

Metal-loaded Liquid Scintillators for Neutrino Experiments

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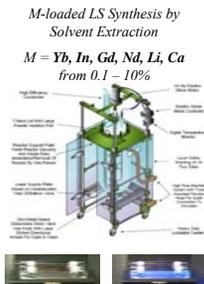
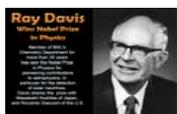
BNL Organometallic Liquid Scintillator Center – A program designated for neutrino experiments

Abstract

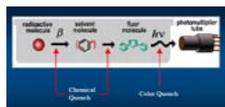
After the first direct observation of neutrino flavor transformations at the Sudbury Neutrino Observatory, future planned neutrino experiments are focusing on the understanding of the neutrino oscillation mechanism by determining key neutrino parameters, such as the mass differences and mass hierarchy, the mixing angles, and the possibility of CP violation. Organic liquid scintillators (LS) have been the detection medium of choice for neutrinos since the early discovery experiment of Reines and Cowan. For the delayed neutron-capture signal, the advantages of adding a metal element to the LS (to form M-LS) are significant. Chemically, there are challenges to adding inorganic salts of metal, such as directly to the LS. Key aspects of the metal-loaded LS (M-LS) for neutrino detection are (a) long-term chemical stability, (b) high optical transparency, (c) high photon production by the LS, and (d) ultra-low impurity content, mainly of natural radioactive contaminants, such as U, Th, Ra, and Rn. BNL Neutrino & Nuclear Chemistry group holds a long history of neutrino research since Ray Davis's pioneering Homestake experiment and has developed new chemical techniques of loading metals, such as In, Yb, Gd, Nd, and currently Li and Ca, in organic liquid scintillator that can be used for low-energy solar neutrino, reactor neutrino, or double-beta decay experiments. Metals at different concentrations in a series of liquid scintillators have been studied systematically at BNL. We have successfully prepared many metal-doped scintillators, with long attenuation lengths (10-15 m) and high light yields. These have been stable for long period of time since synthesis (>2 years for Gd-LS and Nd-LS, and >3 years for In-LS), a crucial characteristic in experiments that are planned to run for at least 3 years. Our chemical-doping technologies and the performance of different organometallic liquid scintillators for different experiments will be presented.

50 Years of BNL Chemistry and Neutrinos

- ❖ **HOMESTAKE** Radiochemical Detector
615 tons of C_2Cl_6 ; $^{37}Cl + \nu_e \rightarrow ^{37}Ar + e^-$ (~40 years)
- ❖ **GALLEX** Radiochemical Detector
30 tons of Ga; $^{71}Ga + \nu_e \rightarrow ^{71}Ge + e^-$ (1986 - 1998)
- ❖ **SNO** Water Čerenkov Detector (SNOlab)
1k tons of ultra-pure D_2O (1996 - 2006)
- ❖ **LENS** Real-time Detector (R&D)
100 tons of ~8% ^{113}In in Liquid Scintillator (began ~2000)
- ❖ **BNL-AGS NEUTRINO FACTORY (VLBN0)**
500k tons Water Čerenkov Detector? (began ~2002)
- ❖ **Reactor θ_{13}** Real-time Detector (R&D)
200 tons of 0.1% Gd in Liquid Scintillator (began ~2003)
- ❖ **SNO+ Low-energy Solar ν Double Beta Decay**
1k tons of 0.1% Nd Liquid Scintillator (began ~2005)
- ❖ **Geo-neutrino or Double-beta decay**



M-loaded LS Synthesis by Solvent Extraction
 $M = Yb, In, Gd, Nd, Li, Ca$ from 0.1 - 10%



LS selection and M-LS characterization

1. Long-term Stability
2. Long attenuation length
3. High quantum yield
4. ES&H

Material-Compatibility Testing

1. Impacts of material to liquid
2. Impacts of liquid to material
3. Over 150 (polymers, SS, coating, liners, etc) were tested in Water, LS, M-LS



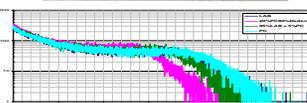
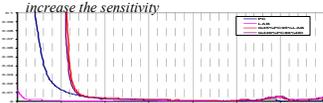
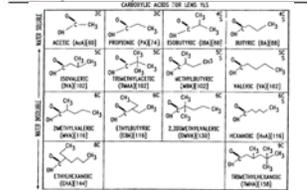
Organometallic LS Synthesis

Preparation of high-quality M-LS must be Reliable and Reproducible: **long-term stability and long attenuation length**

At BNL:

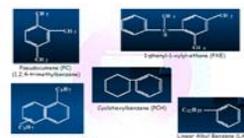
- ❖ We solvent-extract organometallic compound into LS as a function of pH with carboxylic acid, RCOOH
- ❖ We study effects of inclusion in organic phase of OH, H_2O , free carboxylic acid, Cl
- ❖ We characterize how parameters affect light yield, chemical stability, final [Gd] dissolved in LS, and chemical composition
- ❖ We develop a variety of chemical techniques for purification of all starting materials, which is the key of clean M-LS
- ❖ We remove the U/Th/K from the starting materials to increase the sensitivity

Extraction	Gd forms in LS	Comparison
alcohols	$GdCl_3 \cdot 6H_2O$	$GdCl_3$ easily dissolved in the mixture of alcohol and PC; low light yield and not stable possibly due to quenching effect and high vapor pressure
β-HD compounds	Gd(carboxylate) ₃ , PCl_2 , $(CH_2)_2$, $(CH_2)_3$, $(CH_2)_4$, $(CH_2)_5$, $(CH_2)_6$	high extraction efficiency; moderate attenuation length; not stable for long term (some phases might interact with other compounds, CMOZ experience of NO ₂)
carboxylic acid	Gd(carboxylate) ₃ , PCl_2 , $(CH_2)_2$, $(CH_2)_3$, $(CH_2)_4$, $(CH_2)_5$, $(CH_2)_6$	high extraction efficiency; long attenuation length; high light yield; very stable (>14 months for Gd-LS and over 2 years for In-LS since synthesis)



