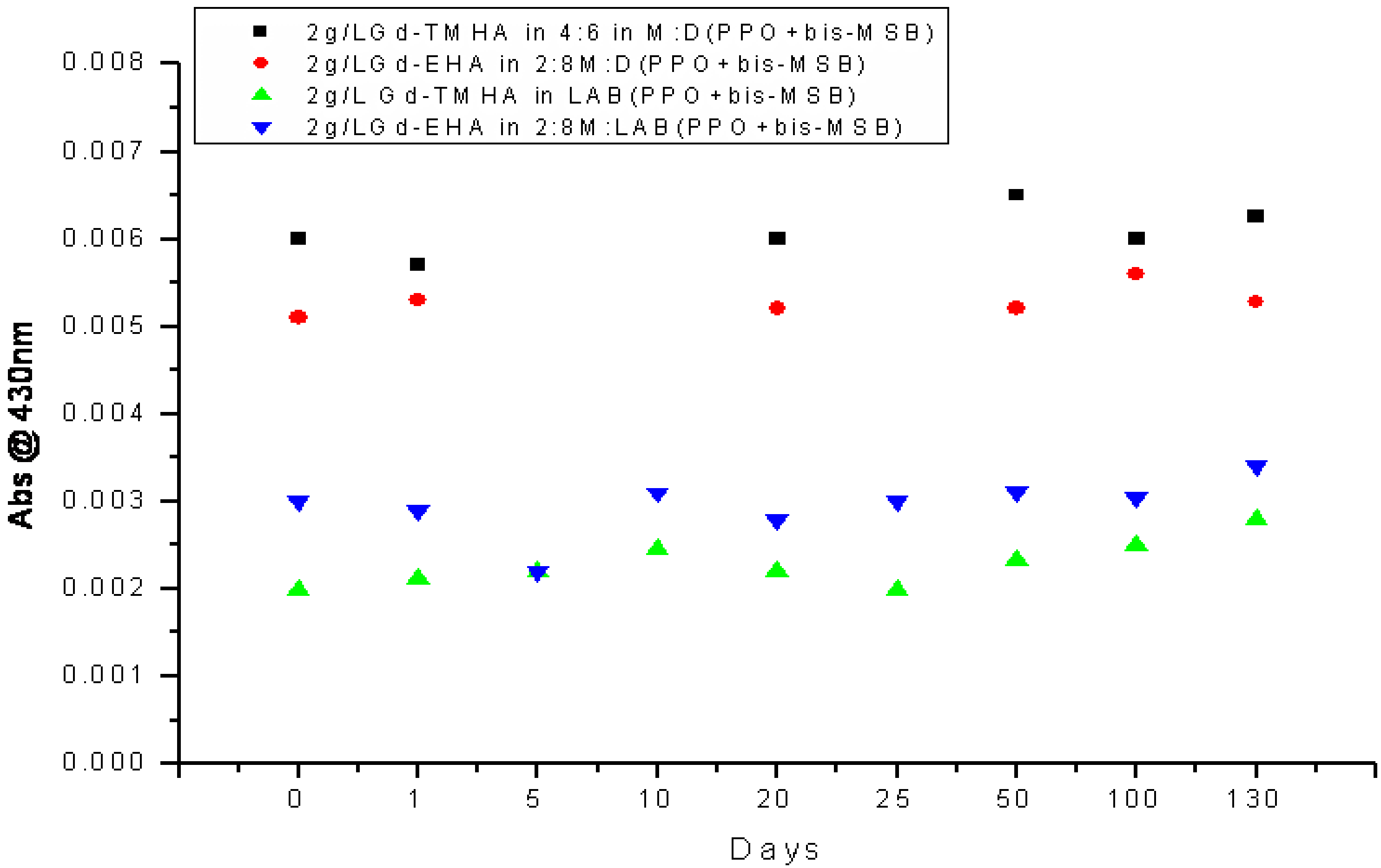
Four Gd-LS samples have been prepared

Gd (g/L)	Ligand	Fluors	Solvents
1.5	TMHA	PPO5g/L bis-MSB 10mg/L	LAB
2.0	TMHA	PPO5g/L bis-MSB 10mg/L	LAB
1.5	EHA	PPO5g/L bis-MSB 10mg/L	$V_{mesitylene}:V_{LAB}=2:8$
2.0	EHA	PPO5g/L bis-MSB 10mg/L	$V_{mesitylene}:V_{LAB}=2:8$

Daya Bay-IHEP Transparency

sample name	Absorption Measurement		Vertical LED System
	Abs @ 430nm	Calculated Attenuation Length/m	Attenuation Length/m
LAB (unpurified)	0.0053 ±0.0002	8.2±0.3	8.7±0.6
LAB (filtered)	0.0054 ±0.0003	8.0 ±0.4	9.0±1.0
LAB+PPO +bis-MSB (filtered)	0.0062 ±0.0001	7.0±0.1	7.1±0.4

Daya Bay-IHEP Stability



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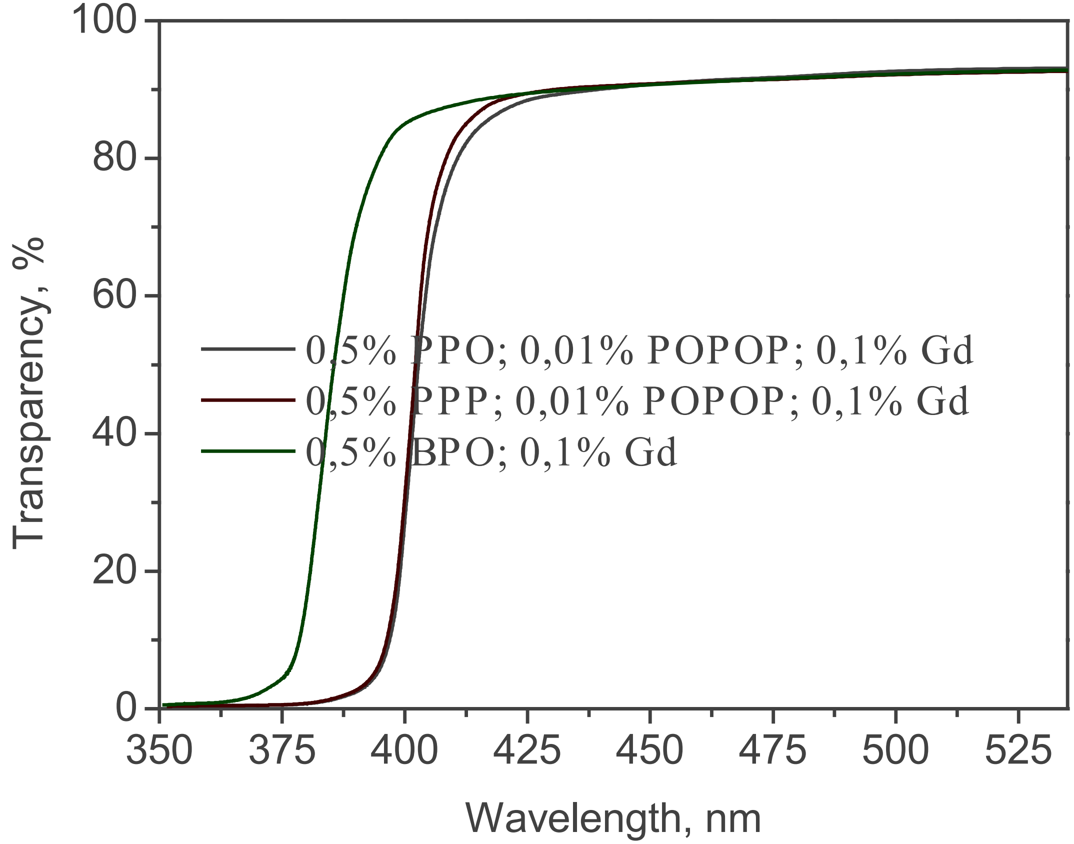
Daya Bay-JINR Gd-LS

The final composition of the Gd-loaded scintillator:

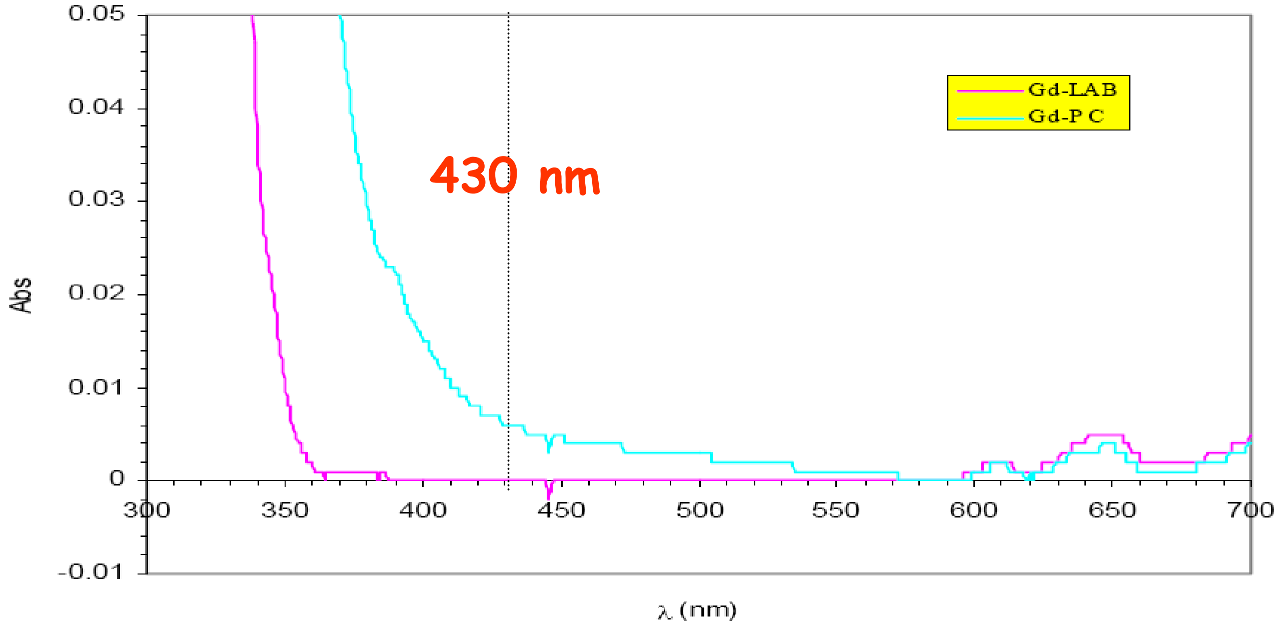
- LS: PCH (60%vol.) + TBP (40%vol.);
- primary shifter – PPO (0,5%);
- light shifter – POPOP (0,01%);
- Gd-containing dopant – $Gd[HMPA]_3Cl_3$ (0,1% Gd)

Daya Bay-JINR Transparency

Gd-loaded scintillators measured by 1-cm cell



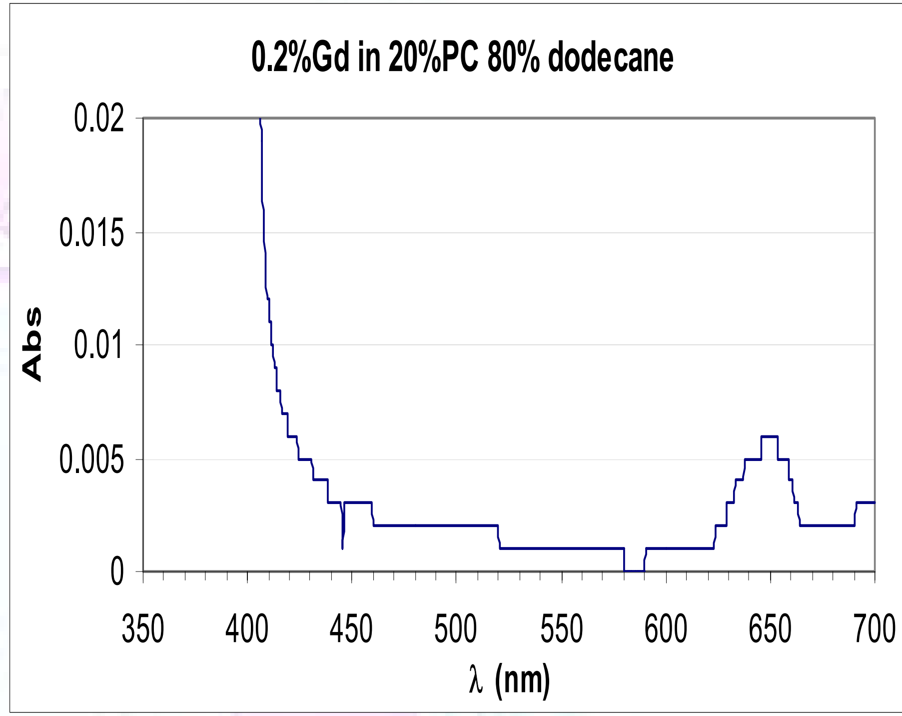
Daya Bay-BNL Gd-LS



- Few percents of Gd can be loaded into pure LAB or pure PC, respectively, using BNL carboxylate recipe.
- Both LS needs additional organic solvent:
  - Gd-LAB needs diluent (PC) to dissolve certain shifters.
  - Gd-PC needs inert diluent (MO or dodecane) to improve compatibility.