Scintillator Selection

- ❖ high density, flash point, low toxicity, and low cost
- ❖ chemical compatibility with acrylic
- ❖ high light yield and long attenuation
- ❖ Able to load organometallic compound



	Gd Loading	d (g/cm ³)	UV Abs. ¹³⁰ before/after	Abs at 260 nm	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	0.983	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 $C_{27}H_{54}$	No	0.85	0.002 - 0.001	1	-1.46	~	$6.73 - 8.00 \times 10^{22}$	215 C
Dodecane	No (<20%)	0.75	0.001 - 0.000	1	1.422	~	6.89×10^{22}	71 C

Neutrino Oscillation Parameters

Solar Neutrino Experiments: $\Delta m_{21}^2 = (7.8^{+0.6}_{-0.3}) \times 10^5 \text{ eV}^2$; $\theta_{12} = (32^{+4}_{-3})^\circ$

Atmospheric Neutrino Experiments: $\Delta m_{32}^2 = (2.4^{+0.4}_{-0.3}) \times 10^3 \text{ eV}^2$; $\theta_{13} \approx 45^\circ$

What do we learn from the past experiments?	
Known Oscillation Parameters	$\Delta m_{31}^2 \approx \Delta m_{32}^2 \gg \Delta m_{21}^2$ θ_{12} and θ_{33} are large
Unknowns need to be solved	$\sin^2 2\theta_{13}$ & δ sign of Δm_{32}^2

Reactors: Disappearance ($\bar{\nu}_e \rightarrow \bar{\nu}_i$) at $\Delta m_{32}^2 \approx 2.5 \times 10^3 \text{ eV}^2$

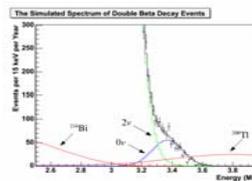
$$P(\bar{\nu}_e \rightarrow \bar{\nu}_i) = 1 - \sin^2 2\theta_{13} \sin^2 \frac{\Delta m_{32}^2 L}{4E} + \text{very small terms}$$

Reactor experiments provide the only clean measurement of $\sin^2 2\theta_{13}$; look for non- $1/r^2$ behavior of the $\bar{\nu}_e$ rate with no matter effects, no CP violation, almost no correlation with other parameters.

Double Beta Decay Isotopes in LS: search for neutrinoless double beta decay in Nd-LS

$$\bar{q} \equiv (C^{100} |A^{100}|^2) \times 10^{13} \quad ^{150}Nd$$

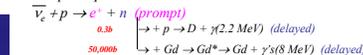
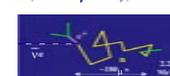
Isotope	\bar{q}
⁴⁸ Ca	0.54
⁷⁶ Ge	0.73
⁸² Se	1.70
¹⁰⁰ Mo	10.0
¹¹⁴ Cd	1.30
¹³⁶ Xe	4.20
¹³⁴ Xe	0.28
¹⁵⁰ Nd	57.0



- 3.37-MeV endpoint
- $(9.7 \pm 0.7 \pm 1.0) \times 10^{18} \text{ yr } 2\nu\beta\beta$ half-life measured by NEMO-III
- 1 kton organic liquid scintillator
- good energy resolution needed, but statistics could compensate for poor resolution
- Endpoint shape distortion measure instead of peak search
- For 0ν , 1000 events per year with 1% natural Nd-loaded liquid scintillator

Reactor θ_{13} Experiment: measure reactor ν in Gd-LS

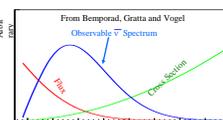
IBD (inverse β -decay) events in Gd-doped liquid scintillator:



Time- and energy-tagged signals to suppress background events.

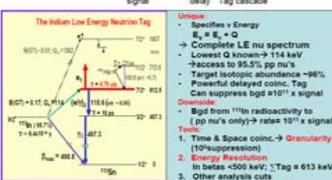
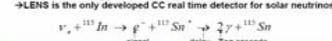
❖ Energy of $\bar{\nu}_e$ is given by:

$$E_{\bar{\nu}_e} \approx T_{e^+} + T_n + (m_n - m_p) + m_{e^+} \approx T_{e^+} + 1.8 \text{ MeV}$$



Low Energy Neutrino: detection of pp, Be, CNO and PP solar ν in In-LS. Luminosity of Sun

Tagged ν -capture reaction in Indium



Summary

- ❖ BNL neutrino group has capability and facility for developing organometallic liquid scintillator covering (1) synthesis, (2) characterization and (3) purification of M-LS
- ❖ Successfully developed organometallic Liquid Scintillator technologies for loading a variety of metal into liquid scintillator at different concentrations (0.1 - 10%)
- ❖ Capable of large-scale production (in tons) of M-LS ($A^{100} > 10m$) for different neutrino experiments (LENS 125-ton In-LS, Daya Bay 200-ton Gd-LS, and SNO+ 1000-ton Nd-LS).
- ❖ M-LS are stable for long period of time (>3.5 yrs for 10% In-LS; >2 yrs for 1% Nd-LS; and >2 yrs for 0.1% Gd-LS).
- ❖ Attenuation length of M-LS is improved from few meters to tenths meters using varying purification schemes
- ❖ The R&D of Li-loaded LS and Ca-loaded LS for future geo-neutrino and double-beta-decay experiments. Preliminary results are very promising by loading few % M in LS using crownether technology.