BNL Proposed Gd-LS (1)

Gd in LS	Gd-Carboxylate
Liquid Scintillator	20% PC + 80% dodecane
Loading Methods	Solvent-solvent Extraction
[Gd]	1 g/L
Density	~0.79 g/mL
L_{λ}^{Att}	> 15m
Stability	> 1.5 years
Future Study	Compatibility Test



Light yield is ~80% of pure PC

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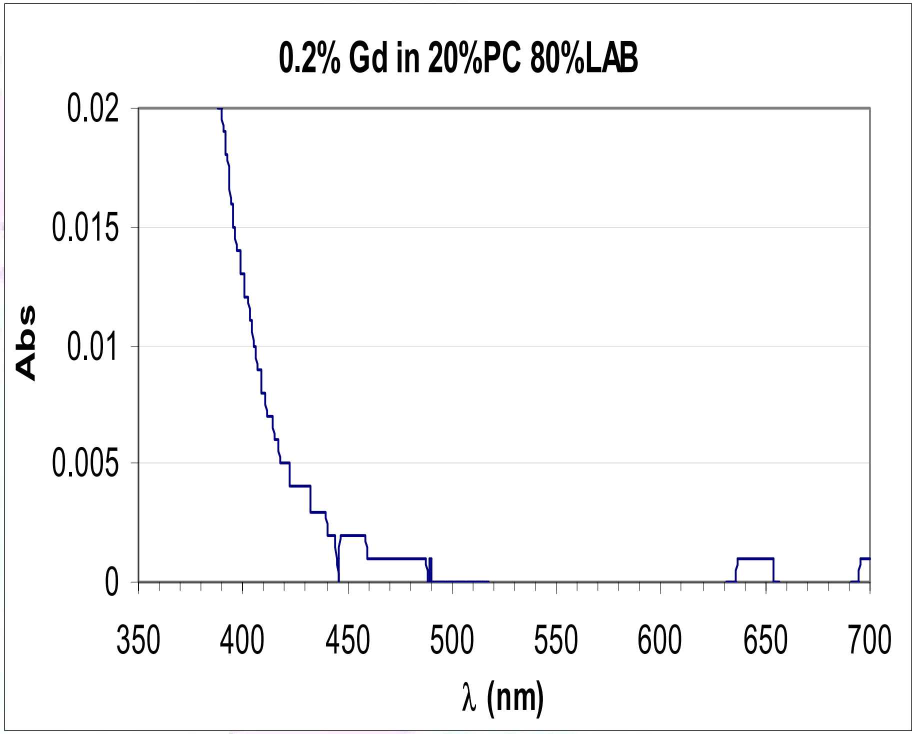
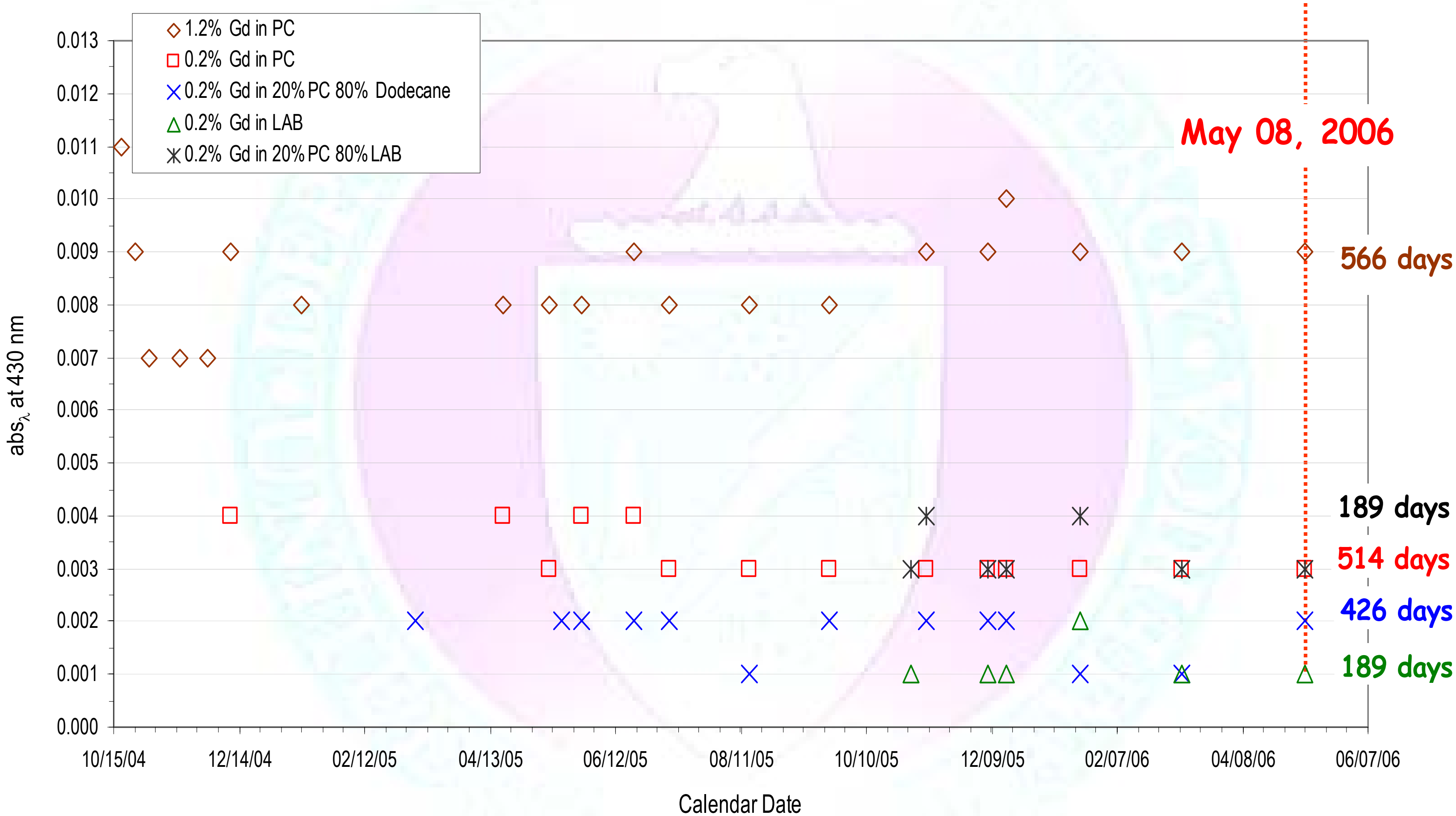
BNL Proposed Gd-LS (2)

Gd in LS	Gd-Carboxylate
Liquid Scintillator	10~20% PC + LAB
Loading Methods	Solvent-Solvent Extraction
[Gd]	1 g/L
Density	~0.87 g/mL
L_{λ}^{Att}	> 15m
Stability	> 8 months
Future Study	Compatibility Test QC monitoring



Light yield is ~100% of pure PC

Stability of Gd-LS in PC (10 cm cells)



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Something relating to Gd-LS and LS that we need to think of

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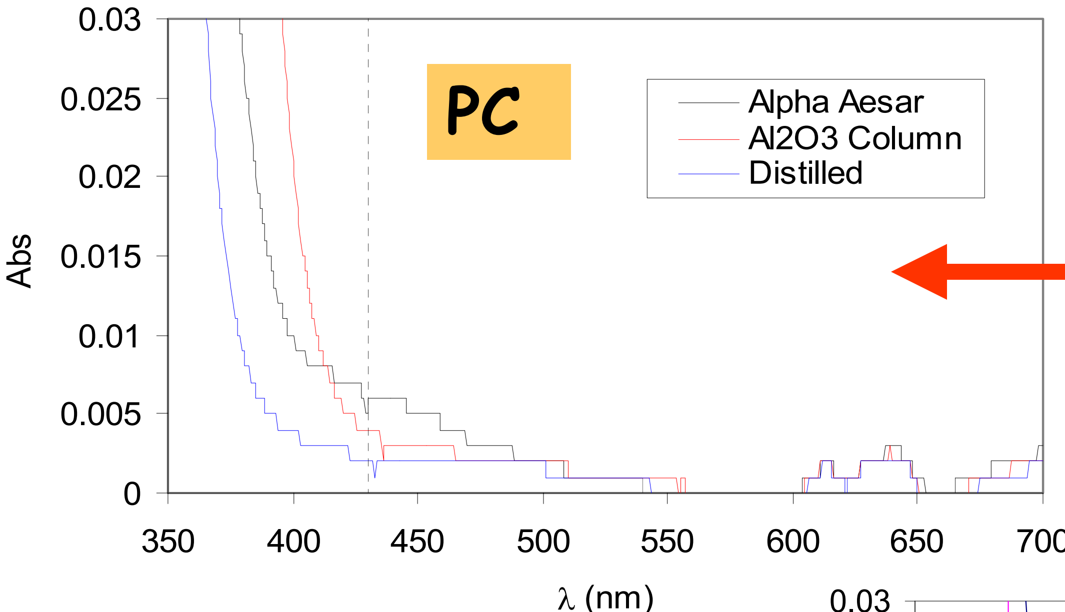
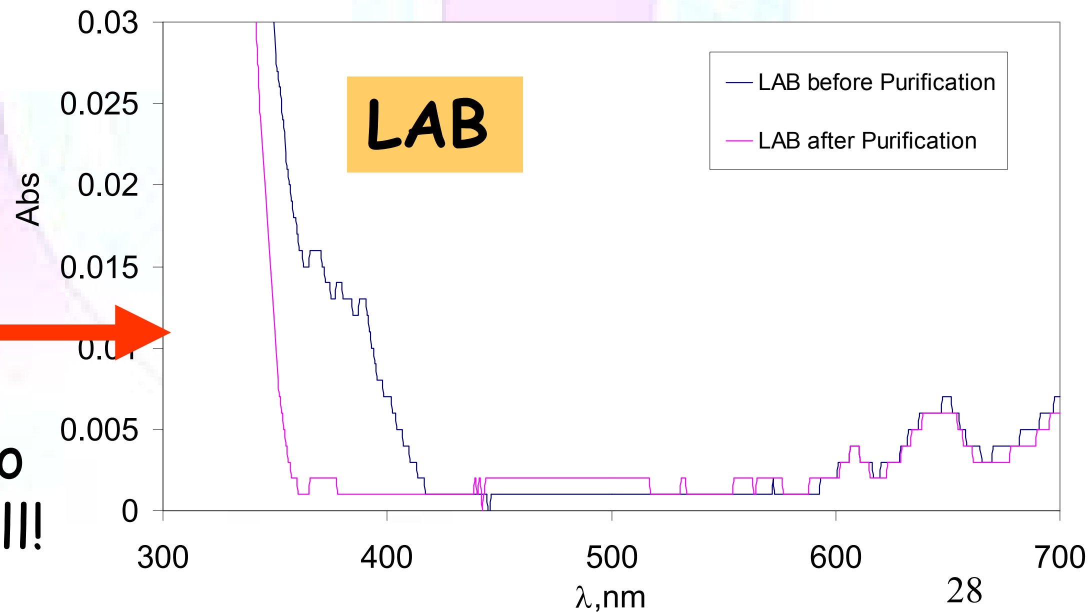
LS Purification



Vacuum Distillation improves the abs$_{\lambda=430nm}$ from 0.006 (~7 m) to 0.002 (~21m)

Column removes the absorption bump below ~410 nm and pushes the baseline even lower at 0.000 ~ 0.001 that attenuation is too small to be measured by 10-cm cell!



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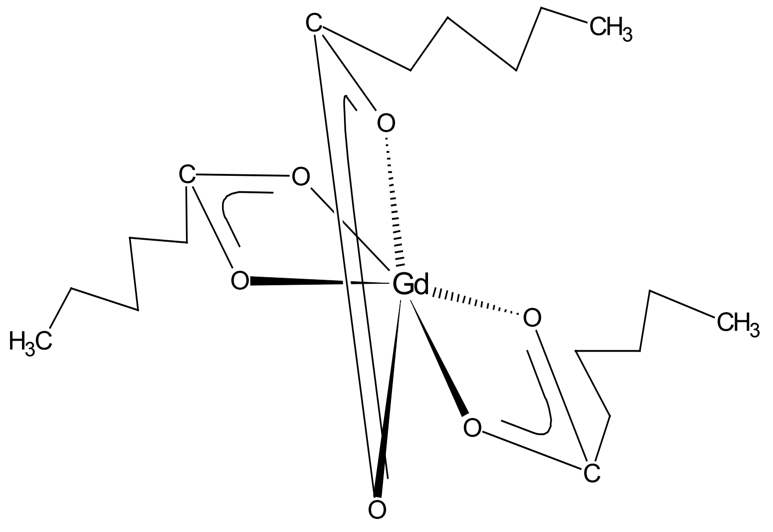
Gd-LS Characterization

Inner-sphere organometallic complex



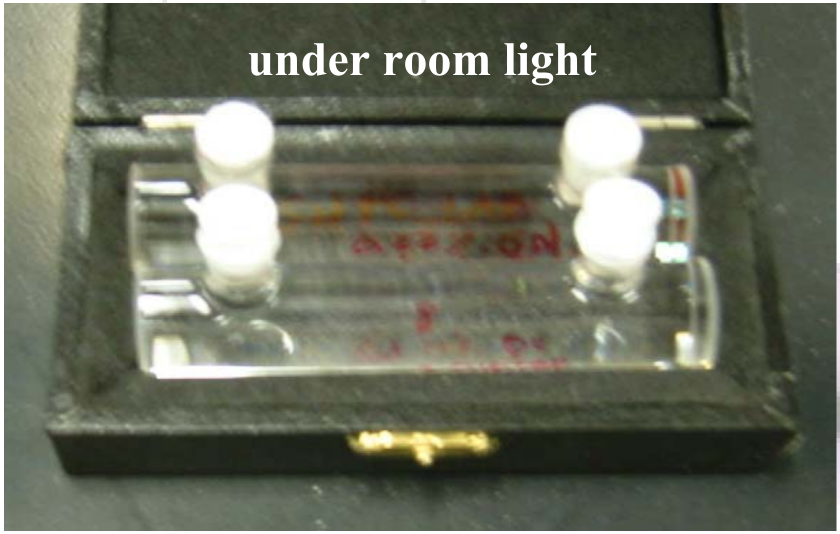
Outer-sphere ligands associated with Gd-LS: [H_2O], [OH^-], [NH_4^+] and [Cl^-]

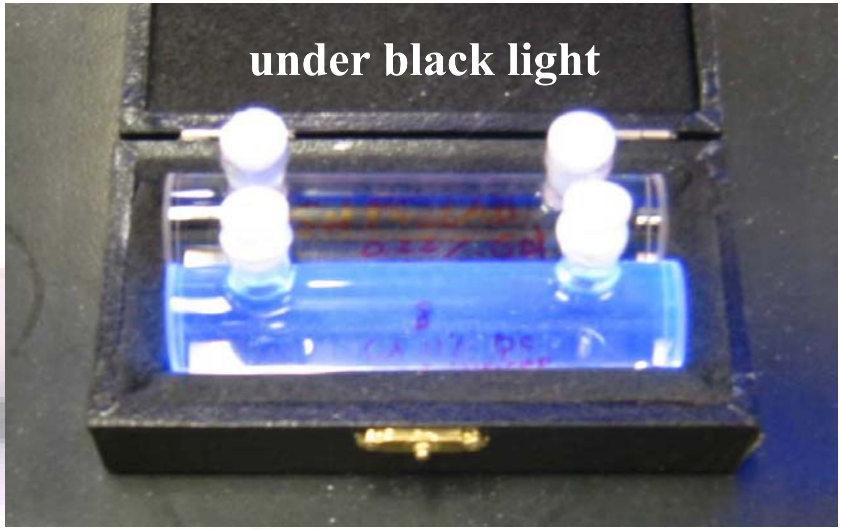
Analytical Facilities at BNL

- $L_{1/e}$ (attenuation length) by 10-cm UV-Vis, dual-beam, blue laser system, new 2-m LED vertical.
- Light Yield (S%)
- [Gd^{3+}] by colorimetric method
- [RCOOH]$_{total}$ by acid-base titration
- [RCOOH]$_{free}$ by IR
- [Gd species]$_{PC}$ by IR
- [H_2O] by Karl-Fischer titrator
- [NH_4^+] and [Cl^-] by electrochemistry

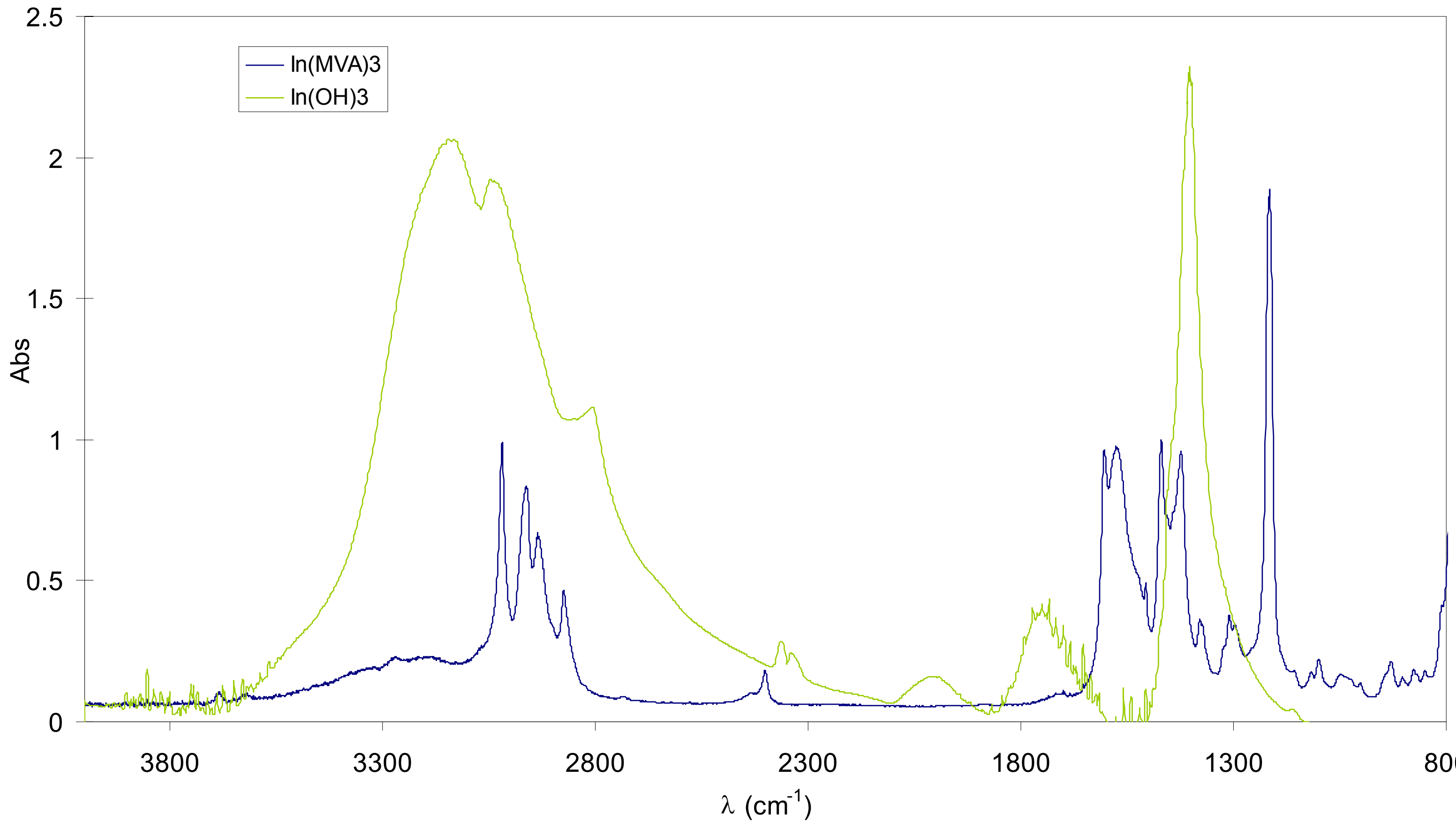
Attenuation Length Measurements

- 1- and 10- cm cells in Shimadzu UV-1601 spectrometer (200 – 1100 nm)
- 1-m horizontal cell, blue laser system (442 nm)
- 2-m vertical, pulsed LED variable wavelength system (350 – 700 nm)





IR spectra - good and bad M-LS



Selection of Fluors

Wavelength Shifter (shift the UV light to the visible region)			
Primary		Secondary	
Concentration (1.5 ~ 6 g/L)	t_d (ns)	Concentration (15 ~300 mg/L)	t_d (ns)
butyl-PBD	1.1	bis-MSB	1.6
PPO	1.5		
p-TP	0.95	POPOP	1.5
PBD	1.1		

Filling several detectors simultaneously is not trivial

- All the detectors are the same – fill them from **one** batch!

Ex: 200 tons

~ 2×10^5 L or 200 m^3

~ 7.5-m diameter spherical tank

~ W×L×H cylinder tank that can be accommodated

- From Surface or Underground?

Summary

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- For unloaded LS, safety, radioactive and chemical purity, availability, transparency, light yield, compatibility, density and H-atoms are important.
  1. PC or PXE + dodecane
  2. LAB
- For Gd-loaded LS, stability stability stability is the most important! Several techniques are developed and look good at small scale:
  1. Gd-BDK
  2. Gd-carboxylate
  3. Gd-PO
- LS Purification and Gd-LS characterization are the keys of success.
- Several technical issues, LS blending, storage and transportation, filling into different detectors simultaneously, and Gd/H ratio, need to be addressed.